

Selective absorption of a surface electromagnetic wave propagating on a metal in the presence of a thin dielectric film

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We report observation of a maximum of the absorption of a surface electromagnetic wave by a dielectric film on the surface of a metal near the longitudinal frequency of the oscillations of the dielectric.

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Surface electromagnetic waves (SEW) of the infrared band, propagating over a surface of a conducting medium, have become observable after the advent of lasers of sufficient power, although the discussion of whether SEW are real dates back to the beginning of this century.^[1] Recent experiments^[2] have shown that such a wave attenuates quite slowly as it propagates over the surface of a metal: it can be observed as a distance of several centimeters from the point of excitation. In contrast to a plane wave incident on a metal surface, the amplitude of an SEW is maximal on the surface. These properties of SEW make them particularly attractive for spectral investigations of very thin dielectric films (10 – 1000 Å) on surfaces of well-conducting metals. A noticeable SEW attenuation can be attained in this case not on account of the film thickness but on account of the large propagation length of the SEW over the metal. Variation of the laser wavelength would then make it possible to trace the character of the selective attenuation of the SEW on account of the elementary excitations of the film material.

Our measurements have shown that the SEW propagation length depends essentially on the method used to prepare the metal layer and on the state of its surface.^[3] Knowing the degree of the influence of these effects, we have

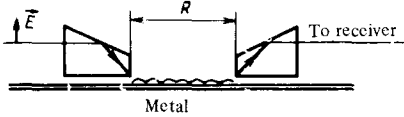


FIG. 1. Paths of rays through prisms.

proceeded to study the frequency-dependent attenuation of SEW in materials having intense selective absorption in the tuning band of the CO₂ laser (943 - 1078 cm⁻¹).

A theoretical analysis of the spectroscopy of films with the aid of SEW^[4,5] has shown that in the presence of longitudinal-transverse splitting the largest SEW attenuation should occur at the longitudinal oscillation frequency ω_{LO} . In experiments performed by the same group (private communication), selective absorption of SEW in a cellulose-acetate film was observed. However, the investigated band has no noticeable longitudinal-transverse splitting whatever. We have therefore chosen substances having much more intense absorption bands in this region: silicon monoxide and apatite. Apatite is a uniaxial crystal that is weakly anisotropic in the region of interest to us, having transverse and longitudinal oscillation frequencies $\omega_{TO} \sim 1040$ and $\omega_{LO} \sim 1080$ cm⁻¹, respectively. Silicon monoxide films are usually amorphous, so that when we refer to transverse-oscillation frequencies we shall have in mind the frequency at which the imaginary part of the dielectric constant is maximal (~ 980 cm⁻¹), and the longitudinal frequency will be taken to be the one at which the imaginary part of the reciprocal dielectric constant is maximal (~ 1130 cm⁻¹). [7]

To excite and register SEW we used a modified attenuated-total-internal-reflection method with two prisms (Fig. 1). When the light beam is incident at an angle $\theta > \theta_{crit} = \sin^{-1}(1/n)$ (n is the refractive index of the prism), an exponentially-attenuating field is produced under the prism. The projection of the wave vector k on the propagation direction is

$$k = n \frac{\omega \sin \theta}{c} > \frac{\omega}{c} .$$

By choosing the angle we can satisfy the momentum-conservation condition. Under these conditions an SEW can be excited by placing the investigated surface at a certain ($\sim \lambda$) distance from the film. The second prism makes it possible to receive the SEW that moves over the surface and attenuates exponentially. The shape of the prism is chosen such that the rays entering the first prism and leaving the second are on the optical axis of the setup. By measuring the signal emerging from the second prism as a function of the distance between the prisms we can determine the SEW propagation length. The dispersion equation of a surface polariton for a metal coated with a dielectric film of thickness d is [5]

$$\left(1 + \frac{\beta_1}{\beta_2}\right) \left(1 + \frac{\beta_2}{\beta_3}\right) - \left(1 - \frac{\beta_1}{\beta_2}\right) \left(1 - \frac{\beta_2}{\beta_3}\right) e^{-2\kappa_2 d} = 0, \quad (1)$$

where $\beta_i = \tilde{\epsilon}'_i / \kappa_i$ and $\kappa_i = \omega / c \sqrt{\tilde{\epsilon}'_i - \tilde{\epsilon}_i}$; the subscripts 1, 2, and 3 stand for air, dielectric, and metal, respectively.

