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Ultrafast optical control of terahertz surface plasmons in subwavelength hole arrays at room temperature

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We demonstrate optical control of surface plasmon enhanced resonant terahertz transmission in two-dimensional subwavelength metallic hole arrays fabricated on gallium arsenide based substrates. Optically pumping the arrays creates a conductive layer in the substrate, reducing the terahertz transmission amplitude of both the resonant mode and the direct transmission. Under low optical fluence, the terahertz transmission is more greatly affected by resonance damping than by propagation loss in the substrate. An ErAs:GaAs nanoisland superlattice substrate is shown to allow ultrafast control with a switching recovery time of ~10 ps. © 2009 American Institute of Physics. [DOI: 10.1063/1.3168510]

Extraordinary optical transmission through subwavelength metallic hole-arrays has been an active research area since its initial demonstration.¹ Optical transmission efficiency of such arrays can be higher than unity when normalized by the holes’ areas and this anomalously high transmission has been explained as a result of resonance excitation of surface plasmon polaritons (SPP) or surface plasmons at the metal-dielectric interface. Surface plasmons have already been demonstrated to have high impact in integrated photonics, nanolithography, tunable filters, nonlinear optics, and biosensors.²–⁶ The scalability of the surface plasmon allows it to operate at almost any desired frequency, including terahertz,⁷–¹¹ by appropriate design of the hole shape and periodicity. Besides the interesting physical phenomena, a large portion of terahertz surface plasmon research is aimed at terahertz device applications. The frequency selective resonance properties of subwavelength metallic hole arrays and metamaterials have potential use in functional terahertz devices such as filters, modulators, and switches.¹²–¹⁴ Ultrafast switching or modulation of the resonant behavior of the terahertz through metallic hole arrays is of particular interest for high speed communication and sensing.

Recently, switching of the SPP-assisted resonant terahertz transmission has been demonstrated using magnetic, electric, optical, and thermal techniques.¹²,¹³,¹⁵–¹⁷ In these demonstrations, either the dielectric properties of the substrates or the metallic hole arrays were modified by external controls. For example, the conductivities of the semiconductor hole arrays were controlled by either temperature or the applied optical excitation. In another demonstration, a magnetic field was applied to change the dielectric constant of the liquid crystal in the metal array.¹⁶ Most of these methods do not support modulation on an ultrafast time scale. Hendry et al.¹⁸ has applied an optical technique on a silicon-based metallic grating that supports fast modulation but only at cryogenic temperatures (20 K). Also, their resonance recovery, or switching time, was about 100 ps at 20 K. In the case of optical modulation, switching time is mainly determined by the carrier lifetime of the substrate semiconductors. At room temperature, bulk silicon has a carrier lifetime of about a millisecond which severely degrades switching speed. In this work, we present the room temperature, ultrafast modulation of SPP-assisted terahertz transmission through a metallic subwavelength hole array fabricated on a ErAs:GaAs superlattice substrate using ultrafast optical excitation. Due to the direct bandgap at 800 nm and the short carrier lifetime of this superlattice, the SPP resonance can be modulated with much lower optical fluence than in Ref. ¹⁸ and can fully recover within ~10 ps at room temperature.

The first sample used in this work was a two-dimensional (2D) metallic hole array fabricated on semi-insulating (SI) GaAs with a carrier lifetime on the order of nanoseconds. The second sample used the same metallic hole array fabricated on a superlattice consisting of alternating layers of GaAs and ErAs nanoislands grown on SI-GaAs. Both substrates have a direct band gap across which carriers can be excited by 800 nm light. Standard photolithography was used to fabricate the aluminum (200 nm thick) hole arrays on the substrates. The cross-sectional view of the samples is shown in Fig. 1. The rectangular holes of dimension 50×25 μm² were patterned in a square array with a lattice constant of 100 μm. The transmission of terahertz radiation through the metallic hole arrays was measured using a millisecond which severely degrades switching speed. In this work, we present the room temperature, ultrafast modulation of SPP-assisted terahertz transmission through a metallic subwavelength hole array fabricated on a ErAs:GaAs superlattice substrate using ultrafast optical excitation. Due to the direct bandgap at 800 nm and the short carrier lifetime of this superlattice, the SPP resonance can be modulated with much lower optical fluence than in Ref. ¹⁸ and can fully recover within ~10 ps at room temperature.

