Reversible modulation and ultrafast dynamics of THz resonances in strongly photoexcited metamaterials

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We demonstrate an ultrafast reversible modulation of resonant terahertz (THz) response in strongly photoexcited metamaterials. The transient spectral-temporal response of the dipole transition ~1.6 THz exhibits a distinct non-monotonic variation as a function of pump fluence. The transition energy shift, strength, spectral width and density-dependent ultrafast relaxation manifest a remarkable re-emergence of the resonances after initial quenching. Our simulation, incorporating the first-order diffraction from the photoinduced transient grating, reproduces the salient features, providing a new avenue for designing nonlinear and frequency-agile THz modulators.

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Artificially subwavelength structured materials, so-called metamaterials, attract strong current interest due to their exceptional properties, such as simultaneously negative permittivity \( \varepsilon(\omega) \) and permeability \( \mu(\omega) \), which implies a negative refractive index \( n \) not available in natural materials. In addition, artificial magnetism, super focusing and specifically-tailored structures responding to ultra-broadband electromagnetic radiation, from gigahertz (GHz), terahertz (THz) to visible range, set them among the most promising candidates for next generation optoelectronic devices and large-scale photonic functional systems.

Recently, there has been significant interest in understanding ultrafast THz responses of nonlinear metamaterials, e.g., split ring resonators (SRRs) patterned on substrates exhibiting substantial nonlinearities. The resulting components and schemes allow for constructing controllable active THz devices with dynamical tunability of amplitude and phase, which fills the demand for THz technology gap. Prior experiments using time-domain THz spectroscopy have demonstrated dynamic tuning of both magnetic and electric dipole resonances of SRRs on semiconductors or superconductors. More complex structures of coupled resonators on GaAs substrates have been proposed and experimentally studied, revealing frequency-agile THz modulation, e.g., a resonance shift with increasing photoexcitation. However, thus far all ultrafast studies of THz materials have been concentrated on the relatively low photoexcitation, where a reduction or quenching of the resonant absorptions is observed. Two outstanding issues are still poorly understood: first, there lack insights on how high density photoexcited carriers modify the dynamic responses of metamaterials; second, the decay pathway of photoexcited metamaterials after femtosecond (fs) excitation is not explicitly studied, and, particularly, the temporal evolution of the transient states have only been understood as a density-independent relaxation back to the equilibrium.

In this letter, we demonstrate an ultrafast reversible modulation of resonant THz absorption in strongly photoexcited metamaterials by optical pump and THz probe spectroscopy. This process directly manifests itself via a remarkable reemergence of the originally quenched THz resonance above a crossover density \( N_c \sim 4.7 \times 10^{16} \text{ cm}^{-3} \) after femtosecond photo-excitation. Increasing the excitation from below to above the transition density, we identify two distinctly different relaxation pathways of the THz resonance with opposite dynamics. A model calculation of transient spectra incorporating the first order diffraction mode from the photoinduced transient grating reproduces the salient features, which is further corroborated by their dependence on the unit cell lattice constant. The revealed scheme represents a relatively simple and generic approach to achieve nonlinear and frequency-agile functions in THz device without particularly complex structures.

The samples used are double SRRs on GaAs, illustrated in the inset of Fig. 1 (a), which are patterned 6 \( \mu \text{m} \) copper rings on high resistivity GaAs of 630 \( \mu \text{m} \) thick. We mainly focus on two SRR samples with unit cell lattice constant of 50 \( \mu \text{m} \) and 45 \( \mu \text{m} \), and the outer dimension of an individual SRR is 36 \( \mu \text{m} \). Our optical pump and THz probe spectroscopy setup is driven by a 1 kHz Ti:sapphire regenerative amplifier with 40 fs pulse duration at 800 nm. One part of the output is used to excite the sample, while a small fraction is used to generate and detect THz pulses. Phase-locked THz field transients are used as a probe, which is generated via optical rectification and detected by electro-optic sampling in 1 mm ZnTe crystals (supplementary material). Copper apertures of 2.5 mm are placed in front of the sample and a GaAs reference to ensure a uniform illumination and faithful comparison. The THz field transmis-
The static transmission spectra are shown in Fig. 1(a) (black dotted) with a THz probe normally incident on the sample plane. The THz probe is parallel to the magnetic dipole resonance induced by the circulating electric currents generated by the incident electric field.

