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SUPERCONDUCTOR TERAHERTZ METAMATERIALS

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A thesis submitted to the Nanyang Technological University in partial
fulfillment of the requirement for the degree of Doctor of Philosophy

2018

Statement of Originality

I hereby certify that the work embodied in this thesis is the result of original research done by me except where otherwise stated in this thesis. The thesis work has not been submitted for a degree or professional qualification to any other university or institution. I declare that this thesis is written by myself and is free of plagiarism and of sufficient grammatical clarity to be examined. I confirm that the investigations were conducted in accord with the ethics policies and integrity standards of Nanyang Technological University and that the research data are presented honestly and without prejudice.

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Authorship Attribution Statement

This thesis contains material from 3 paper(s) published in the following peer-reviewed journal(s) where I was the first author.

Chapter 3 is published as Y. K. Srivastava, M. Manjappa, L. Cong, W. Cao, I. Al-Naib, W. Zhang and R. Singh, "Ultrahigh-Q Fano resonances in terahertz metasurfaces: strong influence of metallic conductivity at extremely low asymmetry" *Advanced Optical Material*, 2016, 4, 457. DOI: 10.1002/adom.201500504.

The contributions of the co-authors are as follows:

- I together with Prof. Ranjan Singh conceived the idea and initial project direction.
- I prepared the manuscript drafts. The manuscript was revised by Dr. Longqing Cong, and Prof. Ibraheem Al-Naib.
- All the simulations and experimental plan was designed by me.
- Manukumara Manjappa performed theoretical modeling.
- Dr. Wei Cao and Prof. Weili Zhang fabricated the samples and performed terahertz characterization.
- Prof. Ranjan Singh supervised the whole work and edited the manuscripts drafts.

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The contributions of the co-authors are as follows:

- I conceived the idea and prepared the manuscript drafts. All microscopy, including sample preparation, was conducted by me in the Facility for Analysis, Characterization, Testing and Simulation.
- Manukumara Manjappa and Dr. Harish N. S. Krishnamurthy assisted me in sample characterization, and in revising manuscripts.
- Prof. Ranjan Singh supervised the whole work and edited the manuscripts drafts.

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- I conceived the idea and prepared the manuscript drafts.
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Abstract

Terahertz metamaterials showcase tremendous potential for plethora of applications ranging from biochemical sensing, high-speed wireless communication, and efficient modulators. Conventional terahertz metamaterials are made of noble metals. Incorporating superconductors in terahertz metamaterials potentially provide unprecedented opportunities to control resonance features which has huge implications in the real-world technologies. This thesis reports on new applications of superconducting terahertz metamaterials for ultra-low loss operation, ultrafast manipulation of electromagnetic radiation and novel quantum effects.

This work experimentally and numerically demonstrates that the quality factor of a low asymmetry Fano resonant asymmetric split ring metamaterial is extremely sensitive to the conductivity of the constituent materials. At low asymmetry regime even high conductivity noble metals fail to support Fano resonance excitation. The metamaterials designed using high- T_c superconductors provide access to the extremely low asymmetry regime and excites extremely sharp Fano resonances. Highest Q factor achieved with superconducting resonator (167) found to be two-fold in comparison to metallic resonators (84). There is a clear advantage of superconducting resonators sustaining high Q Fano resonances compared to conventional metals in designing low loss metamaterials at terahertz frequencies.

Further, all-optical *dual-channel* ultrafast switching of sharp Fano resonances excited in superconducting terahertz asymmetric split ring resonators have been shown. Upon irradiation with optical pump, the ultrasensitive Cooper pairs in cuprate superconductor undergo dual dissociation-relaxation dynamics within a single superconductivity restoration cycle and lead to dual switching windows at picoseconds timescale. The high sensitivity of

Cooper pairs to external perturbations combined with the strong field confinement in Fano resonators enables access to such unique dual switching features. Moreover, we discover that superconducting metamaterials of thickness 25 nm ($\lambda/40,000$, where λ is the Fano resonance wavelength) show well evolved Fano resonances while metallic samples of identical thickness do not show any resonance excitation. Ultrathin superconducting metamaterials provide solution for low threshold switching and inductance enhanced devices.

Moreover, a niche approach to realize Meissner effect at terahertz frequencies in quantum metamaterial system to achieve the quantum level switching between flux exclusion and flux penetration in a metal-superconductor hybrid quantum metamaterial system is developed. The observed dynamic switching behavior is attributed to the reconfiguration of the total inductance of the system due to magnetic flux exclusion and penetration. Most importantly, upon irradiation with the external optical pump, the quantum metamaterial undergoes ultrafast switching between the flux penetration and flux exclusion states at very low pump fluences and thereby lead to low-loss, ultrasensitive, frequency agile, switchable quantum photonics devices.

Overall, in this thesis we demonstrated ultra high- Q resonances in superconductor metamaterials, which could be actively tuned in the ultrafast timescale using extremely low threshold optical pump fluences. We further demonstrate the existence of the Meissner effect phenomenon at terahertz frequencies which lead to the realization of novel quantum metamaterials. Thus the superconductor metamaterials presented in this thesis manifests great prospects and pathways for new generation terahertz high-speed photonics and electronics. The field of terahertz quantum metamaterials could be further enriched by including Josephson junctions in the geometry of the metamaterials.

Chapter 1

Introduction

1.1 Introduction to superconductivity

The phenomenon of superconductivity was discovered by H. Kamerlingh Onnes, in 1911.¹ He noticed abrupt disappearance of electrical resistance of Mercury while cooling it below the boiling temperature of Helium. The accidental discovery of vanishing electrical resistance below a certain temperature was later termed as superconductivity and became an astonishment in the field of condensed matter physics. The temperature below which superconductivity exist is termed as critical phase transition temperature (T_c). A superconductor is referred as an ideal conductor with complete absence of the scattering below T_c and often have largest scattering above T_c .^{2,3} Before 1986, the superconductivity was found in several materials but most of them were metals or simple binary metallic compound with relatively very low T_c (usually $< 20\text{K}$) and termed as conventional or low-temperature superconductors. The field of superconductivity took an uprise turn in 1986 with the discovery of a new class of materials where superconductivity persists above liquid nitrogen temperatures.⁴⁻⁶ Such materials are called high- T_c or unconventional superconductors and generally have complex material structures. Mostly they include copper oxide (CuO_2) layers sandwiched between other materials. Some of the widely studied high T_c superconductors are $\text{YBa}_2\text{Cu}_3\text{O}_7$ ($T_c \sim 80\text{-}93\text{ K}$), $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ ($T_c \sim 105\text{ K}$), and $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8-\delta}$ ($T_c \sim 133\text{ K}$ at ambient pressure and 164 K at high pressure).^{5,7} MgB_2 is the metallic superconductor with highest known T_c of 39 K .⁸

The abrupt disappearance of electrical resistance below T_c is not the only unusual property of superconductors but they also expel externally applied weak magnetic field from the interior of the material except from a thin outer layer. The magnetic field strength decays exponentially at a length of London penetration depth (λ) due to screening by the lossless electrical current. This phenomenon is called the Meissner-Ochsenfeld effect.^{2,9} Hence, the basic properties which characterize superconductivity are the simultaneous existence of perfect electrical conductivity and perfect diamagnetization. However, on the application of high magnetic fields (greater than the critical field strength), flux lines penetrate in the interior of superconductors, and the material behaves like an ordinary conductor. The critical value of the magnetic field at which the magnetic flux lines penetrate inside a superconductor is called a critical magnetic field (H_c). Similarly, on the application of electrical currents, the permanent currents persist below a certain amount of current density called critical current density (J_c). Hence, superconductivity only exists below certain critical thresholds such as critical phase transition temperature (T_c), critical current density (J_c), and critical magnetic field (H_c).

The superconductors are loosely divided into two categories, Type-I and Type-II, depending on their behavior and response to an external magnetic field. Superconductors of the first kind referred as Type-I superconductors, expel the magnetic field up to a maximum critical value H_c , and assume normal conducting state at higher field values (as shown in Figure 1(a)). The value of the H_c depends strongly on the temperature, it is highest at 0 K and reaches zero at the transition temperature. The superconductor of the second kind, referred as Type-II superconductors show perfect diamagnetism below the lower critical magnetic field H_{c1} and turn to a normal conductor at a much higher magnetic field called upper critical magnetic field H_{c2} . In between H_{c1} and H_{c2} , magnetic fields partially penetrate into the

sample as schematically presented in Figure 1(b). The shielding currents flowing in the superconductor concentrate the magnetic field lines such that a system of flux lines is generated. The material system in between two critical fields is called “Shubnikov Phase” and system of flux lines is called Vortices or Abrikosov Vortices.^{2,10} These vortices are quantized in nature and generally arrange themselves in the form of a triangular lattice. The magnetic field penetrating each vortex contains a single quantum of flux and is given by $\phi_0 = \frac{h}{2e} \approx 2.07 \times 10^{-15} \text{ Tm}^2$, where h is the Planck’s constant and e is the electronic charge.² With increasing magnetic field, number of vortices entering sample increases and

Figure 1.1: Schematic illustration of the response of (a) type I and (b) type II superconductor to a static magnetic field. (c) The magnetic field in the interior of the superconductor as the function of the applied magnetic field in type I and type II superconductor.

finally at the upper critical magnetic field, entire material assumes normal state allowing full penetration of applied magnetic field.

There have been significant efforts to understand the extraordinary properties of superconductors and several theoretical models have been established which successfully explain conventional superconductors. Among the theoretical models, Ginzberg-Landau's intuitive macroscopic theory of superconductors and BCS (John Bardeen, Leon Cooper, and Robert Schrieffer) microscopic theory of superconductivity is found to be of most importance.^{11,12}

1.1.1 London equations

London equations were formulated in 1935 by London brothers, which govern microscopic electromagnetic field in and around a superconductor.¹³ This was a first meaningful mathematical description of superconductivity which was able to explain the Meissner effect phenomenon. The two London equations relate current density (J_s) to electric ($\vec{E}$) and magnetic ($\vec{B}$) component of the electromagnetic field individually and are given as

$$\frac{\partial J_s}{\partial t} = \frac{n_s e^2}{m} \vec{E} \quad \dots \dots \dots 1.1 (a)$$

$$\nabla \times J_s = -\frac{n_s e^2}{m} \vec{B} \quad \dots \dots \dots 1.1 (b)$$

where, e is the elementary charge of an electron, m is the mass of the electron, and n_s is the number density of the superconducting carriers. Equation 1.1 (b) could be manipulated using amperes law ($\nabla \times B = \mu_o J$) as

$$\nabla^2 \vec{B} = \frac{1}{\lambda^2} \vec{B} \quad , \quad \lambda = \sqrt{\frac{m}{n_s \mu_o e^2}} \quad \dots \dots \dots 1.2$$

where λ is the London penetration depth and depicts a characteristic length over which the magnetic field is suppressed exponentially in the superconductor. The value of λ varies with temperature as the superconducting carrier density n_s depends strongly on the temperature. Its value is lowest at 0 K and goes to infinite at T_c thus allowing penetration of the magnetic field in the superconductor at and above T_c . The value of penetration depth at a finite temperature T ($\lambda_L(T)$) in terms of penetration depth at 0 K ($\lambda_L(0)$) is given by

$$\lambda_L(T) = \frac{\lambda_L(0)}{\sqrt{1 - \frac{T^4}{T_c^4}}} \quad \dots \dots \dots 1.3$$

1.1.2 BCS theory

In 1957 Bardeen, Cooper, and Schrieffer presented a theory to explain the origin and basic understanding of superconductivity in conventional superconductors.¹¹ According to BCS theory, despite the repulsive nature of Coulomb forces between two electrons, the electron-phonon interaction may lead to an attractive force between them below T_c . The attractive force leads to the pairing of electrons (making a Cooper pair) of opposite momenta and spins. However, the transition temperature is dependent on Debye phonon energy, effective pairing potential and electron density of the state at Fermi level.^{2,14} The effective pairing potential consists of both attractive ($e-ph$ interaction) as well as repulsive (Coulomb) nature of electrons. The energy required to break a Cooper pair at 0 K is called superconducting gap energy and represented as 2Δ . The superconducting gap strongly depends on temperature and is predicted to increase from zero at $T = T_c$ to a limiting value $2\Delta(0)$ at $T = 0$ K. Temperature dependence of superconducting gap predicts that at $0 < T < T_c$, Cooper pairs and quasiparticles exist simultaneously. At $T = T_c$, the superconducting gap disappears, and Cooper pairs cease to exist. The binding energy of the Cooper pair is typically of the order

few meV and falls in terahertz part of electromagnetic spectrum, which makes terahertz spectroscopy an ideal tool to investigate features associated with the non-equilibrium dynamics of Cooper pairs.¹⁵⁻¹⁷

BCS theory provides adequate information about the conventional metallic superconductors but it lacks to account for the origin of high- T_c observed in unconventional superconductors. The crystal structure of high T_c superconductors is rather complex and generally have high anisotropy layered perovskite form. Unlike conventional superconductors, which have direction independent “s-wave” superconducting gap, unconventional superconductors have direction dependent “d-wave” superconducting gap with nodal regions in k-space.¹⁸ The BCS theory also does not explain the “d-wave” nature of the superconducting gap in high- T_c superconductors.

1.1.3 Two-fluid model of superconductivity

Once a superconductor is cooled below its T_c , the charge carriers consist of two parts, single electrons and paired electrons. The single electrons are called quasiparticles and are affected by the scattering mechanism. The population density of quasiparticles is represented by n_n and is responsible for the losses. The other part which consists of paired electrons (Cooper pairs) with population density n_s , are immune to the scattering losses and hence leads to a dissipation-less flow of the currents. Considering the effect of these two types of the charge carriers, Gorter and Casimir in 1934 proposed a two-fluid model of the conductivity of a superconductor. According to this model, the currents flowing in a superconductor has two available paths, one through the normal single electrons (n_n) and other through the superconducting electrons (n_s). The ratio of the charge carriers at a given temperature T is given by

$$\frac{n_s}{n} = 1 - \left(\frac{T}{T_c}\right)^4 \quad \dots \dots \dots 1.4$$

where n is total carrier density and is given by $n = n_n + n_s$. At temperature $T = T_c$, $n_s = 0$ and at 0 K, $n_s = n$. At DC and low frequencies, the conductivity resulted from the Cooper pairs (σ_s) is much larger than the conductivity due to the normal carriers (σ_n) and thereby leads to negligible resistance. However, σ_n play a significant role in the conducting properties of superconductor at higher frequencies.

The contribution due to both kinds of the carriers to the currents is additive and is given by

$$J_T = J_n + J_s \quad \dots \dots \dots 1.5$$

where J_T , J_n , and J_s denote the total current, normal current and superconducting current densities, respectively. The J_n is governed by Ohm's law whereas J_s is governed by London's equations (equation 1.1).¹³ The complex optical conductivity can be expressed in the form of current and electric field (E) as

$$J_T = J_n + J_s = \sigma E = (\sigma_n + \sigma_s)E \quad \dots \dots \dots 1.6$$

Drude model describes the contribution to the conductivity (σ_n) by normal carriers and is given as

$$\sigma_n = \frac{n_n e^2 \tau_n}{m^* (1 - i\omega \tau_n)} \quad \dots \dots \dots 1.7 (a)$$

and superconducting contribution to conductivity (σ_s) is given by

$$\sigma_s = \frac{in_s e^2}{m^* \omega} \quad \dots \dots \dots 1.7 (b)$$

Where τ_n and m^* are the relaxation time and effective mass of the normal particles, respectively. Equation 1.7 reveals that the real part of conductivity entirely originates from the normal carriers while the imaginary part accounts for the contribution from both kind of carriers. However, at low frequencies and low temperatures, the imaginary part of conductivity is dominated by superconducting carriers and give rise to most of the peculiar properties of the superconductors.

1.2 Introduction to YBCO

Yttrium barium copper oxide (YBCO) is one of the most famous high- T_c superconductors with transition temperature above liquid nitrogen temperature. Typically, YBCO shows T_c of 80-93K.¹⁹ The general formula for YBCO compound is $YBa_2Cu_3O_{7-\delta}$, which is also known as Y123. YBCO compounds show superconductivity only at a doping level $\delta \leq 0.655$ and behave as an insulator or antiferromagnetic material at higher δ values.²⁰ The critical temperature of YBCO also decreases with increasing δ . The schematic of the crystal structure of YBCO is shown in Figure 1.2 which depicts tetragonal lattice structure for $\delta = 1$ (non-superconducting state) and undergo phase transition to orthorhombic phase on decreasing doping level ($\delta = 0$, superconducting state). The phase transition from tetragonal to orthorhombic phase occurs due to the formation of copper-oxygen chains and is believed to be primarily responsible for the onset of superconductivity.²¹ The superconducting gap in YBCO shows d -wave symmetry which has nodal regions with zero superconducting gap. Another striking feature of the YBCO is the presence of a pseudo gap at temperature $> T_c$, which arises due to the presence of the reduced density of states in the vicinity of the Fermi level. The detailed phase diagram showing all the phases present in the YBCO is shown in Figure 1.3.¹⁶

Figure 1.2: The YBCO crystal structure in a) tetragonal phase ($\delta = 1$) and b) orthorhombic phase ($\delta = 0$).

Figure 1.3: Phase diagram of the YBCO superconductor.¹⁶

1.3 Applications of superconductors

Attempts to utilize the superconductivity in the real-world devices are on since the discovery of the phenomenon itself. The Kamerlingh-Onnes predicted that the persistent currents in superconductors would help to generate the magnetic field in highly efficient and economical ways. These days, the large volume, stable and high power superconducting magnets are the backbone of applications such as magnetic resonance imaging (MRI) machines, nuclear magnetic resonance (NMR) spectrometers, particles accelerators, mass spectrometers, low noise amplifiers and Large Hadron Collider (LHC).²²⁻²⁴ Despite the requirement of the cryogenic cooling, three important properties of superconductors namely low-Joule losses, nonlinear response to the oscillating currents below superconducting gap frequencies and Josephson effect, also found vital applications in the technological world. The rapid single flux quantum technology based digital circuits, RF, and microwave filters have enriched the low-loss electronics world. Superconductors have also shown potential for several possible applications such as superconducting maglev trains, highly sensitive magnetometers and magnetic levitation devices, smart grid, smart power storage devices, superconducting magnetic refrigerators and ultrafast electronics.^{23,25}

1.4 Cooper pair dynamics

Before discussing the Cooper pair dynamics, it is important to understand the charge carrier dynamics of metals on optical irradiation. On irradiation with an optical pulse, the incident photons are absorbed by free electrons of metal. The excited electron, often called hot electron, have much higher energy than other unexcited free electrons. The excited hot electron relaxes via interaction with neighboring electrons by an extremely fast electron-

electron ($e-e$) interaction process.^{26,27} Because of the $e-e$ interactions, electron and phonon subsystems have different energies and they try to reach a local equilibrium state via relaxation through electron-phonon ($e-ph$) interaction at a higher temperature. Finally, the excess energy is dissipated via a relatively slower process mediated by heat diffusion and phonon escape from the sample into the substrate and thus restore the whole system to the initial state.^{14,28}

The mechanism of the charge carrier relaxation of superconductors upon photoexcitation is different from hot electron relaxation mechanism in metals.²⁹⁻³³ The schematic diagram of the Cooper pair dynamics is systematically depicted in Figure 1.4. Initially, most of the charge carriers are in the form of Cooper pairs and occupy the single quantum ground state. The ground state level occupied by the Cooper pair is separated by the superconducting gap (2Δ) from the quasiparticle energy level as shown in Figure 1.4 (a). When a typical superconductor is irradiated with an optical pulse with energy higher than the binding energy of the Cooper pairs, a fraction of Cooper pairs get dissociated into highly energetic quasiparticles, as shown in Figure 1.4 (b). The excited quasiparticles interact with other Cooper pairs in the vicinity to lose their excess energy and this cascading process continues till the energy of the originally excited quasiparticles reaches nearly equal to the superconducting gap as shown in Figure 1.4 (c). The number of directly excited quasiparticles increases with increasing fluences of the incident pump, which reduces the number of available Cooper pairs for secondary dissociation via interaction with excited hot quasiparticles. When most of the Cooper pairs are dissociated into quasiparticles, the hot quasiparticles relax via the $e-ph$ mechanism. Both of the above processes of quasiparticle relaxation to the vicinity of quasiparticle energy level (superconducting gap level) occurs in the extremely short timescale of sub-picosecond to few picoseconds. When most of the

Figure 1.4: Schematic of the Cooper pair dynamics upon irradiation with an optical field of energy greater than the superconducting gap.

quasiparticles are relaxed to the vicinity of the superconducting gap level, they start recombining to form Cooper pairs by emitting 2Δ phonons, as shown in Figure 1.4 (d). The 2Δ phonons only exist in the superconductors and once emitted, have energy greater than the superconducting gap and thus results in further dissociation of Cooper pairs until they attain energy nearly equal to the superconducting gap. This effect is called phonon bottleneck effect. As a final step, these 2Δ phonons escape to the substrate and bring the whole system to equilibrium state (see Figure 1.4 (e)). The processes shown in Figure 1.4 (c), (d) and (e) are significantly slower and occurs on the time scale of 10-100 picoseconds. There have been huge efforts to study the charge carrier dynamics as it can unearth the underlying physics and Cooper pair formation phenomenon which always has been a trending topic of investigation

since the discovery of high- T_c superconductors. Figure 1.4 presents the general picture of the photoresponse of the a superconductor.

The information revealed by the charge carrier dynamics in superconductors is dependent on the several factors such as the Cooper pair binding energy, thickness of the superconductor, optical pump fluences. To study the Cooper pair dynamics, various characterization techniques could be employed such as terahertz-pump terahertz-probe (TPTP), optical-pump-terahertz-probe (OPTP), and optical-pump optical-probe (OPOP). A review of the various studies on charge carrier dynamics of the Cooper pairs in cuprate superconductors has been detailed by W. Li et. al.³¹

1.5 Kinetic Inductance

Kinetic inductance is essentially the inductance arises due to the inertial mass of the current carriers. Kinetic inductance is observed in high carrier mobility conductors and superconductors at very high frequencies. The kinetic inductance arises naturally in the Drude model of conductivity when the scattering rate τ becomes non zero. According to the Drude model, the conductivity of the metals is given by

$$\sigma = \frac{ne^2\tau}{m(1 + i\omega\tau)} = \frac{ne^2\tau}{m(1 + \omega^2\tau^2)} - i \frac{ne^2\omega\tau^2}{m(1 + \omega^2\tau^2)} \quad \dots 1.8$$

where m is the mass of the charge carrier, n is the number of charge carriers, τ is the collision time and ω is the angular frequency. In the normal metals, the collision time is $\sim 10^{-14}$ seconds which makes the term $\omega^2\tau^2$ insignificant at lower frequencies such as microwaves and terahertz. Therefore, the kinetic inductance in metals is significant only at higher

frequencies. However, in a typical superconductor where the collision time is very large ($\tau \sim \infty$), the kinetic inductance show significant values.

The kinetic inductance of a thin superconducting strip of the thickness t , length l and width w are given as $L_K = \frac{\mu_0 \lambda_L^2 l}{wt}$, where μ_0 is the vacuum permeability, λ_L is the London penetration depth, respectively.³⁴⁻³⁶ However, for the superconducting strips of thickness higher than the London ($t > \lambda_L$), the kinetic inductance is given by $L_K = \frac{\mu_0 \lambda_L l}{w}$.

1.6 Metamaterials

The response of an incident electromagnetic radiation in a material medium is described by the Maxwell's equation

$$\vec{\nabla} \cdot \vec{D} = \rho \quad \dots \dots \dots 1.9 (a)$$

$$\vec{\nabla} \cdot \vec{B} = 0 \quad \dots \dots \dots 1.9 (b)$$

$$\vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \quad \dots \dots \dots 1.9 (c)$$

$$\vec{\nabla} \times \vec{B} = \mu \vec{J} + \epsilon \mu \frac{\partial \vec{E}}{\partial t} \quad \dots \dots \dots 1.9 (d)$$

where $\vec{E}$ and $\vec{B}$ are the electric field intensity and magnetic flux density, respectively. The quantities ρ , $\vec{J}$, ϵ and μ are free charge density, current density, electric permittivity of the medium and magnetic permeability of the medium, respectively. The above Maxwell's

Figure 1.5: Characterization of material properties by electric permittivity (ϵ) and magnetic permeability (μ)

equations suggest that the sign of ϵ and μ plays a fundamental role in governing wave propagation behavior within a material medium. Based on ϵ and μ values of the medium, we can conveniently characterize electromagnetic materials in four classes as shown in the Figure 1.5. Most of the dielectrics, metals above plasma frequency falls in the region I of the ϵ - μ diagram where both $\epsilon > 0$ and $\mu > 0$ and consequently electromagnetic radiation can propagate in forward direction through these media. In region II and IV of the ϵ - μ diagram where $\epsilon < 0$, $\mu > 0$ and $\epsilon > 0$, $\mu < 0$, respectively, all the electromagnetic waves decay evanescently within the medium and therefore no propagating wave could sustain. The materials with both $\epsilon < 0$ and $\mu < 0$ belong to region III of the ϵ - μ diagram and does not exist naturally. However, this class of materials have been shown with artificial construct called “Metamaterials.”^{37,38} Since both effective medium parameters are negative, this class of

materials show unusual properties such as backward wave propagation, negative refraction, and reversal Snell's law.

Metamaterials are composites of specially designed sub-wavelength unit cells known as meta-atoms.^{38,39} Just as the properties of natural materials are essentially derived from the chemical constituents and bonds in the material, the electromagnetic properties of a metamaterial arise from the geometry of individual meta-atoms. These sub-wavelength scale, composite structured materials are designed to have specific structure dependent electromagnetic properties and could be engineered to show electric as well as magnetic resonance response at the specific predetermined frequency.^{39,40} Due to the subwavelength size of the meta-atom, the incident radiation only interacts with the effective polarization and/or magnetization developed in the composite medium instead of individual meta-atoms. Hence, at macroscopic level, the composite medium can be characterized by effective medium parameters such as electric permittivity $\epsilon(\omega)$ and magnetic permeability $\mu(\omega)$.

