Full Photonic Band Gap for Surface Modes in the Visible

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(Received 23 May 1996)

We report the first observation of a full photonic band gap for surface modes. An experimental study has been made of the propagation of surface plasmon polaritons on a silver surface that is textured with a hexagonal array of dots with a periodicity of 300 nm. We find that propagation is prohibited in all directions for modes with energies between 1.91 and 2.00 eV. [S0031-9007(96)01261-6]

PACS numbers: 42.70.Qs, 71.36.+c, 73.20.Mf

An excited atom can only relax via the spontaneous emission of a photon if its energy matches that of an available optical mode. This result is central to cavity QED and has led to the recognition that spontaneous emission by atoms is not necessarily a fixed property but can be modified by imposing boundary conditions on the electromagnetic field. The effect of controlling the density of optical modes in this way has been investigated in a number of simple geometries including planar mirrors [1] and wavelength scale cavities [2,3].

An alternative approach is to use materials that are periodically modulated on the scale of the wavelength of light. The interaction between light and such materials leads to photonic band gaps [4–6] which are similar in origin to the band gaps that occur for electrons in crystals. If such a gap coincides in energy with the excited state of an atom within the material then spontaneous emission will be inhibited. Conversely, at the edge of the gap there is an increased density of states, and the spontaneous emission rate is actually enhanced [7].

For maximum effect a full photonic band gap is required, one that will prohibit the propagation of all modes over a certain energy range, irrespective of polarization or propagation direction. This generally requires a dielectric structure that is periodically modulated in all three dimensions. Such structures have been demonstrated both theoretically and experimentally, mainly in the microwave regime [6]. For many applications, however, one would like a band gap that operates in the visible part of the spectrum. Typically, it is in this spectral range that processes such as spontaneous emission are of most importance. Fabricating a material that is modulated in 3D with the required submicron periodicity is a considerable challenge and has restricted progress in this field. However, there is much that can be done using surface modes rather than bulk modes. It is this case that we address in this work.

Surface modes provide a simpler system in which to study photonic band gaps, one that is accessible to both experimental and theoretical investigation. Since such modes are, by their very nature, bound to an interface, mode propagation can be completely blocked by a photonic band gap that operates in just the remaining two

dimensions. This can be achieved by periodically modulating only the interface, removing the need for a fully three-dimensional structure. For a surface mode this modulation may take the form of either a modulated refractive index or a textured interface. We have chosen the second of these alternatives since suitably textured surfaces with a submicron periodicity are readily fabricated using standard holographic techniques.

The surface mode that we have used is the surface plasmon polariton (SPP) which is a nonradiative transverse magnetic (TM) mode that propagates at the interface between a metal and a dielectric [8]. The fact that there is only one polarization to consider further simplifies the requirement for a full band gap.

If the metal/dielectric interface is periodically modulated with a Bragg vector (equal to 2π /period) that is twice the wave vector of the SPP, then modes propagating in the direction of the Bragg vector scatter from the surface to form standing waves. It is straightforward to show that there are two possible standing waves with different energies so that there is an energy gap in the mode dispersion [9–11]. In our work to date, we have studied how these gaps depend on the surface profile [12] and have demonstrated that they can be used to inhibit the spontaneous emission from dye molecules adsorbed onto the surface of the metal [13]. However, we have so far restricted ourselves to surfaces that are modulated in only one direction. Such a surface can clearly only prohibit mode propagation over a narrow range of directions. In order to produce a full band gap one needs to consider a surface with a more complex periodic structure. The most promising candidate is a hexagonal array of scatterers. In this letter we present an experimental study of the propagation of SPPs on such a surface and demonstrate that it exhibits a full surface photonic band gap in the visible regime.

Figure 1 is a scanning electron micrograph of the surface that we have studied. It consists of a near hexagonal arrangement of dots. The repeat distance is 300 nm and the dots have a radius of around 100 nm. The symmetry is not precisely hexagonal: Figure 2 gives the dimensions. A holographic technique was used to fabricate this structure from a layer of photoresist

FIG. 1. A scanning electron micrograph of the hexagonal array of dots. The dots are composed of photoresist on a glass substrate. The surface has been coated with a thin film of silver to support the propagation of SPPs.

deposited onto a glass substrate [14]. A thin (40 nm) silver film was evaporated onto the surface to support the propagation of SPPs. In this Letter we report on the SPPs that propagate on the silver/air interface.

In order to probe the SPP gap optically, some form of momentum matching is required to couple the nonradiative SPPs to photons [8]. We have used a standard prism coupling technique to achieve this [8,12]. A matching fluid was used to give good optical contact between the uncoated side of the glass substrate and a prism. Monochromatic light incident on the prism at a suitable internal angle θ resonantly excites SPPs that propagate on the silver/air interface. This absorbs energy from the beam, reducing the reflectivity. For a given photon energy, resonant coupling occurs when the component of the photon wave vector in the plane of the silver/air interface (k_x) matches the SPP wave vector (k_{SPP}) . That is, when θ satisfies

$$
nk_0 \sin \theta = k_{\rm SPP}, \tag{1}
$$

FIG. 2. A schematic of the surface showing the dimensions determined by measuring the diffraction of 457.9 nm wavelength light from the surface. The angles are accurate to $\pm 0.2^{\circ}$ and the lengths to ± 0.2 nm.

where *n* is the refractive index of the prism and k_0 is the wave vector of the incident photon in vacuum. Since $k_{\text{SPP}} > k_0$, this coupling occurs for angles beyond the critical angle. Thus the SPPs are coupled to photons by the evanescent field that penetrates the silver. It is for this reason that the silver film has to be optically thin.

