Long-Range Coupled Surface Exciton Polaritons

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An entirely new type of coupled surface electromagnetic mode has been observed, the existence of which depends upon a large imaginary component of the dielectric constant of a thin layer. It is a long-range mode, giving a very sharp reflectivity dip for transverse magnetic polarized radiation coupled to it using the attenuated-total-reflection technique. Because its existence relies on the presence of a highly absorbing layer it may open up new avenues for nonlinear optics experiments.

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Surface polariton modes exist at the interface between two materials; they are basically electromagnetic waves coupled to the polarization charge at the interface. These have been thoroughly studied in the case of the surface plasmon polariton which exists at the interface between a metal and a dielectric. Hitherto little attention has been paid to the surface mode which may exist at a boundary between two dielectrics, one of which has a large absorption coefficient. Consider such a boundary between a material specified by ϵ_2 $(=\epsilon_{r2}+i\epsilon_{i2})$ and a pure dielectric with only a real dielectric constant, ϵ_1. For a strongly absorbing material a frequency ω may be chosen for which ϵ_{r2} is zero. In this case solving Maxwell's equations for the surface mode gives for the wave vector along the surface, $\kappa_{s\infty}$ $(=\kappa_{r\infty}+i\kappa_{i\infty})$,

$$\kappa_{r\infty}=\frac{\omega}{c}\sqrt{\epsilon_1}\left(\frac{\epsilon_{i2}[\epsilon_{i2}+(\epsilon_1^2+\epsilon_{i2}^2)^{1/2}]}{2(\epsilon_1^2+\epsilon_{i2}^2)}\right)^{1/2}<\frac{\omega}{c}\sqrt{\epsilon_1} \tag{1}$$

and

$$\kappa_{i\infty}=\frac{\omega}{c}\sqrt{\epsilon_1}\frac{\epsilon_1\sqrt{\epsilon_{i2}}}{\{2(\epsilon_1^2+\epsilon_{i2}^2)[\epsilon_{i2}+(\epsilon_1^2+\epsilon_{i2}^2)^{1/2}]\}^{1/2}}. \tag{2}$$

Since $\kappa_{r\infty}<(\omega/c)\sqrt{\epsilon_1}$, then if radiation of vacuum wave vector $\kappa=\omega/c$ is incident from the ordinary dielectric, this surface mode may directly be excited. One of the few reports of the observation of such a mode at room temperature[1] shows a broad resonance typical of such an absorbing system. Because of the breadth of this resonance very little work has been performed with this type of surface mode. No one appears to have examined the situation where two of these surface modes are brought close enough together to couple. It is this situation which is addressed in this Letter.

Consider the simple arrangement of Fig. 1. The active medium of dielectric constant ϵ_2 is surrounded symmetrically by dielectrics of constant ϵ_1. For the coupled surface mode which is antisymmetric in surface charge,[2]

$$\tanh(\tfrac{1}{2}\alpha_2 d)=-\epsilon_1\alpha_2/\epsilon_2\alpha_1\,, \tag{3}$$

and for the symmetric mode,

$$\tanh(\tfrac{1}{2}\alpha_2 d)=-\epsilon_2\alpha_1/\epsilon_1\alpha_2\,, \tag{4}$$

where

$$\alpha_j=(\kappa_s^2-\epsilon_j\kappa^2)^{1/2},\quad j=1,2\,, \tag{5}$$

with

$$\kappa_s=\kappa_r+i\kappa_i\,. \tag{6}$$

These equations are readily evaluated for a highly absorbing thin layer for which $\epsilon_2=i\epsilon_{i2}$ (i.e., $\epsilon_{r2}=0$). [This condition is realizable in a system having a strong resonance, for example, as found in ZnO (Ref. 3) which near the transverse-exciton frequency has an ϵ_r value of zero with $\epsilon_i\approx 49$.] At small d (i.e., $d\ll\lambda$) it follows from Eq. (4) that for the symmetric mode

$$\kappa_r\approx\sqrt{\epsilon_1}\kappa[1+0.125\epsilon_1(d\kappa)^2] \tag{7}$$

and

$$\kappa_i\approx\sqrt{\epsilon_1}\frac{\kappa}{4}\frac{\epsilon_1^2}{\epsilon_{i2}}(d\kappa)^2\,. \tag{8}$$