Historically, the idea of the artificial subwavelength structures and composites could be tracked down to J. C. Bose (Figure 1.6 (a) and (b)) but the field of modern electromagnetic metamaterials unleashed with the prediction of a hypothetical material with simultaneous negative permittivity and negative permeability by V. G. Veselago.^{37,41} However, the first theoretical and experimental demonstration of the simultaneous existence of negative permittivity and negative permeability was demonstrated by J. B. Pendry et al. and D. R. Smith et al., respectively, which ultimately lead to negative refractive index materials.⁴²⁻⁴⁵ Since then there has been tremendous growth in the field of metamaterials and plasmonic devices, especially in its design, understanding, and applications.^{39,46} Although the metamaterials are mostly associated with negative refraction^{42,47}, but this is only one manifestation of the possible intriguing functionalities, by carefully designing the meta-

Figure 1.6: Early examples and initial building blocks of the Metamaterials. a) and b) Linear polarizer consisting of a twisted jute polarizer and book and interleaved tin foils, respectively, discovered by J. C. Bose.⁴¹ c) an array of infinitely long wires of radius ' r ' and periodicity ' a ' acting as low frequency plasma. d) schematic of the split ring resonator proposed by Sir J. Pendry.⁴⁵ Array of infinitely long wires give rise to negative permittivity while split ring resonator results in negative permeability.

atoms, several exotic phenomena such as cloaking⁴⁸, artificial magnetism⁴⁹, sub-wavelength imaging⁵⁰, perfect absorption⁵¹, and perfect lens^{44,50} have been demonstrated across the electromagnetic spectrum which are impossible/ difficult to achieve with natural materials.

The field of metamaterials is getting matured to the point where its applications are pursued in the frequency regime ranging from zero to optical.

On-demand electromagnetic properties of metamaterials have also shown great promises for applications in various areas such as flat optics, flat lasing devices^{52,53}, sensing⁵⁴⁻⁵⁷, slow light⁵⁸⁻⁶⁰, and enhanced light-matter interaction. Most of these real-world applications demand for high quality (Q) factor resonances either for enhanced light matter interaction by high localization of the incident fields or for higher spectral resolution. The Q -factors of the subwavelength structures are limited by the inherent losses incurred due to the structural parameters and constituent materials. One of the grand challenge in the metamaterials is to minimize the losses. There are two kinds of losses in metamaterials namely radiative and non-radiative which limit sharpness of the resonances. The radiative losses arise due to the radiative nature of the subwavelength structure and could be readily engineered by altering geometrical parameters. Non-radiative losses arise due to the inherent ohmic losses of the constituent materials and could be engineered by changing the conducting properties of the constituent materials. Conventional metamaterials are made of noble metals and significantly curb non-radiative losses owing to the excellent conducting properties in the terahertz part of the frequency spectrum and therefore results in strongly resonant metamaterial structures.^{61,62} Metals support sharp resonances but lacks dynamic tunability as their electromagnetic properties are least influenced by external perturbations. However, dynamic tuning or resonance switching has been achieved by integrating phase change materials such as semiconductors, perovskites, liquid crystals, 2D layered materials into metamaterial structure.^{40,63-72} On application of external stimuli, the phase change materials modify the metamaterial embedded environment locally and thereby influence the overall optical response. The tunable nature of optical response could be achieved by mechanical tuning of

microelectromechanical systems/ nanoelectromechanical systems (MEMS/NEMS) based structures, but their fabrication requires complex fabrication techniques.⁷³⁻⁷⁵ The dynamic tunability could also originate from the materials composing the resonator structures itself, in contrast to the tuning metamaterial environment. The superconductors could be the ideal material to construct tunable artificial structures as their complex conductivity exhibit tunability over a wide range of values, through application of electric current, magnetic field, optical irradiation and temperature variation.⁷⁶⁻⁷⁸ Dynamically tunable, sharp resonance features are highly desirable for multifunctional applications such as lasing, ultrasensitive sensing, nonlinearity and slow light.

1.7 Superconductors in metamaterials

In this section, we review the development and application of superconductors in metamaterials. The superconductors are highly utilized in the field of metamaterials due to several unique properties such as zero DC electrical resistance, excellent tunable platform, inherent nonlinearity to the oscillating currents, perfect diamagnetism and quantum effects such as Josephson tunneling and flux quantization.^{76,77,79} However, it is to be noted that the superconductors show extraordinary properties only below a certain finite frequency determined by the superconducting energy gap.² As the frequency of operation extend beyond microwaves, the overall conductivity of a superconductor decreases. According to the two-fluid model, the real (σ_{re}) and imaginary (σ_{im}) part of the frequency dependent complex conductivity of superconductors is expressed as

$$\sigma_{re} = \frac{n_n e^2 \tau_n}{m^* (1 + \omega^2 \tau_n^2)} \text{ and } \sigma_{im} = \frac{n_s e^2}{m^* \omega} + \frac{n_n e^2 (\omega \tau_n)^2}{m^* \omega (1 + \omega^2 \tau_n^2)} \quad \dots \dots \dots 1.10$$

where n_n and n_s are the normal carrier and Cooper pair densities, respectively. Here τ_n is the relaxation time of the normal carriers, m^* is the effective mass of the normal carriers, e is electronic charge, and ω represents the frequency of the incident electromagnetic wave.^{2,80} Equation 1.9 suggests that the superconductors have finite ohmic resistance at non-zero frequencies and it increases with increasing frequencies thereby their application in metamaterials/ photonic devices is restricted to the devices operating at frequencies lower than the superconducting gap ($2\Delta/h$). Beyond this, the electrodynamics of the superconductor approaches to the poor metallic character and is dominated by normal charge carriers, whose abundance increases with increasing frequency and temperature. Typically, the superconducting gap in high- T_c superconductors is in the range of 2-75 meV, which falls in terahertz part of the electromagnetic spectrum.¹⁵⁻¹⁷ Superconductors are mainly used for the following applications in metamaterials/photonic devices

- i) Low loss metamaterials**
- ii) Actively tunable and switchable metamaterials**
- iii) Nonlinear metamaterials**
- iv) Perfect diamagnetic metamaterial**
- v) Quantum metamaterials**
- vi) Squid and Josephson junction metamaterial**

As the field of superconducting metamaterial advances, tremendous attempts have been made to utilize superconductors for low loss, nonlinearity, and dynamic switching applications. The conventional Drude metal component of metamaterials suffers from a large increase in the loss as their geometrical dimensions are reduced to obtain the desired effect for non-trivial applications. The extraordinary features of metal made metamaterials, such as negative refraction, the ability of a split-ring resonator to create artificial magnetism, and evanescent

wave amplification became highly sensitive to even small amount of losses.⁸¹⁻⁸³ This makes conventional metals unproductive for any application involving shrinking of the structural dimensions. Most of the loss reduction schemes employs superconductors, i.e. materials exhibiting zero DC electrical resistance to replace lossy metallic component of the metamaterial structures.⁷⁶⁻⁷⁸ However, the fruitful results of severe loss reduction are only achieved in the metamaterials operating at low frequencies. M. C. Ricci et al. has demonstrated low-loss metamaterials operating at GHz frequency regime using Nb based split ring resonators (SRR) and achieved extremely high quality (Q) factors of 4.5×10^4 below T_c , while no such resonance feature was observed for the identical metallic (Cu) based structures.⁸⁴ C. H. Zhang et al. has demonstrated 24 times enhancement in the unloaded Q factors of NbN based metamaterials in comparison to identical gold structures at terahertz frequencies.⁸⁵ After this, there have been several demonstrations of lossless features at radio and GHz frequencies, however clear distinction of lossless resonances at terahertz frequencies remains puzzling.^{78,86-88} Superconducting metamaterial structures are also proven to be an envoy for the Drude metals in the plasmonic limit and found to show very robust artificial magnetism. The superconductors also have small surface resistance in comparison to Drude metals up to microwave frequencies.⁸⁹ The low temperature superconductor Nb typically show the surface resistance of few nano-ohms whereas high- T_c superconductors show surface resistance in the micro-ohm range at microwave frequencies.^{89,90} The small surface resistance of superconductor allows overcoming scaling limitations in the metamaterials. Reducing the dimension below London penetration depth (λ) enhances the kinetic inductance and thus allowing further miniaturization.³⁴ The kinetic inductance is the inductance associated with the kinetic energy of the Cooper pairs. A detailed review on the loss-less features of superconductors can be found in reference^{78,78}.

The origination of intrinsic nonlinearity is due to the extremely sensitive nature of superconducting phase to the external stimuli, and that has led to the realization of actively tunable metamaterials, which is the most fascinating aspect of superconducting metamaterials. On the application of external stimuli such as temperature^{87,88,91-94}, magnetic field^{86,95,96}, electrical current⁹⁷⁻⁹⁹ and optical excitation¹⁰⁰, the optical response of metamaterials could be tuned and eventually switched on and off. Several groups have demonstrated methods to control the resonances in superconducting plasmonic hole arrays, LC resonators, and asymmetric split ring resonator, respectively, by means of temperature dependent properties of superconductors.^{87,88,92} M. C. Ricci et al., B. Jin et al. and D. Wang et al. have accomplished magnetic field dependent superconducting properties of Nb and NbN to demonstrate magnetic control of terahertz resonances.^{86,96,101} Actively controlled electromagnetically induced transparency (EIT) and slow light devices were discussed by various groups using superconductors.^{93,102} All-Optical ultrafast tuning of LC resonant metamaterial systems with switching speed of few picoseconds was also demonstrated.¹⁰⁰ The ultrafast dissociation and formation of Cooper pairs at such ultra-short timescale have been utilized for enhanced terahertz radiation generation and detection.¹⁰³ Along with the intrinsic nonlinearity upon interaction with external stimuli, giant nonlinearity has been achieved by V. Savinov et al. by introducing weak links in the geometry of the metamaterials.¹⁰⁴ The intentionally created weak links in the otherwise superconducting wire results in the resistive heating thereby give rise to strong nonlinearity.^{104,105}

The Josephson junctions (JJ) and superconducting quantum interference device (SQUID) has significantly enriched the field of superconducting metamaterials, especially at microwave and radio frequencies.^{106,107} A JJ is primarily a weak link sandwiched between two superconductors, which allows tunneling of Cooper pairs without any external voltage. On

Figure 1.7: Unconventional superconducting metamaterials a) a schematic of DC magnetic metamaterial¹⁰⁸ b) DC magnetic cloaking¹⁰⁹ and c) wood-cut superconducting quantum metamaterial for flux exclusion¹¹⁰.

adding a chain of SQUIDS formed by superconducting loops containing JJs, the inductance of the system could be reconfigured actively. Application of external magnetic field provides an opportunity to actively tune the resonance frequency as the inductance of the JJs is the periodic function of the magnetic field threading the superconducting loop. The quantum mechanical nature of the superconductors in which the Cooper pair density itself varies with

the applied magnetic field, current, and temperature, provides an opportunity to tune, not only the kinetic inductance but also the Josephson inductance. This approach has been utilized both theoretically¹¹¹ and experimentally¹¹² to provide wideband tunability and intrinsic nonlinearity by externally applied magnetic field (both DC and RF), current and temperature and thus resulting in an appealing alternative of frequency agile metamaterials.⁷⁹

The astonishing features brought about by incorporation of superconductors in the metamaterial structure are not only limited to above discussed realizations but also include several other types of artificial structures such as Zero frequency metamaterials^{108,113}, DC magnetic cloaking^{109,114}, and concentrations¹¹⁵. There have been attempts to realize quantum metamaterials to demonstrate flux exclusion in a resonator system at microwave (GHz) frequencies.¹¹⁰ However, the switching between flux exclusion and flux penetration regime could not be observed due to experimental limitations.¹¹⁰

1.8 Scope and overview of this thesis

Manipulation of light-matter interaction lies in the focal point of the modern science and technology of next generation photonics devices for the applications in high-speed communication, routing, and sensing. Various materials and their composites have been adopted over time to efficiently control electromagnetic radiation and lead to everything from electronics to communication systems. Evolution and advancement of metamaterials have provided an ideal platform for understanding of light-matter interaction and practical solutions such as cloaking, superlensing, sensing, and nonlinear electromagnetic response. In particular, terahertz (0.2-10 THz) metamaterials are of immense interest due to their applications in bio-chemical sensing, and efficient modulation of terahertz radiation. Terahertz radiation lies between microwave and infrared part of the frequency spectrum and

acts as a bridge between electronics (microwave) and photonics (infrared). One of the most promising applications of terahertz spectrum is in non-destructive, non-ionizing and fast imaging and sensing modalities. Terahertz metamaterials, whose electromagnetic responses are determined by the designer shape and size of the subwavelength inclusions, could provide many practical solutions for an array of industries. The subwavelength inclusions are conventionally made of metals that have certain losses. The inherent loss in metallic resonators results in a reasonably broad spectrum. However, several practical applications require high- Q factor resonances with extremely high electromagnetic field confinement at nanoscale for enhanced spectral resolution in spectroscopy and to enhance the sensitivity of the sensors. Superconductors with negligibly small losses have emerged as an excellent alternative for terahertz metamaterials. This thesis is concerned with exploring pathways to achieve new applications of superconducting metamaterials for fundamental studies as well as technological aspects of terahertz subwavelength structures and led to the demonstration of limitations imposed by metallic resonant structures in terms of maximum achievable Q factors, achieving ultra-high- Q resonant devices using high T_c superconductors, ultrafast terahertz modulators, dual-channel ultrafast switches, low-threshold broadband modulators, thinnest metamaterials and flux exclusion metamaterials. In this chapter, we have discussed fundamentals of superconductors, important equations describing superconductivity, high T_c superconductors, and an overview of electromagnetic metamaterials. We also discussed the potential application of superconductors in metamaterials and an overview of superconducting metamaterials.

Chapter 2 discusses the main techniques used in this thesis for design, fabrication, and characterization of metamaterial samples studied for the work presented in this thesis, which

include numerical simulations for designing metamaterial samples, pump-probe spectroscopy setup and optical cryostat employed for low temperature terahertz spectroscopy.

Chapter 3 describes the effect of metallic conductivity on the optical response of the Fano resonant metamaterials. At terahertz frequencies, the optical properties of metals can be described by various dispersion models such as PEC, Drude and DC conducting metals and all these models result in a similar resonant response of metamaterials. However, our study reveals that Fano resonances, in the low structural asymmetry regime, are highly sensitive to the conductivity of metal and they show significantly different resonance response in terms of line-widths, amplitude, and Q factors for all three dispersion models. We demonstrate that the DC conductivity model closely matches to the resonance response of the experimentally fabricated Fano resonant metamaterial devices at terahertz frequencies.

Chapter 4 explains the significance of using low loss high- T_c superconductors over metals to achieve high Q resonances at terahertz frequencies in different asymmetry regimes. This study demonstrates that superconductors can access extremely low asymmetry regime of the Fano resonators and support excitation of ultra-high Q Fano resonances at very low values of structural asymmetry regime. Such extremely low asymmetry regime is inaccessible by identical metallic Fano resonators. We also observe a significant line-width narrowing of Fano resonances in superconducting structures in comparison to metallic structures.

Chapter 5 describes the research on ultrafast superconducting metamaterial switch. This study reveals that upon photoexcitation with an optical pulse of energy higher than the binding energy of the Cooper pairs, superconductors show the existence of dual dissociation-relaxation channels within a single superconductivity restoration cycle. This peculiar

observation leads to the realization of superconducting dual-channel, ultrafast switchable devices operating in picoseconds timescale.

Chapter 6 describes the discovery of ultrathin terahertz metamaterials. This research reveals that ultrathin superconducting metamaterials support excitation of strong resonances while identical conventional metallic metamaterials fail to support excitation of resonance mode below their skin depth thicknesses. Furthermore, ultrathin superconducting metamaterials are characterized using optical pump-terahertz probe spectroscopy which demonstrates extremely low threshold switching of resonances at an ultrafast timescale.

In Chapter 7 we present preliminary results on finding of a new quantum metamaterials system which demonstrates the existence of Meissner–Ochsenfeld or Meissner effect at terahertz frequencies. We report ultrafast quantum level switching between flux exclusion and flux penetration regime in the optical response of the hybrid metal-superconductor coupled metamaterial system. The observed dynamic switching behavior is explained using the total inductance of the system.

Chapter 8 gives a summary of the achieved results during this thesis and presents future research directions in the superconducting metamaterials.

Chapter 2

Experimental Methods: Simulation, Fabrication and Characterization Techniques

2.1 Numerical simulations

This section gives a brief introduction to the commonly used numerical simulation principles to model metamaterial resonators at terahertz frequencies. Numerical modeling is a powerful tool for identifying new designer meta-atoms for novel applications and provide several important information. We use a commercially available software, Computer Simulation Technology Microwave Studio (CST-MWS) to numerically simulate the optical response of metamaterials structure. CST-MWS is a Maxwell equation solver that employs Finite Integral Techniques (FIT) or the Finite Difference Time Domain (FDTD) technique with appropriate boundary conditions. The Maxwell's equations in the integral form inside a material medium of charge density ρ and current density $\vec{J}$ is given by¹¹⁶

$$\oint_A \vec{D} \cdot d\vec{A} = \int_V \rho dV \quad \dots \dots \dots 2.1 (a)$$

$$\oint_A \vec{B} \cdot d\vec{A} = 0 \quad \dots \dots \dots 2.1 (b)$$

$$\oint_s \vec{E} \cdot d\vec{s} = - \int_A \frac{\partial \vec{B}}{\partial t} \cdot d\vec{A} \quad \dots \dots \dots 2.1 (c)$$

$$\oint_s \vec{H} \cdot d\vec{s} = \int_A \left(\frac{\partial \vec{D}}{\partial t} + \vec{J} \right) \cdot d\vec{A} \quad \dots \dots \dots 2.1 (d)$$

The CST-MWS provides two kinds of solvers namely “frequency domain solver” and “time-domain solver”, solvers that employed different approximations and boundary conditions to solve infinitesimal Maxwell’s grid but result in the identical output. CST-MWS is equipped with “Computer Aided Design” (CAD) to perfectly design the desired artificial structure. Most of the commonly used materials such as metals (gold, aluminum, silver, chromium, nickel, copper etc.), dielectrics (silicon, quartz, alumina), and polymers (polyamide) used for metamaterial design are available in the CST material library. However, CST-MWS allows a broad range of user defined materials by assigning material parameters such as frequency dependent dispersion, dielectric constant, Drude metal parameters, conductivity, thermal and mechanical parameters. A detailed flowchart of the steps involved in the design of an object and numerical simulation is given in Figure 2.1.

Figure 2.1: Flowchart showing steps involved in designing and solving EM wave equations for the metasurface using CST-MWS simulator.

Figure 2.2: Schematic of the asymmetric metamaterial array: Font and side view of the unit cell boundary conditions.

In our case, we used frequency domain solver for the numerical simulations. The boundary conditions which best mimic response of a metamaterial array in the frequency domain simulation are "Unit Cell", "open" and "open + add space" in the X-Y plane, $Z_{\min}$, and $Z_{\max}$ direction, respectively. This unit cell boundary conditions replicate an infinite array of designed structures considering all possible edge effects, while open boundary in "Z" direction ensures uniform excitation of the sample as it depicts the direction of propagation of the excitation signal. An extra space in the $Z_{\max}$ direction is essential to avoid the effects of the fringing field generated by metallic surface upon excitation signal. Figure 2.2 depicts a schematic of the unit cell boundary conditions applied on an asymmetric metamaterial array.

To solve the integral form of Maxwell's equations (equation 2.1), the unit cell is divided into small grids call "mesh". Frequency domain solver adopts tetrahedral geometry of the grids

and needs to be smaller than the smallest dimension in the design. CST-MSW allows automatic as well as manual meshing scheme. Figure 2.3 represents schematic of meshing of metamaterial structure. In the frequency domain simulation, integral form of Maxwell equations (equation 2.1) is Fourier transformed, resulting in the replacement of time derivative operator by its Fourier equivalent:

$$\frac{\partial}{\partial t} = i\omega \quad \dots \dots \dots 2.2$$

and resultant equations are solved separately for each frequency in the entire frequency domain using the interpolative method. Thus, the frequency domain solver is most efficient to simulate narrow resonance features. The computed complex transmission and reflection output components are stored in the scattering matrix “S Matrix” called “S-parameters” which relates outgoing wave to the incoming wave. The complete solution can be easily obtained from the matrix equation given by

$$\begin{bmatrix} O_1 \\ O_2 \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix} \begin{bmatrix} I_1 \\ I_2 \end{bmatrix} \quad \dots \dots \dots 2.3$$

where subscript 1 and 2 represents port 1 and 2, respectively. The coefficients of S Matrix are interpreted as following

S_{11} = reflection coefficient at port 1,

S_{12} = transmission coefficient at port 1,

S_{21} = reflection coefficient at port 2,

S_{22} = transmission coefficient at port 2.

Figure 2.3: Schematic of mesh adoption on an asymmetric metaatom.

The CST-MWS also give information about 2D and 3D distribution of surface current densities, electric field, magnetic field, and power at the region of interest. The electric field, magnetic field, and charge distribution help to explain electromagnetic response of the metamaterial structure and provide adequate information about the nature of the resonance mode.

2.2 Sample fabrication

The desired dimensions of the metamaterial structures which are designed to resonate in the terahertz part of the electromagnetic spectrum are on the micron scale, which is most

commonly fabricated by UV illumination-based photolithography or electron beam lithography technique. In this thesis, three kinds of metamaterial structures are designed and fabricated employing conventional UV-illumination based photolithography technique. These structures include conventional metallic metamaterials, superconducting metamaterials, and superconductor-metal hybrid metamaterial structures. The detailed fabrication process of which is elaborated below.

2.2.1 Metallic metamaterial samples

Normal planer metallic metamaterials usually consist of the periodic array of metallic structures deposited on a high resistivity silicon substrate. In our case, we used double side polished p-type silicon substrates of thickness 500 μm and resistivity higher than 5000 $\Omega\text{-cm}$. The detailed fabrication process is given below

- 1) Silicon wafers are cleaved in the desired size and thoroughly cleaned using isopropyl alcohol (IPA) and acetone solution. These cleaned substrates are then dried in the dry nitrogen flow to get rid of any remnant solvent or moisture.
- 2) A positive photoresist (AZ5214E) of thickness 1.5 μm is deposited on the silicon substrate using spin coating technique. The desired thickness (1.5 μm) of photoresist is achieved by spin coating it at 4000 rpm speed for 40 seconds.
- 3) The photoresist coated silicon substrates are pre-baked on a hotplate at 105°C for 1 minute to harden the photoresist.
- 4) The prebaked substrates are then exposed to the UV illumination after proper mask alignment using mask-aligner MJB4. For better resolution and clear sidewall profile, we use hard contact mode during the UV exposure.

Figure 2.4: Schematic illustration of the fabrication process flow. Figure (a)-(c) represents various steps involved in the photolithography, (d) represents the metal deposition and (e) depicts lift-off of the undesired metal.

5) Further, the UV illuminated substrates are soaked in the developer solution (AZ developer) for 30 seconds and then rinsed in the deionized (DI) water. The developer solution removes the part of the photoresist which is exposed to UV light, thus leaving behind the desired pattern.

6) The aluminum metal of thickness 200 nm is deposited using thermal evaporation technique on the patterned substrates. Finally, the substrates are soaked in acetone to liftoff undesired metal and the photoresist from the substrate and thus resulting in the desired metallic metamaterial structures.

This fabrication process flow is shown in Figure 2.4 and optical microscopy images of the fabricated samples are shown in Figure 2.5.

Figure 2.5: Optical microscopy (OM) images of fabricated metallic samples on a) silicon and b) sapphire substrate.

2.2.2 Superconducting metamaterial samples

Superconducting metamaterial samples are fabricated using the UV illumination-based photolithography and wet chemical etching technique. We used commercially available YBCO films deposited on *r*-cut sapphire substrates, purchased from Ceraco (Ceraco ceramic coating GmbH) Germany. The YBCO films of thickness 50, 100 and 200 nm are grown on a 500 μm thick *r*-cut sapphire substrate, whereas the 25 nm thick films are grown on 2 mm thick *r*-cut sapphire substrate. Cerium Oxide (CeO_2) of thickness 30 nm is in-situ deposited as a buffer layer between YBCO film and sapphire substrate to improve YBCO film growth quality and avoid reactions of YBCO with substrate material at high temperatures. The properties of the YBCO films used in this thesis are given in Table 2.1.

Table 2.1: Superconducting phase transition temperature (T_c) and critical current density (J_c) of the YBCO films.

YBCO thickness	Film Type	T_c (K)	J_c (MA/cm ²)
25 nm	M-Type	81.2	0.6
50 nm	M-Type	84.9	2.3
100 nm	M-Type	85.1	2.6
200 nm	M-Type	85.4	2.9

The YBCO films are thoroughly cleaned using acetone and IPA and dried with dry nitrogen flow to get rid of any contamination. A positive photoresist (AZ5214E) of thickness 1.5 μm is spin-coated over YBCO films. These photoresist coated films are prebaked at 105° C on a hotplate and exposed with UV illumination after proper mask alignment. The UV illuminated

substrates are then soaked in the developer solution (AZ developer) for pattern development. The patterned films are rinsed in DI water and dried with nitrogen gas flow. Remaining photoresist after pattern development acts as an etching protective layer. The undesired part of YBCO film, which is uncovered with photo-resist, is wet etched using 0.04% nitric acid solution. The etching time is optimized for different thicknesses of the YBCO films. The samples are finally rinsed in DI water and dried with nitrogen flow. The optical microscopy (OM) images of the few fabricated samples are shown in Figure 2.6.

Figure 2.6: Superconducting metamaterial samples fabricated by wet etching of 100 nm YBCO film on *r*-cut sapphire substrate.

2.2.3 Hybrid (superconductor-metal) metamaterial samples

The metal-superconductor hybrid sample is fabricated by the combination of the above two process. First, the YBCO film is patterned to get superconducting structures using the fabrication method described in section 2.2.2. The sample containing superconducting structures again lithographically patterned to form metallic structures using the fabrication method described in section 2.2.1. The separation between YBCO and metallic patterns is maintained by properly overlapping alignment marks in the predefined photomask. The optical microscopy image of the fabricated superconductor-metal hybrid sample is shown in Figure 2.7.

