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Surface Plasmon Nanooptics in Plasmonic Band Gap Structures: Interference of Polarization Controlled Surface Waves in the Near Field

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Nanoscopic emission from periodic nano-hole arrays in thick metal films is studied experimentally. The experiments give direct evidence for SP excitations in such structures. We show that the symmetry of the emission is governed by polarization and its shape is defined the interference of SP waves of different diffraction orders. Near-field pattern analysis combined with the far-field reflection and transmission measurements suggests that the SP eigenmodes of these arrays may be understood as those of ionic plasmon molecules.

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Ever since Ebbesen et al. [1] discovered enhanced light transmission through thick metal films punctured with periodic nano-hole arrays, there has been an upsurge of interest in such nanostructures. Recent experimental and theoretical works have attributed the enhanced transmission to the excitation of surface-bound waves - surface plasmons (SP) [2–4] in terms of which the spectral positions and strengths of the transmission resonances are determined [1,5–8,11–18]. In addition, these structures display characteristics of two-dimensional band gap materials, and therefore they are frequently referred to as plasmonic band gap structures. However, the detailed role of SP in physics of transmission enhancement is currently under much debate [18]. Specifically, the eigenmodes of these plasmonic bandgap structures, their relation to band gap formation and the microscopic origin of the line shapes of the transmission spectra still need to be identified. Here, optical near-field experiments combining spatial and spectral resolution are relevant, as they allow for a direct mapping of the eigenmodes of such subwavelength waveguide structures [19,20]. First, near-field experiments with off-resonant excitation [6] show well-defined localized transmission peaks centered at the nano-holes, as one would expect for a single apertures [21,22]. Yet, these experiments give no direct evidence for SP excitations.

In this Letter, we study near-field emission patterns in plasmonic band gap structures under resonant excitation conditions. Experiments performed near the air-metal [1,0] SP transmission resonance give unambiguous evidence for SP excitations in periodic nano-hole arrays. The symmetry of the near-field pattern is governed by the incident polarization and its spatial shape is described as a coherent superposition of SP waves of various diffraction orders. These patterns are therefore representative of the Bloch wave eigenmode of plasmonic band gap structure. In contrast, at the sapphire-metal [1, 1] SP resonance, localized light emission around the holes is seen. This gives first ex-

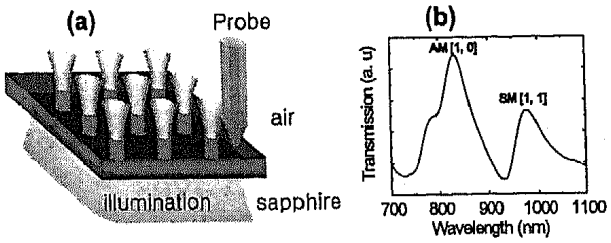

FIG. 1. (a) Schematics of our experiment (b) Transmission spectrum of a gold sample with period=760 nm, hole size=250 nm, and film thickness=200 nm.

perimental evidence for recently proposed theoretical models that describe the SP veigenstates on the two metal surfaces as those of ionic "plasmonic molecules" [14], in much the same way as electronic state of isolated atoms combine to form molecular levels.

Fig. 1(a) is the schematics of our experiments. We employ a near-field scanning optical microscope [23,24] in the transmission geometry. The sample is a metal nano-hole array grown on a sapphire substrate. A Ti:Sapphire laser excites the sample on the sapphire-metal side near normal incidence and a metal-coated fiber-tip with a sub-100 nm aperture collects light on the air-metal side. A far-field transmission spectrum of a sample with a 250 nm hole diameter and a period of 760 nm in a 200 nm thick gold film is displayed in Fig. 1(b). It shows transmission peaks at SP resonances corresponding to surface charge oscillations at the air-metal (AM) and sapphire-metal (SM) interfaces, respectively. The 820 nm peak corresponds to the air-metal SP resonance with its symmetry along the x or y axes defined as horizontal and vertical, respectively. These are termed AM [1,0] or [0,1] modes. The 980 nm peak is the SM [1,1] sapphire-metal SP resonance with its symmetry along the axis diagonal.