Note that both the real and imaginary parts are quadratic in $d\kappa$. As $d\kappa$ tends to zero κ_r approaches $\sqrt{\epsilon_1}\kappa$ and the mode has momentum above that available in the surrounding dielectrics. (To couple to this mode some form of momentum enhancement, such as attenuated total reflection, is therefore needed.) Also, as $d\kappa$ tends to zero κ_i tends to zero giving therefore a very weakly attenuat-

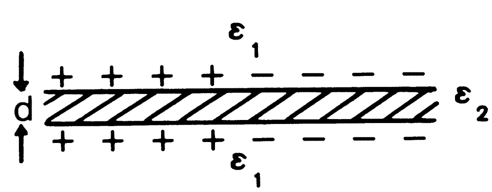

FIG. 1. Symmetric arrangement of thin active medium, of dielectric constant ϵ_2, trapped between two ordinary dielectrics, of dielectric constant ϵ_1.

ed propagating mode which we label the long-range surface exciton polariton, the LRSEP, because of the requirements of a large ϵ_i which may be found at the transverse-exciton resonance frequency in a range of materials.

An interesting point about κ_r is that the only information contained from the film is its thickness. In this thin-film limit the dielectric constant of this highly absorbing layer does not influence the momentum of the resonance, only its width. A further point to note is that the larger ϵ_{i2} the narrower the resonance.

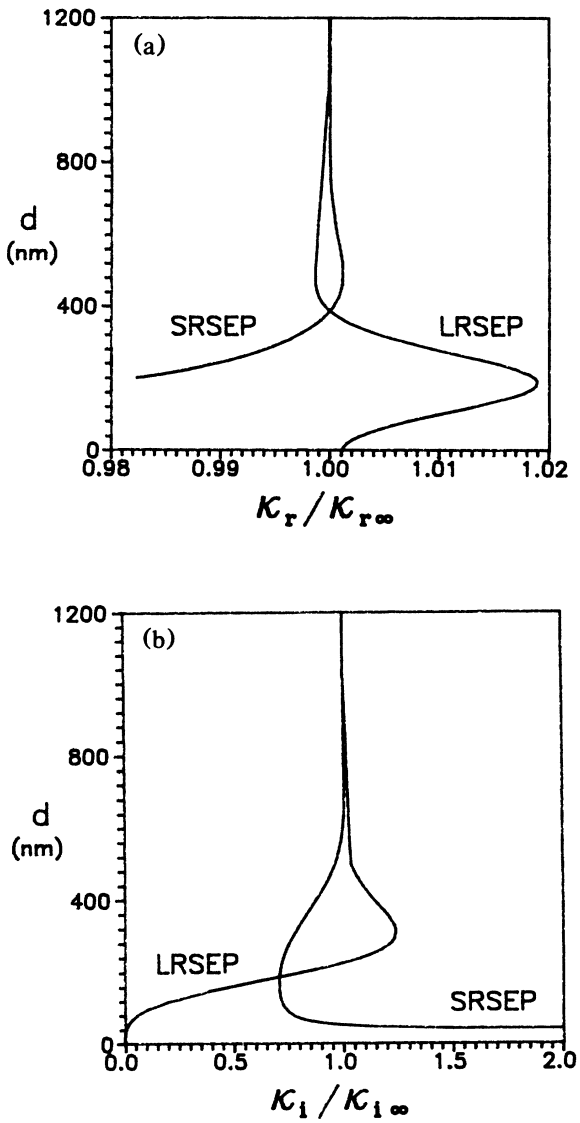

FIG. 2. Thickness dependence of the coupled-surface-mode wave-vector components for both the long-range (LRSEP) and short-range (SRSEP) surface exciton polariton. $\epsilon_1=2.2176$, $\epsilon_2=40i$, and $\kappa=1.853\times10^6$ m^{-1}. The scaling factors $\kappa_{r\infty}$ and $\kappa_{i\infty}$ are the values obtained for the single-interface exciton polariton. (a) Real component; (b) imaginary component.