Figure 2.7: Metal-superconductor hybrid metamaterial sample consisting of 200 nm Al metal (ring) and 100 nm YBCO (disk) on *r*-cut sapphire substrate.

2.3 Terahertz (THz) Spectroscopy

2.3.1 Terahertz wave

Terahertz (THz) radiation commonly occupies the region between the microwaves and infrared frequencies of the electromagnetic spectrum which is also known as the “terahertz gap”. Thus, it is loosely defined as the frequencies extending from 0.1 -10 THz (Figure 2.8)

Figure 2.8: Electromagnetic spectrum depicting the terahertz band.(source: internet)

thereby bridging the gap between high-speed electronics and photonics. The corresponding wavelength and energies, period and wavenumbers are as follows,

Wavelength: $\lambda = c/f = 0.03\text{-}3 \text{ mm}$ (1 THz = 0.3 mm)

Period: $T = 1/f = 0.1\text{-}10 \text{ ps}$ (1 THz= 1 ps)

Photon energy: $h\nu = 0.41\text{-}41 \text{ meV}$ (1 THz = 4.1 meV)

Wavenumber: = $3.335\text{--}333.5\text{ cm}^{-1}$ ($1\text{ THz} = 33.35\text{ cm}^{-1}$)

The meV energy scale of terahertz wave makes them important for studying intraband transitions in semiconductors, lattice vibrations in solids, superconductors and vibrational motion of biological specimens. The terahertz possess several advantages such as non-ionization, non-destructive and spectral fingerprinting which makes it highly important for spectroscopy.^{117,118} Another greatest advantage of the coherent terahertz time domain spectroscopy is that they facilitate direct measurement of amplitude and phase information of a sample without application of Kramers-Kronig relations. Hence, it leads to direct measurement of the material parameters such as complex refractive index, complex permittivity, and complex conductivity. The spectroscopic and imaging techniques did not cover the terahertz frequencies until recently due to the lack of efficient terahertz generation, detection sources and optical components, but this gap is now rapidly explored after the breakthrough in inventing photoconductive antenna¹¹⁹⁻¹²² and nonlinear frequency mixing¹²³⁻¹²⁶ based terahertz generation sources.

2.3.2: Terahertz time domain spectroscopy

2.3.2.1 Photoconductive antenna based THz-TDS system

The photoconductive antenna is the widely used component for terahertz generation and detection.¹¹⁹⁻¹²² Figure 2.9 gives the pictorial representation of terahertz generation and detection by optical irradiation of a voltage biased photoconductive antenna. A femtosecond laser pulse of wavelength 800 nm is split into two parts using a beam splitter. First part is being used for terahertz generation and another for terahertz detection. The emitter of the photoconductive antenna consists of two metallic electrodes of width $w = 10\text{ }\mu\text{m}$, separated

Figure 2.9: Schematic diagram of terahertz generation and detection through GaAs photoconductive antenna based THz-TDS set up.

by a distance $L = 80 \mu\text{m}$ coated on a low-temperature GaAs substrate. To generate terahertz pulses, a DC voltage of 70 V is applied across the electrodes. When the GaAs substrate is irradiated with an ultrafast optical pulse of energy greater than its band gap, the photo-induced free charge carriers are generated. The photoinduced carriers generated in the gap between two metal electrodes are accelerated by a biased electric field (E_b) and give rise to a time varying photocurrent density ($J(t)$) as given by,

$$J(t) = N(t)e\mu E_b \quad \dots \dots \dots 2.4$$

where $N(t)$ denotes is the density of photo induced charges, e is the electronic charge and μ is the mobility of the electrons. Since the holes usually have much lower mobility than electrons, their contribution in photocurrent can be neglected. The shape of laser pulse and carrier lifetime determines the format of generated photocarrier density $N(t)$ and thereby also determine the photocurrent density. The electromagnetic (terahertz) pulse generated by $J(t)$ in the far field region as given by

$$E_{THz} = \frac{Ae}{4\pi\epsilon_0 c^2 z} \frac{\partial N(t)}{\partial t} \mu E_b \quad \dots \dots \dots 2.5$$

where term A represents the illuminated area between metallic electrodes, z is the distance between terahertz source and far-field probe, ϵ_0 is the permittivity of vacuum, and c is the speed of light. From equation (2.5), it is clear that the energy of the terahertz pulse is derived from the electric energy stored across the gap instead of the energy of the optical excitation pulse. The femtosecond optical pulse triggers the release of energy stored between the gaps in the form of terahertz wave. The polarization of the generated terahertz wave is along the biased field and thus could be switched by changing the polarization of the biasing field.

The photoconductive antenna-based detectors work on a similar principle as the terahertz emitter, where generated terahertz field acts as a bias voltage instead of an externally applied voltage. A current source is connected across the two metallic electrodes, which are deposited on a low-temperature gallium arsenide (LT-GaAs) or ion implanted silicon on sapphire (SoS) substrate. As shown in Figure 2.9, the time delay between optical pump pulse and generated terahertz pulse is controlled by a delay stage. The electric field at any point in time is mapped by sampling photocarriers generated due to optical pumping between the two electrodes of the detector. The photocurrent ($\bar{J}$) induced by terahertz field is given by

$$\bar{J} = \bar{N}e\mu E(\tau) \quad \dots \dots \dots 2.6$$

where $\bar{N}$ is the photocarrier density, τ is the temporal delay between the optical probe and terahertz pulse and $E(\tau)$ is the temporally mapped terahertz electric field. By recording $E(\tau)$ as a function of τ , terahertz pulse waveform can be obtained. Mostly, a current amplifier is used to amplify the photocurrent of the order of nanoampere to the measurable amount. An optical chopper synchronized with a lock-in amplifier is used to improve the signal-to-noise ratio of the recorded data. Direct mapping of $E(\tau)$, instead of the intensity of the terahertz

waves allows direct measurement of the amplitude and phase information and thus facilitate in deriving complex optical constants of the sample. The recorded time-domain and corresponding Fourier transformed frequency-domain spectra of the terahertz pulse is shown in Figure 2.10.

Figure 2.10: (a) Measured time-domain terahertz transmission signal in the dry N_2 environment using photoconductive antenna-based THz-TDS setup. (b) Corresponding frequency-domain terahertz transmission signal obtained using Fast Fourier transformed (FFT) of time domain signal.

2.3.2.2 Non-linear crystal based THz-TDS system

Non-linear crystal based terahertz generation and detection has also acquired great interest due to the unique advantage of longer bandwidths and high peak powers of the terahertz pulse in spite of its inferior signal to noise ratio. Here, a non-linear crystal such as ZnTe or LiNbO₃ is pumped by an intense laser pulse of high energy with low repetition rate and emit terahertz waves due to optical rectification process. Optical rectification, which is a second-order nonlinear optical phenomenon, is a difference-frequency generation with negligible frequency difference. A single optical pulse is sufficient to stimulate optical rectification from an electro-optic (EO) crystal, thereby making the entire process extremely simple and efficient. Once an optical pulse is incident on a nonlinear non-centrosymmetric crystal, it generates transient polarization, and which emits terahertz pulses. Schematic of a typical ZnTe crystal based terahertz spectroscopy setup is shown in Figure 2.11.

Figure 2.11: Schematic of ZnTe crystal based THz-TDS setup.

The Maxwell's equation in a material medium of Polarization P , Magnetization M is given by

$$\nabla \times E = -\mu_0 \frac{\partial H}{\partial t} - \mu_0 \frac{\partial M}{\partial t} \quad \dots \dots \dots 2.7 (a)$$

$$\nabla \times H = \varepsilon_0 \frac{\partial E}{\partial t} + \frac{\partial P}{\partial t} + J \quad \dots \dots \dots 2.7 (b)$$

$$\nabla \cdot H = -\nabla \cdot M \quad \dots \dots \dots 2.7 (c)$$

$$\nabla \cdot E = -\frac{1}{\varepsilon_0} \nabla \cdot P + \frac{1}{\varepsilon_0} \rho \quad \dots \dots \dots 2.7 (d)$$

where J and ρ are current and charge densities, respectively. In a dielectric medium,

$$\nabla \cdot H = 0 \quad \dots \dots \dots 2.8 (a)$$

$$\nabla \cdot E = 0 \quad \dots \dots \dots 2.8 (b)$$

Using equation 2.7 and 2.8, the inhomogeneous wave equation for induced terahertz light can be derived as

$$\nabla \times \nabla \times E_{THz} = -\mu_0 \nabla \times \frac{\partial H}{\partial t} \quad \dots \dots \dots 2.9 (a)$$

$$\frac{\partial}{\partial t} (\nabla \times H) = \varepsilon_0 \frac{\partial^2 E_{THz}}{\partial t^2} + \frac{\partial^2 P^{NL}}{\partial t^2} \quad \dots \dots \dots 2.9 (b)$$

$$\nabla \cdot (\nabla \cdot E_{THz}) - \nabla^2 E_{THz} = -\frac{1}{c^2} \frac{\partial^2 E_{THz}}{\partial t^2} - \mu_0 \frac{\partial^2 P^{NL}}{\partial t^2} \quad \dots \dots \dots 2.10$$

The equation 2.10 transform to an inhomogeneous wave equation on using equation 2.7

$$\nabla^2 E_{THz} - \frac{1}{c^2} \frac{\partial^2 E_{THz}}{\partial t^2} = \mu_0 \frac{\partial^2 P^{NL}}{\partial t^2} \quad \dots \dots \dots 2.11$$

The induced polarization could be expressed as Taylor's expansion of the form

$$P(t) = \chi^1 E(t) + \chi^2 E(t)^2 \quad \dots \dots 2.12$$

where χ^1 and χ^2 represent the first and second order electric susceptibility tensor. On substituting $E(t) = A(t)\cos(\omega_0 t)$ in equation 2.12, polarizability can be expressed as

$$P(t) = \chi^1 A(t) \cos(\omega_0 t) + \chi^2 \left(\frac{A(t)^2}{2} + \frac{A(t)^2 \cos(2\omega_0 t)}{2} \right) \quad \dots \dots 2.13$$

The coefficient of χ^2 leads to transient polarization which is responsible for the terahertz generation for an optical pulse of pulse width 50-100 fs.

The terahertz pulses are detected using electro-optic sampling (EOS) technique which is based on the influence of the terahertz electric field on the optical properties of a second birefringent nonlinear ZnTe crystal via Pockel's effect. The strong electric field could induce anisotropy in the ZnTe crystal which in turn changes the polarization of the low intense optical beam passing through the crystal. A ZnTe crystal is cubic in nature hence its optical properties are isotropic in the absence of electric field. The terahertz wave is polarized in the x'-y' plane and oriented at an angle 45° to the x' axis and modifies the indices of refraction as

$$n_{x'} = n_0 + \frac{1}{2} n_0^3 r E_{THz} \quad \dots \dots 2.14 (a)$$

$$n_{y'} = n_0 - \frac{1}{2} n_0^3 r E_{THz} \quad \dots \dots 2.14 (b)$$

$$n_{z'} = n_0 \quad \dots \dots 2.14 (c)$$

where n_0 is the refractive index without electric field incidence, r is the electro-optic constant and E_{THz} represents the electric field of the terahertz radiation. The change in $n_{x'}$ and $n_{y'}$ are equal in magnitude and opposite in sign. More interestingly, the change in magnitude is

proportional to the E_{THz} and thus the optical response leads to direct measurement of the electric field passing through the crystal.

The femtosecond optical beam of wavelength 800 nm is synchronized to terahertz beam before passing through the analyzer crystal and thus will experience a change in polarization due to Pockel's effect. The change in polarization is detected by transmitting probe beam through a quarter-wave plate followed by polarizing prism which splits the beam into two orthogonal components that are measured using individual photodiodes. The signal from the photodiodes is electronically subtracted and referenced to a signal in the absence of the terahertz electric field, thus allowing direct measurement of terahertz field.

In addition, a branch of laser pulse can also be used to optically pump the sample and thus helps to realize robust optical-pump terahertz-probe spectrometer. The optical beam of higher intensity is typically used for optically pumping the sample to excite it to the non-equilibrium state and the corresponding effects are measured using terahertz probe. The time delay between the arrival of pump and probe beam is controlled by a delay stage thus allows for the terahertz beam to probe the response at various stages of non-equilibrium excitation. By having terahertz pulse in fixed small steps before, during and after the optical excitation pulse, the transmission of terahertz under the influence of optical excitation could be determined as a function of time. In our course of study, we employ the OPTP setup to probe the excitation and relaxation dynamics of Cooper pairs in YBCO superconductors.

2.4 Optical Cryostat

Most of the metamaterial structures studied in this thesis are made of YBCO superconductor and thus needed to be cooled to cryogenic temperatures for characterization. A continuous

helium flow open cycle cryostat is incorporated in the terahertz setup to cool down the samples down to 4 K. The cooling is achieved by transferring liquid helium from supply dewar to the cold stage of the refrigerator using a transfer line. A transfer line is a vacuum-jacketed, flexible hose made up of stainless steel and provide low heat loss transfer of the liquid helium. On externally pressurizing the supply dewar using helium gas, transfer line establishes the controlled continuous flow of liquid helium and maintain a constant temperature at the cold stage. The flow of liquid helium is controlled by externally applied pressure in the supply dewar and a flow meter connected to the cryostat. This cryostat can maintain any temperature between 4 K to 400 K in the presence of sufficient liquid helium flow. We also employed a closed cycle optical cryostat, as shown in Figure 2.12 to cool down some of the superconducting samples. In this system, the cooling is achieved by cooling down the helium vapors which cools the metallic cold finger. In this process, the refrigerator of the cryostat is kept in a vacuum in order to eliminate convection heat transfer

Figure 2.12: (a) Optical cryostat and (b) terahertz time domain spectroscopy setup used for cryogenic measurements¹²⁷

to the cold internal parts to maintain low temperatures. A radiation shield is used to minimize the radiant heat load on the cold stage and is cooled by helium exhausting from the heat exchanger. A temperature controller and thermocouple are used to control and monitor the temperature of the sample holder, respectively.

Chapter 3

Conductivity Engineered Fano Resonance:

Ultrahigh- Q Resonances in Terahertz Metasurfaces

This chapter deals with the conductivity engineered Fano resonances and limitations of the metallic Fano resonant metamaterials in terms of maximum achievable Q -factors and figure of merit.

3.1 Introduction

The recent progress in the field of metamaterials^{37,43,45,128} has given a major impetus to the manipulation of the terahertz waves which thus led to the realization of various terahertz photonic devices for applications such as bio-sensing, fingerprint chemical sensing, switches, high-speed modulators, non-linear devices for wireless communication systems and many more^{53,63,65,68,71,75,129}. Most of the principle applications of terahertz photonic devices demands for high quality (Q) factor resonance response either for enhanced light-matter interaction and localization of incident radiation for high sensitivity sensors or enhanced spectral resolution in spectroscopy. The Q factor of resonance response in a metamaterial device is limited by the inherent losses due to constituent materials and geometrical parameters. The loss mechanisms in subwavelength metamaterials are classified broadly into two categories namely radiative loss and non-radiative (ohmic) loss. The radiative loss arises due to the radiative nature of the resonator structures while non-radiative loss arises mainly due to the finite resistance of the materials constituting resonators. Hence, engineering of the structural geometry and constituent materials of the meta-atom for curbing losses has become

an important tool of the metamaterial design to achieve ultra-high- Q resonances for the potential applications in real-world devices.^{93,94,130-132} Total quality factor (Q_T) of metamaterial resonators in terms of radiative and non-radiative losses is given by

$$\frac{1}{Q_T} = \frac{1}{Q_R} + \frac{1}{Q_O} \quad \dots \dots \dots 3.1$$

where Q_R and Q_O represent the quality factors due to the radiative and non-radiative component, respectively.¹³³ Therefore, it is essential to probe the interplay between the radiative and Ohmic losses in the resonator system to achieve maximum possible Q factors.

Most of the metamaterial structures are made up of the metallic constructs to support strong resonances due to high conductivity. However, significantly high losses in metals have restricted the possibility of efficient devices in optical, and infrared part of the electromagnetic spectrum. Metals at terahertz frequencies are conventionally treated as near perfect electric conductors (PEC) and therefore give rise to low ohmic losses. However, we systematically show in this chapter that the assumption of metal as PEC becomes highly inaccurate for the case of high- Q asymmetric Fano resonant metamaterial structures. We observed that the high- Q Fano resonances are highly sensitive to the conducting properties of the resonators. High conductivity at terahertz frequencies helps to minimize ohmic losses, but the Q factors are still limited by the radiative losses.^{61,62,133,134} There is three key hypothesis to describe the dispersion properties of metals which are widely used in metamaterials in the terahertz frequency domain: PEC, Drude model and DC conductivity of the metal.^{62,134,135} The details of each hypothesis are as follows,

3.1.1 Perfect electrical conductor (PEC)

Often metals are treated as perfect conductor (PEC) in the terahertz part of the electromagnetic regime. PEC is an idealized material with zero electrical resistance or equivalently infinite electrical conductivity. However, PEC does not exist in the realism, but its hypothesis is useful in the analysis of the systems which possess negligible electrical resistances. The electrical currents flowing through PEC does not experience any decay and dissipate no energy. The electrical resistance is responsible for the generation of heat, and thus PEC does not experience any heating effect. Here it should be stressed that the PEC is different from superconductor as superconductors not only possess zero DC electrical resistance but also show zero magnetic field in the core of the material. In the case of PEC, magnetic field remains constant and can attain non-zero values also. While performing electromagnetic simulations, structures modeled as PEC could be simulated faster as the components of the equations consisting of finite conductivity could be neglected.

3.1.2 Drude model of conductivity

The conducting properties of the metals could be very well described by the Drude model that describes the frequency dependent conductivity and dispersion of a metal. According to the Drude model, the dielectric function of a metal can be written as

$$\varepsilon(\omega) = \varepsilon_{\infty} - \frac{\omega_p^2}{\omega^2 + i\omega\omega_{\tau}} \quad \dots \dots \dots 3.2$$

The real (ε_r) and imaginary (ε_i) parts of the dielectric function could be separated as

$$\varepsilon_r = \varepsilon_{\infty} - \frac{\omega_p^2}{\omega^2 + \omega_{\tau}^2} \quad \dots \dots \dots 3.3 (a)$$

$$\varepsilon_i = \frac{\omega_p^2 \omega_\tau}{\omega(\omega^2 + \omega_\tau^2)} \quad \dots \dots \dots 3.3 (b)$$

where ε_∞ is the high frequency dielectric constant, ω_p is the angular plasma frequency, and ω_τ is the angular damping frequency of the charge carriers . The angular plasma frequency in terms of the carrier density is given as $\omega_p = \sqrt{\frac{ne^2}{m\varepsilon_0}}$. The complex conductivity could be calculated using equation 3.3 as

$$\varepsilon(\omega) = \varepsilon_r + i\varepsilon_i = \varepsilon_\infty + \frac{i\sigma(\omega)}{\varepsilon_0\omega} \quad \dots \dots \dots 3.4$$

where, $\sigma(\omega)$ is the complex conductivity. The complex conductivity and its real ($\sigma_r(\omega)$) and imaginary part ($\sigma_i(\omega)$) could be expressed as

$$\sigma(\omega) = \frac{\sigma_{DC}}{(1 + i\omega\tau)} \quad \dots \dots \dots 3.5(a)$$

$$\sigma_r(\omega) = \varepsilon_0\omega\varepsilon_i(\omega) = \frac{\sigma_{DC}}{1+\omega^2\tau^2} \quad \dots \dots \dots 3.5(b)$$

$$\sigma_i(\omega) = \varepsilon_0\omega(1 - \varepsilon_r(\omega)) = \frac{\sigma_{DC}\omega\tau}{1 + \omega^2\tau^2} \quad \dots \dots \dots 3.5(c)$$

where
$$\sigma_{DC} = \frac{ne^2\tau}{m}$$

For a Drude metal, high frequency dielectric constant ε_∞ is considered as 13.7. The quantities ω_p and ω_τ are calculated from the measured dielectric constants from various experiments. We calculated Drude parameters for Aluminum metal obtained by fitting the experimental data given in reference 134.¹³⁶ The obtained values of ω_p and ω_τ are 22657.17 and 123.15 THz, respectively. The real and imaginary parts of the conductivity calculated using equation 3.4 is shown in Figure 3.1.

Figure 3.1: Calculated real and imaginary part of the conductivity of Aluminum metal using Drude model.

3.1.3 DC conductivity

The DC conductivity model presents frequency independent conductivity of a material. In the low frequency regime where $\omega\tau \ll 1$, equation 3.5 reduces to

$$\sigma(\omega) = \sigma_{dc}$$

The DC conductivity of the Al metal is taken as 3.56×10^7 S/m at room temperature.

Using assumption $\omega\tau \ll 1$ and $\epsilon_\infty \sim 0$ in equation 3.4 and 3.5, the permittivity can be expressed as

$$\epsilon \approx -\frac{\sigma_{dc}}{\Gamma\epsilon_0} + \frac{i\sigma_{dc}}{\omega\epsilon_0} \quad \dots \dots \dots 3.6$$

Hence, in the DC conductivity assumption, the conductivity is frequency independent in the terahertz frequency range, but the imaginary part of the permittivity is still an implicit function of frequency.

3.1.4 Metallic Fano resonant metamaterials

In most of the previous reports, metallic subwavelength resonators are either constructed using their DC conductivity or as PEC at terahertz frequencies. Although both assumptions result in the similar resonance response and agree closely with the measurements, but here we discovered that the resonance response of Fano resonant terahertz asymmetric split-ring resonator (TASR)^{130,137-142} is significantly different for all the three chosen conductivity models. This difference is observed in terms of line width and amplitude of the resonance response and is more prominent at low structural asymmetry in the Fano resonator which results in the sharp resonances due to the low radiative loss. The Fano resonance broadens at larger asymmetry due to the enhanced radiative loss, and in this realm, the response of the TASRs for the three assumed conductivity models is almost similar. Our terahertz transmission measurements on identical fabricated samples reveal that the experimental Fano resonance linewidth and amplitude is in good agreement with the DC conductivity model. Here, the Fano resonance is excited due to the destructive interference between a broad continuum (bright mode) and a narrow discrete mode (dark mode) that gives rise to a high- Q asymmetric line shape resonance.^{131,132,139,142} One of the most important characteristic of the Fano resonance is the remarkable confinement of electromagnetic energy in the subwavelength structures. However, on increasing the structural asymmetry, the sub radiative nature of confined electromagnetic energy gradually becomes radiative, thereby leads to the lowering of the Q factor of the resonances. At low structural asymmetry in the structure, the radiative losses are suppressed and become comparable to the ohmic losses which thus leads

to significantly different behavior of the Fano resonance for PEC, Drude metal, and the DC conductivity models of metals.

3.2 Design of the metamaterial structure

Here we probe the strong influence of conductivity on the Fano resonant metasurfaces (TASR) with different structural asymmetries at terahertz frequencies. We carried out numerical simulations for each metamaterial design with metallic conductivity governed by various conductivity models, i.e. PEC (PEC-TASR), Drude conductivity (Drude-TASR), and DC conductivity (DC-TASR) using finite integral technique based frequency domain solver of commercially available software CST microwave studio. Figure 3.2 depicts the schematic diagram and optical microscopy (OM) image of a double gap asymmetric split-ring resonator (TASR) structure. The terahertz field impinges at normal incidence on the metamaterial array. The TASR structure consists of 200 nm thick metallic arms structured over 500 μm thick silicon substrate. We chose aluminum (Al) metal for the design and experiment due to its high conductivity at terahertz frequencies and the stability of the samples. The asymmetry is introduced in the symmetric structure by moving one of the split gap away from the central vertical axis by a distance ' d ' as shown in Figure 3.2 (a). For this study, the asymmetry is varied from $d = 0.3 \mu\text{m}$ to $10 \mu\text{m}$.

We also defined the asymmetry parameter as the ratio of the difference between the two-arm length to the total length of both arms of the resonator and is given as

$$\alpha = \frac{l_1 - l_2}{l_1 + l_2} \times 100\% \quad \dots \dots \dots 3.7$$

where l_1 and l_2 are the length of two metallic arms that forms the TASR structure.¹⁴³ The

Figure 3.2: Schematic diagram of the asymmetric split-ring resonator array: a) All the geometrical dimensions are depicted in the unit cell. “ d ” is the asymmetry parameter, which is the displacement of one of the split gaps away from the central vertical axis. b) Optical microscopy (OM) image of the small portion of the fabricated sample.

Fano resonance is excited by incident terahertz field polarized (E_x) perpendicular to the gap side arm of the resonator. The Fano resonance excitation is forbidden in the perfectly symmetric structure and thus it is called a trapped mode resonance.¹³² Introduction of the structural asymmetry leads to the excitation of the extremely sharp narrow linewidth asymmetric line shape Fano resonance.

