Terahertz Control of Plasmon Induced Opacity in Photoexcited Metamaterial on GaAs

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Abstract: Strong THz pulses instantly restore the sharp resonance of coupled plasmonic oscillators quenched by photoexcitation. The THz control of plasmon induced opacity is applicable to sensitive sensing and ultrafast switching in plasmonic devices.
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A plasmonic metamaterial consisting of radiative elements coupled with subradiant elements exhibits plasmon-induced transparency (PIT), where a narrow transmission resonance emerges within a broad absorption band due to destructive interference between the bright and dark modes [1]. The extreme dispersion of PIT in the narrow spectral region signifies the high sensitivity of the metamaterial to external perturbations. Optical excitation of PIT metamaterials turns off the plasmon resonances, injecting photocarriers in the insulating area of the composite media. The optical control of PIT has demonstrated novel applications to terahertz (THz) sensing and modulation [2]. On the other hand, high-field terahertz (THz) pulses have been used to manipulate photocarriers in semiconductors [3], which has potential applications to ultrahigh-speed optoelectronics [4,5]. In this study we demonstrate that intense THz pulses transiently restore the plasmon resonances of THz metamaterials quenched by femtosecond photoexcitation.

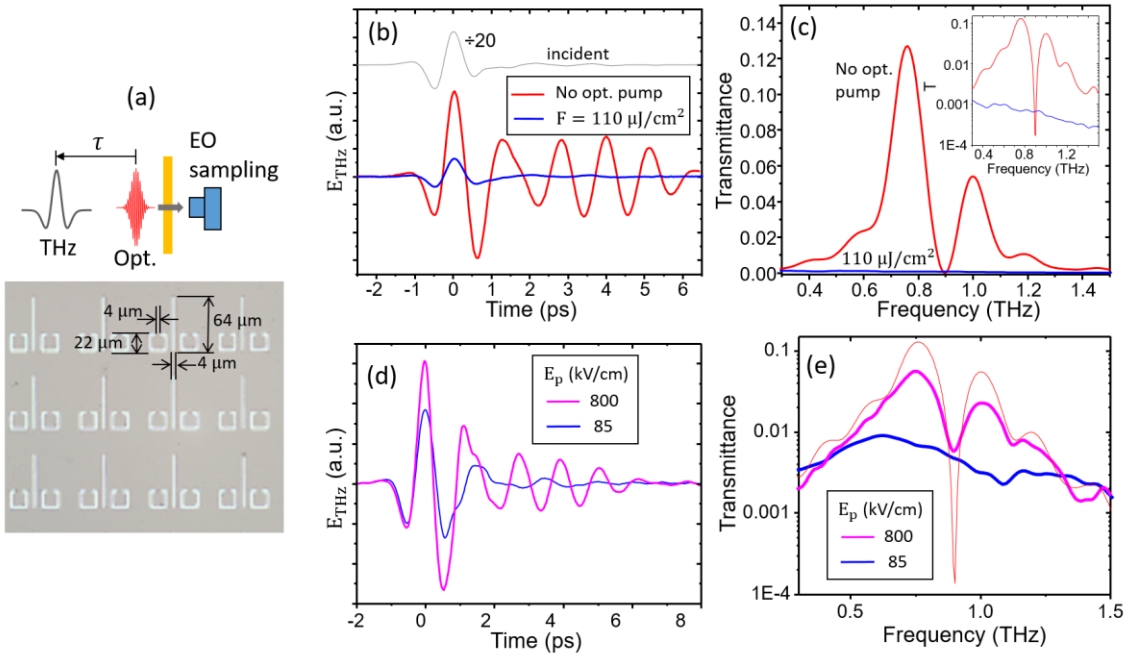

Fig. 1. (a) Microscopic image of the THz plasmonic metamaterial consisting of radiative slot antennas (length, 64 μm; width, 4 μm) and subradiant split-ring resonators (length, 22 μm; gap, 4 μm) (b) THz waveforms transmitted through the metamaterial with (blue) and without (red) optical excitation (pump fluence, $F = 110\ \mu J/cm^2$; time delay, τ=-150 ps). The incident THz waveform is in gray (the field amplitude is reduced by a factor of 20). The peak field amplitude of the incident THz pulse E_p is 85 kV/cm. (c) Transmittance spectra of the waveforms in (b). Spectra in log-scale are shown in the inset. (d) Transmitted THz waveforms for E_p=800 (violet) and 85 (blue) kV/cm at $F = 12\ \mu J/cm^2$. (e) Transmittance spectra of the waveforms in (d) in log-scale.