Fig. 2(a) shows an atomic force microscopy image of our gold sample described above. Fig. 2 (b), (c) and (d) show the near-field emission pattern for horizontal, diagonal, and vertical polarizations respectively, indicated by the arrows. The excitation wavelength is 780 nm, near the AM [1,0] resonance. Very unlike non-resonant excitation [6], these images show stripe-like emission patterns, clearly extending outside the aperture onto the flat metal surface. The symmetry of the stripes is perpendicular to the incident polarization, reflecting the longitudinal nature of SP excitations.

We now discuss the wavelength dependence of the emission patterns on a different gold sample with a period of 650 nm and a hole size of 150 nm. Fig. 3(a) shows the far-field transmission spectrum in the region of the air-metal [1,0] resonance. The arrows indicate the spectral positions: 740, 750, 780, and 800 nm at

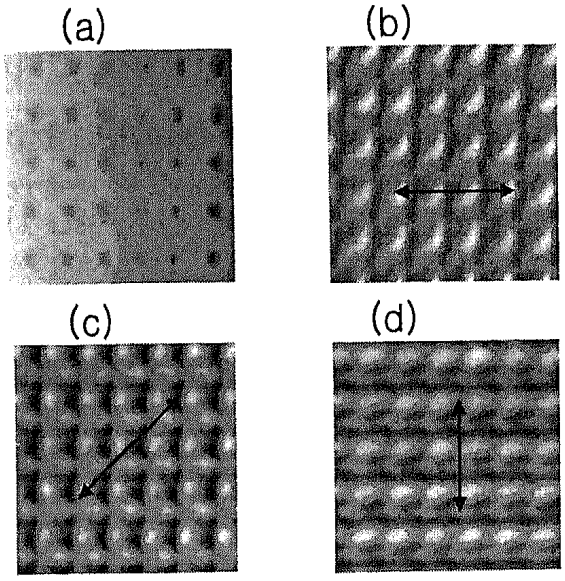

FIG. 2. (a) Topography of the gold sample with 760 nm period (b), (c), (d) Near-field images at different polarizations (arrows). The scanning area is roughly 5 μm × 5 μm.

which the near-field images Fig. 3(b), (c), (d), and (e) were taken, respectively. The polarization was vertical in all images. The images again show stripe-like patterns that run perpendicular to the polarization direction. The details of the images are highly sensitive to the excitation wavelength. Fig. 3(b), e.g., shows two dominant stripes within one period along the vertical direction. On the other hand, Fig. 3 (c), (d), and (e) have one dominant stripe in one period, with localized emission around the holes. Often, a fine structure exists, such as the dark spots near the center of the aperture, within the localized emission areas in Fig. 3(c).

In Fig. 3(d) and (e), the emission pattern changes significantly. Again we see strong patterns centered on the metal surface, in between the apertures. In addition we now observe a pronounced emission that is clearly centered at the apertures, and more strongly localized than the emission spots in Fig. 3(c).

The complicated, yet periodic emission patterns and also their strong wavelength dependence imply that the patterns arise through interference between coherently propagating SP's associated with different diffraction orders, characterized by the wave numbers $\pm\frac{2\pi}{a_0}(m,n)$ for integers m and n. The relative amplitudes and phases of the various wavevector components depend sensitively on wavelength, and should also critically depend on such sample parameters as the size of holes. For instance, the smaller the hole sizes, the larger its spatial frequencies are [21,22]. Therefore, in samples with smaller hole sizes the field inside the apertures is composed of a broader distribution of wavevectors. Hence more SPs diffraction