The numerical simulations are performed using CST microwave simulation software as discussed in section 2.1. A high resistivity silicon with dielectric constant of 11.7 is used as a terahertz transparent substrate. The DC conductivity of Aluminum is taken to be 3.56×10^7 S/m to design DC-TASR. Similarly, the Drude model is used to calculate the frequency

dependent conductivity to design and perform numerical simulations of Drude-TASR. The plasma frequency $\omega_p/2\pi = 3606$ THz and the damping frequency $\Gamma/2\pi = 19.6$ THz, respectively, is used to calculate the conductivity of Aluminum metal and is shown in Figure 3.1.¹³⁶

3.3 Discussion

3.3.1 Perfect electrical conductor based metasurfaces

Figure 3.3 (a)-(f) depicts the simulated amplitude transmission spectra of the PEC-TASR with asymmetry $d = 0.3, 0.5, 1, 3, 5$ and $10 \mu\text{m}$, respectively. Although, we also carried out simulations for the structural asymmetries below $d = 0.3 \mu\text{m}$ but we were unable to resolve the resonance mode as the observed transmission amplitude of the Fano resonance is very low (<0.01 or $<1\%$). Hence, in this work we restrict our studies to the minimum asymmetry d of $0.3 \mu\text{m}$. These transmission curves are recorded when the electric field of the incident THz wave is polarized perpendicular to the gap side arm of the TASR. Here, it should be noted that these TASR structures are made up of PEC, and thus the loss mechanism is only radiative in nature. The highest Q factor of the Fano resonance in PEC-TASR is 328 for the structure with asymmetry $d = 0.3 \mu\text{m}$. Here, it is essential to note that on increasing structural asymmetry, the Q factor of the Fano resonance decreases gradually.^{130,137,138} On increasing structural asymmetry d , the linewidth of the Fano resonance broadens, whereas the transmission amplitude increases gradually. The transmission amplitude of Fano resonance is calculated as the difference between peak to dip of the transmission at resonance in the transmission spectra. The Q factors are obtained by fitting the transmitted intensity spectrum using standard Fano equation given as

$$T_{Fano} = \left| \left(a_1 + ja_2 + \frac{b}{\omega - \omega_o + j\gamma} \right) \right|^2 \quad \dots \dots \dots 3.8$$

where coefficient a_1 , a_2 and b are constant real numbers, ω_o is the Fano resonance frequency, and γ is the damping rate of the resonance.^{144,145} By fitting T_{Fano} Vs ω using equation 3.8, we obtained the values of ω_o and γ while a_1 , a_2 and b are kept free parameters. Finally, the Q factor is estimated as

$$Q = \frac{\omega_o}{2\gamma} \quad \dots \dots \dots 3.9$$

Figure 3.3: (a)-(f) Fano resonance spectra of the PEC-TASR at different structural asymmetry $d = 0.3, 0.5, 1, 3, 5$, and $10 \mu\text{m}$, respectively.

3.3.2 Comparison of PEC, Drude and DC conductivity model

We further probed the influence of conductivity on the Fano resonance profiles by comparing resonance response of the TASR for the three different conductivity models at varying structural asymmetry parameter d . Figure 3.4 (a)-(c) shows the resonance response of the PEC-TASR, Drude-TASR, and DC-TASR, respectively, at extremely low structural asymmetry of $d = 1 \text{ } \mu\text{m}$. PEC-TASR excites extremely sharp Fano resonance with a significant amplitude of transmission, while no Fano resonance was observed in the case of Drude-TASR and DC-TASRs. The Q factor of the Fano resonance for PEC-TASR at $d = 1 \text{ } \mu\text{m}$ is estimated to be about 280. The absence of Fano resonance for the case of Drude-TASR and DC-TASR reflects that the excitation of Fano resonance mode itself requires a critical minimum conductivity at a given specific structural asymmetry. The asymmetric metamaterials structures with lower structural asymmetry require a higher critical conductivity value to give rise to the Fano resonance mode. It is due to the fact that at lower structural asymmetry, the non-radiative and radiative losses are comparable and hence even a subtle difference in the conductivity give rise to a large change in the surface currents flowing in two unequal arms of the TASR structure, which thus results in large difference in the Q factors and amplitudes of Fano resonances in the different cases of conductivity models. Similarly, Figure 3.4 (e)-(g) depicts the resonance response of the PEC-TASR, Drude-TASR, and DC-TASR for an asymmetry of $d = 3 \text{ } \mu\text{m}$. We observed largest resonance amplitude for the PEC-TASR, followed by Drude-TASR and DC-TASR. The Q factors ($Q = 46$) of the Fano resonances are equal for Drude-TASR and DC-TASR, whereas, for PEC-TASR, it is more than five times ($Q = 245$) greater than other two cases. Figure 3.4 (i)-(k) shows the transmission spectra for the TASR structures with structural asymmetry $d = 5 \text{ } \mu\text{m}$, where the Q factor of the Fano resonance for PEC case is $Q = 131$, for Drude case is $Q = 29$,

and for DC case $Q = 26$. We observed that the Q factor for the Drude-TASR and DC-TASR are nearly the same.

Figure 3.4: Comparison of amplitude transmission spectra of PEC, Drude, DC and experimental TASRs. Figure 3(a)-(d) depicts the transmission spectra for TASRs with asymmetry $d = 1 \mu\text{m}$, (e)-(h) for asymmetry $d = 3 \mu\text{m}$ and (i)-(l) for asymmetry $d = 5 \mu\text{m}$. PEC governs the metallic conductivity in TASR's in Figure 3(a), (e) and (i), Drude model for (b), (f) and (j), and DC conductivity for (c), (g) and (k). Figure 3(d), (h) and (l) represents experimentally measured transmission spectra.

3.3.3 Experimental Fano resonances in metallic structures

We carried out transmission measurements on the experimentally fabricated sample of structural dimensions based on the simulated design. These samples were fabricated on a 500 μm thick high resistivity ($> 5000 \Omega\text{-cm}$) double side polished p -type silicon wafers using standard UV illumination-based photolithography technique. The aluminum metal of thickness 200 nm was thermally evaporated over patterned substrates and desired metallic structures was achieved by removing undesired metal by lift-off techniques. A 10 mm thick silicon substrate of identical properties was optically attached at the rear side of the sample to eliminate the Fabry-Perot reflections from the back surface of the substrate and thus enabling longer scan up to 200 ps. A longer scan of the terahertz pulse is required to capture high Q resonances as the spectral resolution of the transmission spectra is inversely proportional to the scan length in timescale. The terahertz time domain scan was performed through the fabricated sample and an identical silicon substrate and later in the post-processing steps the transmission spectra is obtained by estimating $|\tilde{T}(\omega)| = |\tilde{E}_s(\omega)/\tilde{E}_R(\omega)|$, where $\tilde{E}_s(\omega)$ and $\tilde{E}_R(\omega)$ are the Fourier transformed electric field of the sample and reference substrate pulses, respectively. Figure 3.4 (d), (h) and (l) depicts the measured transmission spectra of the samples with asymmetry $d = 1, 3$ and $5 \mu\text{m}$, respectively. The measured transmission spectra have a good agreement with the corresponding DC-TASR spectra. The Fano resonance appears at slightly higher frequency in experimental spectra in comparison to the simulated spectra, which could be due to the small difference between the fabricated and simulated TASR structures.

3.3.3 Fano resonance profiles at low and high asymmetry

We further compare the Fano resonance response of the PEC-TASR, Drude-TASR and DC-TASR in the low and high structural asymmetry regimes in terms of the amplitude and resonance linewidth. Figure 3.5 (a) and (b) depicts the simulated transmission spectra at the symmetry of $d = 3 \text{ }\mu\text{m}$ and $d = 10 \text{ }\mu\text{m}$, respectively, for all three conductivity models. The amplitude of the Fano resonance in the transmission spectra is highest for PEC-TASR and lowest for DC-TASR. At low structural asymmetry $d = 3 \text{ }\mu\text{m}$, the Q factors of the Fano resonances for DC-TASR and Drude-TASR are the identical ($Q = 46$), while it is more than 5 times larger ($Q = 245$) for PEC-TASR. However, at larger structural asymmetry of $d = 10 \text{ }\mu\text{m}$, the difference between amplitude and linewidth of the resonance for three conductivity cases is not as drastic (see Figure 3.5 (b)) as for lower asymmetry values. The estimated Q factor for the PEC, Drude and DC-TASRs drop down to 29, 8 and 10, respectively. Hence, the Fano resonance show contrasting behavior in the low and high structural asymmetry regimes for the three conductivity models.

3.4 Electric-field distribution in the metamaterials for different materials

To further highlight the difference between the Fano resonance profiles of the TASRs of PEC, Drude and DC conductivities of metal, we simulated the electric field distribution in the meta-atom at the Fano resonance for all the three different conductivities at asymmetry $d = 3 \text{ }\mu\text{m}$ as shown in Figure 3.5 (c), (d) and (e). We observe large electric field confinement in the capacitive gaps of the PEC-TASRs, followed by the Drude and the DC-TASRs. The electric field confinement in Drude and DC TASRs are much weaker compared to the PEC-TASRs.

The tight field confinement in subwavelength structures is extremely important for its various multifunctional applications such as ultrasensitive sensors and efficient cavities.

Figure 3.5: Comparison of Fano resonance profiles at low and high asymmetry. (a) and (b) depicts the transmission spectra for asymmetry $d = 3$ and $d = 10 \mu\text{m}$, respectively. Red, black and blue curves corresponds to PEC, DC and Drude metals, respectively. The green curve represents the experimentally measured transmission spectrum. (c)-(e) shows the simulated electric field distribution at the dip of Fano resonance for PEC, DC, and Drude-TASR, respectively, at an asymmetry of $d = 3 \mu\text{m}$.

3.5 Two coupled oscillator model

The coupling refers to the transfer of energy between two or more physical system. Here the transfer of energy takes place between the resonators and coupling mechanisms between the resonators could be modeled as equivalent circuit modeling^{146,147}, Lagrangian formalism¹⁴⁸, coupled mode theory¹⁴⁹ or by using the coupled oscillator model¹⁵⁰. Here, we theoretically model the observed Fano resonance features using the two coupled oscillator model. The 'bright mode' that directly couples to the incident electromagnetic radiation interferes with the 'dark mode' which is decoupled from the incident waves. The equations governing two-oscillator model are as following,

$$\ddot{x}_b(t) + \gamma_b \dot{x}_b(t) + \omega_b^2 x_b(t) + \Omega^2 x_d(t) = \frac{QE_0 e^{i\omega t}}{M} \quad \dots \dots \dots \quad 3.10$$

$$\ddot{x}_d(t) + \gamma_d \dot{x}_d(t) + \omega_d^2 x_d(t) + \Omega^2 x_b(t) = 0 \quad \dots \dots \dots \quad 3.11$$

where x_b and x_d represent the displacement amplitude for bright and dark mode respectively. Q and M are the charge and mass associated with the bright mode and E_0 is the electric amplitude of the incoming EM radiation. The solution of equation 3.10 and 3.11 for x_b and x_d , leads to the complex susceptibility expression given by,

$$\chi = K \left(\frac{(\omega^2 - \omega_d^2) + i\omega\gamma_d}{\Omega^4 - (\omega^2 - \omega_b^2 + i\omega\gamma_b)(\omega^2 - \omega_d^2 + i\omega\gamma_d)} \right) \quad \dots \dots \dots \quad 3.12$$

The real part of the susceptibility describes the dispersion behavior of the coupled system while the imaginary part defines the dissipation of the system. The parameters ω_b and ω_d are the angular resonance frequencies of the bright and dark modes, respectively, whereas the term γ_b and γ_d represents the linewidth of the bright and dark mode, respectively. K is the

Figure 3.6: (a) and (b) The simulated amplitude transmission and corresponding analytical fit using coupled oscillator model for PEC-TASR (red), DC-TASR (black) and Drude-TASR (blue) with asymmetry $d = 3$, respectively.

proportionality constant which depends on the Q and M . The term Ω represents the coupling between bright and dark mode and depends on the separation between both modes and the asymmetry parameter (α). For the values of proportionality constant $K = 3.6 \times 10^{24}$, $\omega_d = \omega_T$, and $\omega_b = 3.8 \times 10^{12}$ rad/sec, the Fano resonance curves obtained for PEC-TASR ($\Omega = 0.45 \times 10^{12}$ rad/sec, $\gamma_d = 1 \times 10^8$ rad/sec), DC-TASR ($\Omega = 0.57 \times 10^{12}$ rad/sec, $\gamma_d = 3 \times 10^{10}$ rad/sec) and Drude-TASR ($\Omega = 0.52 \times 10^{12}$ rad/sec, $\gamma_d = 8 \times 10^9$ rad/sec) are shown in the Figure 3.6(b) for an asymmetry of $d = 3 \mu\text{m}$, which shows a good agreement with the numerically simulated results shown in Figure 3.6(a). We observe that the $\gamma_{b,d}$ and Ω depends inversely on the conductivity value. We also note that the Ω depends directly on the α while the $\gamma_{b,d}$ is independent of α . Figure 3.6(b) also represents that the Q factor and the amplitude of the Fano-resonance depend directly on the conductivity parameter. For instance, PEC-TASR

shows higher Q factor and larger amplitude compared with the DC-TASR for the same asymmetry parameter (α).

3.6 Q Factor and figure of merit

From the discussion in the section 3.3.4, it is clear that the behavior of the PEC-TASRs is significantly different from DC and Drude-TASRs, especially at the lower structural symmetry. We have investigated this behavior for all three different conductivity models in terms of the Q -factor and figure of merit (FoM) at varying asymmetry. Figure 3.7(a) depicts the Q factors of PEC, Drude, DC and experimental TASRs for Al metal. We observe extremely high Q factors for PEC-TASR at low asymmetries, which reduces drastically with increasing asymmetry in the structure and show saturation at about $d = 7 \text{ }\mu\text{m}$. The Q factors of the Fano resonances for DC-TASR and Drude-TASR are comparable and show good agreement with experimentally measured results. At lower structural asymmetry, measured Q factors are slightly higher because the Fano resonance is not very accurately measured due to the limited spectral resolution in the measurements which could result in a different linewidth of the Fano resonance.

Figure 3.7(b) depicts the FoM, which is defined as the product of the Q factor and the transmission intensity (ΔI) of the resonance peak.¹⁴³ At the asymmetries below $6 \text{ }\mu\text{m}$, the FoM of the PEC-TASR is one order of magnitude larger than the Drude and DC-TASR. The FoM values for Drude-TASR are slightly higher than the DC-TASR case due to the slightly higher intensity of the Fano resonances in Drude-TASR. This is due to the fact that DC model is a approximation of Drude model in the low frequency range and higher scattering rate. Higher scattering rate in DC metals leads to lower resonance amplitudes in comparison to the Drude metals. The FoM values peak at structural asymmetry of $d = 4 \text{ }\mu\text{m}$, $6 \text{ }\mu\text{m}$ and 4

μm for PEC-TASR, DC-TASR and Drude-TASR, respectively. The peak values of FoM for PEC, DC, and Drude-TASR's are 169.43, 5.80 and 9.10, respectively. The FoM for experimentally fabricated samples are in close agreement with the DC-TASR case. Thus, in

Figure 3.7: Q -Factor and FoM comparison for simulated spectra for three different conductivity models and measured spectra, at varying asymmetry: (a) and (b) shows Q factors and FoM of Fano resonance, respectively, estimated from simulated and experimental transmission spectra with increasing asymmetry d in the structure.

the low structural asymmetry regime (below $6\text{ }\mu\text{m}$), the conductivity plays an important role in determining the Fano resonance linewidth and amplitude due to the low radiative losses. However, at higher structural asymmetries (above $6\text{ }\mu\text{m}$), the resonance response is mainly determined by the radiative nature of the losses.

3.7 Conclusion

In this chapter, we discussed three scenarios of metallic conductivities in which we have PEC, frequency dependent conductivity described by Drude model, and DC conductivity of the metal. The DC conductivity case is the approximation of Drude conductivity model in the low-frequency regime. The experimental result closely matches with DC conductivity case. We observe that the conductivity of the constituent materials plays a critical role in determining the Q factor and the FoM of the Fano resonances at low structural asymmetry in the terahertz asymmetric metamaterials. High conductivity materials are highly sought to excite sharp resonances at low structural asymmetry. At larger structural asymmetries of Fano resonators, the Fano resonance does not depend strongly on the conductivity parameter as the radiative losses become dominant in comparison to the ohmic losses. Overall, our results manifest that the material conductivity becomes an important parameter for designing ultra high- Q metamaterials for sensing, lasing and non-linear devices at terahertz frequencies.

Chapter 4

Accessing the High- Q Resonances with High- T_c

Superconductor Metamaterial

This chapter deals with the superiority of high- T_c superconductors in accessing high- Q resonances by curbing non-radiative losses in the low asymmetry regime of Fano resonant asymmetric split ring resonators at terahertz frequencies. We also present a comparative study of the role of superconductors in Fano and Lorentzian (LC) resonant metamaterials in achieving maximum Q -factor and figure of merit and show that the superconducting structures provide larger Q factor and figure of merit than the identical metallic structures.

4.1 Introduction

As discussed in chapter 3, one of the major challenges in the field of plasmonics and metamaterials has been to engineer the radiative and non-radiative (ohmic) losses which limits the performance of the subwavelength resonators in terms of the achievable quality (Q) factors. The Q factor governs the light matter interaction in a subwavelength metamaterial system. The non-radiative losses arises because of the resistive nature of the constituent materials of the resonator, whereas radiative losses result from the radiative nature of energy stored in the sub-wavelength structures. Therefore, there have been constant attempts to curb the inherent losses either through geometry engineering^{130,132,138,151-156}, use of low-loss materials^{78,86-88,93,94,157} or by integrating resonator structure with the gain materials.

In chapter 3 of this thesis, we discovered that even the highly conducting Drude metals fails to excite the sharp Fano resonances¹⁴² at lower asymmetries. Here, the objective is to excite high- Q Fano resonances using high- T_c superconductors in a simple terahertz asymmetric split ring resonator (TASR) identical to the metamaterial structure shown in Figure 3.2. In classical systems, asymmetric line shape Fano resonance typically evolves due to the coupling between a broad continuum and a narrow discrete mode.^{131,137-139,142} Here, the broad dipolar mode arising in a perfectly symmetric structure interferes with the narrow discrete mode due to the tiny break in the structural symmetry. The linewidth and amplitude of the Fano resonance mode are determined by the incurred losses, where the ohmic loss is contributed by the materials used to fabricate the resonator structure. The electromagnetic energy stored in the subwavelength resonator structure give rise to the radiative losses. The intensive design engineering could reduce the radiative component of the losses. However, the Q factors of the metamaterial structure are still limited by the significant radiative losses.^{61,133} In chapter 3, we systematically probed the strong influence of material conductivity at the evolution of Fano resonance in low structural asymmetry TASR metamaterials. We observed that the metallic structures fail to excite sharp Fano resonances at extremely low asymmetry realm of the TASR in spite of high conductivity of Drude metals at terahertz frequencies. The critical value of asymmetry in the TASR metamaterial at which Fano resonance mode could be excited is governed by the conductivity of the constituent materials. However, it is demonstrated previously that the Q factors of the Fano resonance are governed by the structural asymmetry and decreases exponentially with increasing structural asymmetry.¹⁴³ Hence, it is extremely important to access the low asymmetry realm of the Fano resonators to achieve high- Q resonances and this could be achieved by designing the resonator structures with a material which behave like a perfect

electrical conductor (PEC), so that the Ohmic losses tends to zero. Therefore, here we chose a high T_c superconductor as a realistic material to probe that whether such a dissipation less material supports evolution of sharp Fano resonances at extremely low asymmetry realm which is inaccessible by the identical metallic structures at terahertz frequencies.

In this chapter, we used high- T_c superconductor yttrium barium copper oxide (YBCO) to access the low asymmetry regime of the Fano resonators to excite extremely sharp and dark resonances, which is inaccessible with the metallic conductivity. In a superconductor below its T_c , the intrinsic loss is minimum due to the presence of Cooper pairs. Thus, by using superconducting properties of the YBCO material below their T_c of 86 K, sharp resonances could be excited at extremely low values of asymmetry. However, above T_c , YBCO behaves as a poor metal and does not support excitation of Fano resonance mode.

4.2 Metamaterial design and simulation

We choose a metamaterial structure identical to the design used in chapter 3 and shown in Figure 4.1, which depicts schematic of the two-dimensional metamaterial array of the subwavelength asymmetric split ring resonator. The asymmetry in the structure is introduced by displacing one of the split gaps away from the central vertical axis by a distance ' d ', as shown in Figure 4.1. Here, the asymmetry is varied from $d = 1$ to $15 \mu\text{m}$. The numerical simulations are performed using software CST Microwave Studio. The detailed simulation method is given in section 2.1. Sapphire with permittivity of 11.7 is considered as terahertz transparent substrate. The frequency dependent complex permittivity of YBCO, as shown in Figure 4.2, obtained using terahertz transmission and phase measurements is used to define the dispersion behavior of superconductor at terahertz frequencies. The complex permittivity

Figure 4.1: Schematic of the asymmetric split ring resonator array. Asymmetry ' d ' is the displacement of the upper split gap from the central vertical axis. All the geometrical dimensions are revealed in the unit cell.

values of YBCO are measured at 27 K, which is much below their T_c of 87 K.¹⁵⁸ Identical metallic TASR structures are designed using aluminum metal with the DC conductivity of 3.56×10^7 S/m. The thickness of the Al and YBCO is considered identical to be 200 nm. In the numerical simulations, 'unit cell' boundary conditions are used in x-y plane to achieve an infinite metamaterial array, while open boundary conditions are used in the z-direction. The linearly polarized terahertz wave propagating in z- direction is illuminated on the sample at normal incidence. Such TASR structures result in the Fano resonance mode by exciting it with the incoming terahertz beam polarized perpendicular to the gap bearing arm (E_y) of the resonator.

Figure 4.2: Real and imaginary part of the complex permittivity of YBCO measured using terahertz time domain spectroscopy at 27 K.

4. 3 Accessing low asymmetry regime of Fano resonator

Numerically simulated amplitude transmission spectra of the metallic TASR (Al-TASR) and superconducting (YBCO-TASR) metamaterial at an extremely low asymmetry of $d = 1.5 \mu\text{m}$ are shown in Figure 4.3. The asymmetry could also be expressed in the % as $\alpha = \frac{l_1 - l_2}{l_1 + l_2} \times 100$, where l_1 and l_2 are the length of the two arms of the TASR. An asymmetry of $d = 1.5 \mu\text{m}$ corresponds to extremely low value of $\alpha = 1.3\%$. From Figure 4.3, we observe that the YBCO-TASR excites well evolve Fano resonance at 0.51 THz, whereas Al-TASR does not show any Fano resonance feature. The excitation of Fano resonance in YBCO-TASR could be understood in terms of suppressed ohmic losses at extremely low asymmetries. YBCO below its T_c has lower non-radiative (Ohmic) losses in comparison to the metals. The Fano

Figure 4.3: Simulated transmission spectra of Al-TASR and YBCO-TASR with asymmetry

$d = 1.5 \mu\text{m}$.

resonance dips of YBCO-TASR appears at lower frequencies in comparison to the Al-TASR due to the kinetic inductance of the superconductors. However, we intentionally blue shifted the resonance of YBCO-TASR by 46 GHz for a better comparison between the superconductor and metallic TASR.

We comprehensively compared the simulated transmittance spectra of the YBCO-TASR and Al-TASR at different asymmetries to clearly demonstrate the importance of superconductors over the conventional Drude metals. Figure 4.4 depicts that the Fano resonances in the YBCO-TASR can be observed at an asymmetry as low as $d = 1 \mu\text{m}$ ($\alpha = 0.85\%$). On decreasing the asymmetry in the TASR metamaterial, the Fano resonance line-width decreases gradually. From Figure 4.4, we also observe that the Al-TASR excites Fano resonances at only a slightly larger structural asymmetry of $d = 2 \mu\text{m}$ and above. Figure 4.4 (a) and (b) depicts that the linewidth of Fano resonances of YBCO-TASR are narrower in

comparison to Al-TASR. The narrow linewidths of resonances in superconducting metamaterials are due to the lower resistive losses in YBCO in comparison to Al metal. Thus, we clearly demonstrate that the YBCO-TASR excites narrow linewidth Fano resonance at extremely low asymmetries, while metallic TASRs fail to excite any such Fano resonance mode. It is observed that the transmission amplitude of Fano resonance increases gradually with increasing asymmetry which is typical of such asymmetric split ring resonators.

Figure 4.4: Comparison of simulated transmittance spectra of YBCO-TASR (a) and Al-TASR (b) with asymmetry $d = 1, 1.5, 2$ and $3 \mu\text{m}$ respectively.

4.4 Comparison of superconducting and metallic Fano resonance evolution

We further compared the response of YBCO-TASR and Al-TASR at various asymmetries to estimate the critical asymmetry for both materials at which the Fano resonance begins to evolve. From Figure 4.5, we observe that for the case of Al metal with DC conductivity of

Figure 4.5: Transmission spectra of YBCO-TASR and Al-TASR at different asymmetries. (a) and (b) represent the amplitude transmission spectra for YBCO-TASR and Al-TASR for extremely low asymmetry $d = 1.5$ and $1.8 \mu\text{m}$, respectively. (c) and (d) depicts the transmission spectra for YBCO-TASR and Al-TASR for higher asymmetry $d = 5$ and $10 \mu\text{m}$, respectively.

3.56×10^7 S/m, the minimum asymmetry required to excite the Fano resonance is $\sim 1.8 \mu\text{m}$ ($\alpha = 1.5\%$). We call this value of asymmetry as critical asymmetry. Figure 4.5(a)-(b) and 4.5(c)-(d) depicts the contrasting behavior of the Fano resonance feature in the low and high asymmetry regimes. We observed that the response of the TASR for YBCO and Al materials is different mainly in terms of the line-width of the Fano resonance mode even at higher asymmetries, where the radiative losses are much higher. At higher asymmetry, the linewidth of the Fano resonance is slightly narrower for YBCO-TASR in comparison to the Al-TASR. The Q factors of the asymmetric lineshape Fano resonances are estimated as $Q = \frac{\omega_o}{2\gamma}$, where ω_o is the central angular resonance frequency and γ is the damping rate of the resonance. The values of the ω_o and γ is obtained by fitting the transmitted intensity spectrum using standard Fano equation 3.8. The highest Q factors observed for YBCO-TASR and Al-TASR are 167 and 84, respectively as shown in Figure 4.5(a) and 4.5(b). This suggests that the YBCO superconductor supports extremely sharp Fano resonances. The YBCO-TASR results in almost two-fold higher Q factors in comparison to the metallic counterparts.

