Effect of interface resistance on terahertz pulses in bimetallic microparticles

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We describe time-domain studies of terahertz (THz) pulse propagation in random bimetallic media composed of Au-coated Cu microparticles. Despite the similar electrical properties of Cu and Au, coating the Cu microparticles with Au nanolayers increases the effective resistivity of the particles by up to two orders of magnitude, resulting in large attenuation and phase modulation of the transmitted THz pulses. The experimental results are in reasonable agreement with simulations based on a damped harmonic oscillator.

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Surface electromagnetic waves have been of research interest for over a century, starting with Zenneck's formulation of a solution to Maxwell's equations for a wave propagating on a surface with finite losses.[1] Such surface waves have since been studied on a wide range of geometries and at frequencies spanning from the visible down to the radio spectral range. Recently, surface electromagnetic waves confined on metallic surfaces, commonly referred to as surface-plasmon waves,[2–7] have been investigated for their potential technological application in fields such as microelectronics,[2,3] communications,[4–7] and biosensing.[3] In the visible spectral range, surface-plasmon waves are excited either on very thin metal surfaces or on nanoparticles composed of a noble metal (Ag, Au, or Cu).[2–7] In search for new materials with tailored electromagnetic functionality, a class of hybrid plasmonic media, composed of more than one metal species, is being explored.[8–16]

Interestingly, bimetallic surfaces have unique and diverse surface-plasmon properties dependent on the constituent metallic species. In particular, surface waves on bimetallic surfaces can show vastly different behavior, depending on the exact composition and distribution of the two metals. Bimetallic alloyed nanoparticles typically exhibit a plasmonic response that can be described by an average dielectric function of the parent metallic species.[8–10] Increased diversity of plasmonic behavior is observed in bimetallic *layered* nanoparticles (also known as "*core-shell*" nanoparticles). Such bimetallic surfaces exhibit electromagnetic properties that significantly differ from those of the constituents and cannot be described through a combination of the dielectric functions of the parent metals.[11–16] It has been speculated that these extraordinary electromagnetic properties can be attributed to the interfacial metallic contact.[17,18] However, the influence of the interface on the electromagnetic properties of bimetallic media remains unresolved. Despite this unsettled issue, research on the interaction between surface electromagnetic waves with bimetallic surfaces holds promise for a multitude of photonic applications not otherwise achievable using monometallic systems.

With the growing attention devoted to surface-plasmon waves in the visible regime, there has been a concomitant rise in the investigations of surface-plasmon waves in the neighboring far-infrared and terahertz (THz) regimes. Such interest is stimulated, in part, by the advantages offered by THz time-domain spectroscopy, which permits spatial and temporal electric-field measurements of surface-plasmon waves. To date, surface-plasmon waves at THz frequencies have only been studied on monometallic surfaces[19] and subwavelength particles,[20] while more complicated and diverse surface-plasmon behavior on bimetallic surfaces are awaiting to be explored.

In this work, we present an experimental investigation of THz surface electromagnetic wave propagation on bimetallic layered surfaces. THz time-domain spectroscopy is employed to investigate THz pulse propagation through random ensembles of Cu microparticles that are partially coated with Au nanolayers. Cu and Au are ideal metals to study interfacial effects because they both have simple Fermi surfaces and their optical and electronic properties are well characterized.[21] Access to both the amplitude and phase of the transmitted THz electric field, $E_{\mathrm{trans}}(t)$, enables unprecedented time-domain visualization of THz pulse propagation through the bimetallic medium. Remarkably, despite the similar optical and electrical properties of Au and Cu, covering the Cu microparticles with Au nanolayers results in large amplitude attenuation and phase modulation of $E_{\mathrm{trans}}(t)$. The dramatic attenuation is ascribed to an increase in the surface resistivity of the Au-coated Cu particles by two orders of magnitude. Our experiments provide strong evidence that the large resistivity increase originates from interface resistance at the boundary between the Au nanolayer and Cu core. Not only are these findings important for elucidating the nature of surface-plasmon losses in bimetallic media, but they will open the door to far-infrared photonic applications using nanolayered microparticles.

