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Properties of Surface Polaritons in Layered Structures

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We present the theory of surface polaritons which propagate in a film of a surface-active material placed on a substrate. The theory provides a description of the new mode observed recently by Evans, Ushioda, and McMullen in the Raman spectrum of a GaAs film placed on a sapphire substrate.

Quite recently, Evans, Ushioda, and McMullen (EUM)[1] have observed a new mode in the Raman spectrum of visible light scattered from a GaAs film grown on a sapphire substrate. The mode lies between the LO and TO frequencies of the GaAs film, where its dielectric constant is negative. This suggests that the mode is a surface polariton, since the wave vector $k_{\parallel}$ of the mode lies to the right of the light line appropriate to the vacuum or the sapphire substrate. This paper presents the theory of the mode responsible for the feature observed by EUM, and examines its properties. We begin by deriving the dispersion relation of the mode.

The geometry is illustrated in Fig. 1. Region I ($z > 0$) is the vacuum, region II ($0 > z > -d$) is a thin dielectric film with isotropic dielectric constant $\epsilon(\omega)$, and region III ($-d > z > -\infty$) is an anisotropic substrate described by a dielectric tensor whose only nonzero elements are $\epsilon_{xx} = \epsilon_{yy}$

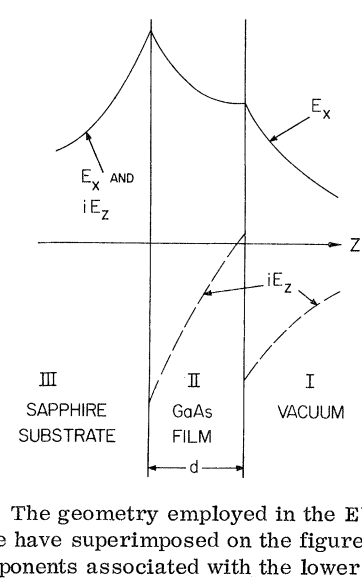

FIG. 1. The geometry employed in the EUM experiment. We have superimposed on the figure the electric field components associated with the lower branch of the dispersion curve with $k_{\parallel} = 8\omega_T \epsilon_{\parallel}^{1/2}/c$.

$=\epsilon_\perp$ and $\epsilon_{zz}=\epsilon_\parallel$.

We study propagation of electromagnetic surface waves in this structure, in particular, waves which propagate in the x direction, with electric vector in the xz plane (TM waves). In the present geometry, there are no surface polaritons of TE character. The α Cartesian component of $\vec{E}$ is

$$E_\alpha(x,t)=E_\alpha(z)\exp(ik_\parallel x)e^{-i\omega t}, \tag{1}$$

with similar expressions for $\vec{D}$ and $\vec{H}$ which is parallel to $\hat{y}$. Maxwell's equations give

$$-\frac{d^2E_x}{dz^2}+ik_\parallel\frac{dE_z}{dz}=\frac{\omega^2}{c^2}D_x, \tag{2a}$$

$$ik_\parallel\frac{dE_x}{dz}+k_\parallel{}^2E_z=\frac{\omega^2}{c^2}D_z, \tag{2b}$$

$$H_y=\frac{c}{i\omega}\left(\frac{dE_x}{dz}-ik_\parallel E_z\right). \tag{2c}$$

We look for solutions localized in the vicinity of the GaAs film, with fields that fall to zero as $z\to\pm\infty$. The results are summarized as follows. In region I, with $\vec{D}=\vec{E}$,

$$E_x(z)=A^{(1)}\exp(-\alpha_0 z)=\frac{\alpha_0}{ik_\parallel}E_z(z)=\frac{ic\alpha_0}{\omega}H_y(z), \tag{3}$$

where $\alpha_0=[k_\parallel{}^2-(\omega^2/c^2)]^{1/2}$, and we require $ck_\parallel>\omega$ for α_0 to be real. In region II, $\vec{D}=\epsilon(\omega)\vec{E}$, and

$$E_x(z)=B^{(1)}\exp(\alpha_1 z)+B^{(2)}\exp(-\alpha_1 z), \tag{4a}$$

$$E_z(z)=\frac{k_\parallel}{i\alpha_1}[B^{(1)}\exp(\alpha_1 z)-B^{(2)}\exp(-\alpha_1 z)]=\frac{ck_\parallel}{\omega\epsilon(\omega)}H_y(z), \tag{4b}$$

where now $\alpha_1=[k_\parallel{}^2-\epsilon(\omega)(\omega^2/c^2)]^{1/2}$ is real when $\epsilon(\omega)<0$.