As the thickness of the layer is increased these simple expressions fail and it is necessary to solve the dispersion relations numerically. This we have done for a range of thicknesses, the results being shown graphically in Fig. 2. In these figures the scaling factors are $\kappa_{r\infty}$ and $\kappa_{i\infty}$, the components of $\kappa_{s\infty}$ for the semi-infinite layer. There are, as expected, two coupled modes, which become in the thin-film limit ($d/\lambda<0.1$) the weakly damped LRSEP and the highly damped short-range surface exciton polariton, the SRSEP.

The primary reason for the existence of the LRSEP is the exclusion of the electric field from the thin layer through the presence of the symmetric surface charge distribution. This results in virtually no power propagating in the thin layer, as is clear from Fig. 3 in which is shown the calculated Poynting-vector amplitude for the LRSEP. In this calculation, using $\kappa=1.853\times10^6$ m^{-1}, the active medium is 40 nm thick with $\epsilon_2=40i$ and $\epsilon_1=2.2176$.

It may be argued that the zero in the real part of ϵ_2 is a special constraint but it is easy to show using numerical evaluation that a small value of ϵ_{r2} has no significant effect upon the resonance provided $|\epsilon_{r2}|<\epsilon_{i2}$. Therefore this hitherto undiscovered coupled surface mode is readily physically realized by studies of a thin layer of material at a frequency which satisfies this constraint.

We have confirmed the existence of the LRSEP using attenuated-total-reflection coupling to a thin layer of vanadium (the experimental configuration is shown in the inset of Fig. 4) at a wavelength of 3.391 μm. At this wavelength the gap between the coupling prism and the active layer, for optimum coupling to a surface mode, may be typically 5–10 μm, which is easy to achieve. In the present study this coupling gap is filled with a fluid, a mixture of hexachloro-1,3-butadiene and tetrachloroethylene, that has negligible absorption at this wave-

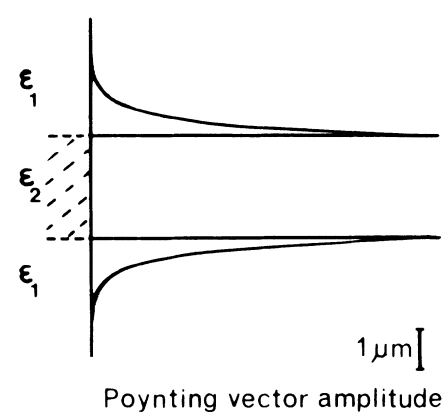

FIG. 3. The Poynting-vector amplitude for the LRSEP corresponding to the fully symmetric system. The active layer has a thickness of $d=40$ nm, with $\epsilon_2=40i$, $\epsilon_1=2.2176$, and $\kappa=1.853\times10^6$ m^{-1}. In the figure the thickness of this layer is scaled by 625× relative to the remainder of the system.