We study the THz transmission through polydisperse, randomly oriented Cu-core/Au-layer(Cu/Au) spherical microparticles having a diameter D ranging from 56 to 107 μm (Fig. 1). Because of the random nature and polydispersity of the particles, any geometrical electromagnetic resonances in the sample are precluded.[20] The metallic microparticle samples (2 mm thick) are excited by linearly polarized, 1 ps wide, THz pulses generated by femtosecond photoexcitation of a 100 μm SI-GaAs photoconductive gap. The particles are densely packed such that ballistic straight-line THz light propagation through the sample is not possible. However, it has been shown that polarized THz light propagates through the densely packed microparticle samples via near-field coupling of surface electromagnetic waves.[20] The transmitted

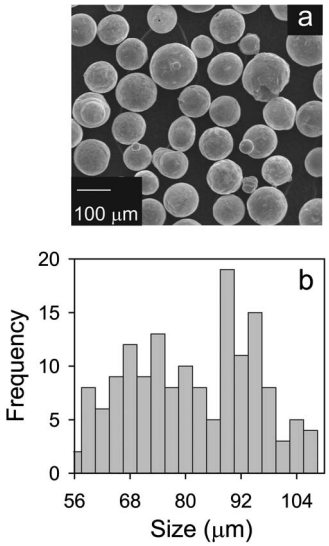

FIG. 1. (a) A scanning electron microscope image of the Cu microparticles and (b) their size distribution.

THz electric field is detected via an electro-optically sampled ⟨111⟩ ZnSe crystal.[22]

The THz electric field pulse, $\boldsymbol{E}_{\text{inc}}$, incident on the particles excites localized conduction electron oscillations within the skin depth, δ, of the particle surface. Since the size of the particle is much smaller than the wavelength, an approximately uniform field accelerates electrons at the surface of the particle collectively, generating a surface charge oscillation that is coupled to a low-frequency surface-plasmon wave.[20] The current associated with the charge oscillations can be described by $\boldsymbol{j}_p = \boldsymbol{E}_{\text{total}}/\rho_{\text{eff}}$, where ρ_{eff} is the effective resistivity of the particles at the surface and $\boldsymbol{E}_{\text{total}} = \boldsymbol{E}_{\text{inc}} + \boldsymbol{E}_{\text{depol}}$ is the total surface electric field consisting of contributions from $\boldsymbol{E}_{\text{inc}}$ and the depolarization electric field $\boldsymbol{E}_{\text{depol}}$ from the induced charges. We have previously shown that the surface wave associated with the charge oscillations on individual particles couple via nearest-neighbor interaction across the particle collection and radiate into the far field at the edge of the sample.[20] By means of nearest-neighbor coupling of the surface charge oscillations, changes in ρ_{eff} within δ of the particle surface are directly mapped onto a transmissivity modulation. In a situation where metallic particles are covered with layers of a dissimilar metal, it is expected that the interface resistance at the boundary between the metals will impede the surface charge oscillations. Consequently, the transmission will attenuate due to increased electron scattering from roughness of surface topography, interface alloying, and/or potential steps at the interface.[18] Of particular interest are the latter two contributions to interface resistance, which are sensitive to the electronic properties of the metals. Interfacial alloying results in the formation of an intermediate interfacial layer composed of a mixture of the two metallic elements. Potential steps, on the other hand, arise from the difference in the work functions between the two constituent metals. When two such metals are brought into physical contact, conduction electrons flow from the lower work-function metal into the higher work-function metal until an equilibrium chemical potential is achieved. As a result, an electrostatic charge built up at the interface creates a potential difference from which electrons can scatter.[23–25] Interface resistivity thus augments the effective resistivity of the particle. For independent scattering processes, ρ_{eff} can be described by Matthiessen's rule

$$\rho_{\text{eff}} = \rho_{\text{Au}} + \rho_{\text{Cu}} + \rho_{\text{interface}} = \frac{m^*}{Ne^2\tau_{\text{eff}}}, \tag{1}$$

where $\rho_{\text{Au,Cu}}$ is the resistivity contribution from Au (Cu), $\rho_{\text{interface}}$ is the intrinsic interface resistivity between the metals, τ_{eff} is the effective electron scattering time, and m^*, N, and e are the electron effective mass, the electron density, and the electron charge, respectively.

