Subsurface probing of terahertz particle plasmons

K. J. Chau, K. M. Rieckmann, and A. Y. Elezzabi
Ultrafast Photonics and Nano-Optics Laboratory, Department of Electrical and Computer Engineering, University of Alberta, Edmonton, Alberta T6G 2V4, Canada

(Received 29 November 2006; accepted 25 February 2007; published online 30 March 2007)

Here, the authors exploit the potential barrier at the interface between dissimilar metals to probe frequency dependent subsurface charge induction on metallic microparticles excited with terahertz radiation. The authors’ experimental data and model show that terahertz electromagnetic charge induction on the microparticles occurs over a distance comparable to the skin depth. This work provides a technique to probe subsurface terahertz charge induction in subwavelength metallic structures and may open research avenues of low-frequency plasmonic behavior. DOI: 10.1063/1.2718504

Subwavelength scale metallic nanoparticles play a key role in the continuing development of nanophotonic applications such as biosensing, imaging, waveguiding, and random lasing.1–5 These structures are unique because light incident upon the nanoparticles can resonantly drive electrons throughout the nanoparticle volume, creating a resonant particle plasmon. For spherical particles, the resonance condition corresponds to the frequency ω, where the permittivity of the metal, εr(ω), satisfies the condition Re[εr(ω)] = −2. When this condition is fulfilled, the polarization of the nanoparticle approaches infinity. While much work has been conducted in the visible frequency range, there has been increasing interest in the study of plasmonlike phenomena in neighboring terahertz frequencies.6–12 By scaling the metallic particle dimensions from the nanometer to the micrometer scale, it has been suggested that a nonresonant dipolar particle plasmon effect, analogous to the particle plasmon behavior observed in the visible range, exists in the terahertz range.5 At terahertz frequencies, the resonant particle plasmon response of subwavelength microparticles is inaccessible since Re[εr(ω)] = −10^5. This large permittivity also dictates that incident terahertz radiation cannot infiltrate deeply into the metallic medium. However, due to the subwavelength size of the particles, the finite penetration distance of the incident terahertz field, E_{THz}(ω), into the subwavelength particle can produce a plasmonic response (collective induction of conduction band electrons) whose nonradiative decay is highly sensitive to the surface conditions. Within a finite distance into the particle, ξ(ω), the formation of the terahertz particle plasmon is governed by several mechanisms: (1) the terahertz radiation incident on the particle surface penetrates tens of nanometers into the metal where it induces charge motion (or current) and (2) the depolarizing dipolar electric field is formed by charge accumulation at the particle’s surface.2 Notably, it remains a challenge to experimentally probe these mechanisms within the nanoscale region below the metallic surface especially for nonplanar geometries. The development of sensitive experiments to access the electromagnetic interactions beneath the surface of metals would not only provide insights into the nature of terahertz radiation penetration in subwavelength structures but also offer additional clues into the mechanism of terahertz particle plasmon formation and their associated nonradiative losses.

In this letter, we introduce a unique noninvasive method to probe the subsurface charge motion of terahertz particle plasmons excited on metallic subwavelength particles. Here, induced charge motion within a depth ξ(ω) is investigated by exploiting the potential barrier that develops at the interface between dissimilar metals in intimate contact. When two dissimilar metals are placed in contact, electrons flow from the metal with the lower work function into the metal with the higher work function until their Fermi levels are aligned. This charge accumulation (confined at the interface within the Thomas-Fermi length) creates a potential barrier, which impedes the flow of electrons across the interfacial region. By overlaying subwavelength particles with spatially varying series of alternating metallic nanolayers, the interfacial potential barriers between the layers locally augment the nonradiative losses of electromagnetically induced current. The interfaces, therefore, provide extremely spatially localized probes of charge induction. The scale length of terahertz induced charge motion within the microparticles is extracted by varying the number of nanolayers, N, and their thicknesses over nanoscale distances from the surface. The findings not only show that terahertz electromagnetic charge induction in metallic particles is largely governed by the skin depth but also provide a direct glimpse into subsurface decay mechanisms of terahertz plasmons.