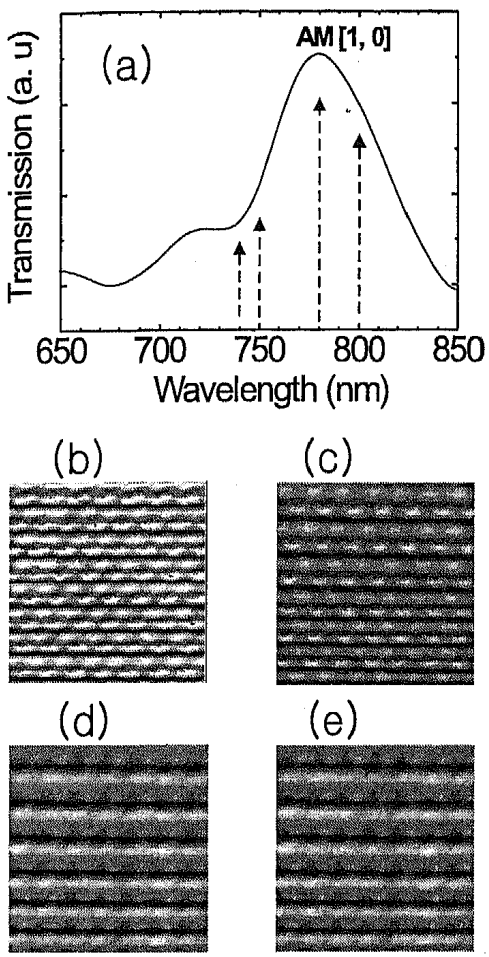

FIG. 3. (a) Transmission spectrum of a gold sample with period= 650 nm, hole size=150 nm, and film thickness=200 nm (b), (c), (d), and (e): Near field emission patterns at wavelengths of 740, 750, 780, and 800 nm, respectively.

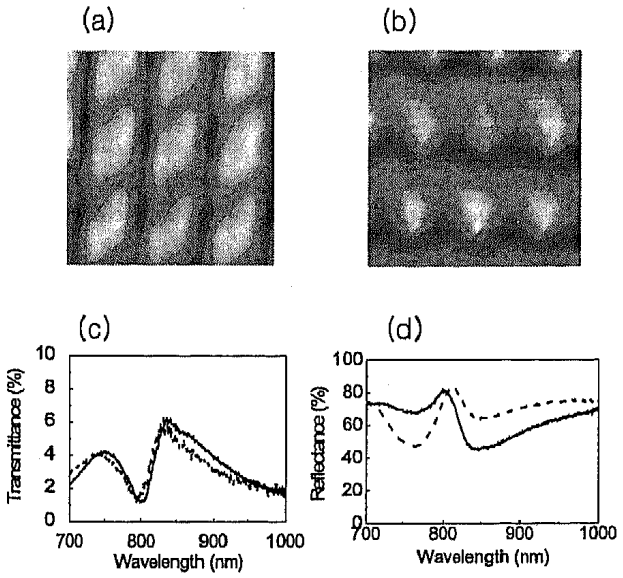

FIG. 4. Comparison of air-silver [1,0] mode and sapphire-silver [-1,1] mode. Holes with 250 nm diameter were made on 200 nm thick silver films with a period of 600 nm. Emission patterns of the air-metal [1,0] mode using $\lambda = 730$ nm. (b) Emission patterns of the sapphire-metal [1,1] resonance using $\lambda = 830$ nm. (c) Far-field transmission spectrum for the said silver sample. Dotted lines: Transmission spectrum when the incident light is from the air side. (d) Far-field reflection spectrum taken from the sapphire side in the same case as (c). Dotted lines: Reflection spectrum when the incident light is from the air side.

orders are expected to be excited when the field inside the aperture couples to SP excitations along the metal-air interface. Comparing images shown in Fig. 2 and 3, with 250 and 150 nm hole sizes respectively, qualitatively supports this trend.