We carried out time-resolved high-field THz spectroscopy of an optically excited THz metamaterial. We employed tilted-pulse-front optical rectification in a Mg:$LiNbO_3$ prism to generate intense single-cycle THz pulses. The field amplitude of the broadband THz pulses (central frequency, 1 THz; bandwidth, 1 THz) reached 1 MV/cm at an optical pulse energy of 1 mJ. The THz waveforms were traced by electro-optic sampling in a 1-mm ZnTe crystal. Figure 1a

shows a microscopic image of the metamaterial formed by a periodic arrangement of a slot antenna (SA) and two split-ring resonators (SSRs). The plasmonic elements were etched out in a 500-nm-thick Al film deposited on a GaAs substrate. The plasmonic resonances, depending on the dimension of the resonators (Fig.1a), are tuned to 0.9 THz. The incident THz fields, perpendicular to the SAs, are radiatively coupled with the antennas, while the SSRs become dark elements in this polarization configuration. The THz waveform transmitted through the metamaterial (red in Fig.1b) shows plasmon oscillations after a single-cycle THz pulse (gray line in Fig.1b) excites the coupled resonators. Its transmittance spectrum (red in Fig.1c) exhibits a sharp opacity dip at 0.9 THz, the SSR resonance frequency. The plasmon-induced opacity (PIO) emerges from the coupling between the bright mode of SAs and the dark mode of SSRs. Femtosecond optical excitation injects photocarriers in the GaAs substrate, quenching the plasmon oscillations. The THz pulses transmitted through the optically excited sample (blue in Fig.1b) are negligibly small, showing no plasmon oscillations. The plasmon resonances collapse as shown in the transmittance spectrum (blue in Fig.1c). Strong THz fields partially restore the plasmon oscillations in the optically excited metamaterials, as shown in Fig.1d. The transmittance spectrum of the strong THz pulse (violet in Fig.1e) presents the opacity dip, while that of the weak THz pulse is small and flat in the spectral window. The instant revival of the plasmon oscillations is accounted for by THz field induced reduction of conductivity in GaAs. The dominant mechanisms of the conductivity reduction in photoexcited GaAs are field-driven intervalley scattering and interband tunneling [4].

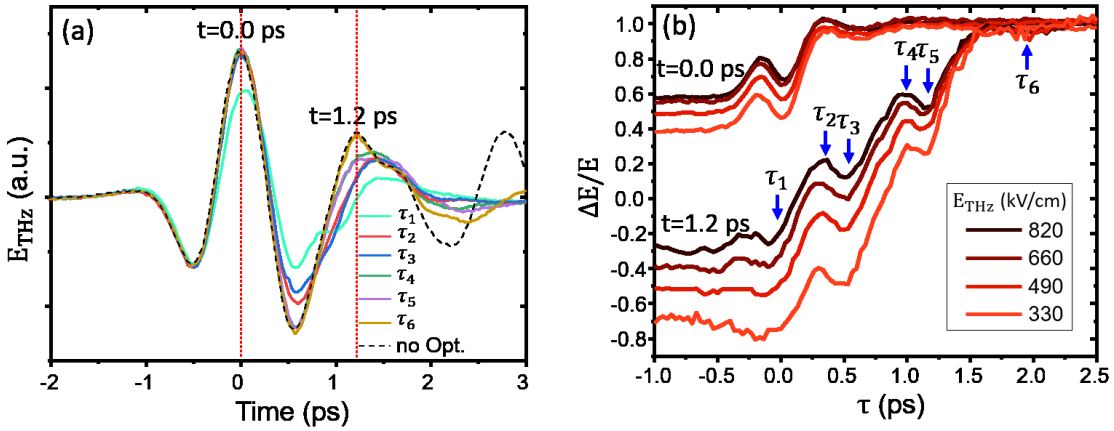

Fig. 2. (a) Transmitted THz waveforms at time delays between optical and THz pulses, $\boldsymbol{\tau_{1-6}}$=-0.07, 0.37, 0.53, 1.00, 1.17, and 1.93 ps at F = 61 μJ/cm^2. (b) Normalized differential amplitude ΔE/E versus time delay $\boldsymbol{\tau}$ at t=0.0 and 1.2 ps for the peak fields of the incident THz pulse, $\mathbf{E_p}$=820, 660, 490 and 330 kV/cm.

We observe the ultrafast dynamics of the interplay between photoexcitation and plasmon oscillations, monitoring the changes in THz transmission as varying the time delay τ between optical and THz pulses. Figure 2a shows the transmitted THz pulses at τ=-0.07, 0.37, 0.53, 1.00, 1.17, and 1.93 ps. The main peak at t =0.0 ps is strongly attenuated at τ_1=-0.07 ps, while hardly changes at the positive time delays, τ_{2-6}. Figure 2b shows that the differential transmission at the peak widely fluctuates between τ =-0.5 and 0.3 ps, showing an oscillatory behavior in the time domain. In Fig.2a, the second peak at t = 1.2 ps undergoes more complex modulation consisting not only of attenuation, but also of phase shift and broadening. Figure 1b presents the oscillatory modulations of the second peak as a function of τ. The rapid and large-scale fluctuations of the transmitted THz fields, which are absent in a bare GaAs substrate, indicate strong interference between photocarriers and plasmon oscillations.

In summary, our experimental study demonstrates that optical excitations quench the plasmon oscillations in coupled THz metamaterials, while strong THz pulses instantly restore the sharp plasmonic resonance. The high-field THz effects exhibit the ultrafast dynamics of photocarriers interacting with plasmon oscillations. The experimental scheme presents potential applications to ultrahigh-speed control of metamaterial and plasmonic devices.

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