The experimental measurements consisted of recording the reflected intensity as a function of the photon energy and k_x . A white light source and a computer controlled spectrometer were used to produce a collimated TM-polarized monochromatic beam in the wavelength range 400 to 800 nm. The angle of incidence was controlled by placing the sample on a computer controlled rotation stage capable of 0.01° resolution. The intensity of the light reflected from the prism was monitored by a photomultiplier tube, and a reference detector placed before the sample corrected for variations in the source intensity. The beam was mechanically chopped and the signal extracted using phase sensitive detectors.

Reflectivity data were recorded as function of the photon energy and k_x , the computer automatically setting the angle of incidence θ and the wavelength of the spectrometer to the required values for each point. Figure 3 shows a typical set of data. The regions of low reflectivity (dark) are a result of photons that have been absorbed through the resonant excitation of SPPs. Since these photons match both the energy and the momentum of the SPPs, the dark bands in Fig. 3 directly map out the dispersion curve of the SPP. There is a clear gap in the dispersion curve, centered at 1.98 eV.

FIG. 3. A sample set of reflectivity data recorded as a function of the photon energy and k_x , the component of the photon momentum in the plane of the silver/air interface. This data is for a propagation direction ψ of 100 $^{\circ}$ (inset). The light regions represent high reflectivity and the dark regions correspond to low reflectivity. The dark triangle in the bottom right corner is an artifact of the measurement technique. Inset: The propagation direction ψ is defined with respect to one of the principal Bragg vectors. Experimentally, this angle is determined by diffracting the 457.9 nm wavelength beam from an argon ion laser.

In order to map out the entire band structure of the surface mode, dispersion curves were obtained for the full range of propagation directions. SPPs excited via prism coupling propagate in the direction defined by the plane of incidence of the photons. The propagation direction is determined, therefore, by the angle ψ between the plane of incidence and a particular Bragg vector in the textured surface (see inset in Fig. 3).

The reflectivity data also exhibited extra features which, for some propagation directions, lay within the SPP gap. In a further experiment we coated the sample with more silver so that it was optically thick (about 200 nm). As a result, light incident through the prism was unable to penetrate the silver and so could not excite SPPs on the silver/air interface. When reflectivity data such as Fig. 3 were then repeated, the features corresponding to the SPPs vanished, but the additional features within the gap remained. We conclude, therefore, that they correspond to modes on the silver/photoresist interface. The nature of these modes is not yet clear, but for the present we note that they do not propagate on the silver/air interface. They do not, therefore, affect any conclusions that we may reach concerning the propagation of SPPs on that interface.

The dispersion curve for each propagation direction exhibits a clear gap, the energies of the upper and lower branches depending on ψ (Fig. 4). Data is shown only for ψ between 0° and 180°. From the symmetry of the surface, the results will repeat themselves in the range 180° to 360° . It is clear that there is a full gap between 1.91 and 2.00 eV: there are no propagating modes within this energy range in any direction on the silver/air interface.

In each direction the energy gap occurs when the wave vector of the SPP intersects the zone boundary. That is, when

$$
k_{\rm SPP} \cos \psi = \frac{G}{2},\qquad (2)
$$

FIG. 4. The energies of the upper and lower branches of the SPP energy gap plotted as a function of the propagation direction ψ . The angles are accurate to $\pm 0.2^{\circ}$ and the energies to ± 0.01 eV. There is a full gap between 1.91 and 2.00 eV.

where G is the magnitude of the appropriate Bragg vector. Since the energy of the SPP is, to a reasonable approximation, linearly related to its wave vector, we can plot the data in Fig. 4 on polar axes to reveal the symmetry of the first Brillouin zone (Fig. 5). Figure 5 shows the expected hexagonal shape.

From Eq. (2) the energies of both branches should vary as $1/\cos \psi$. The lines in Fig. 5 are plotted according to this equation. They show excellent agreement with the experimental points, except in the corners where the experimental measurements are consistently lower than the simple prediction. At these points the SPPs are strongly scattered from two distinct Bragg vectors, and one might anticipate that this would distort the energies of the stationary states. They will no longer be simple standing waves. Further work is needed to investigate the detailed form of the curvature at these points. With the current sample geometry the natural width of the resonance between the SPPs and photons limits the resolution, thus making such an experimental investigation difficult.

In conclusion, we have reported the first observation of a system that exhibits a full photonic band gap for surface modes in the visible spectrum. We have studied the surface plasmon polaritons that propagate on a hexagonally textured silver surface and have directly measured the dispersion curve for the full range of propagation directions. It should now be possible to study many of the interesting phenomena associated with photonic band gaps in the optical regime using surface modes. These would include inhibited and en-

FIG. 5. The data from Fig. 4 plotted on polar axes (circles). In each propagation direction the band gap occurs when the wave vector of the SPP intersects the first Brillouin zone boundary. Since the energy of the SPP is approximately linearly related to its wave vector, plotting the data in this fashion reveals the symmetry of the Brillouin zone. The lines are plotted according to this simple assumption and show good agreement with the data.

hanced spontaneous emission [4,13] and the role of defect states [6].

The authors are grateful to the EPSRC for supporting this research and to Geoff Bradberry for assistance with data acquisition.

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