Terahertz Plasmonic Random Metamaterial

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***Abstract*—We explore the terahertz electromagnetic properties of a composite metamaterial consisting of subwavelength-sized dielectric particles with metallic inclusions. We observe group refractive index exhibits atypical which cannot be explained by conventional effective medium theory. It is shown to arise from near-field particle-plasmon coupling between the metallic particles. Moreover, we provide evidence of electron spin-dependent terahertz plasmonic transport through both ferromagnetic and ferromagnetic/nonmagnetic metamaterials.**

I. Introduction And Background

Electromagnetic interaction with a medium is dependent on the ratio of its dimensions to the wavelength of radiation. As the relevant dimensions of the medium approach the mesoscopic regime, the electromagnetic properties of the medium can be different from the bulk electromagnetic properties of the constituent material. To date, the interaction of electromagnetic radiation with metamaterials so far has focused on investigating artificially fabricated, subwavelength-ordered structures, such as split-ring resonators [2] and some ordered metamaterials exhibiting negative refraction [3]. However, a general and a more interesting class of metamaterial would be a random one. An important question to be raised: is order or periodicity a prerequisite for observation of peculiar electromagnetic properties in metamaterials?

First, we explore the terahertz electromagnetic properties of a simple composite metamaterial consisting of a random ensemble of subwavelength-sized dielectric particles with metallic inclusions. By adding metallic particles to the dielectric ensemble, plasmonic interactions amongst the metallic elements lead to a marked atypical refractive index and thus group velocity relative to that in the constituent dielectric ensemble.

Next, we introduce new phenomena in the interaction of THz radiation with magnetic metals, giving rise to a new field of Spinplasmonics [4,5]. We show that the THz electromagnetic properties of metallic particle ensembles can be modulated via electron spin-dependent plasmons, analogous to optical and solid state systems. Here, the ferromagnetic particle ensembles can exhibit strong, tunable birefringence at THz frequencies with the application of an external magnetic field. The birefringence is caused by anisotropic magnetoresistance inherent in the ferromagnetic medium. Secondly, another method to achieve magnetically tunable transmission is via electron spin-dependent phenomena in ferromagnetic/nonmagnetic composite media. We demonstrate that an ensemble of ferromagnetic particles coated with nonmagnetic nano-layers shows enhanced magnetic attenuation, delay and pulse re-shaping. The enhanced magnetic effect in the coated particles is attributed to electromagnetically driven electron spin accumulation in the nonmagnetic layer.

II. Results and Discussion

We use THz time-domain spectroscopy to characterize the on-axis and off-axis THz transmission through the metamaterials.

(a) *The plasmonic metamaterial*. Here, the metamaterial employed is made up of a 4.0 mm thick mixture of semi-spherical Co metallic microparticles having a mean dimension, a ~ 74 ± 23 μm, and spherical sapphire dielectric microparticles having a diameter ~ 100 ± 12 μm (Fig. 1). Since skin depth for Co at 1 THz is ~ 100 nm << a, the electromagnetic response of the composite medium cannot be described by effective medium theory.

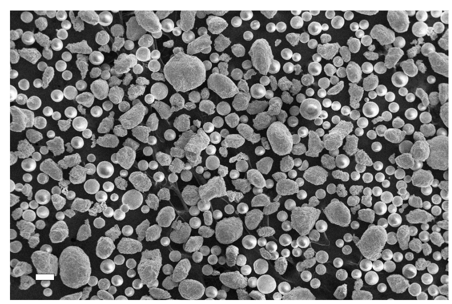

Figure 1. A scanning electron image of a Co/sapphire random metamaterial. The scale represents 100 μm.

Shown in Fig. 2 are time-domain THz electric field waveforms transmitted through the mixture material. When the metallic particle volume fraction, *f*, is increased from *f*=0 to *f*=100, the transmitted pulse amplitude is increasingly attenuated, and the transmitted pulse width is significantly broadened. It is apparent that the addition of metallic particles above *f* = 67 % causes *advancement* of the pulse arrival time (relative to *f* = 0%). However, by examining the arrival times of the intensity peaks, we find as *f* is increased from 0% to 40 %, the pulse is delayed by approximately 1 ps relative to that at *f* = 0 % and that at *f* = 100 % the transmitted pulse intensity is advanced by approximately 2 ps relative to that at *f* = 0 %. The temporal delay is attributed to increased optical path via

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scattering from both the dielectric and metallic particles, while, effectively, the addition of opaque, metallic particles to the metamaterial *reduces* the optical path length through the metamaterial via particle plasmons interaction.