Using terahertz time-domain spectroscopy with the experimental setup described in Ref. 6, we study the terahertz transmission through polydisperse, randomly oriented subwavelength multilayered Cu microspheres having a radius r ranging from 34 to 49 μm. The microparticle ensemble are 2.3±0.1 mm thick and are packed in a polystyrene cell to a filling fraction of ~0.5. As illustrated schematically in a diagram in Fig. 1(a), multilayered microparticle samples are fabricated by sputter coating Cu microspheres with alternating nanolayers (up to five layers) of Au and Cu. The nominal Au and Cu layer thicknesses are t_{Au} = 15 nm and t_{Cu} = 43 nm, respectively. The Au and Cu nanolayers produce a spatially modulated potential beneath the particle surface [Fig. 1(b)], augmenting the nonradiative losses near the surface at the coated regions. We carefully ensure that the various particle samples have identical metallic coverage deposited on their surfaces within ±1%. To achieve precise control of the coverage of the particles during the sputter deposition, the particle samples are masked by a wire mesh having 40

aElectronic mail: kjchau@ece.ualberta.ca
The transmitted terahertz pulse amplitude is strongly dependent on the exterior layer of the multilayers. While the transmitted terahertz pulse amplitude is strongly dependent on the exterior layer of the multilayers, the transmitted terahertz electric field pulses. Surprisingly, the addition of a 43 nm thick Cu layer on top of a 15 nm thick Au layer gives rise to 20±1% transmission attenuation for N>3 [Fig. 1(d)]. This indicates that contact potential barriers lying >73 nm from the surface (corresponding to the total thickness of the N=3 multilayers) do not contribute to the terahertz electric field attenuation.

The time-domain wave forms reveal strong pulse reshaping with increasing N, suggesting frequency dependent attenuation arising from the multilayers. To study this frequency dependence, we determine the configurationally averaged, relative spectral transmission amplitude $\Phi_N(\omega) = \langle E_N(\omega) \rangle / \langle E_0(\omega) \rangle$ through the random samples, where $\langle E_N(\omega) \rangle$ is the average spectral amplitude of the transmission through samples with N (=1–5) layers and $\langle E_0(\omega) \rangle$ is the average spectral amplitude of the nominal transmission through uncoated Cu particles. $\Phi_N(\omega)$ is obtained by averaging the transmission for multiple (>10) random realizations of the ensemble, where the packing fraction and sample thickness are kept constant. The spectral amplitudes are calculated over the transmission bandwidth from 0.2 to 0.4 THz. As shown in Fig. 2(a), the relative transmission amplitude for N=1 varies from 0.89 at 0.2 THz down to 0.68 at 0.4 THz. Adding a 43 nm Cu layer over the top of the Au-layered particles enhances the transmissivity of the monolayered particles. Over the same frequency range, $\Phi_3(\omega)$ is larger than $\Phi_1(\omega)$ and shows less attenuation at higher frequencies. For N=3, $\Phi_3(\omega)$ decreases significantly from 0.2 to 0.4 THz and is as low as 0.25 at the higher frequency end of the spectrum. Interestingly, these results suggest that nonradiative losses arising from the interface are frequency dependent, where the higher frequency components of the pulse are more attenuated than the lower frequency components. Moreover, we observe that the spectral amplitude $\Phi_N(\omega)$ alternates from high to low with the successive addition of layers, similar to the observed trends in the time-domain wave forms. The similarities between $\Phi_1(\omega)$ and $\Phi_3(\omega)$ with $\Phi_2(\omega)$ and $\Phi_4(\omega)$, respectively, point...
out that interfaces lying below the first several layers from the surface contribute minimally to the attenuation.

We develop a model accounting for nonradiative losses at each interface. When $E_{\text{TM}}(r, \omega)$ is incident on an uncoated metallic particle, (1) charge induction creates a transient current density, $j(r, \omega) = j_0(\omega) e^{-r/\delta(\omega)}$, where $j_0(\omega)$ is the current density at the surface and $r$ is the distance from the surface, and (2) surface charge accumulation results in a depolarization field $E_s(r, \omega)$. The nonradiative decay within the particle is described through $\Gamma(\omega) \approx \int j_0(\omega) \cdot E_s(r, \omega) d\nu$, where $E(r, \omega) = E_{\text{TM}}(r, \omega) + E_s(r, \omega)$ is the total electric field and $d\nu$ is the volume element of the particle. Since the current density is related to the total field via $E(r, \omega) = \rho(\omega) j(r, \omega)$, the total nonradiative decay is $\Gamma(\omega) \approx \int \rho(\omega) j_0^2(r, \omega) d\nu$. However, the situation is different for coated microparticles, where the interfaces introduce additional sources of dissipation. For a series of $n$ identical interfaces located at distances $s_1, s_2, \ldots, s_n$ below the surface, nonradiative decay includes the bulk dissipation $\Gamma(\omega)$ in addition to local losses at each interface. The total nonradiative loss for the coated particles can be described via

