Observation of the Kinetic Inductance Limitation for the Fundamental Magnetic Resonance in Ultrasmall Gold v-Shape Split Ring Resonators

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2016

Tobing, L. Y. M., Luo, Y., Low, K. S., Zhang, D., & Zhang, D. H. (2016). Observation of the Kinetic Inductance Limitation for the Fundamental Magnetic Resonance in Ultrasmall Gold v-Shape Split Ring Resonators. *Advanced Optical Materials*, 4(7), 1047-1052.

<https://hdl.handle.net/10356/83716>

<https://doi.org/10.1002/adom.201500739>

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Article type: Original Paper

Title: Observation of the kinetic inductance limitation for the fundamental magnetic resonance in ultrasmall gold ν -shape split ring resonators

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((Optional Dedication))

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The kinetic inductance limitation has been proposed as the fundamental limitation for the attainable magnetic resonance frequency in split ring resonators (SRRs), but its experimental verification remains challenging owing to the stringent dimensional requirements in the resonator nanofabrication. Here, we report the first experimental observation for such saturation effects with saturated frequency at 520 THz using gold ν -shape split ring resonators. The interplay between electric dipole and magnetic dipole as a function of SRR opening angle is theoretically studied and experimentally observed, showing the dramatic increase of intensity buildup and resonance contrast as the dominant mode changes from electric to magnetic.

1. Introduction

Deep subwavelength optical resonators have attracted significant interest for their wide range applications from nanoantennae, chemical sensing, subwavelength resolution lithography, to spectroscopy. [1-13] In the context of these applications, split ring resonator [14] has been widely studied among other geometries [15-16] for its ability to localize light at the nanoscale,

with resonance frequency directly tunable based on its size. However, the magnetic frequency cannot go arbitrarily high as the collective oscillation of electrons plasma (known as plasmons) cannot be faster than the bulk plasma frequency of metal. The kinetic inductance has been proposed as the fundamental limitation for the achievable magnetic resonance in plasmonic nanocavity system [17, 18]. This is a consequence where the electron kinetic energy will dominate over the inductive energy at high frequencies, resulting in the saturation of the magnetic (known as the “LC” mode) resonance frequency. In such a situation, the permittivity of the metal becomes positive and the plasmonic responses of the metal disappear. Another contributing factor is the increase of damping loss associated with interband transition, [19, 20] which greatly suppresses the plasmonic behavior. Thus far, the saturation of magnetic resonance is only demonstrated through numerical simulations, [17, 18] and its experimental investigation is still challenging due to stringent dimensional requirements in the lithographic patterning.

Different approaches have been explored for pushing the magnetic resonance to higher frequencies. Colloidal self-assembly allows nanoresonators to be realized at large scale. [21-25] However, the sizes of the resonators are dictated by the nanosphere dimensions and their location are quasi random with lattice configuration limited to hexagonally closed packed arrays. Another approach is based on the split-ball resonator (SBR), [26] whereby a nanosphere is formed by laser induced transfer, followed by nanogap formation using focused ion beam. Gold SBR with magnetic resonance at 600 nm can be realized by this method, although it comes at the expense of fabrication complexity.

In our previous work, [27, 28] we have demonstrated the smallest reported gold and silver *u*-shape SRRs with magnetic resonances at ~900nm and ~600nm, where SRR with 60-nm size

have been successfully fabricated. However, the reported magnetic resonance wavelengths are still linearly proportional to the resonator size, indicating that the fundamental kinetic inductance has not been reached. On the other hand, fabrication of u -shape SRR smaller than 60-nm size remains practically challenging due to the inherent resolution-throughput trade-off in any lithographic patterning. In this work, we explore the role of a v -shape SRR (v -SRR) structure in further pushing the magnetic resonance to higher frequencies. Importantly, we show that gold v -SRR with magnetic resonance at 578 nm can be achieved through a standard planar fabrication approach. Finally, we report the first experimental observation of the fundamental kinetic inductance limitation in the saturating trend of the magnetic resonance in the mid-Vis frequency range.

