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Near-Field Intensity Correlations in Semicontinuous Metal Films

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Near-field intensity correlations in semicontinuous metal films

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Abstract: Spatial intensity correlation functions of semicontinuous metal-dielectric films of varying metal concentration, p, were obtained from near-field microscopy. The data shows a transition from propagation to localization and back to propagation of optical excitations with increase in p.

OCIS codes: (180.5810) Scanning microscopy; (240.6680) Surface plasmons; (240.6690) Surface waves; (240.5420) Polaritons; (240.0310) Thin films; (160.3900) Metals

Spatial correlations of field and intensity are indicative of the nature of wave transport in random media and have been widely investigated in the context of electromagnetic wave propagation in disordered systems [1,2]. In past studies, correlations in random dielectric systems have been studied theoretically and experimentally [1,2]. However, less is known of near-field intensity correlations in random metallic systems, which can exhibit rich phenomena due the involvement of intrinsic resonance effects-surface plasmons [3-5]. Neither is clear the difference between correlation functions in metallic and dielectric systems, which are expected to differ strongly since the near-field intensity distribution across a planar metallic random structure is extremely inhomogeneous with giant local field maxima whereas dielectric random systems exhibit homogeneous speckle patterns of uniform speckle size.

This paper presents the first experimental study of near-field intensity correlations in metal-dielectric systems in regimes where localization and delocalization are expected. Significant differences are observed between the spatial intensity correlations functions in metal-dielectric systems and those of purely dielectric random media.

In disordered metallic nanostructures, surface plasmon modes are governed by the structural properties of the system and may be strongly localized [3,4]. Recent theoretical studies of metallic nanoparticle aggregates suggest that the eigenmodes of such systems may have properties of both localized and delocalized states [5]. However, it is not clear how such eigenmodes impact the propagation or localization of SPPs excited by impinging light, an issue addressed in this study. In the current experiment, the concentration of metal particles on a dielectric surface was varied over a wide range to control the amount of scattering. Spatial intensity correlations obtained from near-field optical microscopy (NSOM) images show a transition from propagation to localization and back to propagation of optical excitations in planar random metal-dielectric systems with increase in metal filling fraction.

Semicontinuous silver films on glass substrates were synthesized by pulsed laser deposition [6]. Transmission electron microscope (TEM) images reveal that the samples are comprised of individual silver grains with an average size ~20 nm. An increase in deposition time (surface concentration of silver) induces a structural transition from isolated metal grains ($p \leq 0.4$) to interconnected metal clusters ($p \sim 0.6$) and finally to a nearly continuous metal film with dielectric voids ($p \geq 0.8$) [6]. Analysis of TEM data verified that all samples exhibited random structure. In the near-field experiment, samples were illuminated by the evanescent field (in the total internal reflection geometry) of He-Ne lasers operating at 543 nm and 633 nm (p-polarized). The local optical signal was collected by a tapered, uncoated optical fiber and shear-force feedback was used to control the tip-sample separation.

2D Fourier transforms of the near-field intensity distribution (not shown) showed significant elongation in the k-vector distribution along the direction of incident light at low and high metal concentrations whereas the distribution was nearly isotropic at intermediate metal concentrations. This indicates that the scattering strength

changes from weakly scattering to strongly scattering and back again to weakly scattering with increase in metal concentration.

From near-field images, the 2D correlation functions for near-field intensities were computed: C(Δx, Δy) = $C(\nabla x, \nabla y) \equiv \langle I\delta(x+\nabla x, y+\nabla y)\rangle$, where $I\delta(x,y) = [I(x,y) - \langle I(x,y)\rangle]/\langle I(x,y)\rangle$. Figure 1 shows the spatial dependence of C(Δx, Δy) for samples with p = 0.36, 0.65 and 0.83 at λ= 633 nm. Fringes are present along the direction of the incident wave for p = 0.36 and 0.83, but disappear at p = 0.65.

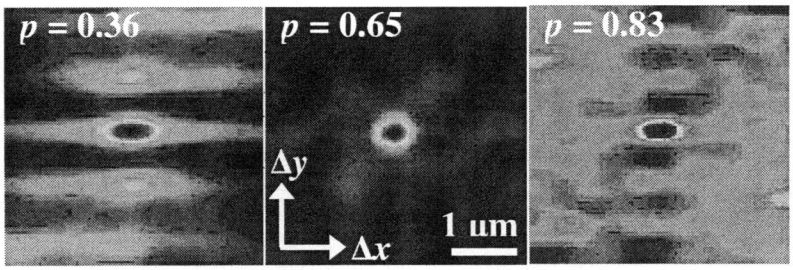

Fig 1. Intensity correlation function C(Δx,Δy) at p = 0.36, 0.65 and 0.83. The incident wave is along the y-axis.

Figure 2 has plots of the intensity correlation functions in the directions parallel and perpendicular to the incident wave vector $k_{\parallel}$, i.e., C(0,Δy) and C(Δx,0). Along $k_{\parallel}$, C(0,Δy) exhibits oscillatory behavior at p = 0.36 with a period of 870 nm. This oscillation is replaced by a monotonic decay at p = 0.65. At p = 0.83, the oscillations reappear with a smaller period of 690 nm. The presence of oscillations in C(0,Δy) is an indication of wave propagation along the y-axis. This propagation is suppressed at p = 0.65, suggesting localization of near-field energy. Therefore, the existence, suppression and reappearance of the oscillations in the near-field intensity correlation function with increasing p correspond to a gradual transition from propagation to localization and back to propagation of optical excitations in the samples. Note that the oscillation periods observed above are always larger than λ, in contrast with purely dielectric media, which exhibit damped oscillations with a period of λ/2 [1,2].

