Ultrafast Pump-Probe Spectroscopy of a Dual-Band Negative-Index Metamaterial

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Abstract

In this paper we study the nonlinear optical response of a dual band negative index metamaterial device with two color pump probe spectroscopy. We measure and numerically reproduce its properties versus probe wavelength, pump fluence and probe polarization. Thereby, we demonstrate its utility as a nanoscale, structurally tunable, subpicosecond all-optical modulator.

1. Introduction

Artificially structured composites, termed metamaterials, consist of sub-wavelength sized structures that exhibit unusual electromagnetic properties not found in nature [1]. While much work has been done in studying their linear properties, the nonlinear optical properties of these structures also offer exciting possibilities, e.g. high-speed modulation & frequency tuning [2]. Here, we fabricate a fishnet structure metamaterial device (an Ag/amorphous-Si (α-Si)/Ag tri-layer perforated by a 2D square periodic array of holes (Fig. 1a) with two negative index resonances [3] at 1.06 μm and 1.58 μm. The fundamental long wavelength resonance is related to the periodicity (*p*) while the secondary short wavelength resonance is related to the diagonal periodicity ($\sqrt{2}\, p$). By photoexciting carriers into the dielectric α-Si layer of the metamaterial, we alter its characteristic response on a sub-picosecond timescale. Then, by probing around the two resonances, we study its non-linear response (Fig. 2c). For a given probe wavelength, polarization and pump fluences of ~1 mJ/cm^2 (experimentally limited), we see pump-probe signals ($\Delta T/T$) as large as 70%. The 600 fs switching time – two orders of magnitude faster than other all-optical devices, large switching ratio and 124 nm thickness demonstrates the utility of our device as a nanoscale, structurally tunable, sub-picosecond all-optical modulator [3]. We numerically reproduce the nonlinear behavior of our device using the Drude conductivity model and a finite integration technique over wide spectral and pump fluence ranges. Thereby, we show that beyond the linear properties of the device, the magnitude of the pump-probe response is completely described by only two material parameters. These results provide insight into engineering various aspects of the nonlinear response of fishnet structure metamaterials.

2. Design and Fabrication of Device

The dual-band metamaterial device reported here is composed of a BK7 glass substrate and a single metal-dielectric-metal (Ag/α-Si/Ag) functional layer with an inter-penetrating two-dimensional square array of elliptical apertures. The geometrical parameters of the device are indicated in Fig. 1a. The orthogonal pitches of the two-dimensional grating are both fixed at 345 nm (p). The thickness of the silver (Ag) and amorphous silicon (α-Si) films are fixed at 28 nm and 68 nm, respectively. The elliptical aperture size ($2a_x$, $2a_y$) is formed with lengths of 0.69·p (238 nm) and 0.47·p (162 nm) in the major and minor axis, respectively as shown in Fig. 1a.

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The normal incidence transmission through the metamaterial device was recorded with a Nicolet Fourier transform infrared (FTIR) spectrometer (Fig. 1b). To understand the linear optical properties of our device, we performed simulations using CST Microwave Studio [18] based on a finite integration technique. We assumed perfect electric conductor (PEC) and perfect magnetic conductor (PMC) boundary conditions between unit cells to stimulate transverse electromagnetic (TEM) plane wave propagation in the z direction. The optical parameters for the constitutive materials were taken as $n_{\text{substrate}} = 1.5$ and $n_{\text{MgF2}} = 1.38$, where MgF_2 is used as the protection layer. The refractive index of α-Si was taken as $n_{\alpha\text{-Si}} = 3.35$. A Drude model for the Ag dielectric function was used with ω_p (plasma frequency) = 9.02 eV and γ_c (scattering frequency) = 0.042 eV. This scattering frequency is increased by a factor of two compared to that of bulk silver to account for additional scattering mechanisms in this polycrystalline thin film, in addition to sample inhomogeneity across the ~mm^2 measurement area associated with the FTIR beam. These parameters produce the best fit between experiment and simulation of the transmission through our device over the wavelength range of interest (1 to 2 μm), as shown in Fig. 1b.

