Surface electromagnetic wave mode and field analysis in a metal–metal-oxide–air system with damping*

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An explanation is provided for the surface electromagnetic wave (SEW) energy ejection from a thin film at the longitudinal and transverse optical-phonon frequencies of the film material. Field ratio calculations identify previously discussed metal–metal-oxide–air SEW modes as those expected from the Berreman configuration, and it is shown that Greenler technique high-incidence-angle-reflectance data on the same system may be similarly interpreted.

I. INTRODUCTION

In a previous paper,[1] we examined the dispersion curves, $1/e$ propagation distance, and Poynting vectors of surface electromagnetic waves (SEW) propagating on a Cu-Cu_2O-air system. The calculation used the full n-media equation[2] and included damping. Structure in the propagation distance L_x as a function of frequency and overlayer thickness was discussed in terms of coupled modes, but no investigation was made into the nature of the modes. In addition, Poynting vector calculations showed that at the transverse (ω_{TO}) and longitudinal (ω_{LO}) optical-phonon frequencies of the film, energy is ejected into the air; i.e., the primary energy transport is in the air. However, no explanation of the discrepancy between this result and predicted field bunching[3] was evinced.

We have subsequently examined these questions, and in Secs. II and III will show that (i) the SEW thin-film energy ejection at ω_{TO} and ω_{LO} can be explained in terms of decoupled modes; (ii) electric field component ratio calculations confirm that the coupled modes are SEW analogues of the usual thin-film transmission-reflection Berreman[4] modes; and (iii) structure observed in reflectance spectra using the Greenler high-incidence-angle-reflection thin-film spectroscopy technique[5] can be similarly explained.

II. ENERGY EJECTION

In Ref. 1, we explained the presence or absence of dips in L_x at ω_{TO} and ω_{LO} by a model which described three-media dispersion in terms of combinations of two-media modes between the layers. A composite of the basic ideas is presented in Fig. 1. The condition for existence of a surface electromagnetic wave at an interface is that $\mathrm{Re}(\epsilon_n) < -\mathrm{Re}(\epsilon_{n+1})$,[6] where ϵ_n is the complex dielectric function of layer n. The figure shows the various combinations of coupled modes between layers 1, 2, and 3 when this condition is met as the frequency is swept. There are regions at ω_{TO} and ω_{LO} where the dielectric function of the film ϵ_2 is negative, but not less than $-\epsilon_3$, the dielectric function of the air. In these regions, there are no two-media modes between layers 1 and 2 or layers 2 and 3. There remains only a (slightly perturbed) mode between layers 1 and 3. Since layer 1, the substrate, is a metal, the field penetration is very small; i.e., only the order of the classical skin depth.[7] Nearly all of the field is in layer 3, the air. The schematic plot of the time-averaged energy flow ratio of air to film at the bottom of Fig. 1 shows this ejection behavior in these regions.

It had been previously predicted[3] that the opposite effect should occur at ω_{LO} ("field bunching"). The error occurred in that the approximate calculations assumed a film mode in this region which, according to the above arguments, does not exist.

III. MODE IDENTIFICATION

If a non-normal incidence reflectance experiment is performed on a metal substrate which has on it a thin film of a material with strong coupling, an absorption band appears in the spectrum at ω_{LO} in addition to or instead of the expected one at ω_{TO}, depending on film thickness. Berreman[4] explained the phenomenon in terms of the fields produced within the thin film and showed that vibrations in the film parallel to the surface are associated with ω_{TO} and those normal to the surface are associated with ω_{LO}, in contrast to the bulk case. In a very thin film on a metal substrate, fields parallel to the metal-film interface, corresponding to ω_{TO}, are very small compared to those normal to the interface, and thus the primary absorption occurs at ω_{LO}, where charge bunching at the interfaces of the film predominates.

