Frequency-agile temporal terahertz metamaterials

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Supporting Information

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Supplementary Figure 1. Electromechanical characterization of released microcantilever

The profile of the released bimorph microcantilever was measured using a Lyncee Tech digital holographic microscope. The microcantilever metamaterial housed on a PCB was fixed on the stage below the microscope with a tape. The bond pads of the PCB were connected to Keithley 2400 source meter to provide the input current. After adjusting for the tilt, intensity and phase offsets with a flat surface, the profile of the microcantilever along the length was measured at ambient conditions, as shown in **Figure S1a**. The length of the microcantilever is 60 μm, and the additional 2 μm in the initial part shows the connecting line which is anchored to the Si substrate. The tip displacement of the microcantilever (δ) upon release (i.e. at 0 mA) is measured to be 9 μm. When the input current was increased, a gradual reduction in δ is observed as shown in **Figure S1a**. At 380 mA, the profile of the microcantilever becomes flat, as it comes in complete physical contact with the Si substrate. The recovery of the microcantilever to the initial position after switching off the input current is shown in **Figure S1b**. After 120 s, the microcantilever goes back to ~90% of its initial value, and complete recovery takes 600 s under ambient conditions. This confirms the repeatable operation of the microcantilever metamaterial under current stimulus.

Supplementary Figure 2. Elucidation of resonances in microcantilever metamaterial and its effect on changing δ , using simulations

Electromagnetic simulations were carried out using commercially available CST software. Simulation model with the normal incidence of terahertz wave with electric field polarized along the length of the microcantilever. Aluminum was modelled as metal with DC conductivity of 3.5×10^7 S/m, aluminum oxide and silicon were modelled as lossless dielectrics with permittivity of 9.9 and 11.7, respectively. [S1, S2] The out-of-plane deformation profile of the microcantilever was realized by bending the straight microcantilever with a specific angle of curvature (θ) to attain desired δ values as shown in **Figure S2a**. The values of δ from specific θ and it clearly shows a cubic dependence. In the simulations, a unit cell of the metamaterial was used with unit cell boundary conditions employed in axial directions

orthogonal to the incident waves. The perfectly matched layers were applied along the propagation of the electromagnetic waves. Plane waves were incident onto the unit cell from the port on the metal side, while the transmission spectrum was determined from the port placed at the other side of metamaterial. Meanwhile, field monitors were used to collect the electric fields, magnetic fields, and the respective surface currents at the specific resonance frequencies. The simulated transmission spectra of the microcantilever metamaterial with varying θ is shown in **Figure S2b**. When $\theta = 20^\circ$, the resonance frequency is at 0.8 THz. At this frequency, the surface current is along the length of the microcantilever and in-phase with the incident electric field of the terahertz wave, which indicates the excitation of electric dipole resonance as shown in **Figure S2d**.^[S3] With decreasing θ , a red-shift in the resonance frequency is observed. The shift in resonance frequency is smaller when the cantilever is further away from the substrate and as it comes closer to the substrate, the value of red shift increases dramatically. At $\theta = 0^\circ$, the resonance frequency is at 0.53 THz. The simulated surface current is in-phase with the electric field of the incident terahertz waves as shown in **Figure S2e**, confirming the electrical dipole nature of the resonance. The resonance frequency tunability from the simulated spectra was calculated for each curvature angle ' θ ' as $\Delta f/f_{20} = (f_{20} - f_{\theta})/f_{20}$ and is shown in the **Figure S2c**. Here, the resonance frequency of the microcantilever metamaterial at $\theta = 20^\circ$ (i.e. f_{20}) is used the reference value, since it defines the zero input current state in the simulations. When θ decreases from 20° , the resonance frequency tunability increases and a maximum tunability of $\sim 35\%$ is achieved. For the measured results, the resonance frequency tunability was calculated for each input current ' I_c ' as $\Delta f/f_0 = (f_0 - f_i)/f_0$, where f_0 and f_i are the resonance frequency of the microcantilever metamaterial with 0 mA and I_c mA of input current, respectively. The measured resonance frequency tunability with increasing input current is shown as inset in **Figure S2c**. Maximum measured resonance frequency tunability of $\sim 30\%$ was achieved.

Supplementary Figure 3. Optical images of the metadvice with varying current stimulus

Optical images of the microcantilever metamaterial was taken using a Nikon Optical microscope (OM) at 5X zoom. The microcantilever metamaterial housed on a PCB was fixed to the OM stage with a thermal tape. The terminals of the PCB were connected to a Keithley 2400 source meter to provide the current input. The optical image was captured 120 s after the applying the input current. The OM images of the microcantilever metamaterial with sequential increase of input current is shown in **Figure S3**. The microcantilevers of the fabricated metamaterial show large out-of-plane deformation, which is evident from the increasing out-of-focus along the length of the microcantilever (i.e. the microcantilever portion closer to the fixed connecting lines is in focus, while the tip of the microcantilever is strongly out-of-focus). The fabricated metamaterial shows a good uniformity of the microcantilever across the entire sample. When the input current is increased, the microcantilevers gradually move downwards, which is observed from increased portion coming in focus and corresponding reduction in the out-of-focus at the tip of the microcantilevers. However, during the operation of the metamaterial, non-uniformity in the deformation of the constituent microcantilevers can be observed. Such variations might occur from the variable grain sizes of the Al layer. This non-

uniformity will translate to the undesired resonance broadening in the terahertz transmission measurement. At 420 mA, the entire portion of all the microcantilevers comes in focus, thereby confirms the physical contact with Si surface.