To validate the simulated results experimentally, we carried out experimental measurement on the samples with geometrical dimensions based on the simulated designs. The samples are fabricated using conventional UV illumination based photolithography and wet etching technique on a commercially available 200 nm thick YBCO film deposited on a 500 μm thick r-cut sapphire substrate. The YBCO film has a T_c of 86.1K and J_c of 2.9 MA/cm². Metallic samples of identical geometry are also fabricated using photolithography. The metallic patterns are achieved by depositing 200 nm thick aluminum metal using thermal evaporation technique. The detailed fabrication technique is given in section 2.2. The experimentally fabricated Al-TASR and YBCO-TASR samples are shown in Figure 4.6 (a) and (b), respectively. The transmission spectra of the YBCO-TASR at a temperature of 27 K

Figure 4.6: Comparison of simulated and experimental Fano resonances. a) and b) represents the optical image of the fabricated Al-TASR and YBCO-TASR, respectively. (c) and (d) depicts the simulated and measured transmission spectra for Al-TASR and YBCO-TASR for an asymmetry of $d = 15 \mu\text{m}$, respectively. Δf is the difference in the linewidth of the superconducting and metallic sample.

being measured using ZnTe based confocal terahertz spectroscopy system.¹²³ The system is incorporated with a continuous flow liquid helium cryostat. The terahertz beam of spot size $\sim 3 \text{ mm}$ is illuminated on the YBCO-TASR array of size $10 \text{ mm} \times 10 \text{ mm}$. The terahertz scans are performed on the sample and the reference substrate. The transmission amplitude is obtained using the relation $|T(\omega)| = |E_S(\omega)|/|E_R(\omega)|$, where $E_S(\omega)$ and $E_R(\omega)$ are the Fourier

transform of the transmitted electric field through the sample and reference substrate, respectively. The YBCO-TASR sample is measured at temperature 27K which is much below the superconducting phase transition temperature of the YBCO. The Al-TASR sample is measured at 295 K as the conductivity of metallic thin films does not change much on cooling down to cryogenic temperature and thus they give rise to identical resonance response on varying the temperature from room temperature to cryogenic temperatures.^{135,157} Numerically simulated and experimentally measured amplitude transmission spectra for an asymmetry of $d = 15 \mu\text{m}$ are shown in Figure 4.6(c) and 4.6(d), respectively. We observe significant linewidth narrowing in the YBCO-TASR in comparison to Al-TASR. In numerical simulations (Figure 4.6(c)) line-width difference between YBCO-TASR and Al-TASR is 8.62 GHz, whereas in the measured (Figure 4.6(d)) case it is 9.14 GHz. Experimental results are in good agreement with the simulated results.

4.5 Electric field confinement in Fano resonant metamaterials

Narrow line width of Fano resonance in superconductors in comparison to metals results in larger field confinement in the capacitive gap of the TASR structures. For better understanding of the electric field confinement in the capacitive gaps of the TASRs, we simulated electric field distribution at Fano resonance frequency in YBCO-TASR and Al-TASR. Figure 4.7 (a) and b) depicts the electric field distribution in YBCO-TASR and Al-TASR, respectively, for an asymmetry of $d = 1.8 \mu\text{m}$, while Figure 4.7 (c) and (d) represent the electric field distribution in YBCO-TASR and Al-TASR, respectively, for an asymmetry of $d = 10 \mu\text{m}$. We observed that at lower asymmetries ($d = 1.8 \mu\text{m}$), the electric field confinement in the capacitive gaps is stronger in YBCO-TASR compared to the Al-TASR. However, at larger asymmetry ($d = 10 \mu\text{m}$) both YBCO and Al-TASRs show tight electric

field confinement at the Fano resonance frequency. Tight field confinement is highly sought for multifunctional applications such as biological and chemical sensing, and efficient cavities.

Figure 4.7: Electric field distribution at low and high asymmetry in the TASR structure: (a) and (b) shows the electric field distribution for an asymmetry of $d = 1.8 \mu\text{m}$ for YBCO-TASR and Al-TASR, respectively. Figure (c) and (d) depicts the electric field distribution in YBCO-TASR and Al-TASR, respectively for an asymmetry of $d = 10 \mu\text{m}$.

4.6 Critical DC conductivity to support Fano resonance excitation

The disappearance of the Fano resonance mode in the TASR metamaterials for conventional metals at lower asymmetries suggests that a critical minimum conductivity is required for supporting excitation of the Fano resonance. In fact, there exists a trade-off between the

material conductivity and the asymmetry parameter. We studied this by performing extensive numerical simulations for the two cases (i) at an asymmetry of $d = 10 \mu\text{m}$, we observe the evolution of Fano resonance by varying material conductivity and (ii) estimated the minimum critical conductivity required to support the Fano resonance excitation at varying asymmetry. Figure 4.8(a) depicts the gradual evolution of Fano resonance on increasing the conductivity values. It is observed that a TASR with an asymmetry of $d = 10 \mu\text{m}$ ($\alpha = 8.5\%$) requires a minimum DC conductivity of $3 \times 10^6 \text{ S/m}$ to support Fano resonance. We further studied the trade-off between the asymmetry parameter and the minimum conductivity required for supporting Fano resonance as shown in Figure 4.8(b), where we observe sharp fall in the minimum critical conductivity with increasing asymmetry parameter. Therefore, with increasing radiative losses in the structure, TASR metamaterial results in Fano resonances of broader linewidth. A Fano resonance with broad line-width, therefore, requires lower conductivity of material which elucidates the decrease in the desired conductivity at larger asymmetry in the TASR structures. For a asymmetry range of $1 \mu\text{m}$ to $20 \mu\text{m}$, we observed an order of magnitude enhancement in the required minimum conductivity of the material to support Fano resonance excitation. We observe an abrupt jump in the required minimum conductivity value below asymmetry $d = 3 \mu\text{m}$ ($\alpha = 2.6\%$), which can be classified as extremely low asymmetry regime (highlighted in yellow colour in Figure 4.8(b)). Such a steep rise in the minimum conductivity required to support excitation of Fano resonance provides an avenue for the other high conductivity materials such as superconductors to play an effective role since the Drude metal lacks to provide such a high conductivity. However, at larger asymmetry, the required minimum conductivity eventually saturates. These results are of great significance in finding an appropriate material for designing the high Q Fano resonant based metamaterial and plasmonic devices. This concept could be extended for the

materials of known conductivity to design Fano resonators by choosing the desired asymmetry in the TASRs.

Figure 4.8: a) Simulated transmission spectra of the TASRs with varying conductivity at a fixed asymmetry $d = 10 \mu\text{m}$. (b) Variation of minimum conductivity of a material required to support Fano resonance with different asymmetry d values. The shaded region represents the sudden increase in the minimum conductivity required to excite Fano resonance at extremely low asymmetry regime.

4.7 Q factor and figure of merit comparison

To further establish the superior role of the YBCO superconductor over metal, we estimated line-width, Q factor and figure of merit (FoM) of the Fano resonance for the Al-TASR and YBCO-TASR with varying asymmetries as shown in Figure 4.9. Figure 4.9(a) clearly depicts that the line-width of the Fano resonance for the YBCO-TASR are narrower for all the asymmetries compared to Al-TASR. The narrow line-widths in YBCO-TASR structure could be attributed to the lower losses arising from the excellent conducting properties of the YBCO superconductor. Figure 4.9(b) shows the estimated Q factors of the YBCO-TASR and Al-TASR with varying asymmetries. At higher structural asymmetries, both YBCO-TASR and Al-TASR results in almost identical Q factors, whereas at lower asymmetries YBCO-TASR clearly give rise to high Q factor resonances. The resonance frequencies of the YBCO-TASR are lower as compared to the Al-TASR, and hence, in spite of narrower line-width, Q factors are almost identical for both the materials at higher asymmetries. The maximum Q factors achieved for YBCO-TASR is 167, which is almost two-fold higher than the highest Q factor of Al-TASR. Figure 4.9 (c) represents the comparison of the FoM for the YBCO-TASR and Al-TASR. The FoM is defined as the product of the Q factor to the intensity (ΔI) of the resonance peak. YBCO-TASRs structure results in larger FoM values than Al-TASRs at all the asymmetries. FoM peaks at an asymmetry of $d = 5 \mu\text{m}$ with the highest value of 6.90 for YBCO-TASR, while FoM for Al-TASR peaks at an asymmetry of $4 \mu\text{m}$ with the highest value of 5.80, as shown by red and blue curves in Figure 4.9(c), respectively. Thus, we observe that at lower structural asymmetries below $3 \mu\text{m}$, the material conductivity plays a significant role in determining the Fano resonance amplitude and linewidth due to relatively low radiative losses.

4.8 Loss tangent of superconductor and metal

The Q factor of the subwavelength metamaterial structures is inversely proportional to the loss tangent. The loss tangent is defined as

Figure 4.9: Line-width, Q Factor and FoM of Fano resonances for YBCO-TASR and Al-TASR at varying asymmetry: (a) (b) and (c) represent the line-width, Q factors and FoM, respectively, with increasing asymmetry d in the YBCO and Al-TASR. Dotted area represents low asymmetry regime accessible by YBCO-TASR but inaccessible by Al-TASR.

$$\tan\delta = \frac{\epsilon''}{\epsilon'} \quad \dots \dots \dots 4.1$$

where ϵ' and ϵ'' is the real and imaginary part of the permittivity. The loss tangent in terms of the real (σ') and imaginary (σ'') part of the conductivity is given by

$$\tan\delta = \frac{\sigma'}{(\omega\epsilon_0 - \sigma'')} \quad \dots \dots \dots 4.2$$

Equation 4.2 shows that the resonant properties of the metamaterial structure depends on real as well as imaginary part of the conductivity of the material. Low real part and high imaginary part of the conductivity in the case of YBCO in comparison to the Aluminium metal leads to the lower loss tangent value.¹⁵⁹ Since Q factors are inversely proportional to the loss tangent, YBCO-TASR support higher Q factor resonances.

4.9 Effect of conductivity on Lorentzian resonant terahertz metamaterials

In this section, we study the impact of conductivity on terahertz metamaterials which have different geometry and loss mechanism compared to TASR. For this study, we selected a single gap split ring resonator (SRR), where the whole current loop ignoring the gap acts as an inductor (L) and split gap acts like a parallel plate capacitor (C) and results in Lorentzian (LC) line-shaped resonances.^{160,161} The LC resonance in SRR is well know and it is described by an equivalent RLC circuit. High circulation current in the SRR perimeter is essential for strong resonance excitation but it is limited by the resistance of the material that forms the SRR. The LC resonance is a fundamental magnetic resonance that couples strongly with the free space and results in a higher radiative loss. Since it shows higher radiative losses, and thus hinders the enhancement of Q factor in LC resonators. Therefore, unlike the TASR

metamaterials where the dominant loss mechanism is determined by the asymmetry parameter, the loss mechanism in LC resonators is governed by the radiative losses. The schematic of the SRR array and the unit cell is shown in Figure 4.10. On excitation with a terahertz wave polarized (E_x) parallel to the gap side arms, SRR results in super-radiant Lorentzian LC resonance.¹⁶⁰ The asymmetry ' d ' in the structure is introduced by moving the split gap away from the central vertical axis. The structural dimensions of the SRR are chosen in such a way that they result in the LC resonance at about 0.5 THz, which is identical to the Fano resonance frequency of the TASR. In the numerical simulations, the metallic (Al-SRR) and superconducting (YBCO-SRR) structures are defined by the DC conductivity of aluminum and experimentally measured permittivity of YBCO, respectively, as described in section 4.2.

Figure 4.10: Schematic of the single gap split ring resonator array (SRR); all the structural dimensions are revealed in the unit cell. “ d ” is the asymmetry, which is the displacement of the split gap from the central vertical axis.

Figure 4.11: Simulated amplitude transmission spectra obtained from SRR for the incident THz field polarized parallel to the gap arm of the structure: a-d) represents the simulated transmission spectra with asymmetry $d = 0, 2, 5$ and $10 \mu\text{m}$ in SRR, respectively. Colours of the curves represent the superconductor (red) and metal (black) materials used to design SRR.

The numerically simulated amplitude transmission spectra of Al-SRR and YBCO-SRR for x polarization of incident THz field with varying asymmetry $d = 0, 2, 5$ and $10 \mu\text{m}$ are shown in Figure 4.11(a)–(d), respectively. Al-SRR and YBCO-SRR structures result in almost similar resonance response in terms of transmission amplitude at all the asymmetries. Furthermore, the superconducting resonances are slightly sharper as compared to the metallic

counterpart because of its lower ohmic loss. The kinetic inductance of Cooper pairs in superconductors lead to red spectral shifting of the resonances in the YBCO-SRR in comparison to identical Al-SRR structures. We further studied the Q factor of resonances to gain more physical understanding. The Q factor of the LC resonance is defined as the ratio of the central resonance frequency (f_0) to the full width at half maxima (Δf) of the intensity spectrum. The estimated Q factors of Al-SRR and YBCO-SRR at each asymmetry are shown in Figure 4.11. It is observed that both Al-SRR and YBCO-SRR results in the similar Q factors. However, Al-SRR shows slightly lower Q factors in comparison to the YBCO-SRR due to higher Ohmic losses in the metals. Since superconductor and metal both result in nearly identical resonance response in terms of resonance amplitude and Q factors at terahertz frequencies, we can conclude that as long as the material conductivity is high enough, it is the geometrical parameters that play a dominant role in determining the resonance amplitude and the linewidth of highly radiative LC resonant metamaterials.

4.10: Conclusion

In summary, we have identified that the high- T_c superconductors could play a vital role over normal metals in Fano resonant metamaterials at terahertz frequencies. We showed that the minimum conductivity required for the excitation of sharp Fano resonance increases steeply at low structural asymmetries thereby even high conductivity Drude metals fail to support Fano resonances. Superconductors provide access to this extremely low asymmetry realm, which is inaccessible with the high conductivity metals and results in significantly higher Q factor and FoM of Fano resonances as compared to metals. Moreover, the superconductors results in narrow linewidth resonances at all the asymmetry values. Our observations manifest that along with the structural design engineering, choice of material is vital for

achieving high Q resonances that could find applications in designing ultrasensitive sensors, narrowband filters, and active metadevices in the terahertz regime.

Chapter 5

All Optical Dual-Channel Ultrafast Superconductor Metamaterial Switch

This chapter deals with the ultrafast switching of the high- T_c superconductor based Fano resonant metamaterials. Upon pumping with optical pulse, YBCO metamaterial show existence of dual dissociation–relaxation windows within a single superconductivity restoration cycle which lead to a dual-channel, ultrafast switchable metamaterial devices. Dual switching channels exist due to primary and secondary dissociation of Cooper pairs, which eventually results in two distinct pathways for the modulation of terahertz radiation at picoseconds time scale.

5.1 Introduction

Superconductors are an interesting family of materials that have attracted considerable attraction from both the fundamental and applied physics community. Superconductors possess zero DC electrical resistance and perfect diamagnetism below T_c . Superconductors have enabled various interesting applications such as magnetic resonance imaging (MRI), superconducting maglev trains, magnetometers, superconducting quantum interference devices (SQUIDS), and bolometer. The major impetus to the practical applications of these devices has come from the discovery of the high- T_c superconductors, as the superconductivity in these materials could be observed above liquid nitrogen temperatures.^{4,5} They are excellent conductors below T_c and behave as poor metal/dielectric above T_c . When a superconducting material is cooled below T_c , the normal carriers pair up to form quantum

mechanical Cooper pairs. The exceptional superconducting properties when cooled below T_c , are attributed to the presence of a superfluid – a mixture of normal free carriers and Cooper pairs.^{15,17} At and above T_c , all the Cooper pairs convert to normal quasiparticles.

The Cooper pairs are loosely bound pairs of normal charge carriers and typically have binding energies in the range of 2-75 meV, which thus makes terahertz spectroscopy an ideal tool to investigate the non-equilibrium dynamics of Cooper pairs in superconductors.¹⁵⁻¹⁷ These Cooper pairs show extreme sensitivity to the incident photons of the energy greater than their binding energy. When irradiated with an optical field, the Cooper pairs dissociate and restore again in an ultrafast timescale of few picoseconds (ps).^{32,100,162} In this process, a large reduction in the number of Cooper pairs results in the significant change in the complex conductivity of the superconducting material, thereby leading to an ultrafast switching from an efficient conductor to a poor metal. This active destruction and restoration of Cooper pairs at extremely short timescales make superconductors an ideal material platform for ultrafast switchable photonic and metamaterial devices.

Metamaterials, which are artificially structured subwavelength structures, provide opportunities based on their geometrical structure to manipulate light-matter interactions and have been utilized to show several unusual phenomena such as slow light, subwavelength imaging, flat optics, chirality and perfect absorption.^{43-46,51,53,163} Superconductors are highly sought and extensively employed for the low-loss metamaterials as they effectively curb ohmic losses due to their excellent conductivity in the radio wave, microwave, and terahertz frequency regime.^{76,78,91,102,164} Integrating superconducting materials to the resonator structure also enables dynamic control of the optical response as the superconducting properties are immensely sensitive to the external stimuli, thereby resulting in actively tunable superconducting metamaterial devices using external perturbations such as

temperature^{87,88,92,94,133}, magnetic field^{86,95,96}, electric current⁹⁷⁻⁹⁹ and optical irradiation¹⁰⁰. However, the modulation in the resonance response accomplished by thermal perturbations is very slow, whereas using electrical control requires complicated design and fabrication. Active control of metamaterial response at ultra-fast time scales is an highly sought feature for achieving dynamically tunable devices for on-demand applications. Therefore, optical control of the high- T_c superconductor based metamaterials is an important avenue for realizing ultrafast switchable devices at terahertz frequencies.

In this chapter, we demonstrate an ultrafast switching of Fano resonance^{139,142} through optical irradiation of high- T_c Yttrium Barium Copper Oxide (YBCO) superconductor based terahertz asymmetric split ring (TASR) resonators. When irradiated with an optical pulse with energy higher than the binding energy of Cooper pairs, the Cooper pairs undergo primary dissociation due to the splitting of Cooper pairs into normal quasiparticles. These quasiparticles bind together again to form Cooper pairs in the extremely short timescale of few picoseconds (ps)³². Interestingly, the optical pulse reflected from the back surface of the substrate results in the secondary dissociation of the Cooper pairs. The extremely sensitive nature of Fano resonance^{139,142,156} allows to capture the resonance modulation even for the secondary dissociation of Cooper pairs in the photoexcited superconductor metamaterials, which was inaccessible in the previously studied highly radiative inductive-capacitive (LC) superconductor resonator system.¹⁰⁰ Upon pumping with high fluence of optical field, the Fano resonant metamaterial device shows dual switching channels with different time constants due to the dual dissociation-relaxation of Cooper pairs within a single superconductivity restoration cycle. Apart from the ultrafast switching by optical pumping, we also demonstrate that the similar resonance modulation could be achieved at larger time scale by changing the temperature of the YBCO-TASR. Dynamically tunable high- Q

resonances are desired for applications such as slow light, nonlinearity, ultrasensitive sensing, and precise manipulation of electromagnetic radiation.^{56,104,163,165}

5.2 Design and fabrication of metamaterial sample

To enable active control of sharp Fano resonances using temperature and photo-excitation, we choose the terahertz asymmetric split ring (TASR) resonators as shown in Figure 5.1(a). It is an asymmetric split-ring resonator with dual split gap, where asymmetry is introduced by moving one of the split gaps away from the central vertical axis by a distance ' d '. For this study, we chose an asymmetry of $d = 15 \text{ }\mu\text{m}$, in order to get a Fano resonance of high amplitude for the ease in capturing resonance response in the measurements. However, high Q Fano resonances can be readily achieved by decreasing the structural asymmetry (d) values.¹⁴³ The TASR metamaterial sample is fabricated using conventional photolithography and wet chemical etching of a 50 nm thick YBCO film deposited over a r -cut sapphire substrate of thickness 500 μm . This YBCO film has a T_c of 85.1 K and J_c of 2.9 MA/cm². The detailed fabrication process is given in section 2.2. Optical image of the small area of the fabricated asymmetric metamaterial array is shown in Figure 5.1(b). The inset in Figure 5.1(b) shows the TASR unit cell with all the geometrical parameters. In this TASR metamaterials, Fano resonance mode is excited by incident terahertz pulse polarized perpendicular to the gap bearing arms of the resonator, which also give rise to a broad dipolar resonance at a higher frequency. The observed Fano resonance feature is due to the interference between a narrow discrete (dark) mode arising from the structural symmetry breaking and the broad dipole (bright) mode of the perfectly symmetric structure.^{138,139,156} This YBCO-TASR sample is characterized using a ZnTe nonlinear crystal based confocal terahertz time domain spectroscopy setup. A continuous flow liquid helium cryostat was

incorporated to the system in order to facilitate transmission measurements at cryogenic temperatures. The details of the measurement are given in section 2.3. The near-infrared optical laser beam of wavelength 800 nm (1.55 eV), pulse width of ~ 120 fs with a repetition rate of 1 kHz is split primarily into two parts with one being used for pumping the non-linear

Figure 5.1: Schematic diagram of the YBCO-TASR metamaterial array. (a) The pictorial representation of the OPTP measurements of the YBCO-TASR. (b) optical image of the fabricated YBCO-TASR sample. Inset shows the geometrical parameters of a unit cell with periodicity $p = 75\ \mu\text{m}$, arm length $l = 60\ \mu\text{m}$, arm width $w = 6\ \mu\text{m}$, asymmetry parameter $d = 15\ \mu\text{m}$, and split gap $g = 3\ \mu\text{m}$. (c) The artistic illustration of dissociation of Cooper pairs into quasiparticles due to the photo-excitation with the optical pump.

ZnTe crystal to generate and detect terahertz pulses and the other part of the laser beam is used for photo excitation of YBCO-TASR metamaterial. The photon energy of the photoexcitation pump beam is much larger than the Cooper pair binding energy (20-30 meV) of the YBCO superconductor.¹⁶ The diameter of the optical pump beam is approximately 10 mm, which is much larger than the spot size (4 mm) of the focused terahertz probe beam at the sample position, that ensures a uniform photoexcitation of the YBCO-TASR sample. Two separate motorized delay stage controls the time delay between the terahertz probe and optical pump pulses. Finally, the transmission spectrum is obtained by normalizing spectrum through the sample ($E_S(\omega)$) to the spectrum through reference substrate ($E_R(\omega)$) using the relation $|T(\omega)| = |E_S(\omega)|/|E_R(\omega)|$.

5.3: Optical pump dependent switching of Fano resonance

The Cooper pairs show extreme sensitivity to the external perturbations and are readily dissociated by photoexcitation of superconducting materials with the optical fields of energy above the binding energy of the Cooper pairs. The dissociation of Cooper pairs of a superconductor alters its conducting properties that show strong influence on the optical response of the metamaterial devices. The transmission spectra of the photo-excited YBCO-TASR are measured using an OOTP spectroscopy setup. In the optical pump dependent transmission measurements, the terahertz probe pulse is delayed by 4.9 ps with respect to the optical pump pulse in order to capture the optical response at maximum dissociation of the Cooper pairs. Here, 4.9 ps is the rise time of the Cooper pair dissociation dynamics. Figure 5.2(a) depicts the transmission spectra of YBCO-TASR sample measured on irradiating the sample with different optical pump fluences. The pump fluence is defined as the radiant energy incident per unit surface area ($\mu\text{J}/\text{cm}^2$). In the absence of optical pump pulse, YBCO-

TASR shows strong Fano and dipole resonances at 0.39 and 0.57 THz respectively, as shown by the black colour curve in Figure 5.2(a). On irradiating the sample with low ($\sim 63.7 \mu\text{J}/\text{cm}^2$) fluence of the optical beam, a faint modulation of the Fano and dipole resonances are observed (see red curve in Figure 5.2(a)). On increasing the fluence of the incident optical pump, a large modulation in the Fano and dipole resonance amplitude is observed. The reduction in the amplitude of Fano and dipole resonance is due to the decreased density of the Cooper pairs in the YBCO. The incident optical pulse dissociates a fraction of the Cooper pairs into the normal quasiparticles, thus results in the reduced complex conductivity of the YBCO. The dissociated fraction of the Cooper pair is highly dependent on the fluences of the optical pump beam. At a larger pump fluence of $955.4 \mu\text{J}/\text{cm}^2$, the amplitude of Fano and dipole resonance reduces significantly due to dissociation of a large fraction of Cooper pairs into quasiparticles. However, at this pump fluence ($955.4 \mu\text{J}/\text{cm}^2$), Fano and dipole resonances are not completely switched off due to a smaller fraction of remnant Cooper pairs. The modulation in Fano and dipole resonance amplitudes at various excitation pump fluences are calculated as $(T_{\text{off}} - T_{\text{on}})/T_{\text{off}} \times 100 \%$, where T_{off} and T_{on} , respectively, stand for the resonance amplitudes at ‘off’ and ‘on’ states of the optical pump beam. The amplitude of the Fano and dipole resonance is the difference between the peak and dip of the respective resonance mode in the transmission spectra. The calculated percentage amplitude modulation of Fano and dipole resonance mode at increasing pump fluence of incident optical pump beam is shown in Figure 5.2(b). The utmost amplitude modulation of 86% and 60% is observed for the Fano and dipole resonance, respectively. The Fano resonance shows higher sensitivity to the optical pump at identical fluence of the pump beam in comparison to the dipole resonance, owing to the higher electric field confinement in the split gaps of the TASR at Fano resonant frequency. It is observed that a relatively large optical pump fluence is

Figure 5.2: (a) Measured amplitude transmission spectra of YBCO-TASR at various fluences of the incident optical pump beam. (b) The amplitude modulation versus pump fluence plot for the dipole and Fano resonance.

required to completely switch off the Fano and dipole resonances in a YBCO-TASR sample of thickness 50 nm. However, further engineering of the metamaterial sample with lower YBCO thickness can potentially provide a complete low threshold switching performance at significantly lower optical pump fluences. The fill fraction of superconductor, which is the active resonator material in our TASR structure is only about 23% of the whole metamaterial

area. Focusing the excitation optical pump pulse onto the active (superconducting) area selectively, could also provide significant reduction in the optical pump fluences required for complete switching.¹⁶⁶

5.4: Ultrafast switching

We further capitalize on the ultrafast response observed in the Cooper pair breakup and restoration in the YBCO superconductor upon photoexcitation, which take place in picoseconds timescale to realize ultrafast modulation in the Fano resonance spectra of the YBCO-TASR sample. The black curve in Figure 5.3(a) depicts the Cooper pair dissociation and restoration dynamics of the YBCO-TASR metamaterial (fabricated on 0.5 mm thick sapphire substrate) measured at 5 K under the influence of an optical pump beam of wavelength 800 nm and fluence of $254.8 \mu\text{J}/\text{cm}^2$. Upon illuminating the sample with optical pump beam of energy larger than the Cooper pair binding energy, the superconductivity is destroyed by the dissociation of the Cooper pairs momentarily. The reduced density of the Cooper pairs leads to the sudden reduction in the conductivity thereby increases the differential transmission signal. The quasiparticles bind together again in a few picoseconds timescale to form Cooper pairs and thus results in restoring the superconductivity.