The inset: the SRR unit cell of our sample and the polarization of the normally incident THz radiation. $a=50 \text{ mm}$, $b=36 \text{ mm}$, $d=3 \text{ mm}$, $g=2 \text{ mm}$ and $w=6 \text{ mm}$. (b): The non-monotonic pump fluence dependence (indicated by the gray arrow) of the resonant absorption ~1.63 THz for low (dashed lines) and high (solid lines) photoexcitation. Spectra in both panels are taken at fixed time delay of 20 ps.

The weak and strong photoexcitations lead to significantly different relaxation pathways back to the equilibrium, as shown in the time evolution of $T(\omega)$ in the photoexcited metamaterial of 50$\mu$m lattice constant for (a) 4 $\mu$J/cm$^2$ and (b) 70 $\mu$J/cm$^2$, respectively. These clearly show the opposite relaxation pathways for the weak and strong excitation regimes (see text).

FIG. 2. (color online) Time evolution of negative transmission spectra, $-T(\omega)$, in the photoexcited metamaterial of 50$\mu$m lattice constant for (a) 4 $\mu$J/cm$^2$ and (b) 70 $\mu$J/cm$^2$, respectively.
FIG. 3. (color online) (a) The extracted transient carrier concentration as function of time delay. The left and right gray areas illustrate a crossover from the high to low excitation regime with a crossover density \( N_c \) (black square). The inset plots the extracted transient carrier concentration as function of pump fluence at \( \Delta \tau =20 \) ps, marked together with \( N_c \) corresponding to the density excited by 19 \( \mu \)J/cm\(^2\). (b)-(e): The transient transmission spectra \( T(\omega) \) at various time delays following 70 \( \mu \)J/cm\(^2\) photoexcitation.

sient carrier density. The phase-sensitive nature of the time-domain THz field measurement can directly yield both real and imaginary parts of the optical conductivity in the photoexcited GaAs substrate, on equal footing without any model assumptions. This gives a direct determination of the transient carrier density based on an analysis using the Drude model (see supplementary material). Figure 3(a) plots the temporal evolution of the transient carrier concentration excited by 70 \( \mu \)J/cm\(^2\), which shows an exponential decay with a peak density \( 10^{17} \) cm\(^{-3}\). By correlating the temporal profile to the fluence-dependent carrier concentration at \( \Delta \tau =20 \) ps (inset, Fig. 3(a)), one can define a crossover point at density \( N_c \sim 4.7 \times 10^{16} \) cm\(^{-3}\) at time delay of \( \Delta \tau_c \sim 1.73 \) ns (black square), corresponding to the transient density for 19 \( \mu \)J/cm\(^2\). This naturally divides the transient electronic response of photoexcited metamaterials into two regimes with opposite time and excitation dependence, as illustrated in the shaded areas of Fig. 3(a). Transient spectra at various time delays, as shown in Figs. 3(b)-(e), show a reversal of the THz transmission change once the system is driven cross the boundary, corroborating well with our conclusion, e.g., the THz transmission recovers its strength at \( \Delta \tau =2.2 \) ns after initial quenching at time delays before 1.73 ns.