2. Results and Discussions

The v -SRRs were fabricated by the sonicated cold development process with achievable pitch of ~ 40 nm. [29, 30] After lithographic patterning, the samples were evaporated with 2-nm thick titanium adhesion layer and 30-nm thick gold on ITO-coated glass, followed by lift-off in n -methyl pyrrolidone (NMP) for 10 minutes. Figure 1 shows the examples of the fabricated v -SRRs, which are characterized by the arm length (s) and the opening angle (θ). The left panel shows the v -SRRs with 100-nm arm length and 60° opening angle; while the right panels show the v -SRRs with opening angles varied from $\theta = 180^\circ$ to $\theta = 30^\circ$, for a fixed arm length of 100 nm. The spectral responses of SRRs were then characterized by CRAIC microspectrophotometer (with NA = 0.28 objective lens), where a polarizer was placed in between the sample and the condenser in order to obtain transmission spectra under different polarizations. The transmission characteristics of v -SRRs ($s = 100$ nm) under the X and Y polarizations are shown in Figure 2, where we observe only one resonance dip associated with each polarization. The mode excited under X-polarization is the fundamental magnetic mode (m_0), while the one excited under Y-polarization is the electric mode (e_0). The magnetic mode

refers to the localization of magnetic fields produced by the circulating current, which is excited by electric fields in the horizontal direction through bianisotropic effects (Figure 2a). On the other hand, the electric mode corresponds to a strong net electric dipole in the vertical direction (Figure 2b).

The transmission spectra for the X and Y polarizations are presented in Figure 2c and Figure 2d, respectively. It should be noted that the ν -SRR behaves as both SRR and plasmonic particle, depending on its geometrical features. For the SRR modes, the magnetic (under X-polarization) and electric (under Y-polarization) modes are derived from the fundamental and higher order longitudinal plasmons, respectively, making the resonance wavelength of the magnetic mode always longer than that of the electric mode. On the other hand, for plasmonic particle modes, the resonance wavelength for each polarization is dictated by the resonator size along the direction of the incident polarization. Based on this reasoning, we then deduce whether the observed resonances under the X-polarization are indeed the m_0 modes. This is done by verifying if the resonance wavelength under the X-polarization is longer than that under the Y-polarization when the resonator size is longer in the Y direction. The opposite case would be the plasmonic particle which does not point to the magnetic dipole generation.

It is clear from Figure 2e that the resonance wavelengths of m_0 remain longer than that of e_0 , indicating that the resonances under the X-polarization are indeed the fundamental magnetic resonances. There are in fact many similarities between u -shape SRR and ν -shape SRR in terms of mode characteristics, except for the fact that there is no higher order magnetic mode in the ν -SRR. This is because higher order magnetic mode (m_1) requires the excitation of anti-parallel electric dipole in vertical direction, which then induces the generation of horizontal

electric dipole along the bottom arm of the *u*-shape SRR. As there is no bottom arm in the *v*-shape SRR, the generation of anti-parallel electric dipole is thus not possible.

The resonance shift as a function of the opening angle (from $\theta = 180^\circ$ to $\theta = 0^\circ$) is denoted by the dashed arrows. Prior to the expected blue shift at decreasing angle due to smaller effective mode area, it should be noted that there exists a sudden resonance shift as the resonator changes geometry from a nanorod ($\theta = 0^\circ$ and $\theta = 180^\circ$) into an SRR ($0^\circ < \theta < 180^\circ$). For the X-polarization, a red shift from 908 nm to 924 nm is observed as the opening angle is changed from $\theta = 180^\circ$ (horizontal nanorod case) to $\theta = 150^\circ$, followed by a progressive blue shift at decreasing opening angle. Likewise for the Y-polarization, a blue shift from 723 nm to 636 nm is observed as the opening angle is changed from $\theta = 0^\circ$ (vertical nanorod case) to $\theta = 30^\circ$, followed by a progressive red shift at increasing opening angle. Such a sudden resonance shift is indicative of a mode evolution from a pure electric mode (for $\theta = 0^\circ$, and $\theta = 180^\circ$) into a hybrid magnetic-electric modes in *v*-SRRs, where the resonance behavior is dominantly magnetic when the opening angle is smaller than a certain value. In addition, such a sudden resonance shift can be reproduced by finite difference time domain calculations (Figure S1, Supplementary Information) with a good agreement with the experimental results shown in Figure 2c and 2d.