It is clear that images shown in Fig. 2 and 3 are closely related to the Bloch waves associated with these plasmonic band gap structures. We argue that we are probing eigen functions in these periodic structures. Unlike in solid state physics however, plasmon waves have vectorial properties and a full characterization requires probing of all vector components [19]. We believe that the metal-coated glass fibers that we use are insensitive to $|E_z|^2$ due to the boundary conditions at the air/glass interface. We mostly probe the field polarized along the incident polarization. This is verified by the quantitative agreement between experimental near-field images and 3D FDTD simulations [25].

Thus far, we have concentrated resonant excitations of the air-metal SP resonance. To relate this to excitations of sapphire-metal resonances, similar near-field experiments were performed on a silver sample with a shorter period of 600 nm period and 250 nm hole diameter. In Fig. 4(a) the sample is excited at the AM [1,0] resonance (750 nm). Again, the emission stripes are perpendicular to the polarization. However, at the [1,1] sapphire-metal SP resonance (830 nm), the near-pattern (Fig. 4(b)) is completely different from those at the air-metal SP resonance. Bright regions are located only around the holes and elongated along the lattice diagonal, i.e. the SP propagation direction. Independent of incident polarization, we find an elongation along the diagonal. The failure to form stripe patterns in this case can be understood from the fact that the SM [1,1] mode is resonantly excited at the sappire/metal interface, on the opposite side from the measuring tip. Due to the difference in dielectric constants of sapphire and air, this excitation, when coupled through the apertures, is not in resonance with AM SP excitations. Its wavelength, once exposed to the air-metal interface, is *incommensurate* with the lattice constant. Hence the coherent propagating AM SP waves cannot be excited and the emission is mainly from the holes. Yet, it is very inter-

esting that the localized emission patterns are aligned along the diagonal direction, still reflecting the symmetry of the SM [1, 1] SP mode that is strongly driven at the other interface.

Our interpretation of the different emission patterns for AM and SM modes is largely corroborated by analyzing far-field transmission and reflection spectra. Fig. 4(c) shows the transmission spectra for the same silver sample illuminated from either the sapphire side (solid lines) or the air side (dotted lines). Both spectra show clear maxima at both the AM and SM resonances and their strength is mostly independent of the illumination side.

While the two transmission peaks have similar peak strength, the reflection spectra shown in Fig. 4(b) display two dips of very different strengths. Furthermore, unlike the transmission spectrum peaks, the two reflection dips reverse their relative strengths when the illumination and detection sides are interchanged. In both cases, the larger dip corresponds to the SP excitation at the illuminated interface side.

In the case of the sapphire-side illumination, the AM SP is excited mainly through the coupling provided by the nano-holes. This is why the reflection dip for the air-metal SP mode is much weaker than that for the sapphire-metal mode. On the other hand, once excited at the air-metal side, air-metal SP can propagate, interfere, and can be converted to light very efficiently. This picture is supported when we normalize the transmission peak to the reflection dip. For the AM [1,0] resonance, the strength of the transmission peak divided by that of the reflection dip is approximately 40 %, highlighting the extremely high conversion efficiency. In contrast, the sapphire-metal mode, with transmission peak at about 0.05 and the reflection dip at about 0.4 has much lower conversion efficiency of only 12 %, suggesting that most of its energies is dissipated into heat losses or re-emitted into far-field radiation on the sapphire-metal interface . Once exposed to the AM side, this mode emits light mostly around the holes.

To summarize, we have shown that interference of coherently propagating SP waves generates stripe-like near-field emission patterns that are perpendicular to incident polarization and SP propagation. These images reflect Bloch eigenstates of periodic plasmonic band gap structures, and it appears interesting to relate these findings to bandgap formation in these structures. We explained different emission patterns for air-metal and sapphire-metal SP modes as properties of plasmonic excitations at each interface that are linked through nano-apertures. Our work clearly shows that these metal nano-structures can be a very efficient source of coherently propagating surface waves with possible applications in nano-optic devices. The work in Korea was supported by MOST (NRL program) and that in Germany by DFG, EFRE and SQID.

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