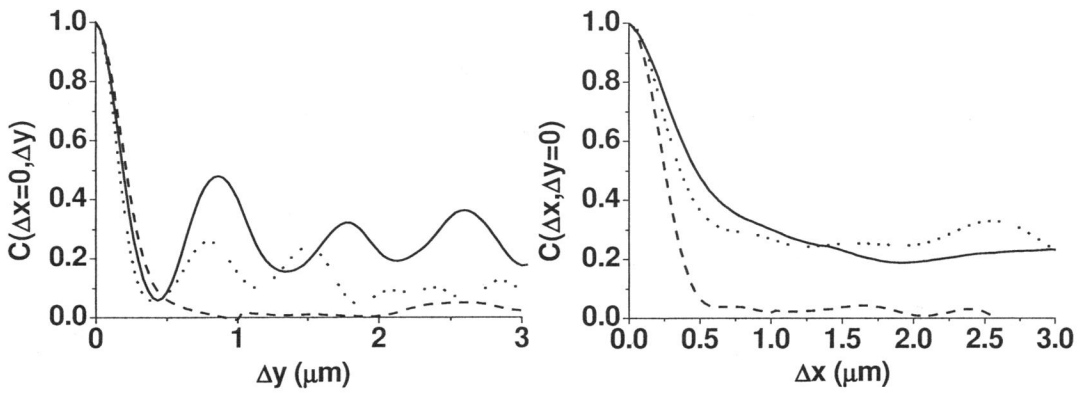

Fig 2. C(0,Δy) and C(Δx,0) at p = 0.36 solid line), 0.65 (dashed line)and 0.83 (dotted line). For comparison, all curves are normalized to a value of unity.

It is clear from Fig. 2 that C(Δx,0) does not oscillate at any metal concentration. Hence, there is no wave propagation in the direction perpendicular to the incident wave. There is a sharp drop in C(Δx,0) at p = 0.65. At p = 0.36 and p = 0.83, C(Δx,0) decreases gradually but does not reduce to zero even at large Δx. The long tail of C(Δx,0) suggests the existence of long-range correlation at low and high metal coverages, which results from a delocalized field. In the intermediate regime of metal concentration where scattering is strongest, the rapid decay of C(Δx,0) to zero indicates that the intensity correlation is short-ranged. This is a sign that the electromagnetic field is localized due to stronger scattering in the presence of anomalous absorption [4]. This result is in sharp contrast with

pure dielectrics where strong scattering enhances long-range correlations, resulting in a longer tail in the correlation function.

Since a *single* propagating wave cannot induce oscillations in the *intensity* correlation function, the oscillations observed in Fig. 2 at low and high metal coverages indicate the existences of at least two propagating waves.

To identify the propagating surface waves, the wave-vectors of the SPPs excited by the impinging light were calculated. At high metal concentrations, the effective medium approximation [4] leads to a dispersion relation which gives two solutions with wave vectors k_1 and k_2, corresponding to SPPs which exist at the silver-air and silver-glass interfaces respectively. The spatial beating between these results in a stationary intensity modulation with a period λ_s. At $p = 0.83$, we estimate λ_s ~640 nm for incident light at 633 nm. The experimentally obtained oscillation period from Fig. 2 is about 690 nm and agrees well with the estimated value of λ_s.

The oscillations of the intensity correlation function at low metal coverage $p \leq 0.4$ once more indicate the interference of two propagating waves, a result that is counterintuitive. The oscillations cannot be explained by backscattering in the sample since the period is significantly different from $\lambda_{\|}/2$ ($\lambda_{\|}$ = 610 nm). Therefore, there exists another propagating wave with a *k*-vector different from the incident one. We propose an explanation for a propagating surface wave based on the collective excitation of surface plasmon modes of individual metal particles and the dipolar coupling between them [7]. At $p \leq 0.4$, the sample consists of individually separated but closely spaced metal grains of nearly uniform size which can be approximated as dipolar oscillators with identical resonant frequencies. Consequently, the surface plasmon resonances of these grains are almost the same and impinging light collectively excites these dipole modes, which oscillate with similar amplitude but different phase, resulting in the formation of a propagating surface excitation wave. This wave is expected to propagate more slowly than the impinging one, with a wave vector $k_p > k_{\|}$.

Unlike dielectric random media, localized modes and delocalized modes are believed to coexist in metallic random systems. Our experimental observation of the localization-delocalization transition illustrates that the influences of these two types of eigenmodes on propagation and localization of SPPs vary with structure and metal content. At low and high metal coverages, delocalized modes dominate the propagation of surface plasmon polaritons. In the intermediate regime of metal concentration, localized modes take over and result in the localization of near-field energy.

In summary, we obtained for the first time, near-field intensity correlation functions in semicontinuous metal-dielectric films with varying metal concentration. As the metal filling fraction was increased, we observed in the near-field a transition from propagation to localization and back to propagation of optical excitations.

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