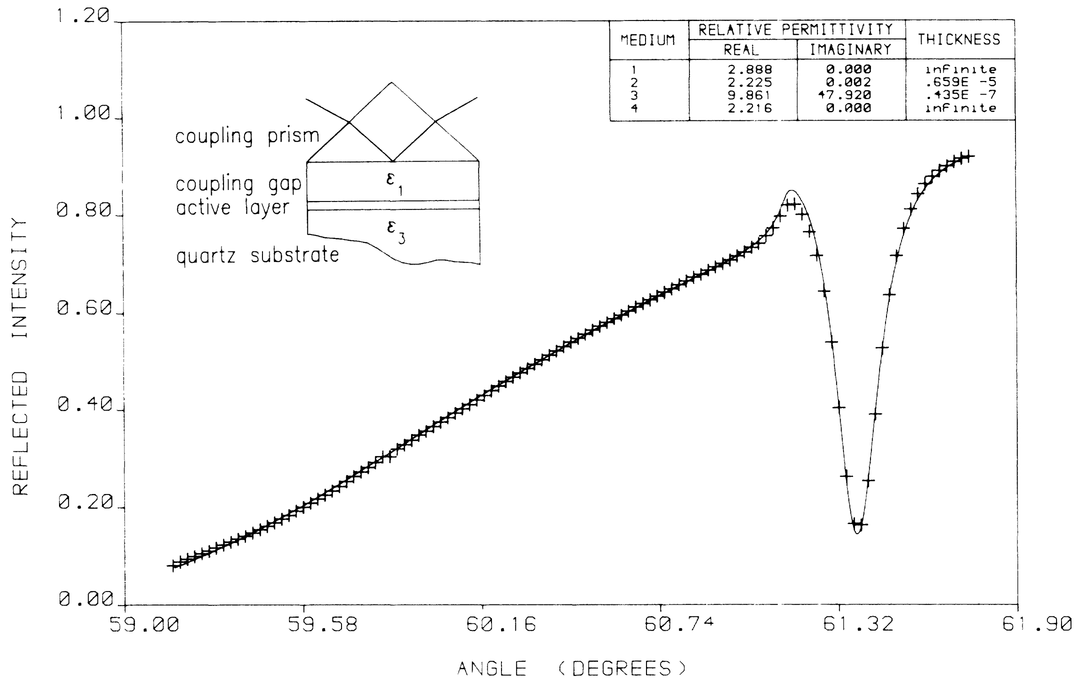

FIG. 4. Data (crosses) for the attenuated-total-reflection signal for transverse magnetically polarized 3.391-μm radiation fitted with Fresnel's theory (full curve). Inset: A sketch of the prism coupling system.

length. The concentration of the mixture is adjusted so that the dielectric constant is very close to that of the quartz substrate, the coupling fluid acting also as the upper dielectric in the now nearly symmetric system. The active layer, a thin film of vanadium, is deposited by vacuum evaporation in a pressure $\approx 10^{-4}$ Pa onto the quartz substrate to a thickness of order 45 nm. At the wavelength used this metal behaves as an excitonic dielectric through the presence of a strong interband transition. This leads to a dielectric constant at 3.391 μm which has a positive real part and a large imaginary part. (From measurements on many vanadium films in other than this present special geometry[4] it is found that $\epsilon_2 \approx 9+48i$.) The reflectivity for transverse magnetic polarized radiation as a function of angle of incidence in the prism is shown in Fig. 4. In this figure the crosses are the experimental data (corrected for reflections at the entrance and exit faces of the prism of index ≈ 1.7) and the full curve is a theoretical fit obtained using Fresnel's equations. The sharp reflectivity maximum at 61.38° is the LRSEP; it is about 0.15° wide, corresponding to a half-width $\Delta\kappa/\kappa$ of 2×10^{-3}.

If we take the fitted parameters for the vanadium film from Fig. 4, then for the LRSEP angle of 61.38° the Poynting-vector distribution shows the clear exclusion of power from the active layer; this is displayed in Fig. 5. There is strong localization of the optical fields at the interfaces with the active medium which results in substantial field enhancement.

In summary, theory predicts that a new type of coupled surface mode, the LRSEP, should exist; it may be very sharp and can be supported by many materials which have strong resonance absorption peaks. This new resonance has been confirmed experimentally for a simple system using 3.391-μm wavelength radiation. There is no doubt substantial scope for using this mode in nonlinear optics applications because of the very strong optical field enhancement available at the surface of the absorbing dielectric. Nonlinear surface mode studies need no longer be constrained to the use of metals or nonabsorbing dielectrics—we await with great interest developments in this area of research.

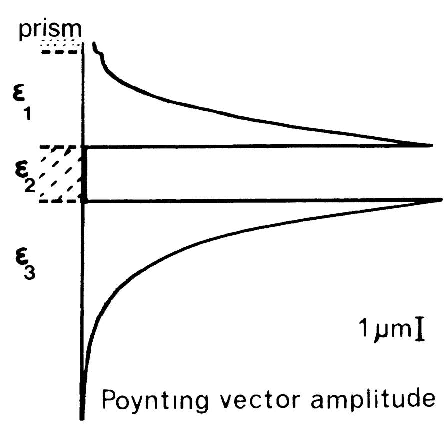

FIG. 5. The Poynting-vector amplitude for the LRSEP at 61.38° in Fig. 4. The thickness of the active layer is scaled by 625× relative to the remainder of the system.

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