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![FIG. 1. (Color online) Cross-sectional schematics of the metallic hole array samples on GaAs and ErAs:GaAs substrates. The substrates in the hole areas, labeled “Photo-conductive layer,” become conductive under illumination.](image-url)
ing terahertz time domain spectroscopy (TDS).

The terahertz TDS used in this experiment utilizes a 1 kHz regeneratively amplified Ti:sapphire laser capable of generating 3.2 mJ, 50 fs pulses at 800 nm. Part of the output laser power is used for terahertz generation and detection using ZnTe crystals via optical rectification and the electro-optic effect. The remainder of the optical power passes through a variable attenuator to form a ~10 mm diameter illumination spot on the sample with an incidence angle of ~10°. Figure 2(a) shows the terahertz transmission, \( |T(\omega)| = |E_{\text{trans}}(\omega)/E_{\text{ref}}(\omega)| \), through the metal hole array on GaAs substrate at various pump fluences normalized to the transmission of a blank unexcited GaAs reference slab. The resonance frequency of the SPP modes of the metallic hole array at normal incidence is approximately given by

\[
\nu_{\text{res}} = (c/L)\sqrt{(n^2 + n_s^2)/\varepsilon_s},
\]

where \( L \) is the period of the hole array, \( \varepsilon_s \) is the dielectric constant of the substrate, \( m \) and \( n \) are mode indices, and \( c \) is the speed of the light in vacuum. Transmission measurements show pronounced surface plasmon resonances of the [1, 0] and [1, 1] modes at 0.8 and 1.12 THz, respectively. In addition to the SPP modes, we also observed periodic oscillation artifacts in the measured transmission spectra due to the truncation of the time domain signals. The truncation procedure was necessary to remove terahertz pulse echoes, arising from the finite thickness of our substrate. These echoes appear before the complete decay of the SPP resonances, which are quite long-lived.\(^7\)

Without optical excitation, the peak transmission of the [1, 0] SPP resonance is 37% relative to the blank unexcited GaAs slab. The transmission drops under photoexcitation because of the creation of a photoconductive layer on the top of the GaAs substrate. The thickness of the photoexcited layer on GaAs for 800 nm light is ~0.8 \( \mu \)m. The dc conductivity of the photoexcited GaAs layer for a fluence of 16 \( \mu J/\text{cm}^2 \) was estimated at about 200 (\( \Omega \) cm)\(^{-1} \) and decays with time.\(^9\) The transmission at the resonance drops to 7.5% with an excitation fluence of 8 \( \mu J/\text{cm}^2 \). The intensity modulation depth can be defined as \( (T_0^2 - T_{\text{Pump}}^2)/T_0^2 \) where \( T_0 \) and \( T_{\text{Pump}} \) are the amplitude transmission without and with pump. We obtained over 95% modulation depth with an optical fluence of 8 \( \mu J/\text{cm}^2 \). We also performed terahertz transmission through a blank GaAs slab as a function of the pump fluence. Figure 2(b) shows the comparison of the fluence dependent transmission of metal hole array on GaAs (dotted line) and blank GaAs slab (solid line) at 0.8 THz. A biexponential decay function is used to fit the measured data. The normalized transmission through the photoexcited slab exhibited a broadband and flat attenuation as shown in the inset of Fig. 2(b). Measurements show that within the low fluence limit (in our case <15 \( \mu J/\text{cm}^2 \)), the transmission amplitude of the hole array sample decays faster than the broadband transmission in the blank substrate. This suggests that SPP resonance damping plays an important role in modulation beyond simple propagation and reflection losses involved in the photoconductive layer. Apparently, only weak illumination is required for the photoconductive layer to damp the SPP resonance across the holes. However, at higher fluence, the GaAs behaves as a metallic substrate and the terahertz transmission through the metallic hole arrays and the bare GaAs both suffer high attenuation. We have not observed any pump induced shift in the resonance frequency, which depends on the permittivity of the dielectric medium. Upon photoexcitation, the permittivity of the dielectric medium changes within the hole region, however, it remains unchanged at the metal-dielectric interface (outside the holes) where the SPP resonance frequency is determined. Further, the thickness of the photoexcited layer inside the holes is small compared to the penetration of the SPP electric field, thus resulting in insignificant shifts in resonance frequencies. Due to the direct bandgap, GaAs requires low fluence for the complete switching compared to the fluence used in Ref. 18 for a grating on a silicon substrate.