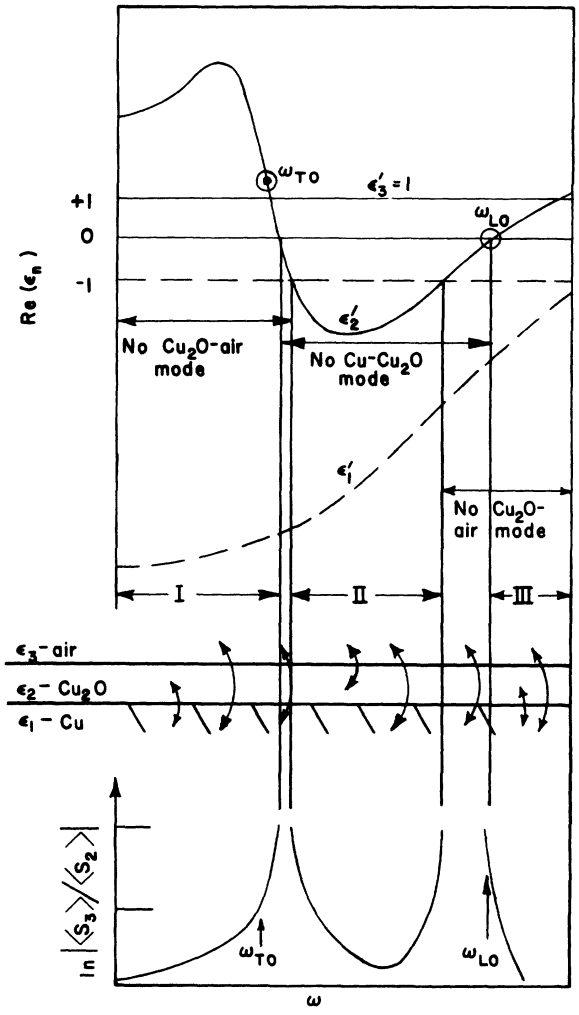

FIG. 1. Schematic of three-media SEW modes using a two-media mode model. The top of the figure shows the exaggerated real part of the dielectric function $\epsilon_n'(\omega)$ for the three layers with the region between $\epsilon_n' = \pm 1$ expanded for clarity. The paired media modes which occur in the various frequency regions are shown in the center of the figure. At the bottom is a representation of the magnitude of the ratio of the time-averaged energy flow in layer 3 to layer 2 as a function of frequency.

The magnitude of the z- to x-component ratio of the electric field in the film (layer 2) of an SEW propagating on a three-media system in the x direction may be written

$$\left|\frac{E_{2z}}{E_{2x}}\right|^2 = \left|\frac{ik_x}{k_{2z}}\left(\frac{-\psi_1 + \psi_2 e^{-2k_{2z}z}}{\psi_1 + \psi_2 e^{-2k_{2z}z}}\right)\right|^2, \quad (1)$$

with

$$\psi_1 = e^{-(k_{3z}+k_{2z})d}(\epsilon_2 k_{3z} - \epsilon_3 k_{2z}),$$

$$\psi_2 = e^{-(k_{3z}-k_{2z})d}(\epsilon_2 k_{3z} + \epsilon_3 k_{2z}),$$

where k_x and k_{nz} are the complex propagation vectors parallel and normal to the direction of SEW propagation for layer n, ϵ_n is the complex dielectric function for layer n, and d is the film thickness. The substrate is defined as layer 1. We used the full n-media equation[2] which includes damping to determine the exact k_x and k_{nz} values.

Figure 2 shows the variation of the magnitude of the z- to x-component ratio of the electric field across the film of an SEW, propagating in the x direction on a Cu-Cu_2O-air system, as a function of frequency for film thicknesses of 500, 1000, and 5000 Å. Also shown is the corresponding variation in $L_x(\omega)$ for the same film thicknesses. At ω_{LO}, the z component is large, and in agreement with the Berreman charge-bunching arguments, absorption by the transverse mode predominates.