It is well known that surface-plasmon waves at THz frequencies are dependent on the metal's conductivity within a relatively large skin depth, $\delta \sim 100$ nm.[26,27] Owing to the large spatial extent of the THz skin depth, the influence of the thin interfacial layer on ρ_{eff} can be easily probed from the THz optical properties of the bimetallic medium. In addition, the range of the THz skin depth relaxes the constraints on the dimensions of the bimetallic layers.

The Cu-core/Au-layer microparticles are fabricated by sputter depositing a 40 nm thick Au layer on dispersed spherical Cu particles. The THz electric field interacts with electrons in both metal constituents and their thin interfacial layer since the thickness of the Au film $d < \delta$. Since the surface wave is coupled to charge oscillations on the outer surface of the bimetallic composite, it is essential to characterize the atomic composition directly at the particles' surface. The surface of the microparticles may consist of pure Au or a Cu/Au alloy due to miscibility and interdiffusion of Cu into Au. Thus, we have performed x-ray-photoelectron spectroscopy[28] (XPS) to determine the exact composition of the microparticle surface. The XPS spectra of the bimetallic samples show no shift of the Au $4f$ core levels relative to the $4f$ core levels in pure Au samples, indicating the absence of Cu/Au alloying directly at the surface of the bimetallic particles. Within the sampling depth of the XPS spectra ($\sim$1 nm), we can conclude with confidence that the surface of the particles consists of a pure Au layer.

Since $\rho_{\text{interface}}$ is proportional to the total interface area between the two metals, THz electric-field transmission measurements are performed with samples having varying percentage surface area covered by Au, f (where 1-f is the percentage uncoated Cu surface area). The Au coverage, or equivalently, Cu/Au interface area, is varied by systematically reorienting the Cu particles during the Au deposition process. The percent Au coverage is quantified by the relative magnitudes of the characteristic Au $4f$ and Cu $2p$ peaks in the XPS spectra where samples with higher Au coverage exhibit larger Au peaks and, correspondingly, smaller Cu peaks. On five bimetallic Cu-core/Au-layer samples, XPS analysis revealed $f = 17$, 24, 32, 44, and 47 %.

Figure 2(a) illustrates $E_{\text{trans}}(t)$ transmitted through the five, 2 mm thick bimetallic Cu-core/Au-layer samples having the same packing fractions, $p = 0.50 \pm 0.02$, along with the reference free space pulse. The transmitted pulse shape evolves from a multilobe, oscillatory pulse for the pure Cu

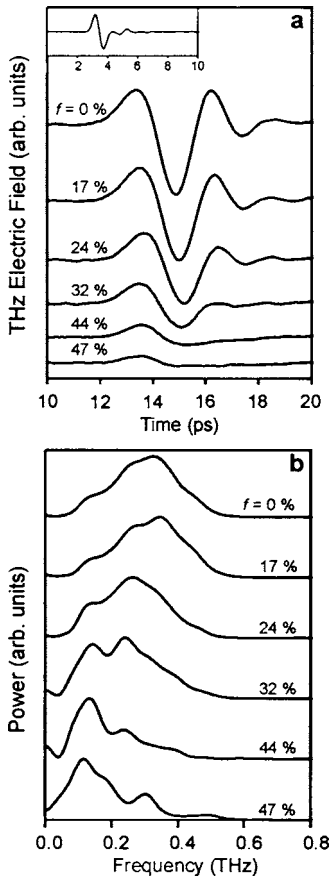

FIG. 2. Experimental (a) time-domain signals and (b) power spectra of the transmitted THz pulses through 2 mm thick Cu -core/Au-layer microparticle samples for varying Au surface coverage f. The inset depicts the reference THz pulse propagated through free space.

