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Localization of surface plasmon polaritons in hexagonal arrays of Moiré cavities

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In view of the progress on the confinement of light, we report on the dispersion characteristics of surface plasmon polaritons (SPPs) on two-dimensional Moiré surfaces in the visible part of the electromagnetic spectrum. Polarization dependent spectroscopic reflection measurements show omnidirectional confinement of SPPs. The resonance wavelength of SPP cavity modes can be adjusted by tuning the propagation direction of SPPs. The results may have an impact on the control of spontaneous emission and absorption with applications in light emitting diodes and solar cells, as well as in quantum electrodynamics experiments. © 2011 American Institute of Physics. [doi:10.1063/1.3529469]

Recent progress in the study of light-matter interaction has stimulated research on the propagation of surface plasmon polaritons (SPPs) on various surfaces. Despite the current data on the improvement of SPP induced emission and absorption in semiconductor devices, the detailed nature of the interaction remains to be understood.1 It is, by now, known that at the atomic scale, the relaxation of atoms may be manipulated by controlling the availability of the final density of optical states.2 Similarly, engineering the available density of states gives the ability to control the group velocity of localized SPPs in cavities which has prompted recent work on cavity-to-cavity coupling, as well as array geometry of the cavities. Because SPPs can be manipulated at nanoscale dimensions and are very sensitive to the changes on the surfaces, they are considered as candidates in the modification of many surface related phenomena.3–10 Since the electromagnetic energy of SPPs are generated and localized between the metal and the dielectric interface, three-dimensional confinement of SPPs is only possible by texturing metal surfaces in two dimensions, which results in strong enhancement of light-matter interaction. Accordingly, the fabrication of two-dimensional grating structures attracted great interest in recent years.3,4,6,10 Efficiency of light emitting diodes have been enhanced using two-dimensional metallic grating.10 This is thought to be due to overlapping of resonantly coupled incident light to SPP excitations with the photoluminescence band of the organic layer. The full plasmonic band gap for a two-dimensional uniform metallic grating has also been demonstrated.3 To date, several examples of plasmonic Bragg reflectors, waveguides, and lenses have been demonstrated using one-dimensional (1D) and two-dimensional (2D) metallic arrays.3,11,12 and propagation of SPPs on these structures has been investigated using near-field optical microscopy.12 Fabrication of 1D Moiré surface containing coupled SPP cavities has been shown.7 By appropriate design of Moiré surfaces, the group velocity of SPPs has been reduced.8 Furthermore, omnidirectional localization in the plane of the 2D Moiré surface is expected to be strong and leads to amplification of the interaction of SPPs with surface processes. However, fabrication and optical characterization of Moiré type 2D SPP cavities have not been reported until now.

The aim of this study is to fabricate 2D SPP cavities and measure the dispersion of SPPs using polarization dependent reflection measurements to determine the degree and directionality of the localization at different azimuthal angles. The fabrication of Moiré surfaces has been achieved using interference lithography (IL), which allows fabrication of 1D and 2D structures on a large scale which is nearly unattainable using other fabrication techniques.3–9

A collimated He–Cd laser beam of 325 nm wavelength was used to form interference fringes onto the photoresist (∼100 nm thick S1800-4 and ∼200 nm thick antireflection coating BARLi) layer spun on a glass substrate. Sequential exposures at slightly different illumination angles generate 1D Moiré surface pattern. To obtain a 2D Moiré pattern, the substrate is rotated by 60° around its normal axis followed by a second exposure. The fabrication of the Moiré surfaces is highly reproducible as attested by routine scanning electron microscope (SEM) or atomic force microscope (AFM) observations. Extreme care, however, needs to be taken to optimize exposure time (∼30 s) of the photopolymer in IL setup, film thickness, and chemical development of the exposed photopolymer for proper delineation of the Moiré profile. In reflection measurements, the depth of the grating has to be properly adjusted to observe the dispersion of SPPs since the dispersion of SPPs vary with the grating depth.9 After developing (∼10 s) the exposed photoresist in a solution of AZ 400K, a thin layer of silver film (∼40 nm) was evaporated directly on the polymer surface to support the propagation of SPPs. The Kretschmann (KR) configuration was established by attaching the glass sample onto a prism using index matching fluid. Reflectivity measurements were performed by varying the incidence angle and monitoring the reflection as a function of wavelength from which dispersion curves of SPPs were obtained.

Figures 1(a) and 1(b) show SEM images of the fabricated 1D and 2D SPP Moiré cavities, respectively. The micrograph in Fig. 1(b) shows a nearly hexagonal arrangement of SPP cavities on the surface. In addition, the structures were further characterized using AFM [Fig. 1(c)]. Line scans across the coupled cavities as a function of azimuthal angle.
show the location of the SPP cavities at each propagation direction [Fig. 1(d)]. It is obvious from the line scans that the size of SPP cavities changes with the azimuthal angle. The profile of 2D Moiré surface can be expressed using the following formula:

\[ S(x, y) = \sin(2\pi x/\Lambda_1) + \sin(2\pi x/\Lambda_2) + \sin[(2\pi(x/2 + \sqrt{3}/2y))/\Lambda_1] + \sin[(2\pi(x/2 + \sqrt{3}/2y))/\Lambda_2], \]

where \(\Lambda_1\) and \(\Lambda_2\) are the periods of the superimposed uniform gratings. The angle between the two different surface modulation directions is 60° for hexagonal SPP cavities.