We also measured the terahertz transmission through a metallic hole array fabricated on an ErAs:GaAs superlattices, which have already proven to benefit many ultrafast applications such as terahertz photoconductive receivers, fast switchable terahertz metamaterials, etc.\(^9\) Their carrier lifetime can be engineered from hundreds of femtoseconds to tens of picoseconds by changing the superlattice period.\(^9\) Our ErAs:GaAs superlattice layer was 2 \( \mu \)m thick to ensure complete absorption of the incident 800 nm light which has a penetration depth of \( \delta_{\text{800 nm}} \approx 0.8 \mu \)m (Ref. 19) and it had a designed carrier lifetime of ~8 ps. The fluence dependent terahertz transmission through the metal hole arrays on ErAs:GaAs is shown in Fig. 3. During these measurements, the optical pump delay was carefully timed to ensure maximum modulation of the transmitted terahertz pulses. Fluence dependent measurements are similar to those of metal hole array on Si-GaAs. A thin layer of the photoconductive ErAs:GaAs damped the surface plasmon across the holes, diminishing the resonance amplitude. The penetration depth of the terahertz SPP inside the photoexcited ErAs:GaAs layer depends on the pump fluence, and for an optical excitation of

![Graph showing terahertz transmission spectra through hole array on GaAs.](image-url)
16 μJ/cm², our estimated penetration depth is $d_{SPP} \approx 5 \, \mu m$ (Ref. 22). We obtained almost 90% modulation depth of the transmitted terahertz intensity when excited with a pump fluence of 8 μJ/cm². Resonance amplitudes kept decreasing with increasing fluence until we observe a complete switch-off of the transmitted terahertz resonance for an optical fluence of $\sim 32 \, \mu J/cm²$.

The ultrafast switching recovery of the surface plasmon resonance in the metal hole array on the ErAs:GaAs superlattice substrate is demonstrated through the measurements of the terahertz transmission as a function of the time delay after photoexcitation. The experimental results are shown in Fig. 4 with an optical excitation fluence of 8 μJ/cm². It clearly reveals the annihilation and then fast recovery of the resonance at 0.8 THz (as well as the higher order resonances). Transmission shows a pronounced resonance peak of amplitude $\sim 0.34$ before the arrival of the optical pulses. It is then strongly attenuated within 1 ps after the optical pulse arrival, but eventually recovers as the time delay increases; after 10 ps the resonance peak amplitude becomes 0.32. We observed a slight reduction in the resonance peak amplitude at $\sim 1.5$ ps compared to that of an unexcited sample shown in Fig. 3. This small reduction is within the limit of our experimental errors and might be due to the presence of photocarriers arising from optical pulse’s prepulse. Unlike bulk GaAs, which has a carrier lifetime of $\sim 2.1$ ns (Ref. 23), the superlattice shows a fast carrier relaxation because ErAs nanoislands trap photocarriers immediately, long before they complete the recombination process. The measurement shows that the resonance recovery time is $\sim 10$ ps, approximately the carrier lifetime in the ErAs:GaAs superlattice. This fast recovery of the resonance demonstrates the ability of such metallic hole arrays to work as ultrafast modulators at room temperature. Shortening the carrier lifetime could further reduce this resonance recovery time.

In conclusion, we have demonstrated ultrafast switching of the SPP resonance on 2D metallic hole arrays. The ErAs:GaAs sample shows an intensity modulation depth of $\sim 88\%$ with an optical pump fluence of 8 μJ/cm² and a SPP resonance recovery (switching) time of $\sim 10$ ps. Our data suggests that SPP resonance damping enhances modulation in the low fluence limit compared to simple propagation and reflection losses in the photoconductive layer. We point out that our hole array on ErAs:GaAs requires an order of magnitude lower optical fluence compared to the work in Ref. 18 and room temperature operation supports future applications.

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