Next, we analyze the transient THz response of photoexcited metamaterials by full-wave numerical simulations. Before we take into account any complex processes, we first simply consider the photoexcitation-induced dynamic response of the GaAs substrate beneath the SRRs. A photoexcited layer is modeled by the well-established Drude model, in which, the frequency-dependent complex conductivity \( \sigma = \varepsilon_0 \omega_p^2/(\gamma - i \omega) \). Here, \( \varepsilon_0 \) is the permittivity of vacuum, \( \gamma \) represents the collision frequency, and \( \omega_p = \sqrt{N e^2/\left(\varepsilon_0 m^*\right)} \) the plasma frequency. Some other parameters include the carrier density \( N \), the free electron charge \( e \), and the effective carrier mass in GaAs, \( m^* = 0.067 m_0 \) (\( m_0 \), the mass of a free electron). Thus, the photoexcited layer has the dielectric function \( \varepsilon(\omega) = \varepsilon_s + i \sigma/(\varepsilon_0 \omega) \), where \( \varepsilon_s \) (=12.7) is the dielectric constant of undoped GaAs. In our simulations, we keep \( \gamma = 1.8 \) THz for all the cases as in Ref. 17.

Figure 4(a) shows the simulated transmission spectra for various carrier density \( N \). The simulation clearly shows the crossover from a reduction to an enhancement of the resonant THz absorption \( \sim 1.6 \) THz as the excitation from low to high density regime. This agrees well with the photoinduced reversible modulation demonstrated in the experiments. Specifically, in the low density below \( 4.0 \times 10^{16} \) cm\(^{-3}\), the THz resonance peak in the transmission curve exhibit clearly weakened transition strength, a blue-shift and spectral broadening. Increasing the carrier density above \( 4.0 \times 10^{16} \) cm\(^{-3}\), the resonant THz peak progressively reemerges, e.g., a clear transmission dip seen for the case \( N = 8.1 \times 10^{17} \) cm\(^{-3}\).
From the simulations, we can attribute the reemergence of the resonant THz absorption at high density to the effect of first-order diffractive waves from photoinduced transient grating. Due to the periodic structure of the SRRs, the first-order diffractive wave is found to propagate in the substrate at high excitation, which strongly influence the fundamental absorption mode. This leads to a new THz transmission dip at \( f_1 = c/(na) \) above a transition density \( \sim 4.0 \times 10^{16} \text{ cm}^{-3} \). Here \( c \) is light speed in vacuum, \( n \) the refractive index of GaAs and \( a \) the lattice constant of the structure. The SRR structure with the dipole resonance close to \( f_1 \) allows to achieve a reversal of resonant THz absorption with increasing photoexcitation, due to a crossover in the absorption from the dipole mode to the diffraction mode.

To corroborate our model, we compare the re-emerged THz resonances from the experiment (Fig. 4(b)) to those from the simulation (Fig. 4(c)) for two different lattice constants, 50 \( \mu \text{m} \) and 45 \( \mu \text{m} \), respectively. The simulations are performed to the structures with the same double-SRR array for the case with the density \( N = 8.1 \times 10^{17} \text{ cm}^{-3} \). It is clearly visible that the simulated transmission dip to higher frequency by decreasing the lattice constant, and the dip positions tightly link to the frequency of the first-order diffraction mode for each case. An extremely good agreement between the simulation and the experiment is found for the transmission-dip position and its shift, which underpins the effect of the high-order diffraction mode in the strongly excited metamaterials.

In summary, we have demonstrated ultrafast photoinduced reversible modulation of resonant THz absorption in strongly photoexcited metamaterials. Increasing the excitation from below to above the threshold density \( N_c \), we observe a crossover from a complete quenching to a reemergence of the THz resonance. Our analysis and theoretical simulations, based on the first order propagation from the photoinduced transient grating, explain the density- and lattice-constant-dependent frequency shift. Our results clearly identify the importance of photoinduced optical modes in metamaterials, besides the electronic resonances, which should be carefully considered in designing future multifunctioning photonic-electronic devices using the artificial periodical structures. The easy implementation of the revealed scheme represents another approach to achieve nonlinear and frequency-agile functions in THz devices.

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