In region III, $D_x(z)=\epsilon_\perp E_x(z)$, $D_z(z)=\epsilon_\parallel E_z(z)$, and

$$E_x(z)=C^{(1)}\exp(\alpha_2 z)=\frac{i\alpha_2\epsilon_\parallel}{k_\parallel\epsilon_\perp}E_z(z)=-\frac{i\alpha_2}{\epsilon_\perp}\frac{c}{\omega}H_y(z), \tag{5a}$$

where $\alpha_2=\{(\epsilon_\perp/\epsilon_\parallel)[k_\parallel{}^2-\epsilon_\parallel(\omega^2/c^2)]\}^{1/2}$. The quantity $\alpha_2>0$ if $k_\parallel>(\omega/c)\epsilon_\parallel{}^{1/2}$.

We presume $\epsilon_\parallel>1$. Then it is only when $k_\parallel>(\omega/c)\epsilon_\parallel{}^{1/2}$ that we obtain a solution localized to the surface of the structure.

Continuity of $E_x(\vec{x}_1,t)$ and $D_z(\vec{x}_1,t)$ at $z=0$ and $z=-d$ yields a set of homogeneous equations for the coefficients in Eqs. (3)–(5) which admit a nontrivial solution only if

$$\left(1+\frac{\alpha_0}{\alpha_1}\epsilon(\omega)\right)\left(1+\frac{\alpha_2}{\alpha_1}\frac{\epsilon(\omega)}{\epsilon_\perp}\right)-\exp(-2\alpha_1 d)\left(1-\frac{\alpha_0}{\alpha_1}\epsilon(\omega)\right)\left(1-\frac{\alpha_2}{\alpha_1}\frac{\epsilon(\omega)}{\epsilon_\perp}\right)=0. \tag{6}$$

Solution of this equation gives ω as a function of $k_\parallel$.

If we take $A^{(1)}$ as the independent amplitude, then

$$B^{(1)}=\tfrac{1}{2}[1-\alpha_1/\alpha_0\epsilon(\omega)]A^{(1)}, \tag{7a}$$

$$B^{(2)}=\tfrac{1}{2}[1+\alpha_1/\alpha_0\epsilon(\omega)]A^{(1)}, \tag{7b}$$

$$C^{(1)}=\exp(\alpha_2 d)\left[\cosh\alpha_1 d+\frac{\alpha_1}{\alpha_0\epsilon(\omega)}\sinh\alpha_1 d\right]A^{(1)}, \tag{7c}$$

where in these expressions, ω and $k_\parallel$ are related by Eq. (6).

When $d\to\infty$, Eq. (6) yields two modes given by

$$\epsilon(\omega)=-\alpha_1/\alpha_0,\quad \epsilon(\omega)=-\epsilon_\perp\alpha_1/\alpha_2. \tag{8}$$

These are the dispersion relations for surface waves which propagate along the vacuum-film and film-substrate interfaces, respectively. These equations may be rewritten in the equivalent forms

$$\frac{c^2k_\parallel{}^2}{\omega^2}=\frac{\epsilon(\omega)}{1+\epsilon(\omega)},\quad \frac{c^2k_\parallel{}^2}{\omega^2}=\frac{\epsilon_\parallel\epsilon(\omega)[\epsilon(\omega)-\epsilon_\perp]}{\epsilon^2(\omega)-\epsilon_\perp\epsilon_\parallel}. \tag{9}$$

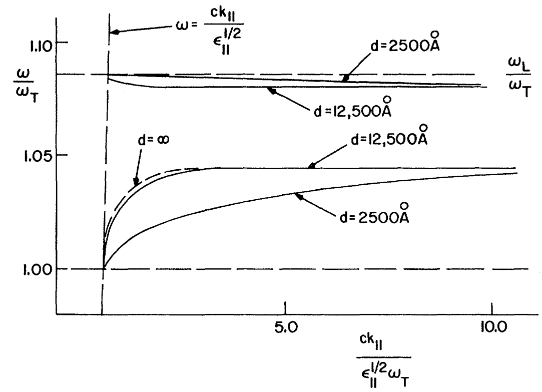

FIG. 2. The dispersion relation for surface polaritons associated with the structure illustrated in Fig. 1, for two values of the thickness d. The dashed curve is the dispersion relation for the "interface polariton" associated with the interface between a semi-infinite GaAs crystal and a semi-infinite sapphire crystal.