The traces of the m_0 and e_0 mode as a function of opening angle are presented in Figure 2e. The resonance strength is then deduced by the resonance contrast (C) defined as the difference between the resonance dip with the nearest peak, i.e., $C = T_{\text{peak}} - T_{\text{dip}}$. Interestingly, the trend for the resonance contrast for decreasing angle goes in the opposite direction for the magnetic and electric modes. It is understood that decreasing SRR angle would shift the resonances closer to the interband transition of gold at ~539 nm, thereby leading to higher

damping loss. However, it is not clear if this translates to resonance contrast in Figure 2c and Figure 2d, since the contrast goes in different direction despite the fact that damping loss increases along with the decreasing resonance wavelengths for the two modes. The more likely explanation comes from the overlapping between the incident polarization and the metal, suggesting that higher contrast would be expected if the incident electric field overlaps more with the metal. In order to verify this, we plot in Figure 2f the resonance contrast of m_0 and e_0 modes as a function of opening angles. It can be seen that the resonance contrast of m_0 mode saturates at large angle, while that of e_0 mode saturates at small angles. This correlates with the incident light overlap with the metal, where the degree of overlapping is proportional to $\sim \cos(\theta/2)$ and $\sim \sin(\theta/2)$ for X-polarization and Y-polarization, respectively.

Another interesting aspect of ν -SRR is presented in Figure 3, showing FDTD calculations of the intensity buildup in the ν -SRR as a function of the opening angle, with its corresponding electromagnetic fields distribution. The typical side and top view of the E_x -field distribution at the fundamental magnetic resonances is shown in the insets of Figure 3a, while the intensity buildup is numerically measured at the point “B” in the simulations. The intensity buildup is rather flat at large angles, but begins to increase sharply below $\theta \sim 50^\circ$. This remains true as the arm length is varied from $s = 100$ nm to $s = 60$ nm. The electromagnetic field distributions of ν -SRR are presented in Figure 3b, showing the evolution from electric-dipole to magnetic-dipole as the opening angle is decreased from $\theta = 180^\circ$ to $\theta = 30^\circ$. At $\theta = 180^\circ$, we have a nanorod situation where the E_x -fields are confined at both tips, with E_y -fields and H_z -fields symmetric with respect to the horizontal axis. In this case, the resonator behaves as a plasmonic particle, with pure electric dipole resonances corresponding to plasmonic oscillation in the direction of incidence polarization. As the angle is decreased, the accumulated electric charges (from the two tips) begin to interact capacitively, skewing the

E_y -field and H_z -field distributions towards the inner side of the ν -SRR. Thus, both electric-dipole and magnetic-dipole are generated in this situation, where the former is due to the conduction current from plasmonic oscillation in the longitudinal direction, and the latter is due to displacement current from the capacitive interaction between the ν -SRR tips. As the gap separation decreases, the ohmic resistance remains unchanged due to a fixed metal length, while the capacitive impedance becomes smaller. This leads to a higher conduction current and stronger E_x -fields in the SRR gap. Thus, the electric field enhancement in the SRR gaps translates to the magnetic dipole strength, suggesting that the excitation of magnetic-dipole is more preferable at small opening angles.