particles to a single lobe followed by a slowly rising tail for $f=47$ %. This is reflected in the frequency domain, where the transmission spectrum peaks at 0.32 ± 0.05 THz for the uncoated Cu particles and as f increases to 47 %, shifts toward 0.10 ± 0.05 THz [Fig. 2(b)]. The most striking feature is the degree of transmission amplitude reduction with increasing f. Remarkably, increasing f from 17 to 47 % dramatically attenuates $E_{\text{trans}}(t)$ by 88 ± 3 %, as shown in Fig. 3. The linear decrease in the electric field amplitude with increasing surface coverage reflects the direct proportionality between the interface area and the interface resistance. The large attenuation in the Cu-core/Au-layer particles is surprising since Cu and Au have similar dielectric functions [$\varepsilon_{\text{Au}}=-1.1\times10^5+7.1\times10^5 i$, $\varepsilon_{\text{Cu}}=-0.51\times10^5+4.3\times10^5 i$ (Ref. 29)] and thus, their optical responses at THz frequencies are expected to be alike. Since $d \ll D$, the attenuation cannot be ascribed to infinitesimal ($8d^3/D^3 \approx 10^{-9}$) volume difference between the Cu and Cu-core/Au-layer particles. Rather, the amplitude attenuation with increasing f arises from resistance at the bimetallic interface through $\rho_{\text{interface}}$, or possibly additional loss that might be associated with the nanoscale Au layer. To investigate the latter possibility, we perform additional experiments with five Cu-core/Au-layer samples having similar f between 18 and 21 %, but with variable $d=20$, 61, 93, 128, 155, and 175 nm. By keeping the Au surface coverage fixed, and thus the interface contact area between the metals constant, the interface resistance is unaltered. This allows us to examine the influence of only the intrinsic resistivity contribution from the Au layer. As shown in Fig. 4, the transmitted pulse shape through the various Cu-core/Au-layer samples does not change as d increases from 20 to 175 nm. In addition, within the error of the experiment (±5 %), we observe negligible amplitude attenuation, indicating that resistivity contributions arising from the Au layer alone are small in comparison to the interface resistivity. Based on the experimental evidence, we can conclude that the large attenuation originates from the interface between the Cu and Au.

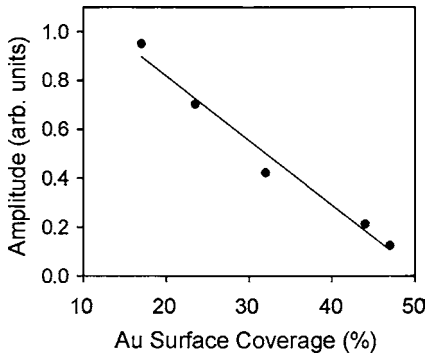

FIG. 3. Peak-to-peak transmitted THz electric-field amplitude vs Au surface coverage f. The electric-field amplitudes are normalized to the transmission through uncoated Cu particles.

To our knowledge, there is no comprehensive theoretical model describing THz surface plasmon dynamics on bimetallic surfaces. However, one can obtain a simple physical understanding of the phenomenon by considering a semiquantitative one-dimensional damped harmonic oscillator model of the charge oscillations driven by the THz electric field, $E_{\text{inc}}(t)$. Since electron motion follows the THz electric field linearly, the use of the harmonic approach is adequate

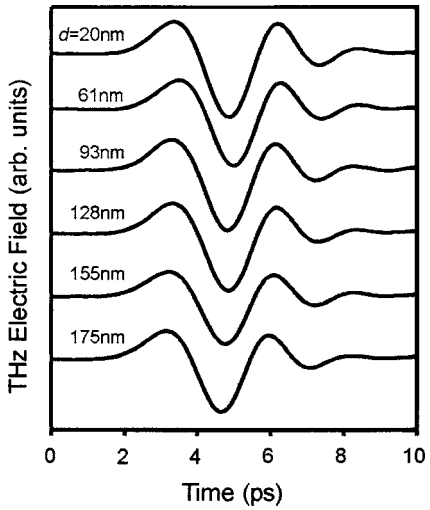

FIG. 4. Experimental time-domain signals of the transmitted THz pulses through 2 mm thick Cu-core/Au-layer microparticle samples for $d=20$ nm to 175 nm.