The effect of the finite film thickness is to couple the solutions associated with each interface [Eqs. (9)]. In general, then, for each $k_{\parallel}$ we expect Eq. (6) to yield two solutions.

As $k_{\parallel} \to \infty$, the asymptotic value of the frequency approached by each of the two branches is deduced by noting that in this limit, α_0 and α_1 approach $k_{\parallel}$, α_2 approaches $k_{\parallel}(\epsilon_{\perp}/\epsilon_{\parallel})^{1/2}$, and also $\exp(-2\alpha_1 d) \to 0$. Thus, for finite d and $k_{\parallel} \to \infty$, one branch of the surface-polariton dispersion relation approaches the frequency given by $\epsilon(\omega) = -1$, the frequency of the surface polariton at the interface between vacuum and a semi-infinite GaAs crystal when $k_{\parallel} \to \infty$ [see Eqs. (9)], and the second branch approaches the frequency given by $\epsilon(\omega) = -(\epsilon_{\parallel}\epsilon_{\perp})^{1/2}$, the frequency of a surface polariton in the limit $k_{\parallel} \to \infty$ at the interface between a semi-infinite GaAs crystal, and the substrate [see Eqs. (9)].

We have solved Eq. (6) numerically, taking for $\epsilon(\omega)$

$$\epsilon(\omega) = \epsilon_{\infty} + (\epsilon_0 - \epsilon_{\infty})\omega_T^2/(\omega_T^2 - \omega^2), \tag{10}$$

where for GaAs,[2] $\epsilon_{\infty} = 11.1$, $\epsilon_0 = 13.1$, and $\omega_T = 270\ \mathrm{cm}^{-1}$. For the sapphire $\epsilon_{\parallel} = 11.6$ and $\epsilon_{\perp} = 9.35$.[3] In Fig. 2, the solid lines give the dispersion relation for two values of the film thickness: $d = 2500$ Å, the value appropriate to the sample used by EUM, and also $d = 12\,500$ Å. We have superimposed on each graph the dispersion relation of the "interface" polariton associated with the interface between a semi-infinite GaAs sample and a semi-infinite sapphire crystal. We show the dispersion relation only in the region $k_{\parallel} > \omega\epsilon_{\parallel}^{1/2}/c$, where real surface modes exist. Also, in Fig. 1, the spatial variation of the electrical field components is plotted for the case where $k_{\parallel} = 8\omega_T\epsilon_{\parallel}^{1/2}/c$.

In their paper, EUM show that the lower branch of the dispersion curves in Fig. 2 provides an excellent fit to their data. They do not observe the upper branch, presumably because throughout the range of $k_{\parallel}$ explored by them, it lies in the low-frequency wing of the LO phonon line, and cannot be resolved from the LO phonon.

Since we are aware of many unsuccessful attempts to observe surface polaritons by the Raman technique, a few comments are in order about the features of the geometry used by EUM which proved to play an important role.

Consider the surface polariton at the interface between a semi-infinite GaAs crystal and vacuum. As we see from Fig. 2, the frequency for which $\epsilon(\omega) = -1$ lies very close to the LO phonon frequency, so at large angles it is difficult to resolve the surface polariton line from the bulk LO phonon line. This will be the case whenever ϵ_{∞} is large, and this difficulty will be present in most crystals employed in Raman studies. In the near forward direction, where $k_{\parallel}$ is small, the frequency of this surface polariton drops toward ω_T. However, a quantitative estimate of the scattering angles required to enter this regime shows them to be very small and inconvenient to realize experimentally. (This is clear from inspection of curve a in Fig. 1 of EUM[1]). Since the scattering volume associated with backscattering of light from bulk LO and TO phonons in opaque crystals is the same as the scattering volume associated with backscattering from surface polaritons, provided the wavelength of the light is large compared to the skin depth (and hence to the penetration depth of the fields associated with the surface polariton), one expects the scattering efficiences in the two cases to be comparable. Indeed, the data of EUM confirm that this is so. However, for the technical reasons just described, the surface-polariton line is hard to resolve.