From such electric field waveforms, the refractive index of the material can be obtained. In particular, for f =20% the effective refractive index of the medium is measured to be =1.92 which is higher that either a pure dielectric particle (n_{eff}^{d} = 1.81) or the metallic particle (n_{eff}^{m} = 1.66) ensemble. Clearly, the random metamaterial exhibits atypical refractive index. Alternatively, propagation velocity of the THz pulses through the metamaterial is markedly enhanced from 0.52c to 0.58c as shown in Fig. 3. This group velocity enhancement coincides with a regime where mutual interactions between metallic particles come into play. By exploring the influence of interparticle separation on the group velocity, we conclude that for f> 27.0 % near-field particle-plasmon coupling between nearest neighbor metallic particles is the dominant plasmonic interaction mechanism in the metamaterial. Furthermore, by applying a magnetic field to the plasmonic metamaterial, the arrival time and phase of the THz pulses can be controlled in a precise fashion.

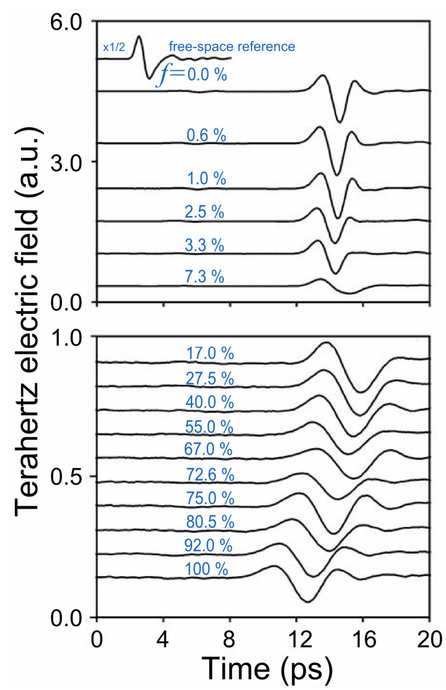

Figure 2. (a) Time-domain waveforms transmitted through an empty sample cell and 4.0 mm thick Co/sapphire particle mixtures for Co f from 0.0 % to 7.3 %. (b) Time-domain waveforms transmitted through the Co/sapphire particle mixtures for percentage volume Co varying from 17% to 100%.

(b) *Spinplasmonics*: The ferromagnetic/nonmagnetic (F/N) metamaterial employed is made up of a 3.0 mm thick of semi-spherical Co metallic microparticles coated with Au nanolayers (Fig. 4). The 45 ± 2 nm thick Au layers are sputter deposited on Co particles ensembles having dimensions ranging from 40 µm to 140 µm and an average dimension of 74 ± 23 µm. A single deposition coats the exposed regions of the Co particles with a thin Au layer.

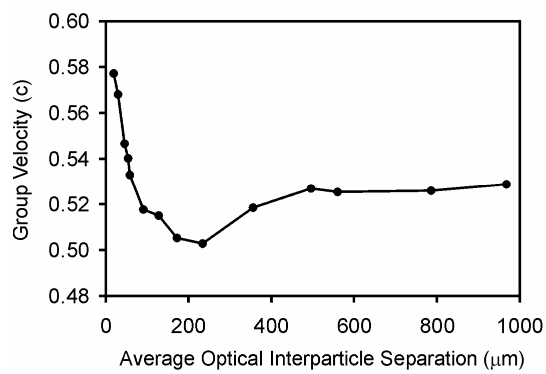

Figure 3. Group velocity versus the average optical separation between metallic particles.

The distributed Au coverage is increased by re-orienting the particle collections after each deposition to expose fresh, uncovered Co particles and re-depositing another 45 ± 2 nm thick Au layer. This procedure is repeated to achieve the desired Au coverage of the particle ensemble. X-ray photoelectron spectroscopy analysis reveals that the percentage Au surface compositions of the three samples are 35 ± 2 %, 42 ± 2 % and 77 ± 2 %.