5.4.1 Dual channel switching

We also observe that at the time delay of around 6 ps from the primary dissociation peak, a secondary peak gradually evolves in the excitation-relaxation dynamics. The secondary peak arises from the secondary dissociation of the Cooper pairs due to the Fabry-Perot etalon pulse from the rear surface of the TASR substrate. The round-trip distance travelled by the optical pulse of wavelength 800 nm inside the substrate before resulting in the secondary

Figure 5.3: (a) Cooper pair dissociation-relaxation dynamics of the YBCO-TASR for the pump fluence of 254.8 $\mu\text{J}/\text{cm}^2$. Red (dashed) curve shows the Cooper pair dynamics of YBCO-TASR on 2 mm thick *r*-cut sapphire substrate. (b) Measured amplitude transmission versus frequency spectra at varying time delays between pump and probe. Inset represents the Fano resonance amplitude modulation for varying pump-probe time delay.

dissociation is given as $2nt_s$ and the corresponding time taken to travel this distance is given as $2nt_s/c$, where n denotes the refractive index of the substrate at optical pump wavelength, t_s is the thickness of the substrate and c is the speed of light in vacuum. For a sapphire substrate of thickness 500 μm and refractive index of 1.76 at wavelength 800 nm, the Fabry-Perot pulse is delayed by 5.9 ps, which matches closely with the experimentally measured time delay. Most importantly, the secondary peak could be engineered to get desired time delay by altering substrate properties which affect reflection of incident optical pulse from the rear surface such as refractive index (n) or thickness (t_s) and provides an interesting approach to manipulate switching speed of the metamaterial device. The red colour (dashed) curve in Figure 5.3(a) shows the Cooper pair dissociation-relaxation dynamics of the YBCO-TASR fabricated on a 2 mm thick sapphire substrate and illuminated with identical fluence of optical pump beam. We note that the secondary dissociation peak evolves at a time delay of 23.6 ps from the primary dissociation peak, which is closely matching with the calculated time delay of 23.4 ps for 2 mm thick sapphire substrate.

The ultrafast response observed in the photoexcitation of the superconductor is reflected in the transmission spectra of the YBCO-TASR sample. Figure 5.3(b) depicts the ultrafast modulation in the terahertz amplitude transmission spectra of the YBCO-TASR sample at various time delay, τ_p denoted as I, II, III and IV of the optical pump pulse depicted in Figure 5.3(a). At the point I, just before ($\tau_p = -2$ ps) the arrival of the optical excitation pulse, the strength of the Fano resonance at 0.39 THz is very strong and is represented by the black curve in Figure 5.3(b). At point II ($\tau_p = 4.9$ ps), where the optical pump results in the maximum dissociation of Cooper pairs, we observe a strong reduction in the Fano resonance strength, owing to the large reduction in the complex conductivity of the YBCO. The quasiparticles resulted due to the dissociation of the Cooper pairs, bind together again to

restore superconductivity thereby results in a gradual enhancement in the Fano resonance amplitude at III ($\tau_p = 9.2$ ps) and IV ($\tau_p = 38.3$ ps). Here, it is important to note that there is a trade-off between the excitation optical pump power and the ultrafast response of the YBCO-TASR. High optical pump fluences lead to the large modulation depth but slow switching speed, whereas low pump fluences result in faster switching speed but relatively lower modulation depth. Therefore, to achieve both ultrafast optical response and remarkable modulation depths from the YBCO-TASR sample, we chose to illuminate the sample with an optical pump fluence of $254.8 \mu\text{J}/\text{cm}^2$. This approach results in a partial modulation of 44% in the Fano resonance amplitude at the maximum dissociation point (II) of Cooper pairs, which could be restored within 40 ps. We also observe that the Fano resonance is not fully restored due to the thermal effects arising due to the optical pump beam.

To observe the secondary dissociation signature in the optical response of the YBCO-TASR sample, we measured the terahertz transmission spectra at various time delays (τ_p) between optical pump and terahertz probe pulse varied in small steps. The inset in Figure 5.3(b) shows the amplitude modulation of Fano resonance at different pump-probe time delays. We observe that initially amplitude modulation in the Fano resonance increases expeditiously and reaches a maximum value at $\tau_p = 4.9$ ps due to the dissociation of the maximum number of Cooper pairs. The amplitude modulation in the Fano resonance reduces on further increase in time delay till the reflected optical pulse hits the sample at $\tau_p = 10.7$ ps. We observe a pronounced peak in the amplitude modulation of Fano resonance in YBCO-TASR at $\tau_p = 10.7$ ps (highlighted in red circle) which arises from the secondary dissociation of the Cooper pairs by reflected optical pulse arriving from the rear surface of the substrate. On further increase in time delay between the the optical pump and terahertz probe, the amplitude modulation reduces gradually. Therefore, the extreme sensitivity of Fano resonances to the

small changes in the conductivity paves the path for the experimental observation of the dual dissociation-relaxation channels in one complete cycle of superconductivity restoration and to realize the dual channel meta-switch.

5.4.2 Switching speed of the metamaterial device

To get an estimate of the restoration time of the superconductivity in the photoexcited sample, we analyzed the Cooper pair relaxation dynamics of YBCO-TASR sample for varying excitation pump fluences. Figure 5.4(a), (b) and (c) depicts the normalized Cooper pair relaxation dynamics of the YBCO-TASR sample measured at 5 K using the OPTP setup at the optical pump fluences of 63.7, 127.4 and 254.8 $\mu\text{J}/\text{cm}^2$, respectively. We observe only a primary dissociation peak for lower optical pump fluence of 63.7 $\mu\text{J}/\text{cm}^2$, while higher pump fluences of 127.4 and 254.8 $\mu\text{J}/\text{cm}^2$ results in an additional secondary dissociation peak along with the primary dissociation. However, both the primary and secondary relaxation dynamics follows bi-exponential relaxation as follows,

$$y = y_0 + A_1 e^{\left(-\frac{(x-x_0)}{t_1}\right)} + A_2 e^{\left(-\frac{(x-x_0)}{t_2}\right)} \quad 5.1$$

where t_1 and t_2 reflect the lifetime of fast and slow relaxation processes, respectively. Whilst the accurate nature of the Cooper pair relaxation dynamics of cuprate superconductors is not fully resolved, but the fast relaxation time (t_1) depicts the electronic transitions, which include the combination of two processes namely relaxation of hot quasiparticles by breaking more remnant Cooper pairs in the vicinity and electron-phonon ($e-ph$) mediated relaxation, while the slow relaxation time (t_2) is attributed to the formation of Cooper pair mediated by phonons of energy $\geq 2\Delta$, where 2Δ is the superconducting energy gap.³¹ These 2Δ phonons are capable of breaking the Cooper pairs again which makes the entire relaxation process slow. This cascading dissociation of Cooper pairs due to the highly energetic phonons is

called phonon bottleneck effect. The detailed description of the Cooper pair dynamics is given in section 1.4. The extracted, fast and slow relaxation time constants for primary and secondary relaxation dynamics of Cooper pairs at all the pump fluences are listed in Table 5.1. The bi-exponential fitting of primary relaxation (shown as red curve in Figure 5.4) reveals that the time scale of the faster decay is around 0.5-1.4 ps for all the pump fluences, whereas the relaxation time constant of the slower decay is around 8-13 ps. Similarly, the bi-exponential fitting of secondary relaxation dynamics demonstrates that the time constants of the faster decay are 2.8 and 3.6 ps, whereas the time constants of the slower decay are 49.7 and 84.2 ps for the optical pump fluences of 127.4 and 254.8 $\mu\text{J}/\text{cm}^2$, respectively, as shown by the green curves in Figure 5.4(b) and (c). It is observed that with increasing optical pump fluences, the relaxation time constant increases which is originated from the increased density of hot quasiparticles that results from the large breakup of the Cooper pairs.³¹ Whilst the switching speed of the metamaterial device is governed by the slower relaxation time constants; the overall operational speed is still within 100 ps. Interestingly, the secondary peak in the Cooper pair dissociation relaxation dynamics offers dual decay windows within

Table 5.1: Relaxation time constants at different optical pump fluences extracted using bi-exponential fitting

Pump fluence ($\mu\text{J}/\text{cm}^2$)	Fitted time constants (ps)			
	Primary relaxation		Secondary relaxation	
	t_1	t_2	t_1'	t_2'
63.7	0.5	8.3	-	-
127.4	1.4	10.3	2.8	49.7
254.8	1.3	13.5	3.6	84.2

Figure 5.4: Normalized Cooper pair relaxation dynamics of the YBCO-TASR sample at incident pump fluences of (a) 63.7 $\mu\text{J}/\text{cm}^2$, (b) 127.4 $\mu\text{J}/\text{cm}^2$ and (c) 254.8 $\mu\text{J}/\text{cm}^2$. Bi-exponential fitting of first and second peak is depicted by red and green curves, respectively.

one complete recovery cycle, with different switching speeds. This phenomenon of secondary dissociation and switching speed manipulation is observed because of the ultra-sensitive nature of the Cooper pairs to the incident optical pump. Two relaxation channels pave the way for dual switching speed with a single incident pump fluence in such metamaterial devices. These designs could lay an advantageous platform for realizing dual channel ultrafast resonant modulators operating at terahertz and microwave frequencies.

5.5: Temperature-dependent modulation of Fano resonance

Besides the optical irradiation based manipulation of the superconducting properties of the YBCO, the quantum mechanical Cooper pairs are highly sensitive to the temperature of the specimen. The thermal control of Fano resonant YBCO-TASR metamaterial was enabled by measuring its terahertz transmission spectra at varying sample temperature. Figure 5.5(a) depicts that the transmission spectra of YBCO-TASR metamaterial show a very strong Fano and dipole resonance features at temperature 5 K, which arises from the superconducting properties of YBCO well below T_c of around 86 K. The amplitudes of the Fano and dipole resonance reduces gradually on increasing the temperature from 5 K and finally vanishes at 80 K, which is very close to the T_c of the YBCO thin film. At and above 80 K, both the Fano and dipole resonance fully disappears due to the phase transition of YBCO from superconducting state into the normal state. This temperature dependent active switching between ‘on’ and ‘off’ state of the resonance features of YBCO-TASR is extremely important for actively switchable metamaterial and plasmonic devices. Moreover, with increase in the temperature, the optical response of the YBCO-TASR sample depicts gradual red spectral shifting of Fano and dipole resonances. The spectral shift of the YBCO-TASR resonance is attributed to the change in the kinetic inductance of the high- T_c superconducting

Figure 5.5: (a) Temperature dependent (5 K-80 K) transmission spectra of YBCO-TASR sample. (b) Fano and dipole resonance frequencies at different temperatures.

YBCO.^{84,167} From Figure 5.5(b) it is observed that on varying the temperature from 5 K to 70 K, the Fano and dipole resonance redshift by 80 GHz and 150 GHz, respectively. Thus, the large change in the imaginary part of complex conductivity of the YBCO superconductor upon photoexcitation results in the modulation in the amplitude of the resonances, whereas the shift in the resonance frequency is due to the large change in kinetic inductance of the

superconducting Cooper pairs. This enable simultaneous control of optical response in terms of resonance frequency tuning as well as amplitude modulation with the change in temperature of the sample.

5.6 Conclusion

In this chapter, we have experimentally demonstrated dual-channel ultrafast modulation of Fano resonances upon irradiation with femtosecond near-infrared laser beam in a terahertz superconducting asymmetric split ring resonator metamaterial. We observed dual dissociation-relaxation dynamics in a single superconductivity restoration cycle which leads to the realization of the ultrafast dual channel meta-switch. Moreover, we also discussed an effective way of controlling and manipulating the secondary dissociation channel by engineering the substrate properties such as refractive index and thickness, which provides an unprecedented control over the overall switching speed of the metamaterial devices. These dual-channel switchable metamaterial devices are highly advantageous for various application in time division multiplexing, superconducting photodetectors, THz high-speed wireless communication and superconducting radiation sensors. The ultrafast response in the Cooper pair relaxation dynamics of the photoexcited superconductors is of immense interest in high-speed electronics and nonlinear metamaterials devices.

Chapter 6

Ultrathin Superconducting Metamaterials

In this chapter, we report an important discovery of ultrathin metamaterials based on high- T_c YBCO superconductor. We show that the 25 nm thick superconducting metamaterials support strong Fano resonance excitation while the metallic resonators possessing sub skin depth thickness fail to support the Fano resonance excitation. Moreover, we also demonstrate that upon irradiation with an external optical field, ultrathin superconducting metamaterials show extremely low threshold, ultrafast, dual-channel switching behavior.

6.1 Introduction

As discussed in the chapter 5 of this thesis, despite the need for the cryogenic cooling, several unique properties of the superconductors such as low Joule losses, nonlinear response to the oscillating currents below superconducting gap frequencies, flux quantization, and Josephson effect have enriched the technological advancement, and further creating new opportunities in photonics.⁷⁶⁻⁷⁹ Superconductors are excellent plasmonic materials as they exhibit negative permittivity.¹⁶⁸ Superconducting metamaterials/photonic devices, provide an excellent platform for the dynamic tunability, as superconductivity is strongly affected by the wide range of external stimulations such as magnetic field, temperature, electric current, and optical irradiation.^{2,86,87,92,95,97,100,101} Under the influence of external perturbation, a fraction of Cooper pairs gets dissociated into the quasiparticles which alter the complex conductivity of the superconducting material and leave its footprints on the optical response of the metadvicees. Once external perturbations reach their critical values such as critical magnetic

field (H_c), critical phase transition temperature (T_c), and critical current density (J_c), the quantum mechanical Cooper pairs disappears, and the superconductor turns to a poor metal.² On irradiation with an optical pulse of energy higher than their superconducting gap energy, the superconductors show an ultrafast transition between the superconducting and resistive state of the material due to the dissociation and restoration of the Cooper pairs on an extremely fast timescale of few picoseconds that lead to the realization of ultrafast switchable metamaterial devices.^{32,162}

In this chapter, we systematically investigate the possibility of utilizing the exceptional features brought about by ultrathin, sub-London penetration depth thicknesses of yttrium barium copper oxide (YBCO) superconductor to the photonic applications. The London penetration depth (λ_L) is defined as the length scale at which the magnetic field below the surface decreases to $1/e$ of its value at the surface of the superconductors. Typically, values of λ_L for YBCO is about 140 nm at 0 K, which increases with increasing temperature and finally becomes infinite at and above T_c .² We experimentally demonstrate superconducting ultrathin, broadband terahertz modulators for ultrafast and low threshold switching applications. On irradiation with an optical field, ultrathin YBCO superconductors undergo dual dissociation-relaxation of Cooper pairs in a single superconductivity restoration cycle and provide an ideal platform for dual channel switching applications. We also used multiple sub-London penetration depth thicknesses (thickness $< \lambda_L$) of high- T_c YBCO superconductor to design double gap terahertz asymmetric split ring (TASR) metamaterials, which supports sharp asymmetric line shape Fano resonances.^{138,139,142,156} The Fano resonance arises due to the interference between a narrow discrete mode arising due to structural symmetry breaking and the broad dipolar mode of the perfectly symmetric structure.^{130,143,156} The observed Fano resonances show extreme sensitivity to the small change in conducting properties of the

constituent materials as they effectively curb radiative losses thereby the resonance response is mainly derived from the ohmic losses incurred by the constituent materials. Surprisingly, we observed that the ultrathin YBCO-TASR of thickness as thin as 25 nm supports excitation of very strong resonances, whereas the resonance features are either absent or extremely faint in metallic structures of identical thickness. Moreover, ultrathin YBCO provides an opportunity to actively tune the resonance features at extremely low threshold of external perturbations. The resonance features could be actively tuned and precisely modulated using significantly low values of electrical current, magnetic field, optical fluences. As a proof of concept, we demonstrated optical switching of the ultrathin YBCO-TASR structures and compared them with structures of higher thicknesses. We demonstrate that the identical degree of modulation in Fano resonance amplitude of 25 nm thick YBCO-TASR could be realized by irradiating the sample with an order of magnitude lower optical pump fluences in comparison to the YBCO-TASR of thicknesses 50 and 200 nm. Once irradiated with optical field, these resonances could be switched 'on' and 'off' at a timescale of 20 ps and provide an opportunity to utilize them as ultrafast, dual-channel photonic switch. Along with low threshold switching nature, sub-London penetration depth thick superconductors give rise to high kinetic inductance, which is related to the inertia of the Cooper pairs and gives rise to the super-inductance.^{84,167} The kinetic inductance of a very thin superconducting wire is calculated as $L_K = \frac{\mu_0 \lambda_L^2}{wt}$, where μ_0 is the vacuum permeability, λ_L is the London penetration depth, t and w are the thickness and width of the superconducting strip, respectively.³⁴ High kinetic inductance in ultrathin superconducting metadevices opens up new prospects in the field of quantum electronic devices, quantum metrology, and information processing. The persistence of strong resonances in the ultrathin YBCO structures have important

technological implications in flat optics, low threshold actively switchable nonlinear scalable meta/photonic devices.

6.2 Ultrathin superconductors for photonic applications

To enrich the field of photonics by the extraordinary features brought about by the ultrathin superconductors, we used 25 nm thick YBCO film deposited over 2 mm thick sapphire substrate. Cerium oxide (CeO_2) of thickness 30 nm was used as the buffer layer between YBCO and sapphire substrate to improve the YBCO film quality and to prevent reactions between the substrate material and YBCO at high temperatures. The CeO_2 is significantly transparent to the optical pump, and terahertz probe frequencies and hence would not affect the optical response of the YBCO film.^{169,170} Figure 6.1(a) depicts the scanning electron microscope (SEM) image of the YBCO film which shows minor outgrowth of the YBCO material. The YBCO thin film has a T_c of 81.6 K and a J_c of 0.6 MA/cm². The thickness of the YBCO film was measured using Atomic Force Microscopy (AFM) technique by preparing a cross section using photolithography method. The measured thickness was found to be 55 nm which include 25 nm thick YBCO and 30 nm CeO_2 as shown in Figure 6.1(b). The terahertz transmission spectra of the YBCO film was measured using a ZnTe nonlinear crystal based terahertz time-domain spectroscopy (THz-TDS) system. A continuous flow liquid helium cryostat was incorporated in the THz-TDS system to perform measurement at cryogenic temperatures. The details of the measurement are given in the section 2.3 of chapter 2. The recorded broadband transmission of the 25 nm thick YBCO film at varying temperatures is depicted in Figure 6.2(a). On varying the temperature from 5 K to room temperature (295 K), thin YBCO film show around 50% modulation in the

Figure 6.1: a) SEM image of the YBCO film deposited over CeO₂ coated *r*-cut sapphire substrate. b) The thickness of the YBCO and CeO₂ film measured using inset figure show AFM image of a cross-section of the film.

broadband transmission owing to a large change in the imaginary part of the conductivity on phase transition from quantum mechanical superconducting phase to resistive phase. Along with the large change in conducting properties of the YBCO on temperature variation, the superconducting Cooper pairs in YBCO are also extremely sensitive to the incident photons of energy larger than the binding energy of their Cooper pairs. The binding energy of Cooper pairs in YBCO is about 20-30 meV.¹⁵⁻¹⁷ On irradiation with a near-infrared femtosecond

Figure 6.2: Terahertz transmission spectra of 25 nm thick YBCO film at a) different temperatures and b) varying fluences of the incident optical pump at 7 K. c) Differential terahertz transmission spectra (Cooper pair dissociation-relaxation dynamics) of YBCO film measured at incident pump fluence of 63.7 $\mu\text{J}/\text{cm}^2$. d) Single and bi-exponential fitting of normalized differential terahertz transmission.

laser pulse of energy 1.55 eV ($\lambda \sim 800$ nm), a fraction of Cooper pairs gets readily dissociated into the quasiparticles.

The fluence of the incident optical pulse and film thickness determines the fraction of Cooper pairs that gets dissociated into quasiparticles.¹⁰⁰ Figure 6.2(b) depicts the broadband (0.3-1 THz) modulation of 25 nm thick YBCO film upon irradiation with extremely low fluences of

optical pump. We observe around 30% modulation in the transmission at 0.8 THz, which is nearly identical to the modulation due to temperature variation from 5 K to 80 K. Most importantly, the hot quasiparticles resulted due to Cooper pair splitting upon optical irradiation recombine again to form Cooper pairs in an extremely short timescale thereby restoring the superconductivity. The large reduction of the conductivity and its restoration in ultrafast timescale provides an opportunity to use high- T_c superconductors for high-speed photonics and switching applications. We recorded the sharp change in the conductivity of the superconducting film in the form of differential transmission ($\Delta T/T$) signal. Figure 6.2(c) represents the Cooper pair dynamics (differential terahertz transmission spectra) of the YBCO film at varying time delays between optical pump and terahertz probe under the illumination of the optical pump with a fluence of $63.7 \mu\text{J}/\text{cm}^2$. Initially, at point A, $\tau_p = 0$, $\Delta T/T$ is zero, which increases rapidly with increasing time delay and goes to a maximum at point B ($\tau_p = 5$ ps) due to maximum dissociation of the Cooper pairs. On further increase in the time delay, there is a sudden reduction in $\Delta T/T$ value, which saturates at higher time delays. We also observe evolution of a secondary peak gradually at a time delay of 29 ps (point C in Figure 6.2 (c)) in the charge carrier dynamics, which arise due to the secondary dissociation of superconducting Cooper pairs by the reflected optical pulse from the rear surface of the substrate. Dual dissociation-relaxation dynamics observed in a complete recovery cycle lead to dual channel switch operating at different switching speed. Switching speed of the dual channels could be readily engineered by changing the thickness and refractive index (see section 5.4.1). To calculate the recovery time of dissociated Cooper pairs in YBCO, we performed bi-exponential fitting of both primary and secondary dissociation using the equation

$$y = y_0 + A_1 e^{(-\frac{(x-x_0)}{t_1})} + A_2 e^{(-\frac{(x-x_0)}{t_2})} \quad 6.1$$

where t_1 and t_2 are the time constants associated with fast and slow relaxation process. More interestingly, on carefully analyzing the fitting data, we observed that the primary dissociation shows identical time constant for fast (t_1) and slow (t_2) process. This peculiar feature is observed due to ultrathin nature of the YBCO films where before the slow relaxation starts dominating, a reflected pulse from the rear surface of the substrate hits the YBCO film again and lead to the dissociation of more Cooper pairs into quasiparticles. However, the bi-exponential fitting of the secondary peak in the dynamics shows different time constants. The detailed mechanism of Cooper pair dissociation-relaxation upon photoexcitation is described in section 1.4. Once a superconductor is irradiated with an optical field of photon energy larger than the superconducting gap (2Δ), it dissociates the Cooper pairs into hot quasiparticles. The percentage of Cooper pairs dissociated into the quasiparticles depends on the fluence (no. of photons incident) of the optical pump, the absorption coefficient of the superconductor and the thickness of the superconducting film. The fast relaxation time (t_1) is attributed to the electronic transitions, which is the combination of two process – (i) relaxation of hot quasiparticles by breaking more Cooper pairs in the vicinity and (ii) electron-phonon (e-ph) relaxation, while the slow relaxation time (t_2) is attributed to Cooper pair formation mediated by 2Δ phonons, where 2Δ is the superconducting energy gap. In the case of very thin superconducting films, most of the Cooper pairs are broken by direct excitation. Thus the fast relaxation process is dominated by *e-ph* relaxation rather than by breaking more Cooper pairs in the vicinity due to the absence of significant number of remnant Cooper pairs. Since electron-phonon relaxation is a slower process than relaxation of hot quasiparticles by breaking more Cooper pairs, thus the thinner superconductor films result in the higher values of fast relaxation time constant. The slow relaxation is determined by Cooper pair formation mediated by 2Δ phonons where 2Δ

phonon dissipates mainly by diffusion into substrates. However, in the case of very thin films, 2Δ phonons need to travel a small distance before reaching to the substrate and thus makes the Cooper pair formation fast. Therefore, due to enhancement in the fast time constant (t_1) and reduction in slow time constant (t_2), we observe almost identical time constants for the primary dissociation. However, in the case of secondary dissociation we observe slightly higher t_2 due to phonon bottleneck effect. The slow relaxation process determines the overall switching speed. The bi-exponential fitting reveals that the switching speed between quantum mechanical superconducting phase to the resistive phase to be around 16 ps. Hence, ultrathin superconductors form an ideal platform for low threshold, ultrafast switching applications.

6.3 Ultrathin superconducting metamaterials

6.3.1 Metamaterial design and fabrication

We systematically probe the ultrathin superconducting metamaterial devices to explicitly demonstrate low threshold switching for photonic applications at terahertz frequencies. We Choose a double gap terahertz asymmetric split-ring resonators (TASR) as schematically shown in Figure 6.3(a), where asymmetry is introduced by moving upper capacitive split gap away from the central vertical axis by a distance ' d ' called asymmetry parameter. Here we choose a high asymmetry of $d = 15 \mu\text{m}$ to get a resonance feature of high amplitude for the ease in measurement. Such TASR structures result in an asymmetric line shape Fano resonance when excited with the incident terahertz wave polarized perpendicular to the gap-bearing arm of the resonators.^{130,171} The designed samples are fabricated using conventional UV-illumination based photolithography technique. The superconducting samples (YBCO-

TASR) are fabricated by wet etching of lithographically patterned YBCO films (thickness 25, 50 and 200 nm) deposited over r-cut sapphire substrates. The small portion of fabricated

Figure 6.3: a) Schematic view of the superconducting TASR sample and OPTP measurement. b) Optical microscopic image of the fabricated TASR sample. The inset depicts one unit cell with geometrical dimensions: periodicity $p = 75 \mu\text{m}$, arm length $l = 60 \mu\text{m}$, width $w = 6 \mu\text{m}$, gap $g = 3 \mu\text{m}$ and asymmetry $d = 15 \mu\text{m}$. c) Transmitted terahertz time-domain pulses with (blue) and without (red) optical pump at 7K.