The reflectivity maps showing the band structure of 2D Moiré surface at each azimuthal angle have been constructed using an ellipsometer WVASE32 (J. A. Woollam Co., Inc., USA) configured as a reflectometer. The azimuthal angles of 2D Moiré surface were changed by rotating the sample with respect to the prism. The schematic representation of the prism coupling technique in the KR configuration used for obtaining reflection measurements is shown in Fig. 2(a). SPPs are excited only when the projection \(k_x\) of the wave vector of the incident light matches that of SPPs at the silver-air interface. The magnitude of \(k_x\) is given by \((2\pi/\lambda)n_p\sin(\Theta)\), in which \(\lambda, n_p\), and \(\Theta\) are the wavelength of the incident light, the refractive index of the prism, and the angle of incidence, respectively. The dispersion curve of 1D Moiré is similar to the one shown in Fig. 2(b). It should be clarified here that for 1D SPP cavities, the dispersion curve is obtained for only one propagation direction which is parallel to the Bragg vector of the grating. The regions in the dispersion curves result from incident light that has been absorbed through resonant excitation of SPPs which propagate on the silver-air interface. As the size of the SPP cavity decreases, coupled resonator optical waveguide type plasmonic waveguide band formation within the band gap region of unperturbed uniform grating can be observed. A nearly hexagonal arrangement of SPP cavities on the surface is shown in the micrograph [Fig. 1(b)]. The distance

![Image of SPP cavities and dispersion curves](image_url)

**FIG. 1.** (Color online) (a) A SEM image of SPP cavities on a 1D Moiré surface. (b) A SEM image of the hexagonal array of SPP cavities on a 2D Moiré surface. The inset indicates the cavity region. (c) An AFM image of the same structure shown in (b). The inset shows the Fourier transform of the image, demonstrating the hexagonal pattern of 2D plasmonic crystal. (d) Line scans across the SPP cavities for each propagation direction. Cavity is located where the depth of the grating approaches to zero.

**FIG. 2.** (Color online) (a) Schematic representation of the KR configuration used to perform reflection measurements. The image used in the scheme is drawn in MATLAB using Eq. (1). The symmetry of the SPP cavities are hexagonal. The plasmonic crystal is mounted on a prism. Dispersion curves of SPPs on 2D Moiré surfaces for azimuthal angles (\(\phi\)) of (b) 0°, (c) 15°, (d) 30°, and (e) 45°. The dispersion curves and outside the dispersion curves show SPP coupled and uncoupled modes, respectively.
between the SPP cavities is around 2.5 μm. There are around 150 nm diameter silver islands with a periodicity of 300 nm within and outside the cavity region. The reflection measurement is shown in Fig. 2(b). The curve clearly shows a plasmonic band gap and a cavity state formation. The band edge and cavity state energies of SPPs on the 2D Moiré surface at 0° azimuthal angle and 1D Moiré surface are comparable.7–9

In order to map out the entire azimuthal distribution of SPPs, dispersion curves were obtained for the full range of propagation directions, i.e., azimuthal angle rotations 15° apart with respect to the prism. Figure 2 shows dispersion curves at 0°, 15°, 45°, and 60° azimuthal angles. The dispersion curve for each propagation direction exhibits a clear SPP band gap and cavity state in which the energies of SPP states depend on the propagation direction [Figs. 2(b)–2(e)]. Due to the sixfold symmetry of the 2D SPP cavities, the results are expected to repeat themselves for each 60° rotation. Therefore, the resonance wavelength of the 2D SPP cavities can be controlled by varying only the propagation direction of the SPPs. With 15° azimuthal angle rotation, the wavelengths of the band edges and the cavity states are blue-shifted with respect to the 0° propagation direction. The dispersion curves of SPPs were obtained for each azimuthal angle rotation and, finally, the upper and lower branches and cavity state wavelengths were drawn on polar axes [Figs. 3(a) and 3(b)]. The band gap and cavity state occur in each propagation direction when the wave vector of SPP intersects the Brillouin zone boundary. This can be achieved when

\[ k_{\text{SPP}} \cos(\phi) = G/2, \]

in which \( G \) and \( \phi \) are the Bragg vector and azimuthal angle, respectively.3 Since the wavelength of the SPP is related to SPPs wave vectors, we can plot the experimental data from dispersion curves (Fig. 2) on polar axes in order to obtain the symmetry of the Brillouin zone (Fig. 3). Using the above equation and taking the values for cavity state (620 nm), the variation of SPP states for all the propagation directions on polar plot axes can be analytically drawn. The analytically calculated and the experimentally obtained data for the cavity mode as a function of the azimuthal angle are shown in Fig. 3(b), where the plot shows good agreement with the experimental data. The band edge and cavity state wavelengths repeat themselves because of sixfold symmetry of the hexagonal lattice. In each propagation direction, the plasmonic band gap occurs when the wave vector of the SPPs intersects the Brillouin zone boundary. It is clear from Eq. (2) that the wavelength of the SPP is related to the propagation direction of the incident light. The wavelength of the SPP states changes with the \( \cos(\phi) \) and the energy of the SPP states varies with \( 1/\cos(\phi) \) since the energy of SPP is linearly related to its wave vector. Therefore, the symmetry of 2D SPP cavities shown in SEM and AFM images in Fig. 1 are further confirmed in the polar plots in Fig. 3. It is clear from the polar plots that the wavelength of the SPP cavity mode can be tuned by changing the propagation direction of the SPPs.

In conclusion, we have investigated dispersion of SPPs in 2D arrays of SPP cavities on Moiré surfaces. Polarization dependent spectroscopic reflection measurements have shown a plasmonic band gap and a cavity state for all the azimuthal angles investigated. The SPP state energies have shown sixfold symmetry as indicated in the reflection measurements. Omnidirectional localization of SPPs on the fabricated 2D plasmonic crystal has been shown. Such two-dimensional arrays of SPP cavities are good candidates, for example, for demonstration of SPP cavity based plasmonic lasers13 and for investigation of SPP cavity based Rabi splitting.14

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