This is indeed depicted by the mode evolution in Figure 3b, where the transition from plasmonic particle (corresponding to electric dipole) to split-ring resonator (corresponding to magnetic dipole) occurs at $\theta \sim 50^\circ$. The lateral sizes of the ν -SRR in the X and Y direction for a given opening angle (θ) and arm length (s) are given by $s_x(\theta) = 2s \sin(\theta/2)$ and $s_y(\theta) = s \cos(\theta/2)$, respectively. Thus, the condition where the resonator sizes are the same in both direction, i.e., $s_x(\theta) = s_y(\theta)$, is satisfied at $\theta_T = 2 \tan^{-1}(1/2) \cong 53^\circ$, which is interestingly close to the transition angle observed in Figure 3a. Thus, from plasmonic particle perspective, the resonance for X-polarization is longer (shorter) than that for Y-polarization at $\theta > \theta_T$ ($\theta < \theta_T$). As the resonance wavelength of m_0 is always longer than that of e_0 (see Figure 2), it then follows that the m_0 resonance behaves more as an electric dipole at $\theta > \theta_T$ and more as a magnetic dipole at $\theta < \theta_T$. This could be the likely cause for the transition from electric-dipole to magnetic-dipole at $\theta \sim 50^\circ$ in Figure 3a.

For completeness, we also present the optical properties of laterally offset ν -SRRs (Figure 4a, 4b) and ν -SRRs with different arm lengths (Figure 4c). The dependence of the resonance

wavelength on the lateral offset illustrates the role of the resonator geometry in further pushing magnetic resonances into the visible spectrum. Increasing lateral offset geometrically transforms the v -SRR into u -SRR (the u -SRR is actually a special case of laterally-offset v -SRR with $\theta = 0$ and $d = s$), and the resonance wavelength is red shifted. Due to strong polarization responses, the m_0 resonance is more sensitive to the lateral shift as compared to the e_0 resonance, where 16 nm of lateral shift lead to a red shift from ~ 647 nm to ~ 821 nm (for m_0 mode) and from ~ 636 nm to ~ 681 nm (for e_0 mode). As the lateral offset increases from $d = 0$ nm (pure v -SRR) to $d = 16$ nm, the resonance contrast for m_0 mode increases by ~ 4 times, while that for e_0 mode remains qualitatively unchanged. This agrees with the fact that the resonance contrast depends mainly on the overlap of incoming light polarization with the metal. In Figure 4c, we show the optical properties for different resonator arm lengths at fixed opening angles ($\theta = 90^\circ$, $\theta = 60^\circ$). The resonance contrasts and wavelengths of m_0 and e_0 modes are expected to increase along with the increase of resonator arm length. On the other hand, upon changing the opening angle from $\theta = 90^\circ$ (square) to $\theta = 60^\circ$ (triangle), we can see that the resonance contrast decreases for m_0 mode and increases for e_0 mode, showing agreement with spectral characteristics in Figure 2.

In order to demonstrate the magnetic resonance frequency saturation resulting from kinetic inductance limitation, we show in Figure 5 the mapping of m_0 mode as a function of inverse resonator size. Here, the resonance positions of the v -SRRs are combined with those of the u -SRRs from our previous work. [23] There are 5 geometric variations for v -SRRs with $s = 100$ nm, while there are 4 geometric variations for v -SRR with $s = 50$ -70 nm. The resonator size (s_{eff}) of v -SRRs is approximated as $s_{\text{eff}} \cong (A_{\text{SRR}})^{1/2}$, where $A_{\text{SRR}} = 1/2 \times (\text{base}) \times (\text{height})$ is the area of the v -SRR with base and height obtained from SEM inspections. The resonator size for the u -SRR structures is defined as the length of the bottom arm. The saturation effect can be seen

clearly at a very small resonator size, where the m_0 mode saturates at ~ 520 THz (578 nm).

The same saturation effect is observed for the e_0 mode (not shown here), where the e_0 mode saturates at ~ 530 THz (565 nm). Note that the observed two saturation frequencies are still lower than the interband transition of gold at 556 THz (539 nm). This is as expected since the plasmonic oscillation is significantly suppressed by the interband transition. The role of interband transition in the magnetic resonance is also investigated numerically (Figure S2, Supplementary Materials), where finite difference time domain calculations are performed based on Drude metals and on gold permittivity according to Johnson and Christy [32].