Now if a thin film of surface-active material [a crystal with $\epsilon(\omega) < 0$] is placed on a substrate with high dielectric constant, a new surface-polariton branch is obtained. This is the lower branch in Fig. 2, which approaches the frequency for which $\epsilon(\omega) = -(\epsilon_{\perp}\epsilon_{\parallel})^{1/2}$, in our theory. Thus, the large substrate dielectric constant produces a new mode which for large $k_{\parallel}$ is shifted down from near the LO mode toward the TO mode of the film. For the parameters used by EUM, this mode lies roughly midway between the GaAs bulk TO and LO phonon frequencies, where it can be observed at large scattering angles unobscured by the wing of the LO phonon line.

If one compares the dispersion relation of Fig. 2 for $d = 2500$ Å with that for the surface polariton at the interface between a semi-infinite GaAs sample and a semi-infinite sapphire substrate (dashed line in Fig. 2), one sees a pronounced and qualitative difference. When $k_{\parallel}$ is decreased from large values, the lower branch of the dispersion relation in Fig. 2 begins to drop toward ω_T far sooner than the dashed curve. When d is finite, there are two distinct sources of the dependence of the frequency of the lower branch on $k_{\parallel}$, and both sources drive the mode toward ω_T as $k_{\parallel}$ decreases. One source is retardation. However, even in the absence of retardation, the frequency of the lower branch varies with $k_{\parallel}$ if the wavelength of the surface polariton and d are comparable. This is the case in the sample employed by EUM and, in fact, the dispersion relation of the surface polariton is very well fitted throughout the range of $k_{\parallel}$ explored by them by a theory which ignores the retardation effects.

In the limit that retardation effects may be ignored, a simple analytic expression for the surface-wave frequencies may be found from Eq. (6). If $c \to \infty$, then $\alpha_0 \to k_{\parallel}$, $\alpha_1 \to k_{\parallel}$, and $\alpha_2 \to (\epsilon_{\perp}/\epsilon_{\parallel})^{1/2} k_{\parallel}$. Then the frequencies $\omega_{\pm}(k_{\parallel})$ of the two modes are readily obtained by noting that the frequency ω now enters Eq. (6) only in $\epsilon(\omega)$. Equation (6) is quadratic in ϵ and easily solved. The frequencies of the surface waves may then be found by using for $\epsilon(\omega)$ the form in Eq. (10). Let

$$\Delta_{\pm}(k_{\parallel}) = \tfrac{1}{2}(1+\bar{\epsilon})\coth(k_{\parallel}d) \pm \tfrac{1}{2}[(1+\bar{\epsilon})^2\coth^2(k_{\parallel}d) - 4\bar{\epsilon}]^{1/2}, \tag{11}$$

where $\bar{\epsilon} = (\epsilon_{\parallel}\epsilon_{\perp})^{1/2}$; then we find

$$\omega_{\pm}^2(k_{\parallel}) = \omega_T^2\left[1 + \frac{\epsilon_{\infty} - \epsilon_0}{\epsilon_0 + \Delta_{\pm}(k_{\parallel})}\right], \tag{12}$$

where $\omega_+(k_{\parallel})$ is the frequency of the upper branch, and $\omega_-(k_{\parallel})$ the frequency of the lower branch of the surface-wave dispersion relation. For $k_{\parallel} > 2\omega\epsilon_{\parallel}^{1/2}/c$, the region where the data of EUM lie, Eq. (12) fits the full result calculated from Eq. (6) to within graphical accuracy.

We conclude with the remark that since the lower branch of the surface polariton in Fig. 2 clearly owes its existence to the presence of the interface between the GaAs film and the sapphire, the details of the dispersion relation and linewidth of this feature in the Raman spectrum should provide information about the quality of the interface, so this mode may prove quite useful.

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