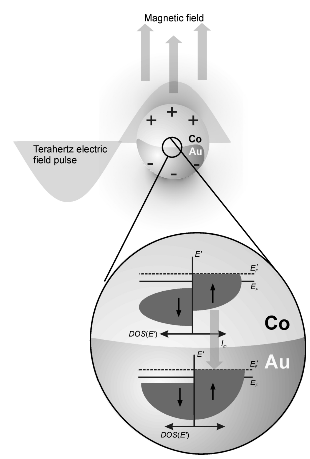

Figure 4. Illustration of a subwavelength spinplasmonic structure consisting of a Co particle coated with Au nano-layers excited with a THz electromagnetic field pulse. The spintplasmonic structure is immersed in a magnetic field so as to align the magnetic domains of the Co particle. The incident electromagnetic pulse induces spin-polarized current from the Co particle to the Au layer, creating a spin-dependent interface resistance.

First, the role of the bimetallic F/N interface on the transmission through the Co/Au particles in zero field is examined. The field, $E_{tr}(t)$, transmitted through Co particle samples having 0 %, 35 %, 42 % and 77 % Au surface coverages (Fig.5) indicates that the Au layer causes drastic

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reduction of the transmissivity independent of magnetic field or electron-spin effects. As Au coverage increases from 0 to 77 %, the amplitude is attenuated by 91 ± 3 %. The attenuation cannot arise from the inherent resistivity of the Au layer, which is much less than that of Co. The marked attenuation that increases with increasing Au coverage is attributed instead to the interface resistance at the Co/Au boundary, which is proportional to the coverage area.

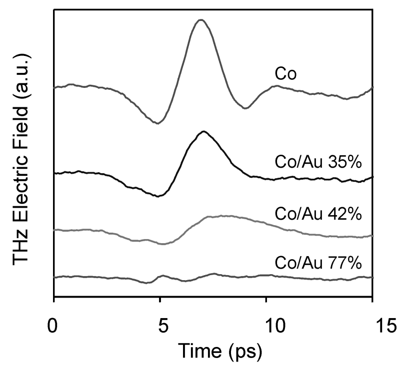

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Figure 5. Time-domain THz transmission signals through 3 mm thick particle ensembles having varying Au coverage, in zero field

The effects of B on the THz waveforms (E_{tr} (t)) transmitted through the composite F/N particles are examined next. Analogous to electrically-driven current in multilayer F/N systems, the electromagnetically driven surface charge on the F/N particles exist in a medium where the resistivity is magnetically influenced via anisotropic magnetoresisitance (AMR) of the Co, spin accumulation effects through Co/Au interface, or both. These contributions can be discriminated by varying the orientation of B. Fig. 6(a) and 6(b) show that the transmission through the Co (0% Au) sample exhibits attenuation and delay due to AMR. In a field $B_{\perp}$ perpendicular to the electric field polarization [Fig. 6(a)] negligible attenuation and negligible pulse re-shaping are observed whereas both substantial amplitude reduction and pulse re-shaping are observed in a parallel field $B_{\|}$ [Fig. 6 (b)].

The THz pulse transmission is studied through Co and Co/Au particles in $B_{\|}$ where both AMR and spin accumulation effects may occur. Figure 6(d) and 5(f) show E_{tr} (t) in the presence of a dc magnetic field $B_{\|}$ = 150 mT (yellow traces) in comparison with E_{tr} (t) in the absence of a magnetic field (blue traces). Pulses transmitted through the Co/Au 35% sample, depicted in Fig. 6(d), show increased magnetically induced attenuation and delay relative to the Co (0% Au) sample [Fig. 6(b)] when $B_{\|}$ is applied. Similar behavior with greater magnetically induced attenuation and delay is seen in the data for the Co/Au 42% sample [Fig. 6(f)]. These B-dependent trends of transmission amplitude and delay for $B_{\|}$ are quantified further. Referring to Fig. 6 (a), for an increase in $B_{\|}$ from 0 mT to 150 mT, $|E_{tr}$ (t)$|$ decreases by 28 ± 3 %, 54 ± 3%, and 73 ± 3% for the Co (0% Au), Co/Au 35%, and Co/Au 42% samples, respectively. The B-dependent electric field attenuation in the Co/Au 42 % sample is about 3 times greater than that in uncoated Co particles or, equivalently, the intensity attenuation is about an order of magnitude greater. Such marked attenuations are accompanied by large temporal delays (relative to their zero-field transmissions). Remarkably, magnetically induced attenuation and delay increase with greater *nonmagnetic* Au coverage of the Co particles. These effects are caused by enhanced AMR in the Co / Au particles.