In the following, the fitting based on the well known LC model [17] is also presented. The fundamental magnetic resonance (the “LC” resonance) is $\omega_{m0} \equiv \omega_{LC} = (LC)^{-1/2}$, where L and C are the total inductance and capacitance of the SRR. Using u -SRR structure for simplicity, the SRR capacitance is $C = \epsilon_0 \epsilon_d w t / g$, where ϵ_0 and ϵ_d are the vacuum and dielectric permittivity, and w , t , and g denote the feature width, thickness, and the gap of the SRR. The SRR inductance is $L = \mu_0 s^2 / t + L_K$, where μ_0 is the permeability, s is the resonator size, and L_K is the kinetic inductance. By assuming $g \sim w$, and defining the kinetic inductance as $L_K = \mu_0 s_K^2 / t$, the ω_{LC} can be reduced to $\omega_{m0}(s_{\text{eff}}) = c(\epsilon_d)^{-1/2}(s_{\text{eff}}^2 + s_K^2)^{-1/2}$, where c is the speed of light in a vacuum. Defining the saturation frequency as the resonance frequency at $s_{\text{eff}} = 0$, i.e., $f_{m0}(0) = c/[2\pi\epsilon_d^{1/2}s_K]$, the fundamental magnetic resonance frequency can be further simplified into $f_{m0}(s_{\text{eff}}) = f_{m0}(0)/(1 + s_{\text{eff}}^2/s_K^2)^{1/2}$. The fittings based on different values of s_K at a fixed saturation frequency are presented in Figure 5. The s_K can be understood as the onset resonator size for the kinetic inductance to dominate over the SRR inductance, where $s_K = 0$ represents the ideal case in which the kinetic inductance limitation does not exist. The best fit parameters are found to be $s_K = 45$ nm and $f_{m0}(0) = 530$ THz. This suggests that

the saturation is only observed when the resonator size is smaller than 45 nm, which is consistent with our previous works where there was no saturation for u -SRR with lateral size of 60 nm.

An interesting comparison can also be made with the theoretical prediction made by Zhou et al, [18] which reported the disappearance of magnetic response ($\mu_{\text{eff}} < 0$) and the saturation of the magnetic resonance frequency at ~ 550 THz in high density multi-cut SRR arrays. The v -SRR differs from the multi-cut SRR arrays in that the magnetic dipole in the v -SRR is generated from the electric excitation through the bianisotropic property of individual SRRs. [31] This is different from the multi-cut SRR structure in that the bianisotropic effect vanishes due to the resonator structural symmetry, thus making the magnetic dipole excitation only possible through magnetic fields. In addition, the resonance characteristics of the v -SRR arrays are determined mainly by the property of the individual v -SRRs, unlike the multi-cut SRR arrays whose resonance characteristics are mainly contributed by the strong coupling between neighboring elements. Despite these differences, the excited magnetic dipole for both structures does exhibit magnetic field normal to the SRR plane.

3. Conclusions

We have presented a comprehensive study of the v -shape split ring resonator and its resonance properties, where the resonant behavior as a mixture of electric and magnetic dipoles related to the resonator opening angle have been numerically and experimentally investigated. By changing the resonator geometry from u -shape to v -shape, we have shown that the magnetic resonance frequency can be further pushed from 330THz (900nm) to 520THz (578nm) under the same dimensional constraint of our nanofabrication process. Furthermore, the saturation of the magnetic and electric resonance frequencies predicted theoretically many years ago [18] has also been experimentally verified, with a remark that

these saturated frequencies are pretty close to the interband transition of gold at 556 THz (539 nm). This suggests the possible role of the interband transition in the observed frequency saturation. We also present theoretical fittings that indicate the onset resonator size for the kinetic inductance limitation, which is found to be 45 nm for 25-nm feature width and 30-nm thickness.