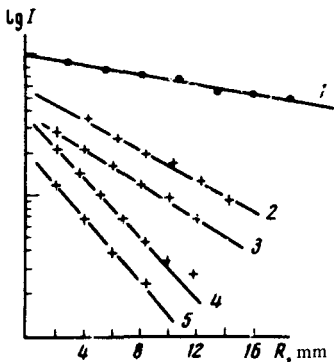


FIG. 2. Dependence of the output signal on the distance between prisms for silver (1) and for silver with apatite film ($d=200 \text{ \AA}$) at frequencies 943 cm^{-1} (2), 770 cm^{-1} (3), 1050 cm^{-1} (4), and 1078 cm^{-1} (5).

The complex wave vector \tilde{k} is contained in the equation implicitly. The propagation length (L) of an SEW in such a system, just as in a pure metal, is equal to

$$L = 1/2 \operatorname{Im} \tilde{k} . \quad (2)$$

Figure 2 shows plots of the output signal against the distance between prisms for silver without a film (1) and for silver with an apatite film 200 \AA thick at various CO_2 -laser emission frequencies (2 – 5). The slopes of the lines drawn through the experimental point (in semilog scale) makes it possible to determine the scattering length L (the distance over which the signal is decreased by a factor e); this distance decreases when the longitudinal frequency of the apatite is approached, but the transverse frequency does not manifest itself in any way. A similar behavior of the propagation length of the SEW is predicted also by calculation (solid lines in Fig. 2).

To eliminate the frequency dependence of the propagation length in the pure metal ($\sim 1/\omega^2$) and to consider only the influence of the film, it is convenient to introduce the coefficient of absorption of the SEW by the film^[4,5]

$$\alpha = \frac{L_0 - L}{L} ,$$

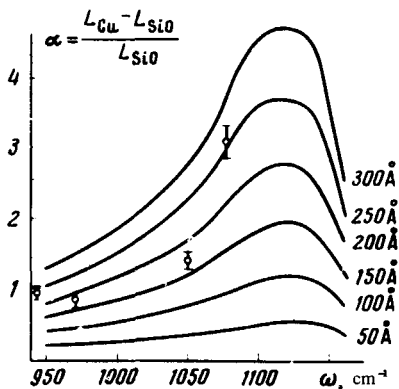


FIG. 3. Frequency dependence of the absorption coefficient on the thickness of a silicon monoxide film on copper. Curves—calculation; points—experimental values for a film 230 \AA thick.

where L_0 is the SEW propagation length in the metal without the film, and L is the length in the metal in the absence of a film. The frequency dependences of this quantity for various thicknesses of silicon monoxide films on copper are shown in Fig. 3. The same figure shows the experimental results for a film 230 Å thick. Just as in the case of the apatite, the absorption coefficient increases as the frequency of the longitudinal oscillations of the film is approached, whereas the frequency of the transverse oscillations remains "unnoticed."

Thus, our measurements confirm the correctness of the theoretical predictions that the absorption of a surface electromagnetic wave by a thin film occurs at the longitudinal-oscillation frequencies. The reason is that the SEW field has both longitudinal and transverse component, but the transverse component interacts weakly with the film, since it is perpendicular to the latter, whereas the longitudinal component is parallel to the film.

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¹R. W. Wood, *Phil. Mag.* **4**, 396 (1902); **23**, 310 (1912); *Phys. Rev.* **48**, 928 (1936); V. Fano, *JOSA* **31**, 213 (1941).

²F. Shoenwald, E. Burstein, and F. M. Elson, *Sol. State Comm.* **12**, 185 (1973).

³G. N. Zhizhin, O. I. Kapustina, M. A. Moskaleva, V. G. Nazin, and V. A. Yakovlev, *Usp. Fiz. Nauk* **117**, 573 (1975) [*Sov. Phys. Usp.* **18**, 927 (1975)].

⁴V. M. Agranovich, *ibid.* **115**, 199 (1975) [**18**, 99 (1975)].

⁵R. F. Bell, R. W. Alexander, C. A. Ward, and J. L. Tyler, *Surface Science* **48**, 253 (1975).

⁶L. C. Kravitz, J. D. Kngseley, and E. L. Elkin, *J. Chem. Phys.* **49**, 4600 (1968).

⁷F. T. Cox, G. Hass, and W. R. Hunter, *Appl. Opt.* **14**, 1247 (1975).