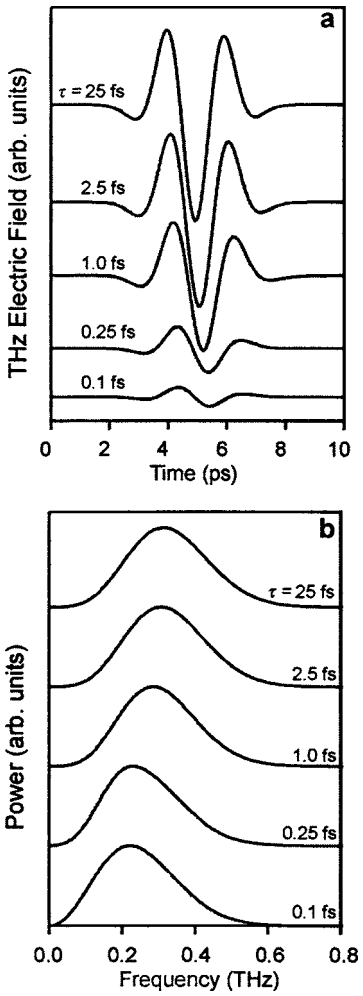

FIG. 5. The calculated (a) time-domain signals and (b) frequency spectra of the transmitted THz pulses in response to single-cycle THz pulse excitation for varying τ_{eff}.

for our discussion to follow. Most importantly, this model enables us to deduce origin of the amplitude reduction and temporal behavior of the plasmon-mediated THz transmission. The collective polarization, $P(t)$, of the particle ensemble is modeled by

$$\frac{\partial^2 P(t)}{\partial t^2} + \frac{2}{\tau_{\text{eff}}}\frac{\partial P(t)}{\partial t} + \omega_{\text{eff}}^2 P(t) \propto E_{\text{inc}}(t) \tag{2}$$

where $\omega_{\text{eff}}^2 = N_{\text{eff}} e^2 / \varepsilon_o \boldsymbol{m}^*$ is the effective plasma frequency of the particle composite, ε_o is the free space permittivity, and N_{eff} is the density of active electrons participating in the plasmon oscillations. It should be noted that since the metal particles fill a volume fraction of 0.50 and the THz pulse interacts only with the electrons within δ of the surface, the effective electron density can be calculated through $N_{\text{eff}} = 6pN\delta/D$.[30,31] Given $N = 10^{28}$ m^{-3}, $p = 0.5$, $m^* \sim 1.49$ m$_0$, $\delta = 100$ nm, and $D = 84$ μm, the effective plasma frequency of the particle composite is $\omega_{\text{eff}} \simeq 10^{14}$ rad s^{-1}. We show in Fig. 5(a) plots of the calculated far-field transmitted electric field $E_{\text{trans}}(t) \propto \partial^2 P(t)/\partial t^2$ in response to single cycle THz pulse excitation for τ_{eff} varying from 25 fs (derived from a Cu

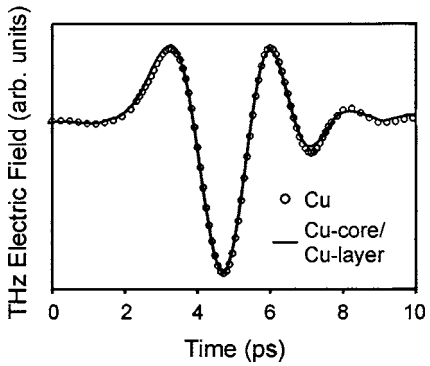

FIG. 6. Transmitted THz pulses through 2 mm thick samples of uncoated Cu particles (circles) and Cu-core/Cu-layer particles (line).

room temperature resistivity $\rho_{dc,\text{Cu}} = 1.7$ $\mu\Omega$ cm) to 0.1 fs. The model results show reasonable agreement with the experimental data despite the fact that the calculated detector response is assumed to be flat over the entire spectral range, which experimentally is not the case. Interestingly, as τ_{eff} decreases, the calculated $E_{\text{trans}}(t)$ is attenuated by 90 ± 1 %, and the frequency spectra redshifts from 0.32 ± 0.02 THz to 0.21 ± 0.02 THz. Furthermore, the $E_{\text{trans}}(t)$ pulse shape evolves from a multilobe pulse to a single-lobe pulse with decreasing τ_{eff}, in agreement with the experimentally observed trend. From our model in conjunction with the experimental data, the 88 ± 3 % electric-field attenuation as f increases from 0 to 47 % represents a reduction in τ_{eff} by two orders of magnitude (corresponding to a two-order-of-magnitude increase in ρ_{eff}). Thus, the damped harmonic oscillator model provides a simple, physical interpretation of the observed phenomenon, but further theoretical work, which incorporates near-field electromagnetic interaction, is required.