$$\Gamma'(\omega) \approx \Gamma(\omega) + \sum_{i}^{n} \int \rho_{\text{int}}(\omega) \delta(r - s_i) j_0^2(r, \omega) d\nu$$

$$= \Gamma(\omega) + \sum_{i}^{n} \rho_{\text{int}}(\omega) j_0^2(s_i, \omega),$$

(1)

where $\rho_{\text{int}}(\omega)$ is the interfacial resistivity between the Cu and Au layers and $f$ describes the average fractional surface coverage of the particles and accounts for the partial coverage of the particles. Thus, the normalized spectral transmission amplitude relative to uncoated particles can be expressed as

$$f \rho_{\text{int}}(\omega) j_0^2 \sum_{i}^{n} \frac{e^{-2s_i/\delta(\omega)}}{\Gamma(\omega)}$$

$$\Phi_N(\omega) = 1 - \frac{[f \rho_{\text{int}}(\omega) j_0^2(\omega) \Gamma^{-1}(\omega)]^n}{\Gamma(\omega)},$$

(2)

where $[f \rho_{\text{int}}(\omega) j_0^2(\omega) \Gamma^{-1}(\omega)] > 0$ is a dimensionless ratio describing the fractional nonradiative losses due to the interface relative to the bulk dissipation. In order to implement the above model to determine $A$ and $\xi(\omega)$, the distances $s_1, s_2, \ldots, s_n$ are obtained from measured values of $t_{\text{Au}}$ and $t_{\text{Cu}}$. As seen in Fig. 2(b), there is excellent agreement between the calculated $\Phi_N(\omega)$ obtained from Eq. (2) and the experimental data. Most notably, the model concurrently describes, with great accuracy, the preferential attenuation of the higher frequency components and the oscillating behavior of $\Phi_N(\omega)$ with the addition of successive Au/Cu nanolayers, in addition to the similarities in $\Phi_N(\omega)$ and $\Phi_N(\omega)$ with $\Phi_N(\omega)$ and $\Phi_N(\omega)$, respectively. An important outcome of the model is the direct characterization of the effective penetration distance of the subsurface charge induction, $\xi(\omega)$. As shown in Fig. 3, $\xi(\omega)$ shows a notable frequency dependency, where it varies from 100 nm at 0.2 THz to 60 nm at 0.4 THz. For a planar geometry, the region of induced charge oscillation precisely corresponds to the skin depth, $\alpha(\omega) = \sqrt{2 \rho / \mu \omega}$, where $\rho$ is the resistivity and $\mu$ is the permeability. In Fig. 3 we plot the classical planar skin depth values for Cu, $\alpha_{\text{Cu}}(\omega)$, and for Au, $\alpha_{\text{Au}}(\omega)$, from 0.1 to 0.5 THz, based on experimental data from Ref. 14. Interestingly, at higher frequencies, $\xi(\omega)$ approaches 72 nm at 0.42 THz, similar to that of $\alpha_{\text{Cu}}(\omega)$. At lower frequencies (<0.25 THz), $\xi(\omega)$ slightly deviates from the 1/\omega dependency of the classical skin depth, suggesting that the validity of the plane wave approximation used to derive the electromagnetic skin depth on a planar surface becomes questionable. It should also be noted that contributions from the anomalous skin effect may further account for the discrepancies between $\xi(\omega)$ and $\alpha(\omega)$. When the electromagnetic field driving the electrons suffers appreciable damping over the distance the electron traverses between collisions, the electron velocity becomes dependent on the field at prior positions along the electron mean free path. This anomalous skin effect results in a decreased skin depth relative to that predicted by the classical Drude model.

In this work, we have employed localized nonradiative losses arising from the interface potential barrier between dissimilar metallic layers to map out $\xi(\omega)$ for terahertz particle plasmons excited on metallic microspheres. We foresee applications of this effect in experiments on terahertz subsurface spectroscopy of metallic media and terahertz characterization of buried metallic junctions.

This work was supported by NSERC.