Supplementary Figure 4. SRIM simulations for ion irradiation

Different energies of ions will penetrate different depths in silicon. **Figure S4** shows the stopping and range of ions in matter (SRIM) simulated plots for the Frenkel defect profile produced by (a) 500 keV, (b) 1 MeV, (c) 2 MeV He⁺ in silicon with depths of approximately 2 μm, 3.5 μm and 7.3 μm, respectively.[S4] In this work, we used 500 keV He⁺ ions with dose of $5 \times 10^{13} \text{ cm}^{-2}$ for the irradiation on silicon substrate with an expected EOR depth of 2 μm.

Supplementary Figure 5. Charge carrier dynamics comparison of pristine and ion-irradiated Si substrate with pump wavelength of 800 nm and 400 nm

Figure S5 shows the charge carrier dynamics of 725 μm thick Si substrate (pristine and $1 \times 10^{14} \text{ cm}^{-2} \text{ He}^+$ ion-irradiated) with (a) 800 nm and (b) 400 nm optical pump, respectively. In the case of 800 nm optical pumping, the photoexcited charge carriers for the Si substrate shows a slower relaxation time, which is expected to be in μs range. [S5] And, the relaxation time for the ion-irradiated Si substrate is almost identical to the pristine one as shown in **Figure S5a**. This is because of the shallower irradiation depth of 500 keV He^+ ions ($\sim 2 \mu\text{m}$) compared to the optical penetration depth of 800 nm optical pump ($\sim 7 \mu\text{m}$) in bulk Si. Hence, the overall dynamics of the ion-irradiated Si substrate is dominated by the response from the pristine Si portion of the substrate for 800 nm optical pump. However, the optical penetration depth of 400 nm pump is around $0.2 \mu\text{m}$, which is much shallower than the ion-irradiation depth of $2 \mu\text{m}$. Hence, we see that for the 400 nm optical pump, the pristine Si shows a slower relaxation time in μs timescale, while the $1 \times 10^{14} \text{ cm}^{-2} \text{ He}^+$ ion irradiated Si substrate shows an ultrafast response of $\sim 300 \text{ ps}$. Hence, 400 nm was chosen for the ultrafast optical control of our ion-irradiated microcantilever metamaterial.

Supplementary Figure 6. Charge carrier dynamics of ion-irradiated Si substrate with variable fluence of 400 nm pump

Figure S6 shows charge carrier dynamics of ion irradiated 725 μm thick Si substrate with dose of (a) $1 \times 10^{12} \text{ cm}^{-2}$, (b) $1 \times 10^{13} \text{ cm}^{-2}$ and (c) $1 \times 10^{14} \text{ cm}^{-2}$ with 400 nm optical pump, respectively. With increasing ion dose, the relaxation time of the photoexcited carriers in Si substrate can be reduced to picosecond timescales. It can also be seen that for a given dose, the value of $|\Delta T/T|$ and hence photoconductivity increases with optical pump fluence. Furthermore, with increasing irradiation dosage, the $|\Delta T/T|$ value reduces for a given pump fluence. Hence, photoconductivity and relaxation time are trade-off parameters and based on the application demands, the irradiation ions, dose and energy need to be selected.

Supplementary Figure 7. CST simulations to study the effect of photoconductivity on the microcantilever metamaterial resonances

Figure S7 shows the simulated results of the effect of increasing photoconductivity of Si substrate at different angle of curvature (θ). As the optical penetration depth of 400 nm pump in Si is 0.2 μm , the conductivity (σ_p) of top 0.2 μm of Si was varied in the simulation, while the remaining Si substrate was modelled as a lossless dielectric. With increasing σ_p , the resonance strength reduced with increased broadening. The presence of highly conductive layer screens the confined resonant field of the microcantilever, thereby leading to a reduced resonance strength and increased linewidth broadening. At sufficiently, high σ_p (~ 10000 S/m) the resonance is completely modulated for all values of θ .

Supplementary Figure 8. Measured terahertz transmission spectra of the microcantilever metamaterial with varying optical pump fluence at different input currents

Figure S8 shows the measured transmission spectra of the microcantilever metamaterial at varying optical stimulus for different input current. The increasing input current shows a clear spectral tunability of resonance frequency and increasing pump fluence modulates the resonance strength at each of the resonance frequency defined by the input current. This provides the experimentally confirmation of ultrafast terahertz resonance modulators with spectral tunability.

Supplementary Figure 9. Influence of microcantilever position on the terahertz resonance modulation under varying pump fluence

Figure S9 shows (a) simulated/(b) measured change in resonance strength ($\Delta RS = RS_{OFF} - RS_{ON}$) with (a) increasing photoconductivity σ_p /(b) pump fluence for various position of microcantilever (a) angle theta/(b) input current. The effect of resonance modulation through screening effect of photocarriers on the surface of Si depends on the distance of microcantilever from the substrate. For 400 nm optical pump, the photocarriers are generated in the top $\sim 0.2 \mu m$ of the Si surface. However, the confined terahertz field spreads exponentially from the metamaterial resonators in the out-of-plane direction. Hence, based on the position of the microcantilever, the interaction of the terahertz field with the photocarriers will be varied. For the case, when the cantilever is on top of the Si substrate ($\theta = 0$), strong terahertz field confinement overlaps with the photocarrier generated region and hence will lead to a strong modulation of resonance strength. However, when the microcantilevers are suspended over the Si substrate, then the interacting terahertz field with the photocarriers becomes weaker as observed from **Figure S9a**. The results from the simulation are also validated from the experimental results shown in **Figure S9b**.

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