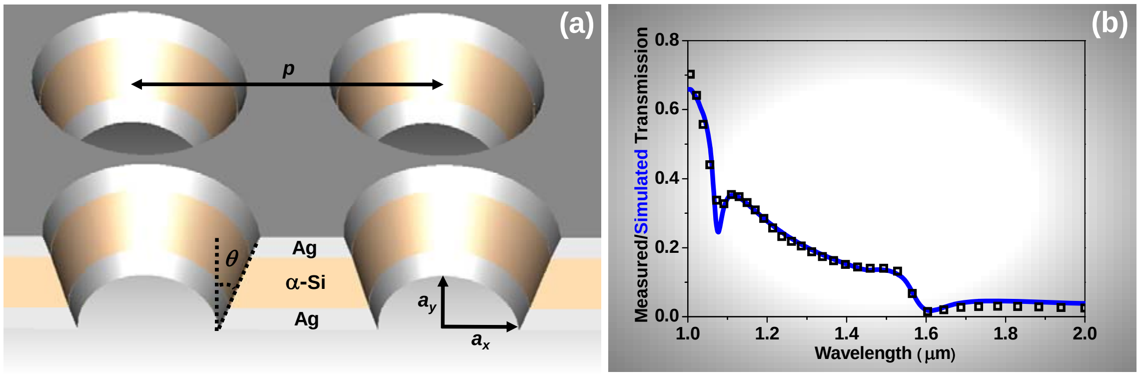

Fig. 1: (a) Schematic view of our negative-index metamaterial (NIM). Geometrical parameters of NIM: $p = 345$ nm; $a_x = 119$ nm; $a_y = 81$ nm; and θ (sidewall-angle) = 18°. The NIM is composed of 28 nm silver (Ag), 68 nm α-Si, and 28 nm Ag layers. The incoming light is polarized parallel to the x-axis. (b) Measured FTIR and 3-dimensional FDTD simulated transmission spectra.

3. Experimental Setup and Results

The ultrafast nonlinear optical response of the device is studied using a sub-100 fs, 590 nm visible pulse to photoexcite carriers above the α-Si bandgap (~1.7 eV), and a near-IR probe pulse to measure the time-resolved change in transmission (ΔT/T), i.e. the switching ratio. We measure ΔT/T as a function of pump-probe delay (Δt), pump fluence, probe wavelength (1.0 – 2.0 μm), and probe polarization. Figs. 2a, b show the switching ratio (ΔT/T) versus Δt for different probe wavelengths at the secondary and fundamental resonances, respectively. In Fig. 2c, we plot the zero-delay switching ratio versus probe wavelength, showing an S-shaped response versus probe wavelength. Fig. 2d shows the peak ΔT/T versus pump fluence for the two resonances at 1640 nm and 1010 nm. Both resonances exhibit an identical square root scaling versus pump fluence. The nonlinear optical response of the metamaterial device can be understood as a change in effective parameters of the metamaterial due to the presence of photo carriers in the α-Si dielectric layer [3]. We model the refractive index of the photoexcited α-Si with the Drude model, and thus numerically reproduce the nonlinear optical response of the metamaterial device. Our simulations show that the nonlinear response is completely determined by the effective mass and Drude scattering rate of the photocarriers in α-Si.