25 nm YBCO-TASR is shown in Figure 6.3(b). The unit cell geometrical dimensions are revealed in the inset of Figure 6.3(b). Identical metallic samples of different thicknesses are fabricated using lift-off method of photolithography and thermal evaporation of Aluminum (Al) metal. The detailed fabrication process is given in section 2.2. The transmission spectra of the TASR samples is measured using Zn-Te based terahertz spectroscopy setup. Figure 6.3(c) depicts the terahertz time-domain pulse passing through 25 nm YBCO TASR at 7 K in

the absence and under the illumination of the optical pump of fluence $76.2 \mu\text{J}/\text{cm}^2$, respectively.

6.3.2 Comparison of metallic and superconducting metamaterials

To get insights into the thickness dependent resonance features of the TASR structures, we fabricated and measured transmission spectra of Al-TASR of thicknesses 25, 50 and 200 nm as depicted in Figure 6.4. Figure 6.4 (a), (c) depicts that the Al-TASR of thickness 25 and 50 nm does not support excitation of Fano resonance mode, whereas dipole resonance mode is extremely faint. However, on increasing Al thickness to 200 nm, we observe well evolved Fano (0.49 THz) and dipole (0.71 THz) resonances as shown in Figure 6.4(e). Contrary to the metallic structures, YBCO-TASRs of thickness as low as 25 nm support excitation of strong Fano and dipole resonances as shown in Figure 6.4(b). On increasing the thickness to 50 and 200 nm, the resonances modes blue shifted. The blue shift in the resonance modes on increasing thicknesses of YBCO is due to decreased kinetic inductance of the YBCO structures. For sub-London penetration depth thick superconductors, kinetic inductance scales inversely with the film thickness and thereby provide an additional control parameter to actively tune the resonance behavior.³⁴ Kinetic inductance enhancement is highly useful in the photonic devices for communication.³⁴ Although Drude metals (e. g. Gold, Silver, and Aluminum) are efficient conductors and widely used for micro-structuring to achieve man-made features at terahertz frequencies, the sub skin-depth metallic layers show significantly reduced conductivities due to the large surface and grain boundary defects in comparison to the bulk state.¹³⁵ The skin depth of a conductor is defined as the penetration length of electromagnetic radiation at which it decreases to $1/e$ of its value at the surface when impinging at normal incidence. The skin depth of metallic conductors is given by

Figure 6.4: Experimentally measured transmission spectra of Al-TASR and YBCO-TASR samples. a), c) and e) represents the amplitude transmission spectra of metallic Al-TASR samples, while b), d) and f) depicts amplitude transmission spectra of YBCO-TASR samples of thicknesses 25, 50 and 200 nm, respectively. F and D represent Fano and dipole resonance mode.

$\delta = \frac{1}{\sqrt{\pi f \sigma_{dc} \mu_0}}$, where f is the frequency of electromagnetic radiation at which the skin depth is calculated, σ_{dc} is the DC conductivity of the metal and μ_0 is the vacuum permeability.¹³⁵

When the thickness of the metallic thin film is lower than 2δ , the surface resistance of the metallic structure increases gradually with decreasing film thickness and is given by $R = \frac{\rho l'}{wt}$, where ρ is the resistivity of the material, l' is the effective length of the TASR arms, w is the

Figure 6.5: Simulated amplitude transmission spectra of TASR samples of thickness a) 25 nm and b) 50 nm with varying material conductivity.

width of arm and t is the thickness of the metallic arm.¹⁶⁷ The skin depth of Al metal at Fano resonance frequency (0.5 THz) is about 115 nm.¹³⁶ The reduced conductivity and high surface resistance of the ultrathin metallic films drastically affect the optical response and are responsible for the absence of Fano resonance and faint dipole resonance at 25 and 50 nm thick Al-TASR. To gain more physical understanding about the impact of sub-skin depth thicknesses of the metallic TASR structures on its optical response, we simulated TASR metamaterial design with identical geometric dimensions as given in Figure 6.3, using a

general material of thickness 25 and 50 nm and varied its conductivity. A sapphire substrate with dielectric constant of 11.7 is used as the terahertz transparent substrate in the numerical simulations. The detailed simulation process is given in the section 2.1. The simulated transmission spectra of TASR of thicknesses 25 and 50 nm and varying conductivity of resonator material is shown in Figure 6.5 (a) and (b), respectively. We observed that the minimum critical DC conductivity required to support excitation of the Fano resonance mode for 25 nm and 50 nm thick structures are 1.3×10^7 and 7×10^6 S/m, respectively. The corresponding surface resistance is about 100 Ω for both the thicknesses. Thus, a TASR structure supports excitation of sharp Fano resonance only if its surface resistance is lower than 100 Ω . Hence at lower thicknesses of the metal films, the minimum critical conductivity to support Fano resonance is significantly higher due to the higher surface resistance. Thermally deposited sub-skin depth thick metallic films show reduced conductivity and hence fail to support excitation of sharp Fano resonances.

6.4 Equivalent circuit model of Fano resonant metamaterials

To better understand the resonance response of TASR metamaterial, we considered equivalent circuit model based on the transmission line assumption.¹⁷² The resonance frequency (ω_0) of individual TASR strip is determined by the effective capacitance (C), inductance (L) and resistance (R) of that strip, and is given by¹⁶⁷

$$\omega_o^2 = \frac{1}{LC} - \frac{R^2}{4L^2} \quad 6.2$$

The geometry of the TASR structure is quite complex and consist of two unequal arms of length l_1 and l_2 . The difference in the length of the two wires plays an important role in determining the resonance response of the TASR structure. When both the arms of the

resonator are of equal in length, the Fano resonance excitation is forbidden, and hence it only results in a broad dipolar resonance mode. Upon introducing asymmetry by moving one of the capacitive gaps away from the center vertical axis by a distance d , which ultimately changes the length of the two arms and give rise to Fano mode along with broad dipole resonance mode.

The excitation of these resonance features could be understood in terms of effective resistance (R_d , R_f), inductance (L_d , L_f) and capacitance (C_d , C_f) of each arm of TASR (see Figure 6.6 (a)). The term R_d and R_f represent the resistance of short arm and long arm, respectively. Resistance R_d and R_f are calculated using the DC conductivity of the material and effective geometric parameters using the expression $R = \frac{\rho l'}{wt}$, where ρ is the resistivity of the material, l' is the effective length of the TASR arm, w is the width of arm and t is the thickness of the resonator arm. For symmetric resonator, L_{gs} represents the geometric inductance of the one arm, while ΔL represents change in geometric inductance because of the asymmetry parameter d . Therefore, $L_{gs}-\Delta L$ and $L_{gs}+\Delta L$ shows the geometrical inductance of shorter and longer strip of the asymmetric resonator (TASR). The geometric inductance of a strip of length l' , width w and thickness t is calculated using equivalent wire loop as $L_{l'} = \frac{\mu_0 l'}{8\pi} + \frac{\mu_0 l'}{2\pi} \left(\log \left(\frac{4l'}{\sqrt{\pi w t}} \right) - 1.75 \right)$. During electromagnetic excitation, the effective inductance of individual strip is further influenced by the near-field coupling of each TASR strip with neighboring strips. When $d=0$, the impedance matches for both strips, which results in parallel current distribution. Because of parallel current distribution, the effective inductance of both strips decreases by same magnitude L_{cs} . As asymmetry is introduced ($d \neq 0$), due to a mismatch in geometric impedance a mismatch also arises in the mutual coupling. This additional mismatch in coupling is considered as L_m , which reduces the

overall inductance of the smaller arm, while it enhances the inductance of the long arm. Considering all these effects, we get effective inductance of shorter (L_d) and longer (L_f) arm. L_{k1} and L_{k2} terms correspond to the kinetic inductance of shorter and longer arm, respectively, only applicable to superconducting TASR.

Similarly, the effective capacitance (C_d , C_f) is obtained by estimating the capacitance of two parallel plate capacitors formed in the TASR structure. C_{gs} is considered to be geometric capacitance with respect to each arm when the resonator is symmetric ($d=0$) and calculated using the capacitance of parallel plate capacitor of area A and width g , $C = \frac{\epsilon_0 \epsilon_r A}{g}$, where ϵ_0 is the permittivity of the vacuum and ϵ_r is the permittivity of the material present in the capacitor. Here the capacitance of the gaps is enhanced due to the substrate in the bottom of the TASR array. The effective capacitance is also influenced by the coupling due to neighboring resonators. We considered two capacitive coupling terms C_{cs} and C_m due to symmetric ($d=0$) and asymmetric ($d \neq 0$) nature of TASR. The coupling term C_{cs} enhances the capacitance of both arms, while coupling term C_m enhances the capacitance of the smaller arm and reduces the capacitance of the longer arm. With these equivalent circuit parameters of the individual strip, Fano and dipole resonant modes are observed in transmittance spectra (defined as the ratio of P_{out}/P_{in}) with respect to incoming radiation. P_{in} and P_{out} represents the input and out powers. In the equivalent circuit terminal '1' and '2' represents incident and transmission ports, respectively. The surface current for these two modes are shown in Figure 6.6(b). The surface currents form a close-loop along TASR perimeter at Fano resonance mode (see Figure 6.6(b)). Thus the energy stored in the TASR is magnetic in nature. However, surface currents are parallel to each other in two arms of TASR at dipole resonance mode (see Figure 6.6(b)) and thus reduces magnetic energy significantly. At dipole

resonance, shorter arm resonates strongly, while at Fano resonance longer arm resonates strongly.

The transmission spectra of the Al-TASR is obtained from the equivalent circuit model by using effective resistance, effective geometric inductance, the effective capacitance of the resonator. By varying the inductive and capacitive coupling parameters, we achieved the transmission curves identical to the measured transmission spectra for various thicknesses of Al metal, as shown in Figure 6.6(c). We observe that the coupling parameters also play a vital role in determining Fano as well as dipole resonance frequencies. Calculated circuit parameters for Al-TASR of thickness 25, 50 and 200 nm using geometry and fitting of the transmission spectra are listed in Table 6.1.

The resonance response of the YBCO-TASR is also influenced by another inductance term which arises due to the kinetic inductance of the superconductors. The kinetic inductance of a thin superconducting strip of the thickness t , length l and width w are given as $L_K = \frac{\mu_0 \lambda_L^2 l}{wt}$, where μ_0 is the vacuum permeability, λ_L is the London penetration depth, respectively.³⁴⁻³⁶ However, for the superconducting strips of thickness higher than the London ($t > \lambda_L$), the kinetic inductance is given by $L_K = \frac{\mu_0 \lambda_L l}{w}$. The calculated kinetic inductance of the short and long arm of the YBCO-TASR for thicknesses 25, 50 and 200 nm is given in Table 6.2. The kinetic inductance increases with decreasing thickness of the superconducting strip. After accounting for the contribution from the kinetic inductance (L_{k1} , L_{k2}) and keeping all other circuit parameters identical to the Al-TASR of different thickness, the calculated transmission spectra for YBCO-TASR is shown in Figure 6.6 (d). The kinetic inductance effectively enhances the inductance of both the arms of the TASR and causes red shift in the Fano and dipole resonances. YBCO-TASR of thickness 25 nm shows highest spectral red

shifting followed by 50 nm and 200 nm thick YBCO structures. The calculated transmission of YBCO-TASR is in good agreement with the measured transmission spectra of the sample with identical thickness. The high kinetic inductance of the ultra-low thicknesses of YBCO open up new pathways of inductance enhanced resonant devices and cavities for high-speed communication applications.

Figure 6. 6: (a) The schematic of the equivalent transmission line circuit model of the TASR. Inset figure shows the unit cell of the asymmetric metamaterial. R_d and R_f represent the resistance of the arms l_1 and l_2 of the resonator. L_d , C_d , and L_f , C_f is the effective inductance and capacitance of the shorter and longer arm of TASR. b) Surface current distribution in the asymmetrically split ring at Fano and dipole resonance frequencies. (c) and (d) depicts the

transmission spectra of Al-TASR and YBCO-TASR, respectively, of thickness 25, 50 and 200 nm obtained using equivalent transmission line circuit model.

Table 6.1: Circuit parameters of Al-TASR calculated using geometry and fitting of the transmission spectra

Thick ness (nm)	Resistance (ohm)		Inductance (pH)						Capacitance (fF)				
	R _d	R _f	L _{gs}	ΔL_{gs}	L _{cs}	L _m	L _d	L _f	C _{gs}	C _{cs}	C _m	C _d	C _f
25	15	22.5	33	4.7	17	3	8.3	23.7	2.4	3.5	1	6.9	4.9
50	8	11.7	30	4.3	15.9	3	6.8	21.4	2.9	3.5	1.8	8.2	4.2
200	2.1	2.8	24	3.7	12.3	3	5	18.4	3.9	3.5	2	9.4	5.4

Table 6.2: Circuit parameters of YBCO-TASR calculated using geometry and fitting of the transmission spectra

Thickn ess (nm)	Resistanc e (ohm)		Inductance (pH)								Capacitance (fF)				
	R _d	R _f	L _{gs}	ΔL_{gs}	L _{cs}	L _m	L _{k1}	L _{k2}	L _d	L _f	C _{gs}	C _{cs}	C _m	C _d	C _f
25	3	4.5	33	4.7	17	3	18	42	26.3	65.7	2.4	3.5	1	6.9	4.9
50	2	2.7	30	4.3	15.9	3	3	15	9.8	34.4	2.9	3.5	1.8	8.2	4.2
200	1.1	1.8	24	3.7	12.3	3	1.45	1.95	6.45	20.4	3.9	3.5	2	9.4	5.4

6.5 Low threshold switching and modulation of ultrathin metamaterials

In addition to the inductance enhanced resonance features of the ultra-thin YBCO metamaterial structures which provide large spectral modulation, they also provide an opportunity for low threshold switching behavior under the influence of external

Figure 6.7: a) The measured transmission spectra of ultrathin YBCO-TASR of thickness 25 nm measured at varying optical pump fluences. b) The degree of amplitude modulation in percentage at varying pump fluences for YBCO TASR of thicknesses 25, 50 and 200 nm. c) Charge carrier dynamics of the 25 nm thick YBCO-TASR measured for an optical pump fluence of 76.20 $\mu\text{J}/\text{cm}^2$.

perturbations. To demonstrate ultra-low threshold switching of the resonance features in YBCO-TASR metamaterial, terahertz transmission spectra under the influence of photoexcitation were measured by an OPTP setup. The time delay between the optical pump and terahertz probe was set to be 5.2 ps, which is the point of maximum dissociation of Cooper pairs in its dissociation-relaxation dynamics. On irradiating the YBCO-TASR sample with a low pump fluence of $19.05 \mu\text{J}/\text{cm}^2$, a faint modulation of the Fano and dipole resonance is observed (see Figure 6.7 (a)). On increasing the fluence of optical pump beam gradually, the modulation of Fano and dipole resonances increases. The modulation in the resonance features is due to the change in the imaginary part of the complex conductivity of the YBCO which arises due to the dissociation of a large number of the Cooper pairs. From Figure 6.7(a), it could be observed that at an optical pump fluence of $76.20 \mu\text{J}/\text{cm}^2$, the Fano and dipole resonances become extremely faint. Figure 6.7(b) shows the comparison of the amplitude modulation of the Fano resonance in YBCO-TASR structures of thicknesses 25, 50 and 200 nm with varying fluences of the incident optical pump beam. The pump dependent modulation of Fano resonance was calculated as $\frac{|T_{nopump} - T_{pump}|}{T_{pump}} \times 100\%$, where T_{nopump} and T_{pump} , respectively, stands for peak to dip amplitude of the Fano resonance during "off" and "on" states of the optical pump. The observed maximum value of Fano resonance modulation for 25, 50 and 200 nm thick YBCO-TASR are about 95, 86 and 70% at optical pump fluences of 76, 955 and $1529 \mu\text{J}/\text{cm}^2$ respectively. Therefore, the TASR structures with reduced YBCO thickness show relatively large modulation at extremely low fluences of optical pump. Low threshold switching of optical response is highly desirable for multifunctional applications such as biological sensing as high power optical pulse could damage the analyte and change its properties. Moreover, we also capitalize on the ultrafast response observed in the Cooper pair dissociation-relaxation dynamics of YBCO to

demonstrate ultrafast switching of the Fano and dipole resonances of YBCO-TASR. The sudden dramatic reduction in the imaginary part of the conductivity of the YBCO upon photoexcitation is reflected in the resonant transmission spectra of YBCO-TASR sample. Figure 6.7(c) depicts the Cooper pair dynamics of the 25 nm thick YBCO-TASR sample irradiated with the optical pump of fluence of $76.2 \mu\text{J}/\text{cm}^2$ measured at 5 K. Figure 6.7(d) shows the measured transmission spectra of the YBCO-TASR at varying time delay between the optical pump and terahertz probe marked as A, B, C, and D as depicted in Figure 6.7(c). The pink curve in Figure 6.7(d) represents the terahertz transmission spectra of YBCO-TASR before the arrival of the optical pump pulse at point A (-1.0 ps) thus results in very strong Fano and Dipole resonances at 0.27 THz and 0.39 THz, respectively. On increasing the time delay between the optical pump and THz probe to be 5.2 ps at point B, the Fano and dipole resonance amplitude becomes extremely faint. This large modulation of Fano and dipole resonances is attributed to the maximum dissociation of the Cooper pairs at point B. The dissociated quasiparticles bind together again to form a Cooper pair in picoseconds timescale and thereby restoring the superconductivity which also results in a gradual strengthening of the Fano and dipole resonances at C (9.1 ps) and D (17.4 ps). However, at a time delay of around 23 ps, a reflected optical pulse from the rear surface of the substrate hits the sample and break the Cooper pair again as shown by the arrow in Figure 6.7(c). This secondary dissociation by the reflected pulse leads to a dual-channel switch which has large technological importance. Here, it is important to note that the resonances are not completely restored due to the phonon bottleneck effect, but it would be completely restored after switching off the incident optical pump. Overall, we observed that the ultra-thin YBCO-TASR structures show an extremely low threshold, ultrafast switching of the metamaterial devices.

6.6 Conclusion

In summary, we have experimentally demonstrated that the ultrathin high T_c superconductors provide an ideal platform for extremely low threshold switching applications. Sub-London penetration depth thick YBCO metamaterial structures support excitation of sharp Fano resonances, while identical sub-skin depth thick metallic metamaterial structures fail to support Fano resonance mode. Furthermore, we developed an equivalent circuit model based on transmission line assumption, which provides a physical interpretation of persistence and spectral position of Fano and dipole resonances in ultrathin superconducting asymmetric metamaterials. We demonstrate an interesting approach to design subwavelength ultrathin superconducting photonic devices with extremely loss-less features to accomplish ultrafast, dual-channel switching and modulation. These ultrathin photonic devices could enable interesting applications such as superconducting radiation sensors, terahertz high-speed wireless communication devices, time-division multiplexing, superconducting photodetectors, and nonlinear metamaterials.

Chapter 7

Meissner Effect in a Coupled Metamaterial System at Terahertz Frequencies

This chapter introduces a new type of metamaterial system which shows the existence of Meissner–Ochsenfeld or Meissner effect at terahertz frequencies. We report an ultrafast quantum level switching between flux exclusion and flux penetration regime in the optical response of the hybrid metal-superconductor metamaterial system.

7.1 Introduction

In the previous chapters, we have shown various phenomena such as ultra-high- Q resonances, ultrafast dual channel metamaterials switch, and ultrathin metamaterials based on the extraordinary conducting properties of high T_c superconductors. Along with the incredible features brought about by the extraordinary conducting properties (electrical effects) of superconductors, we also probe quantum phenomenon arising from the perfect diamagnetism property of the superconductors. The perfect diamagnetism property of the superconductor, in which an applied magnetic field is expelled from the core of the superconductor, is called Meissner effect or Meissner–Ochsenfeld effect.^{2,9} The Meissner effect arises due to the generation of a screening current at the surface of the superconductor. These currents flow within a characteristic length scale called London penetration depth (λ).^{2,9} London penetration depth is defined as a length scale at which the magnetic field decays to $1/e$ of its value at the surface of the superconductor. Meissner effect phenomenon of the superconductor also distinguishes it from perfect electrical conductors. Perfect

electrical conductors prevent penetration of externally applied magnetic field to the core of the material but retain the magnetic field already penetrated inside the body of the material. In contrast to perfect electrical conductors, superconductors not only prevent penetration of externally applied magnetic field but also expels any magnetic field already present in the core of the superconducting material. A detailed discussion about Meissner effect is given in section 1.1.

The response of a superconductor to an AC field is described by two-fluid model of the superconductivity where an AC current is carried out by Cooper pairs as well as the normal electrons.^{2,3} The detailed discussion about the two-fluid model is given in section 1.4. According to the two-fluid model, the superconducting behavior is dominated by the imaginary part of the conductivity which is much larger than the real part of the conductivity. However, the imaginary part of the conductivity decreases inversely with increasing frequency of operation ($\sigma_{img} \propto \frac{1}{\omega}$).^{2,3,168} Thus, at terahertz frequencies, high- T_c superconductors show significant amount of losses due to suppressed imaginary part of the conductivity.

Therefore, at higher frequencies, the response of the superconductor is primarily inductive, resistive and diamagnetic.⁷⁸ The inductive behavior arises due to the generation of screening currents which maintain the Meissner effect in the core of the superconductor. The London penetration depth in which these screening currents flow is given by

$$\lambda^2 = \frac{m}{n_s \mu_0 e^2} \quad \dots \dots \dots 7.1$$

where n_s is the superfluid density, e is the electronic charge, and μ_0 is the vacuum permeability. Thus with decreasing Cooper pair density, λ increases which allow penetration

of a magnetic field to a larger depth. The penetration depth diverges as n_s approaches to zero and in this state magnetic flux penetrates into the superconductor. Also at higher frequencies, the magnetic field can only penetrate via local suppression of the superconductivity as the velocity of the Abrikosov vortices propagation is quite low and it requires a long time to penetrate through the superconductor material in comparison to a single cycle of a high-frequency wave in microwave and terahertz frequency regime.^{173,174} Therefore, the behavior of high- T_c superconductors on the application of a high-frequency magnetic field need to be explored.

In this chapter, we envisioned and experimentally demonstrate the existence of Meissner effect in high-temperature yttrium barium copper oxide (YBCO) superconductor at terahertz frequencies. We employed a metamaterial approach to generate an oscillating pulsed magnetic field and studied its response on a high- T_c superconductor in terms of the optical response of the metamaterial device. The high-frequency flux quantization could be a source of non-linearity in metamaterial and plasmonic devices. Persistence of flux exclusion at higher frequencies has a broad range of implications on the quantum metamaterial and photonic devices operating at microwave and terahertz frequencies. This quantum metamaterial approach does not require for a Josephson junction in its geometry thereby making it much simpler to fabricate it at larger scale.^{111,175} Such quantum metamaterials based on the flux exclusion and flux penetration could find interesting applications for dynamic and efficient modulation of microwave and terahertz radiation.

7.2 Flux quantization phenomenon

Flux quantization phenomenon arises due to the coherent behavior of the Cooper pairs in the macroscopic scale and suggest that the magnetic flux enclosed in a superconducting ring

must be an integer multiple of the flux quantum given by $\phi_0 = \frac{h}{2e} \approx 2.1 \times 10^{-15} \text{W}_b$.^{2,110}

Since its first demonstration by Deaver and Fairbank, the flux quantization is widely accepted as one of the characteristic features of the superconducting state.^{176,177} Flux quantization also forms the basic operation principle for SQUID devices on combining with the Josephson junction and thus became an integral component for numerous applications such as quantum computing, superconductor electronics and high precision magnetometers.^{178,179}

Ginsburg-Landau theory suggests that the Cooper pairs at a position r is described by a single-valued complex wave function given as^{2,3}

$$\psi(r) = \sqrt{n_s(r)} e^{i\phi(r)} \quad \dots \dots \dots 7.2$$

where $n_s(r)$ is the Cooper pair density at point r , and $\phi(r)$ represents the phase of the macroscopic wavefunction. The super current density at point r resulted by Cooper pairs can be expressed as

$$J_s(r) = \frac{2en_s(r)}{m} \left(\frac{h}{2\pi} \nabla\phi - 2eA \right) \quad \dots \dots \dots 7.3$$

where $2e$ is the charge and m is the mass of a Cooper pair, h is the Planks constant, and A is the vector potential due to the magnetic field.

While going around a closed loop, the phase of the wave function can only change by multiples of 2π , therefore integral of the gradient of phase along the superconducting loop results in

$$\oint dl \cdot \hat{l} \nabla\phi = 2\pi k, \quad n = 0, \pm 1, \pm 2$$

Figure 7.1: Magnetic flux threading through a superconducting ring

At sufficiently small magnetic fields and deep inside the body of the superconductor, super current density would be negligible. Thus on integrating J_s along closed superconducting loop will therefore results in

$$\oint dl \cdot \hat{l} J_s = \frac{2en_s(r)}{m} \left(\frac{h}{2\pi} (2\pi k) - 2e \oint dl \cdot \hat{l} A \right) = 0 \quad \dots \dots 7.4$$

On using $\oint dl \cdot \hat{l} A = \phi$, equation 7.3 results in quantized flux, i.e. $\phi = 0, \pm\phi_0, \pm2\phi_0 \dots$, where $\phi = \frac{h}{2e} = 2.1 \times 10^{-15} \text{ W}_b$. Thus the magnetic flux threading through a superconducting loop is quantized. Flux quantization phenomenon is a material independent property of the superconductors and provide a direct relation between electromagnetic intensity and geometry of the closed loop.