Acknowledgements The authors thanked Qihua Xiong for facilitating microspectrophotometer measurements. This work was partially supported by Ministry of Education (RG86/13), A*Star (1220703063), Economic Development Board (NRF2013SAS-SRP001-019), Asian Office of Aerospace Research and Development (FA2386-14-1-0013), the MOE AcRF Tier 2, and the Program Grant (11235150003) from NTU-A*STAR Silicon Technologies Centre of Excellence.

Received: ((will be filled in by the editorial staff))

Revised: ((will be filled in by the editorial staff))

Published online: ((will be filled in by the editorial staff))

Keywords: nanophotonics, plasmonics, split-ring resonator, kinetic inductance limitation

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Figure 1. The ν -SRRs in square lattice configuration. The fabricated SRRs are of 30 nm metal thickness and 25-nm metal width. The opening angles were varied from $\theta = 180^\circ$ (horizontal nanorod) to $\theta = 0^\circ$ (vertical nanorod). The scale bars correspond to 100 nm.

Figure 2. Optical characteristics of gold v -SRRs ($s = 100$ nm). Finite difference time domain calculation of the electromagnetic field distribution for (a) the magnetic mode m_0 and (b) the electric mode e_0 , excited under X-polarized and Y-polarized normal incidence, respectively. The associated electric dipoles and circulating current are shown in solid and dashed lines, respectively. The transmission spectra under (c) X-polarization (m_0 mode) and (d) Y-polarization (e_0 mode). The opening angles are 180° , 150° , 100° , 60° , 30° , and 0° . The blue and orange arrows indicate the transmissions of $\theta = 180^\circ$ and $\theta = 0^\circ$, respectively. (e) The resonance wavelength and (f) resonance contrast of m_0 (red triangles) and e_0 (blue squares) modes as a function of opening angle.

Figure 3. Mode evolution from electric dipole-like to magnetic dipole in ν -SRR. (a) Intensity buildup as a function of opening angle. (b) The evolution of electromagnetic field distributions ($|E_x|, |E_y|, |H_z|$) of ν -SRR along with increasing opening angles.

Figure 4. The role of lateral offset and resonator arm length in v-SRR resonance properties. (a) The schematic of a laterally offset v-SRR. All the scale bars correspond to 100 nm. (b) The resonance wavelengths and contrasts of v-SRRs ($s = 100$ nm, $\theta = 30^\circ$) with different lateral offsets. (c) The resonance wavelengths and contrasts of v-SRR ($\theta = 90^\circ$, $\theta = 60^\circ$) with different arm lengths.

Figure 5. Saturation of the fundamental magnetic resonances in split ring resonator. The fittings based on $s_k = 15$ nm, $s_k = 25$ nm, and $s_k = 45$ nm for a fixed saturation frequency of $f_{m0}(0) = 530$ THz are denoted in solid lines. For all the $u\text{-SRRs}$ and $v\text{-SRRs}$, the feature width and thickness is 25 nm and 30 nm, respectively. At increasing “ $1/\text{size}$ ” order, there are 5 variations for $v\text{-SRR}$ with $s = 100$ nm, and 4 variations for $v\text{-SRRs}$ with $s = 50\text{-}70$ nm. The variations for $s = 100$ nm are (1) $\theta = 90^\circ$, (2) $\theta = 60^\circ$, (3) $\theta = 30^\circ$, $d = 12$ nm, (4) $\theta = 30^\circ$, $d = 8$ nm, (5) $\theta = 30^\circ$, $d = 0$ nm, while the variations for $s = 50\text{-}70$ nm are (1) $\theta = 90^\circ$, (2) $\theta = 60^\circ$, (3) $\theta = 30^\circ$, $d = 14$ nm, (4) $\theta = 30^\circ$, $d = 12$ nm.

Graphical Abstract

The achievable fundamental magnetic resonance frequency in split ring resonator (SRR) is inherently limited by the speed of oscillating electron plasma in the metal. The so-called kinetic inductance limitation was theoretically over a decade ago, but its experimental verification has been hindered due to nanofabrication challenges. Here, we report the first experimental demonstration of magnetic resonance frequency saturation at 520THz in ultrasmall ν -shape SRRs.

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