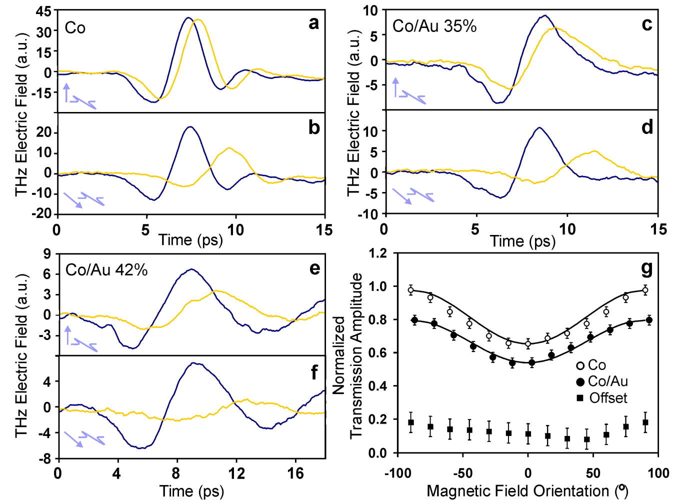

Figure 6. Time-domain THz transmission through (a) Co (0 % Au) (perpendicular), (b) Co (0 % Au) (parallel), (c) Co/Au 35 % (perpendicular), (d) Co/Au 35 % (parallel), (e) Co/Au 42 % (perpendicular) and (f) Co/Au 42 % (parallel) particle ensembles for $|B|$ = 0 T (blue traces) and $|B|$ = 150 mT (yellow traces). The diagrams depict the orientation of the B field (arrow) relative to the electric field polarization. (g) Normalized transmission amplitude through 3 mm thick Co (filled circle) and Co/Au (empty circle) particle samples versus the field angle relative to the incident polarization, where $|B|$ = 160 mT. The transmission amplitudes for both samples have been normalized to their respective zero-field amplitudes, and the data have been fitted with $\cos^2\theta$ functions shown by the solid lines. Filled squares represent the offset between the curves for the Co and Co/Au samples. Within error, the offset is independent of the B orientation.

We explored the interaction of THz radiation with both active and inactive random metamaterials. We observe group refractive index exhibits atypical which is higher than the parent constituents. Moreover, we show evidence of electron spin-dependent plasmonic transport through both ferromagnetic and ferromagnetic/nonmagnetic metamaterials.

REFERENCES

[1] T. J. Yen, W. J. Padilla, N. Fang, D. C. Vier, D. R. Smith, J. B. Pendry, D. N. Basov, and X. Zhang, "Terahertz Magnetic Response from Artificial Materials," *Science*, vol. 303, pp. 1494-1496, 2004.

[2] R. A. Shelby, D. R. Smith, and S. Schultz, "Experimental Verification of a Negative Index of Refraction," *Science*, vol. 292, pp. 77-79, 2001.

[3] H. J. Lezec, J. A. Dionne, and H. A. Atwater, "Negative Refraction at Visible Frequencies," *Science*, vol. 316, pp. 430-432, 2007.

[4] K. J. Chau and A. Y. Elezzabi, "Photonic Anisotropic Magnetoresistance in Dense Co Particle Ensembles," Phys. Rev. Lett., 96, 033903-033906, 2006.

[5] K. J. Chau, Mark. Johnson, and A. Y. Elezzabi, ""Electron-Spin-Dependent Terahertz Light Transport in Spintronic-Plasmonic Media, Phys. Rev. Lett., 98, 133901-133904, 2007.