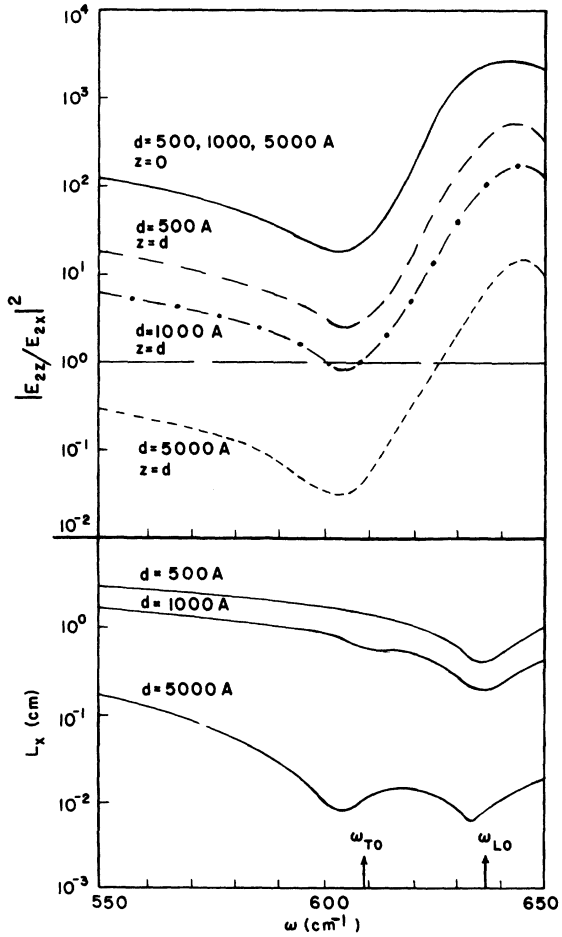

FIG. 2. Magnitude of the ratio of the z to x components of the electric field in the film as a function of frequency and film thickness, and propagation distance $L_x(\omega, d)$ for an SEW on a Cu-Cu_2O-air system. Also shown is the variation of the field ratio as a function of the distance z across a given film. Note that for $z = 0$, the ratio is thickness independent. The optical constants of the Cu and Cu_2O were obtained from Refs. 8 and 9, respectively.

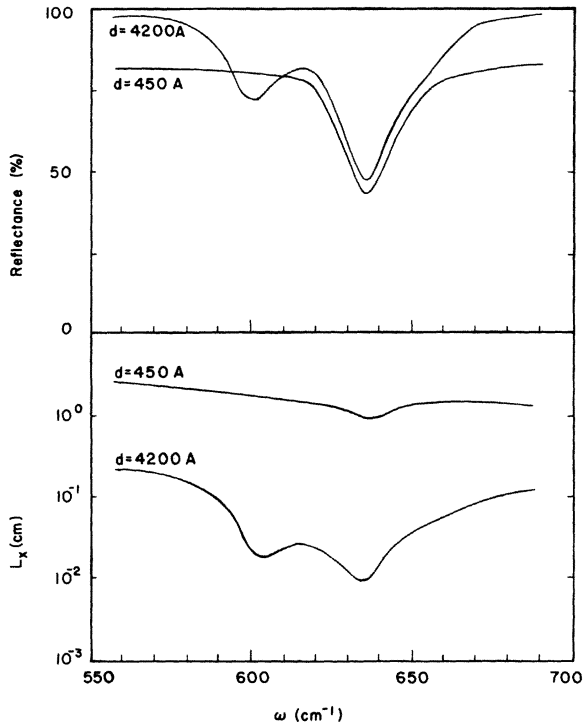

FIG. 3. Percent reflectance for a Cu-Cu_2O-air system from Greenler *et al* . (Ref. 5) compared to the propagation distance L_x of an SEW as a function of frequency for the same system for 450- and 4200-Å-thick Cu_2O films. Note the logarithmic scale for L_x. In experimental practice, the L_x minimum is considerably enhanced by ratioing to the background Cu-air case.

With increasing film thickness, the charge bunching becomes less effective, and the dip in L_x eventually becomes correspondingly smaller.[1] At ω_{TO}, the x component of the field corresponding to the longitudinal mode only begins to predominate at about 1000 Å, and hence ω_{TO} structure in L_x is only observed in film thicknesses greater than this.[1] Thus, purely longitudinal vibrations cannot be observed by SEW in very thin films.

Greenler *et al.*[5] previously examined a Cu-Cu_2O-air system theoretically and experimentally using a high-incidence-angle-reflection-absorption spectroscopy technique. In interpreting their theoretical curves, they observed the absorption band shift for thin films and the appearance of an additional resonance at ω_{TO} for thicker films, but they failed to associate the thin-film dip with ω_{LO}. Figure 3 shows a comparison of the calculations of Greenler *et al.* with our L_x calculations for the same Cu_2O film thicknesses. (In experimental practice, the L_x minimum is considerably enhanced by ratioing to the Cu-air background case.[3]) It is clear that the Berreman mode interpretation applies to their results also. In fact, their case is essentially a higher incidence angle extension of Berreman's results. Greenler *et al.* also show a calculated spectrum for liquid benzene physisorbed on copper which exhibits only a single, nonshifted dip at ω_{TO}. This single dip occurs because of the weakly coupled nature of benzene; i.e., the dielectric function does not pass through zero, and we have observed this behavior experimentally for benzene physisorbed on copper[10] using the SEW method.

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