To further explore the origin of the interface resistance effect on the plasmon-mediated THz transmission, we perform control experiments with monometallic Cu-core/Cu-layer particles. Because of the absence of both a potential barrier and alloying at the interface between the Cu core and the Cu layer, the only source of interface resistance for the monometallic Cu samples is electron scattering from the rough interface topography. We employ samples composed of 40 nm Cu layers, which are sputter deposited onto pure Cu particles (identical to those used to fabricate the Cu/Au samples). These Cu/Cu monometallic particles have a Cu surface coverage of 24 %. In contrast to the bimetallic Cu -core/Au-layer particles, however, the monometallic Cu -core/Cu-layer particles exhibit no measurable attenuation and show no electric field pulse reshaping, as shown in Fig. 6. This supports the conclusion that electric-field pulse reshaping in the Cu-core/Au-layer bimetallic samples arises from electronic dissimilarities between core-layer metallic species and electron scattering from interface roughness is a negligible contributor to the interface resistance.

The phase characteristics and temporal evolution of the experimental $E_{\text{trans}}(t)$ provide further insight into the attenuation phenomenon. In particular, the $E_{\text{trans}}(t)$ phase reveals that the effect of the interface resistance is not purely resis-

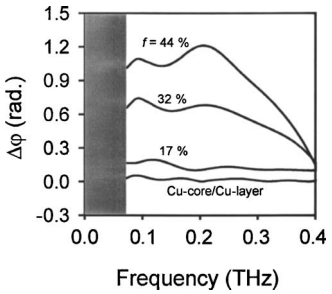

FIG. 7. Differential phase of the transmitted THz pulse through the Cu-core/Cu-layer sample and through the $f=44$ %, $f=32$ %, and $f=17$ % Cu-core/Au-layer microparticle samples. The transmission through the Cu-core/Cu-layer sample shows no significant phase accumulation across the transmission bandwidth.

tive. We show in Fig. 7 the differential phase, $\Delta\varphi(\omega)=\varphi(\omega)-\varphi_{\mathrm{ref}}(\omega)$, where $\varphi(\omega)$ is the phase of $E_{\mathrm{trans}}(t)$, and $\varphi_{\mathrm{ref}}(\omega)$ is the phase of the reference transmitted pulse through the uncoated Cu. The transmission is characterized by significant dispersion; at frequencies below 0.2 THz, $E_{\mathrm{trans}}(t)$ acquires ~ 1 radian as f increases from 17 to 47 %. The additional phase arises from the phase lag between the electric field and the induced currents in the skin layer, which is determined by the microparticles' reactance. That is, increased Au coverage results in a slower polarization response to the driving THz electric field, manifesting as a phase accumulation in $E_{\mathrm{trans}}(t)$. In the temporal domain [Fig. 2(a)], the drastic attenuation of the subsidiary lobes of the pulse as f increases further shows that increasing f not only augments the Joule loss, but also increases the reactance. We believe that the physical origin of the reactive response can be ascribed to dynamical screening at the bimetallic interface.[32,33] Charge screening dephases collective electron motion such that electrons in the particle are no longer synchronous with the driving THz field. Since screening becomes prominent only after the initial rise of the pulse [Fig. 2(a)], we infer that screening dynamics occur on the order of a picosecond.

In conclusion, we have demonstrated large amplitude reduction and drastic phase modification of THz electric-field pulses propagated through Cu microparticles partially coated with Au nanolayers. The experiments show that the pulse reshaping arises from the interface resistance between the metal constituents. Interface contributions from electron scattering due to rough interface topography are shown to be insignificant. Our experimental evidence indicates that the large interface resistance originates from electronic dissimilarities between the two metals, which may result in electron scattering losses due to a potential step at the interface between the dissimilar metals and/or interfacial alloying. Based on a harmonic oscillator model of plasmon dynamics, we infer that the Au nanolayer causes up to a hundred-fold decrease in τ_{eff}, or equivalently, a hundred-fold increase in ρ_{eff}. The strong dependence of the transmission on the nanoscale Au layer makes bimetallic microparticles ideally suited for applications such as tunable passive THz filters and sensors.

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