Propagation of surface polaritons near phase-transition points

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The scattering of surface polaritons by fluctuations of the order parameter may serve as the basis for a method of investigating phase transitions in both transparent and opaque media (e.g., in metals). The intensity of this scattering was determined for polaritons propagating along the surface of the metal coated with a thin dielectric film, near the point of the phase transition in the film.

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The dispersion of surface polaritons is determined, as is well known, by the dielectric properties of the media in contact. In the vicinity of a phase-transition point, however, owing to the fluctuations of the order parameter, the dielectric constant also fluctuates and this uncovers new possibilities of transforming a surface electromagnetic wave into a volume wave (emission of a surface polariton), or else into a surface wave but of different frequency and wave vector (Raman scattering of a surface polariton). In the case when the intensity I of the indicated processes becomes large enough, it is precisely their presence which determines the mean free path L of the surface polariton. It is known[1] that this path is particularly large far from phase-transition points ($L_0 \approx 1$ cm) in the IR region of the spectrum for polaritons propagating along the surfaces of metals. The phenomenon in question can therefore manifest itself even in the very simple situation when we are dealing with the influence exerted on the length L by a phase transition in a thin dielectric film (the film thickness D is much less than the polariton wavelength) deposited on a metal substrate.[1)] In this case the analysis is particularly simple, since the propagation length of the surface IR polariton can be obtained from the results of Mills,[3] who considered the influence of statistical surface roughnesses on the polariton mean free path. Mills[3] defined the deviation of the surface from a plane ($Z=0$) by a random function $\zeta(x,y)$ with $\langle\zeta(x,y)\rangle=0$. The presence of these deviations is formally equivalent to the presence at $Z=0$ of a film whose polarizability per unit area

$$a = \frac{\epsilon - 1}{4\pi}\,\zeta(x, y) \tag{1}$$

is a function of x and y (ϵ is the dielectric constant of the medium). The question of interest to us, on the other hand, can be considered by using the results of[3] if it is recognized that the fluctuating part of the local (per cm^2) polarizability of the film undergoing the phase transition can be expressed in the form

$$a - a_0 = \frac{D}{4\pi}\left(\frac{d\epsilon_0}{d\eta}\right)_0 \delta\eta(x, y)\ , \tag{2}$$

where α_0 is the dc component of the polarizability, which causes no scattering, $\delta\eta=\eta-\eta_0$, $\langle\eta\rangle=\eta_0$, while $\epsilon_0(\eta)$ is the film-material dielectric constant corresponding to the value of the order parameter η. Relation (2) means that we are dealing here with a phase transition in which the η-dependent part of $\epsilon_0(\eta)$ is linear in η at small values of η.

It is shown in[3] that for surface polaritons with frequencies ω much lower than the plasma frequency of the metal (for these frequencies we have $|\epsilon| \gg 1$), the intensity of the scattering by the roughness and the corresponding (partial) mean free path L_1 are determined by the relation

$$I \sim \frac{1}{L_1} = \frac{4}{3\pi}\,\frac{\omega^2 F_\zeta(0)}{c^2|\epsilon|^{1/2}}\ , \tag{3}$$

where $F_\zeta(Q_\parallel)$ is the Fourier component of the correlation function

$$F_\zeta(Q_\parallel) = \int d^2 r_\parallel\, e^{iQ_\parallel r_\parallel} \langle \zeta(r_\parallel)\zeta(0)\rangle\ .$$

The principle process in this case is the scattering corresponding to the termination of the surface waves and their conversion into volume waves.

Comparing (1) with (2) and using (3), we find that in our case

$$I \sim \frac{1}{L_1} = \frac{4\omega^2 D^2\left(\frac{d\epsilon_0}{d\eta}\right)_0^2 F_\eta(0)}{3\pi^2 c^2 |\epsilon|^{5/2}}, \tag{4}$$

so that at $T \approx T_c$ the temperature dependences of the quantities I and $1/L_1$ coincides with the temperature dependence of $F_\eta(0)$. On the other hand, in crystals such as quartz, where $\alpha - \alpha_0 \sim \eta^2$ (cf. (2); at $T < T_c$ we have $\eta^2 = \eta_0^2 + 2\eta_0\delta\eta(x,y)$), the use of relation (3) yields $I \sim 1/L_1 \sim \eta_0^2 F_\eta(0)$. Inasmuch as $\eta_0^2 \sim (T_c - T)$ in this case, the role of the fluctuations becomes minor. In the Landau theory of second-order phase transitions[2)] we have $F_\eta(0) \sim |T - T_c|^{-1}$ (see, e.g., [5]) and this, in accordance with (4), corresponds to a sharp decrease of the partial polariton mean free path L_1. The temperature region in which the effect can be revealed by the decrease of the mean free path $L(1/L = (1/L_0) + (1/L_1))$ is determined by the inequality $L_1(T) \lesssim L_0 \approx 1$ cm. If for some reason this inequality is not satisfied anywhere (fluctuations of the order parameters are suppressed, etc.), observation of the effect calls for direct measurements of the intensity of the light produced by the surface wave in the contact region and scattered by this region. Of course, its spectral composition is also of interest. The mechanism considered above is analogous to that discussed in[5], where, however, the questions of the scattering of surface electromagnetic waves was not touched upon. As to the scattering by grains of the new phase (these are discussed, e.g., in[6]), in light of the remarks made in[7], their role calls for a special estimate. We merely emphasize here that when films are used (see above), size effects of the type discussed in[8] can also appear (this raises, in particular, the question of the boundary conditions for the order parameter).

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[1)]The facts that coating with a thin dielectric film far from the phase transition points has only a negligible effect on L_0 follows from the experiments of[2].

[2)]As shown in[4], in the linear case (see (2)) the Landau theory can be valid over a particularly wide range.

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