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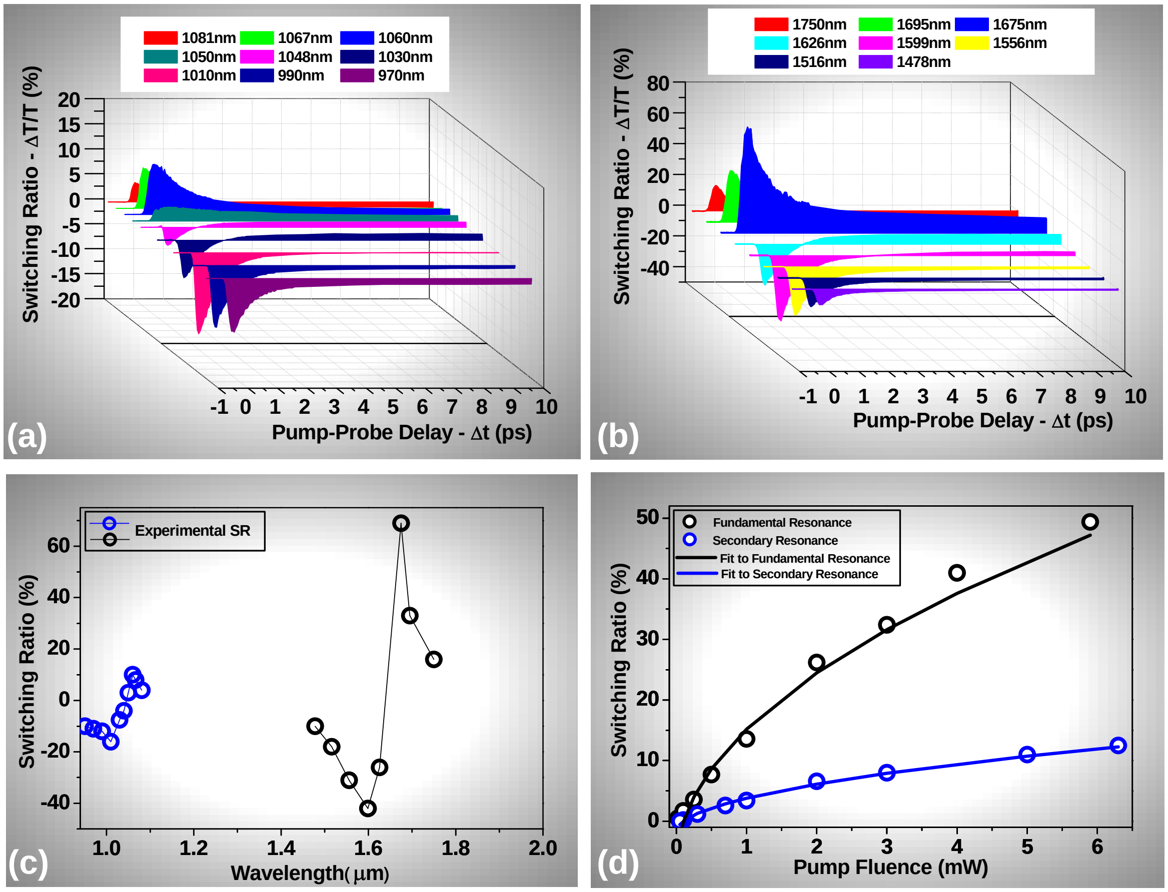

Fig. 2. Measured pump-probe response (ΔT/T) versus pump-probe delay (Δt) for different probe wavelengths around the secondary (a) and fundamental (b) resonances. (c) Zero delay (ΔT/T) versus probe wavelength. We see the S-shaped nonlinear response at the negative index resonance. (d) Square root scaling of ΔT/T versus pump fluence at fundamental (black) and secondary (blue) resonance.

4. Conclusion

In conclusion, we perform two-color pump-probe spectroscopy on a dual-band negative index fishnet structure versus probe wavelength, polarization and pump fluence. With a 70% switching ratio (experimentally limited) and 600 fs switching time, this device performs as a nanoscale, structurally tunable, ultrafast all-optical modulator. We understood and numerically reproduced the experimental data thereby providing deeper insight into the ultrafast nonlinear properties of fishnet metamaterials and their utility for near-IR photonic devices like ultrafast optical switches.

References

[1] Vladimir M. Shavaev, Optical negative-index metamaterials, *Nature Photonics*, vol. 1, p. 41-48, 2007.
[2] Hou-Tong Chen, John F. O'hara, Abul K. Azad, Antoinette J. Taylor, Richard D. Averitt, David B. Shrekenhamer, and Willie J. Padilla, Experimental demonstration of frequency-agile terahertz metamaterials, *Nature Photonics*, vol. 2, p. 295-298, 2008.
[3] Keshav M. Dani, Zahyun Ku, Prashanth C. Upadhya, Rohit P. Prasankumar, S. R. J. Brueck, and Antoinette J. Taylor, Subpicosecond Optical Switching with a Negative Index Metamaterial, *Nano Letters*, vol. 9, p. 3565-3569, 2009.