7.3 Experiment design

To study the existence of Meissner–Ochsenfeld effect at terahertz frequencies, we employed high frequency pulsed magnetic field generated by subwavelength metamaterial structures. For its easiest implementation, we considered the simplest split ring resonator (SRR) as a building block for the two-dimensional metamaterial array as shown in Figure 7.2(a). Upon excitation with an incoming electromagnetic wave parallel to the split gap of the SRR, they

Figure 7.2: (a) Unit cell of the circular terahertz split-ring resonator array. (b) Simulated amplitude transmission spectra of the SRR. Surface current (c) and (d) magnetic field distribution at the dip of LC resonance.

result in a LC resonance.^{160,161} The geometric dimensions are optimized to get a resonance response at about 0.5 THz (see Figure 7.2 (b)). To gain more understanding about the nature of the resonance mode, we simulated the surface current distribution and magnetic field distribution as shown in Figure 7.2 (c) and (d). The surface currents are forming a closed current loop along the SRR perimeter and give rise to the inductance L , split gaps acts like a capacitor and results in the capacitance C . However, the surface currents are highly concentrated at the lower part (opposite to the split gap) of the ring which gives rise to highly concentrated time-varying magnetic field in the vicinity of the lower part of the SRR. Figure 7.2 (d) depicts the presence of large magnetic fields confined in the SRR unit cell. The strength of the oscillating magnetic field in the SRR is governed by the geometry of SRR and the intensity of the excitation wave. We employed time-varying metamaterial magnetic fields at terahertz frequency to design and study a Flux exclusion metamaterial system. For the simplest realization of quantum phenomena of flux exclusion and flux penetration, we enclose a high- T_c superconductor (YBCO) in the resonant SRR structure and systematically probed its interaction with the metamaterial magnetic fields. We also enclose a normal conductor (metal) in the resonant SRR structure to distinguish the effect of the interaction of metamaterial magnetic field to a normal conductor and a superconductor. The geometrical dimensions of the enclosed disk are chosen such that it is non-resonant in the vicinity of the LC resonance mode of the SRR. Thus, in the present case, the resonance response is derived from the outer SRR only. The change in resonance response occurs due to the interaction of the generated oscillating magnetic flux with the centered disk. Based on the simulated design, we fabricated experimental samples using conventional photolithography technique. A r -cut sapphire substrate was selected as a terahertz transparent substrate which also provides better thermal conductivity during cryogenic measurements. The resonant SRRs are

Figure 7.3: Optical microscopy images containing four unit cells of the fabricated a) MR (metallic ring), b) MR-MD (metallic ring-metallic disk) and c) hybrid MR-SD (metal ring-superconducting disk) metamaterial array. Highlighted region reveals a unit cell with geometrical dimensions, ring radius $I_r = 20 \mu\text{m}$; disk radius $I_d = 13 \mu\text{m}$, split gap $g = 3 \mu\text{m}$, resonator width $w = 6 \mu\text{m}$, spacing between ring disk $d = 3 \mu\text{m}$ and the periodicity $p = 48 \mu\text{m}$.

fabricated using aluminum metals in order to get a strong resonance response and eliminate the effect of kinetic inductance. Metals show high scattering losses thus does not give rise to the kinetic inductance. The metallic patterns are formed using 200 nm thick Al metal while YBCO disks are formed using wet chemical etching of the 100 nm thick YBCO film deposited over 0.5 mm thick r-cut sapphire substrate. The detailed fabrication process is given in section 2.2 and, the optical microscopy images of the fabricated samples are shown in Figure 7.3. Overall we designed and fabricated three metamaterials namely a metallic SRR depicted as MR, a metallic SRR enclosing a metallic disk shown as MR-MD and a metallic SRR enclosing a superconducting disk shown as MR-SD, as presented in Figure 7.3 (a), (b), and (c), respectively. Inset in Figure 7.3(b) reveals the geometrical definitions of the unit cell. The separation between the SRR and centered disk was selected minimum ($3 \mu\text{m}$) in order to

get a strong interaction between metamaterial magnetic field and the centered disk. These samples are characterized using a ZnTe nonlinear crystal based OPTP setup. The detailed description of the setup is given in section 2.3. An open cycle continuous liquid helium flow cryostat was incorporated in the measurement setup to allow cooling down the samples to the cryogenic temperatures (5 K). Since the electric conductivity of ultrathin metals does not change much upon cooling to cryogenic temperatures, we characterized metallic samples (MR and MR-MD) at room temperature.^{135,157}

7.4 Flux exclusion and Flux penetration in coupled metamaterial system

The proposed electromagnetic functionality of the quantum metamaterial system is realized using terahertz transmission response of the above described samples. The dashed (black) and solid (green) curve in Figure 7.4(a) depicts the measured amplitude transmission spectra of the MR and MR-MD metamaterials, respectively. It is observed that on enclosing a metallic disk in the metallic ring structure results in the slight reduction in amplitude of the LC resonance mode, while no spectral shifting is observed. This state is called flux penetration state where the magnetic flux generated by the MR metamaterial easily penetrates through enclosed metallic disk. The schematic of the flux penetration scheme in MR-MD metamaterial is shown in the inset of the Figure 7.4(a). This slight reduction in the resonance amplitude is attributed to the modification in the stored magnetic energy of the MR metamaterial at the LC resonance.

We studied the coupling mechanism on enclosing a metallic disk in MR metamaterial using numerical simulations as shown in Figure 7.5. Figure 7.5 a) shows the simulated amplitude

transmission spectra of the MR metamaterial. The resonance frequency (f) of such LC resonators are given by $f \propto \frac{1}{\sqrt{L_R C_R}}$, where L_R and C_R are the inductance and capacitance of

Figure 7.4: Flux penetration and flux exclusion states of the quantum metamaterial. a) Measured amplitude transmission spectra of a MR (metallic ring) and a MR-MD (metallic ring-metallic disk) metamaterial. b) Measured amplitude transmission spectra of a MR (metallic ring) and MR-SD (metallic ring-superconducting disk) metamaterial. Inset in a) and b) show schematic of the side view of the magnetic field distribution in flux penetration and flux exclusion state, respectively.

Figure 7.5: Numerically simulated transmission spectra of the a) MR metamaterial, b) lower half of the metallic disk enclosed in a MR metamaterial c) upper half of the metallic disk enclosed in a MR metamaterial and d) whole metal disk enclosed in MR metamaterial. Inset figures depict the corresponding metamaterial design. L_R , C_R represents the ring inductance and capacitance while L_C and C_C represent the coupling inductance and capacitance, respectively, due to the presence of metallic disk.

the MR metamaterial, respectively. To differentiate the effect of capacitive and inductive coupling because of the enclosed metallic disk in a MR metamaterial, we studied the influence of upper and lower half of the disk enclosure separately. Figure 7.5 (b) depicts the simulated transmission spectra of MR metamaterial in the presence of lower half of the metallic disk. It is observed that enclosed lower half of the metallic disk in MR metamaterial lead to blue spectral shifting of the LC resonance mode. Since the circulating currents in the MR metamaterial are concentrated in the lower part (see Figure 7.2 (c)) thus they strongly couple inductively to the lower half of the enclosed metallic disk and results in blue spectral shifting of LC resonance mode. This blue spectral shifting of LC resonance mode is attributed to the reduction in the inductance due to negative mutual coupling between the metallic disk and MR metamaterial. Therefore, the resonance frequency (f) of LC resonance mode in this system is given by $f \propto \frac{1}{\sqrt{(L_R - L_c)C_R}}$, where L_c is the inductance arising due to the coupling between the metallic disk and MR metamaterial. Figure 7.5 (c) shows the simulated transmission spectra of MR metamaterial enclosed by upper half of the metallic disk. This metallic disk is close to the capacitive split gap of the MR metamaterial where strong electric fields are concentrated which capacitively couples to the metallic disk, thereby resulting in the red spectral shifting. The resonance frequency (f) of LC resonance mode in the presence of upper half of metallic disk is given by $f \propto \frac{1}{\sqrt{(C_R + C_c)L_R}}$, where C_c is the capacitance arising due to the coupling between upper half of the metallic disk and MR metamaterial. When entire metallic disk is enclosed in the MR metamaterial, negligible spectral shift is observed in LC resonance mode as shown in Figure 7.5(d). Here both inductive and capacitive coupling occurs, however, the net effect of both these mechanism results in very minute spectral shifting of the LC resonance.

In contrast to the modification in optical response upon inserting a metallic disk in the MR metamaterial, inserting a superconducting disk results in the different resonance response. A high- T_c superconductor acts like a diamagnetic material (due to the Meissner effect) below their transition temperatures and thereby prevent the penetration of magnetic flux generated by the MR metamaterial. This effect of the magnetic flux exclusion is measured in the form of the change in optical response of the metamaterial system. The dashed (black) and solid (red) curve in Figure 7.4(b) represent the measured amplitude transmission spectra of the MR and MR-SD metamaterials, respectively. Here, the transmission spectra of MR-SD metamaterial sample is measured at 5K. When a superconducting disk (at 5K) is enclosed in the MR metamaterial, it red shifts the LC resonance mode. This red shifting in the LC resonance mode is attributed to the magnetic flux exclusion by the superconductor, and this state of the metamaterial system is called flux exclusion.¹¹⁰ The schematic of the flux exclusion by a superconducting disk enclosed in a metallic ring is shown in the inset of Figure 7.4(b). On applying a magnetic field, unlike metals, a superconducting material generates a shielding current at the surface (in the length scale λ) and thus results in very weak inductive coupling (due to flux exclusion) compared to the metallic disk. Therefore, in this regime, the capacitive coupling is dominant which gives rise to the large red shift. This peculiar behavior of the flux exclusion in superconductor leads to a quantum metamaterial system without the introduction of the Josephson junction.^{111,179}

7.5 Switching between flux exclusion and flux penetration

7.5.1 Thermal switching

To enable the active control of the metal-superconductor hybrid (MR-SD) metamaterial system, we measured amplitude transmission spectra at varying temperatures (ranging from

Figure 7.6: Measured amplitude transmission spectra of MR-SD metamaterial at various temperatures ranging from 5 K to 295 K depicting switching between flux exclusion and flux penetration state.

5K - 293K) as shown in Figure 7.6. We observed that on increasing the temperature, the LC resonance mode of MR-SD metamaterial shows red spectral shift. At the lowest temperature of 5K, MR-SD is in the flux exclusion regime because the superconducting disk opposes the penetration of magnetic flux generated by the MR metamaterial. This occurs due to the generation of the shielding currents at the surface of the superconductor. However, on increasing the temperature from 5 K to T_c , the shielding currents start flowing in a large cross-section area due to increase in the penetration depth of the magnetic flux. This phenomenon gives rise to the enhancement in the kinetic inductance of the superconducting disk, which increases the overall intrinsic inductance of the system that is reflected as the red shift in the LC resonance mode. However, above T_c , the disk undergoes a superconductor (YBCO) to dielectric transition which gives a large enhancement in the total capacitance of

the MR-SD metamaterial and therefore the LC resonance mode further shifts to lower frequencies. We marked this state as flux penetration state as the YBCO disk is not opposing magnetic field penetration. Thus the two resonance states of MR-SD metamaterial, i.e. flux exclusion and flux penetration regime could be thermally switched by varying the temperature.

7.5.2 Ultrafast optical switching

We further capitalize on the ultrasensitive nature of superconductivity to the energy of the incident photons above the binding energy of the Cooper pairs in YBCO superconductor (as discussed in section 5.3 and 5.4) to demonstrate ultrafast switching between flux exclusion and flux penetration states of the MR-SD metamaterial. When irradiated, a fraction of Cooper pairs get dissociated into quasiparticles and thus changes the superconducting properties.^{18,32} Figure 7.7 depicts the measured amplitude transmission spectra of the MR-SD metamaterial upon photo-excitation with varying pump fluence of wavelength 800 nm (1.55 eV) pulsed optical beam. In the optical pump dependent transmission measurements, the terahertz probe pulse was delayed by 5.7 ps with respect to the optical pump pulse in order to capture the change in the optical response, arises due to the optical pulse mediated dissociation of the Cooper pairs (discussed in section 5.3). Upon photoexcitation with a low fluence of $63.7 \mu\text{J}/\text{cm}^2$, the MR-SD metamaterial shows red spectral shift as shown in Figure 7.7. The optical pump pulse dissociates a fraction of Cooper pairs and thus reduces the density of the Cooper pairs. The reduced density of the Cooper pairs increases the kinetic inductance of the superconducting disk and allows penetration of the magnetic flux to a larger extent due to the increased London penetration depth. On increasing the optical pump fluence to $254.8 \mu\text{J}/\text{cm}^2$, the MR-SD metamaterial show further red spectral shift as shown by the green curve in Figure 7.7. The LC resonance mode of the photo-excited MR-SD

Figure 7.7: Measured amplitude transmission spectra of MR-SD metamaterial measured at various optical pump fluence depicting switching between flux exclusion and flux penetration state.

metamaterial with an optical pump fluence of $254.8 \mu\text{J}/\text{cm}^2$ (green curve) matches closely with flux penetration state observed during thermal tuning (black dotted curve). Therefore, the flux exclusion and flux penetration regime of the MR-SD metamaterial could be readily switched using low fluence of optical excitation.

Furthermore, the quasiparticles resulted due to Cooper pair split upon optical irradiation recombine again to form Cooper pairs on an extremely short timescale thereby restoring the superconductivity.^{18,32} This ultrafast switching between the superconducting and resistive phase of the YBCO results in the opportunity for high-speed switching applications in quantum metamaterials and photonics. Upon illumination with the optical pump pulse of energy greater than the superconducting gap energy, the Cooper pairs split into hot

Figure 7. 8: (a) Photoexcitation dynamics of the MR-SD metamaterial measured at optical pump fluence of $127 \mu\text{J}/\text{cm}^2$. b) Normalized charge carrier relaxation dynamics (blue curve) and its fitting (red curve) using bi using bi-exponential decay.

quasiparticles and give rise to large reduction in the conductivity. The hot quasiparticles resulted due to the splitting of Cooper pairs upon optical irradiation recombine again to form Cooper pairs. We recorded dramatic change in the conductivity of YBCO in the form of differential transmission ($\Delta T/T$) signal. Figure 7.8(a) depicts the Cooper pair dynamics of the MR-SD metamaterial measured at 5K under the illumination of the optical pump pulse of 800 nm wavelength with a fluence of $127 \mu\text{J}/\text{cm}^2$. Initially, $\Delta T/T$ is zero at a time delay of 0

ps which increases rapidly with increasing time delay and reaches to a maximum at a time delay of 5.7 ps due to utmost dissociation of the Cooper pairs. With further increase in the time delay, there is a sudden reduction in $\Delta T/T$ value, which saturates at higher time delays. We also observe the evolution of a secondary peak at a time delay of 12.9 ps (marked by a black arrow in 7.8 (a)) in the charge carrier dynamics, which arise due to the secondary dissociation of superconducting Cooper pairs by the reflected optical pulse from the rear surface of the substrate (see section 5.4.1). To estimate the restoration time of dissociated Cooper pairs in YBCO, we performed bi-exponential fitting of primary dissociation using the equation 6.1. The detailed mechanism of Cooper pair dissociation-relaxation upon photoexcitation and related time scales are described in section 1.4. The biexponential fitting of primary dissociation peak reveals fast and slow recovery time constant to be 0.9 and 15.8 ps as shown in Figure 7.8(b). The slow recovery time constant reflects the switching speed of the metamaterial device. Figure 7.8 also reveals that the Cooper pairs are not fully relaxed back due to the phonon bottleneck effect. Therefore using optical irradiation scheme, the flux penetration and flux exclusion regime of MR-SD metamaterial device could be switched on an ultrafast timescale.

7.6 Conclusion

In summary, we demonstrated a niche approach to realize Meissner–Ochsenfeld effect at terahertz frequencies in a quantum metamaterial system. We identified flux exclusion and flux penetration states of a metal-superconductor (Al-YBCO) hybrid quantum metamaterial system. Our results show an effective way of manipulating magnetic flux exclusion and penetration using thermal tuning and optical pumping. Most importantly, upon irradiation with the external optical pump, the quantum metamaterial undergoes ultrafast switching

between the flux penetration and flux exclusion states at very low pump fluences. The observed dynamic switching behavior is attributed to the reconfiguration of the total inductance of the system due to the magnetic flux exclusion and penetration. The existence of Meissner effect at terahertz frequencies has a broad range of implications in realizing quantum metamaterials without Josephson junction. Such quantum metamaterials could find applications in low-loss, ultrasensitive, frequency agile, switchable quantum photonics devices for metrology, efficient radiation sensors and manipulating sub-terahertz waves.

Chapter 8

Summary and Future Research Directions

8.1 Summary and outlook

In this chapter, we summarize the results obtained so far in this thesis and present an outlook for future research directions.

Rapidly growing field of metamaterial getting matured to a point where its applications are pursued by combining metamaterials with a diverse range of sub-disciplines of different research fields to achieve practical solutions for the basic science and advanced technology. The aim of this thesis was to harness the potential of metamaterials by incorporating high- T_c superconductors into its design for real-world applications such as ultrasensitive sensing, ultrafast switching, and efficient modulation of terahertz waves. The work presented in this thesis manifests great prospects and pathways for new generation terahertz meta photonic devices.

Initially, the studies presented in chapter 3 revealed a conductivity engineered approach to excite extremely high Q Fano resonances in terahertz metamaterials. It was discovered that the asymmetric Fano resonant metamaterials are very sensitive to the conductivity of the constituent materials in the low asymmetry region. Furthermore, we have shown that the high conductivity Drude metals fail to excite Fano resonances at extremely low asymmetry regime of the Fano resonators. In chapter 4, we have discovered a method to access the low asymmetry regime of the asymmetric Fano resonators by employing a high-temperature superconductor to excite extremely high- Q Fano resonances. These results would enable

selective engineering of material losses that has crippled the metamaterial community and act as a design principle for metamaterial based active and passive devices that have potential applications in the terahertz frequencies for high sensitivity bio-chemical sensing, non-linearity and lasing.

All the investigations presented in initial chapters are based on low-loss feature of the high-T_c superconductors at terahertz part of the electromagnetic spectrum, while the results presented in the latter part of this thesis are based on the ultrasensitive nature of Cooper pairs. The study presented in chapter 5 depicts all-optical ultrafast dual-channel switching of sharp Fano resonances excited in superconducting asymmetric split ring resonators. These dual-channel switching devices offer prospects as superconducting radiation sensors, time division multiplexers, terahertz high-speed wireless communication applications in the sub-THz bandwidths. Furthermore, ultra-thin superconducting metamaterials devices are introduced in chapter 6. Such ultrathin superconducting films and metamaterial structures provide a solution for ultra-low threshold, ultrafast switching applications. The high kinetic inductance of sub-London penetration depth thick superconductors plays an extremely important role for frequency agile scalable metamaterial devices. Moreover, the flux expulsion metamaterial has been introduced and experimentally studied in chapter 7. Upon irradiation with an optical pulse, this quantum metamaterial shows ultrafast switching between flux exclusion and flux penetration regimes. Therefore, our results presented a new class of superconducting quantum metamaterials.

The results presented in this thesis are proof of concept demonstrations and therefore, there is room for further investigations. Some of the proposed future research directions are listed below.

Superconductors effectively enhance the Q factors of the low asymmetry Fano resonant metamaterials in a regime where radiative losses are effectively suppressed and thus the loss mechanism is dominated by ohmic losses. Thus, on incorporating high- T_c superconductors in a recently developed planar toroidal metamaterials, which effectively suppress radiative losses by forming a mirrored configuration, could potentially further enhance the maximum achievable Q factors.

Ultrathin superconducting metamaterials discovered in this thesis could further enrich the active photonics community as it shows very high kinetic inductance. Incorporating Josephson junctions in an ultrathin metamaterial structure may potentially give rise to a large enhancement in the kinetic inductance and thus further unlock the potential for active photonics.

The demonstration of Meissner effect at terahertz frequencies and flux exclusion metamaterials could potentially unlock geometry dependent nonlinearity and active tunability. By relating the intensity of incident radiation which is driving the flux exclusion and flux penetration to the geometry of the metamaterials could lead to an efficient sensor for electromagnetic radiations.

Finally, the high- T_c superconductors could also provide efficient solutions for low loss, in-plane terahertz waveguides. Thus, active research in this area will be of immense interest for the high-speed photonics and electronics.

Appendix 1

Temperature dependent complex permittivity of YBCO thin films

Figure A.1: Real and imaginary part of complex permittivity of 25 nm YBCO film deposited over LAO substrate measured using terahertz time domain spectroscopy at varying temperature.

Figure A.2: Real and imaginary part of complex permittivity of 50 nm YBCO film deposited over LAO substrate measured using terahertz time domain spectroscopy at varying temperature.

Figure A.3: Real and imaginary part of complex permittivity of 100 nm YBCO film deposited over LAO substrate measured using terahertz time domain spectroscopy at varying temperature.

Appendix 2

Fano fitting

Figure A.4: Transmittance spectra and corresponding Fano fitting to calculate the Q factors at asymmetry $d = 5 \mu\text{m}$ for **a)** DC-TASR **b)** Drude-TASR **c)** PEC-TASR.

Appendix 2

Temperature dependent resonance response of Al-TASR

Figure A.5: Amplitude transmission spectra of Al-TASR metamaterial with asymmetry $d=15\text{ }\mu\text{m}$ measured at varying temperature ranging from 25K to 295 K.

Appendix 3

Publications

Journal publications during PhD candidature

1. **Y. K. Srivastava**, M. Manjappa, L. Cong, W. Cao, I. Al-Naib, W. Zhang and R. Singh, "Ultrahigh-Q Fano resonances in terahertz metasurfaces: strong influence of metallic conductivity at extremely low asymmetry" *Adv. Opt. Mater.* 2016, 4, 457.
2. **Y. K. Srivastava**, M. Manjappa, H. N. S. Krishnamurthy and R. Singh, "Accessing the high-Q dark plasmonic Fano resonances in superconductor metasurfaces", *Adv. Opt. Mater.* 2016, 4, 1875.
3. **Y. K. Srivastava**, R. Singh, "Impact of conductivity on Lorentzian and Fano resonant high-Q THz metamaterials: Superconductor, metal and perfect electric conductor" *J. App. Phys.* 2017, 122, 183104.
4. M. Manjappa*, **Y. K. Srivastava***, L.Cong*, I Al-Naib and R. Singh, "Active photoswitching of sharp Fano resonances in THz metadevices" *Adv. Mater.* 2017, 29, 1603355.
(* equal contribution)
5. **Y. K. Srivastava**, A. Chaturvedi, M. Manjappa, A. Kumar, G. Dayal, C. Kloc, R. Singh, "MoS2 for ultrafast all-optical switching and modulation of THz Fano metaphotonic devices", *Adv. Opt. Mater.* 2017, 5, 1700762.

6. **Y. K. Srivastava**, L. Cong, R. Singh, "Dual-surface flexible THz Fano metasensor" *Appl. Phy. Lett.* 2017, 111, 201101.
7. **Y. K. Srivastava**, M. Manjappa, L. Cong, H.N. S. Krishnamoorthy, V. Savinov, P. Pitchappa, and R. Singh "Superconducting dual-channel photonic switch" *Adv. Mater.* 2018, 30, 1801257.

Co-author contribution

1. L. Cong, **Y. K. Srivastava**, and R. Singh, "Inter and intra-metamolecular interaction enabled broadband high-efficiency polarization control in metasurfaces" *Appl. Phys. Lett.* 2016, 108, 011110.
2. L. Cong, **Y. K. Srivastava**, and R. Singh, "Near-field inductive coupling induced polarization control in metasurfaces", *Adv. Opt. Mater.* 2016, 4, 848.
3. M. Manukumara, **Y. K. Srivastava**, and R. Singh, "Lattice-induced transparency in planar metamaterials", *Phys. Rev. B - Rapid Communications*, 2016, 94, 161103(R).
4. M. Manjappa, S. P. Turaga, **Y. K. Srivastava**, A. A. Bettiol, and R. Singh, "Magnetic annihilation of the dark mode in a strongly coupled bright–dark terahertz metamaterial" *Opt. Lett.* 2017, 42 (11), 2106.
5. M. Gupta, **Y. K. Srivastava**, M. Manjappa, and R. Singh, "Sensing with toroidal metamaterial", *Appl. Phys. Lett.* 2017, 110, 121108.
6. L. Cong, **Y. K. Srivastava**, A. Solanki, T. C. Sum, and R. Singh "Perovskite as a platform for active flexible metaphotonic devices" *ACS Photon.* 2017, 4, 1595.

7. M. Manjappa, **Y. K. Srivastava**, A. Solanki, A. Kumar, T. C. Sum, and R. Singh, “Hybrid lead halide perovskites for ultrasensitive photoactive switching in terahertz metamaterial devices” *Adv. Mater.* 2017, 29, 1605881.
8. R. Yahiaoui, M. Manjappa, **Y. K. Srivastava**, and R. Singh, “Active control and switching of broadband electromagnetically induced transparency in symmetric metadevices”, *Appl. Phys. Lett.* 2017, 111, 021101.
9. L. Cong, **Y. K. Srivastava**, and R. Singh, “Tailoring the multipoles in THz toroidal metamaterials”, *Appl. Phys. Lett.* 2017, 111, 081108.
10. S. Han, M. Gupta, L. Cong, **Y. K. Srivastava**, and R. Singh “Toroidal and magnetic Fano resonances in planar THz metamaterials” *J. Appl. Phys.* 2017, 122, 113105.
11. M. Gupta, **Y. K. Srivastava**, and R. Singh, “A toroidal metamaterial switch” *Adv. Mater.* 2018, 30, 1704845.
12. W. X. Lim, M. Manjappa, **Y. K. Srivastava**, L. Cong, A. Kumar, K. F. MacDonald and R. Singh “Ultrafast all-optical switching of germanium-based flexible metaphotonic devices” *Adv. Mater.* 2018, 30, 1705331.
13. L. Cong, **Y. K. Srivastava**, H. Zhang, X. Zhang, J. Han, and R. Singh, “All-optical active THz metasurfaces for ultrafast polarization switching and dynamic beam splitting” *Light: Science and Applications*, 2018, 7, 28.

14. A. Kumar, **Y. K. Srivastava**, M. Manjappa, and R. Singh, “Color sensitive ultrafast optical modulation and switching of terahertz plasmonic devices”, *Adv. Opt. Mater.*, 2018, 6, 1800030.
15. T. C. W. Tan, **Y. K. Srivastava**, M. Manjappa, and R. Singh, “Lattice induced strong coupling and line narrowing of split resonances in metamaterials” *Appl. Phys. Lett.*, 2018, 112, 201111.

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