Scattering of polaritons by fluctuations of the dielectric constant in thin films

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The scattering of polaritons by fluctuations of the dielectric constant ϵ due to a change in the structure of the electron spectrum upon small changes in the thickness of the film is analyzed. Near exciton lines, the fluctuations in ϵ are more important than direct scattering by irregularities of the film boundaries.

The effect of fluctuations of the dielectric constant ϵ on the propagation of polaritons in films is a topic of independent importance for semiconductor films of inhomogeneous composition. It is also a topic of particular interest for thin films with a pronounced quantum size effect, in which the fluctuations in ϵ are closely related to irregularities of the boundaries. In a study of the scattering of electromagnetic waves by fluctuations in ϵ it is convenient to begin by deriving a perturbation theory for the retarded Greens function of the electromagnetic field, $D_{\alpha\beta}(\omega,\mathbf{r},r')$, which satisfies the equation[1]

$$\left[\frac{\partial^2}{\partial x_\alpha \partial x_\gamma} - \delta_{\alpha\gamma}\Delta - \delta_{\alpha\gamma}\frac{\omega^2}{c^2}\epsilon(\mathbf{r})\right] D_{\gamma\beta}(\omega,\mathbf{r},\mathbf{r}') = -4\pi\hbar\,\delta_{\alpha\beta}\,\delta(\mathbf{r}-\mathbf{r}'), \qquad (1)$$

where $x_\alpha = \{x,y,z\}$, Δ is the Laplacian, and $\epsilon(\mathbf{r})$ takes on the values ϵ_1 and ϵ_2 in the half-spaces $z<0$ and $z>d$, respectively. Since we will be using field values averaged over the thickness of the film below, under the assumption $(\omega/c)d \ll 1$, we write the dielectric constant of the film, which occupies the region $0<z<d$, in the form $\epsilon = \epsilon_0 + \delta\epsilon(\rho)$.

Equation (1) can be written in the integral form

$$D_{\alpha\beta}(\mathbf{r},\mathbf{r}') = D^0_{\alpha\beta}(\mathbf{r},\mathbf{r}') - \frac{1}{4\pi\hbar}\int_0^d dz_1 \int d\vec{\rho}_1 D^0_{\alpha\gamma}(\mathbf{r},\mathbf{r}_1)\frac{\omega^2}{c^2}\,\delta\epsilon(\vec{\rho}_1) D_{\gamma\beta}(\mathbf{r}_1,\mathbf{r}'), \qquad (2)$$

where $D^0_{\alpha\beta}$ corresponds to a film with a constant ϵ_0 (the various components of $D^0_{\alpha\beta}$ are given in Ref. 2). An expression of first order in $\delta\epsilon$ for $D^{(1)}_{\alpha\beta}$ can be found by substituting D^0 for D in the integral on the right side of Eq. (2). Using the function $D^{(1)}$, we can easily find the effective perturbation operator corresponding to the fluctuations in ϵ for the scattering of waves of various types. The mean free path is then found by analogy with the case of the scattering of surface plasmons by irregularities of a metal boundary.[3] For example, assuming that the correlation function of the fluctuations is of the form

$$g(\vec{\rho}) = \langle \delta\epsilon(0)\delta\epsilon(\vec{\rho})\rangle = (\delta\epsilon)^2 \exp\left[-\frac{\rho^2}{a^2}\right] \qquad (3)$$

and assuming that we are dealing with polaritons with wave vectors p satisfying the conditions $(\omega/c) \ll p \ll 1/d$, we find the following expressions for the mean free path l for TM and TE polaritons

$$\frac{1}{l_P^{TM}} \simeq \frac{\pi}{4} p^5 (\delta\epsilon da)^2; \qquad \frac{1}{l_R^{TM}} \simeq p^2 \left(\frac{\omega}{c}\right)^3 (\delta\epsilon da)^2 ,$$

$$\frac{1}{l_P^{TE}} \simeq \frac{\pi}{16} p \left(\frac{\omega}{c}\right)^4 (\delta\epsilon da)^2 , \qquad \frac{1}{l_R^{TE}} \simeq \left(\frac{\omega}{c}\right)^5 (\delta\epsilon da)^2 . \tag{4}$$

Here we have assumed, for simplicity, that the relation $pa \ll 1$ holds, and the subscripts R and P correspond to the decay of the polaritons which is caused by the scattering into photons and that which is caused by the scattering into other polaritons, respectively. The TM polaritons, which we are discussing here, exist under the condition $1 \ll -\epsilon \ll 1/(\omega/c)d$, while TE waves exist under the condition $1/(\omega/c)d \ll \epsilon \ll -1/(\omega/c)d^2$. A detailed description of polaritons in thin films can be found in Ref. 4.

In a comparison of (4) with expressions for the decay of polaritons due to scattering by irregularities of the film boundaries, given in Ref. 2, we find that if we take B to be the average height of the irregularities and α their average width [the same as in (3)], then the scattering by fluctuations in ϵ is stronger in the case $\delta\epsilon > \epsilon_0(b/d)$. If the change in ϵ is caused by a change in the thickness d, this condition can be satisfied when the frequency ω is close to one of the resonant frequencies of the film, ω_0, e.g., the frequency of an exciton transition. In this case ϵ becomes

$$\epsilon = \epsilon_{\text{background}} + \frac{A}{\omega(\vec{\rho}) - \omega - i\Gamma} , \tag{5}$$

where

$$\omega(\vec{\rho}) = \omega_0 + \Delta\omega(\vec{\rho}) ,$$

$$\Delta\omega(\vec{\rho}) \simeq 4\omega_p \frac{d - d(\vec{\rho})}{d} , \qquad \omega_p = \frac{\hbar\pi^2}{d^2 m^*}$$

d is the average thickness of the film, m^* is the effective mass of an exciton, and Γ is the width of the exciton level. If the relations

$$A/\Gamma \gg 1, \qquad \omega_p \gg \Gamma, \qquad \Delta\omega_{max} \gtrsim \Gamma \tag{6}$$

hold, then it can be seen from (5) that the quantity $\epsilon(\omega)$ may fluctuate significantly, even if $b/d \ll 1$. Since the change in ϵ for frequencies near ω_0 is on the order of ϵ in this case, we have $\delta\epsilon d > \epsilon b$. The scattering of light and surface polaritons with such frequencies will be determined primarily by the fluctuations in ϵ, not by the direct interaction with irregularities. This statement means that in this case the film boundaries can be assumed planar, and only the change in ϵ need to be taken into account, as was assumed in Eqs. (1) and (2). Accordingly, in analyzing the scattering of polaritons with wave vectors $p \gg \omega/c$, without considering retardation effects [i.e., without considering their scattering into photons, which is much weaker than the scattering of polaritons into each other by virtue of (4)], we find the problem of a two-dimensional system with a random potential. In studying the localization of longitudinal surface plasmons at an irregular metal surface we run into a definitely three-dimensional situation.

In a two-dimensional disordered system, we know that all states must be localized (Ref. 5, for example). Since the mean free path l_P is large in comparison with the polariton wavelength λ_P, however, this localization occurs over distances much greater than λ_P. The states become strongly localized when the fluctuations in ϵ are such that the relation $l_P \sim \lambda_P$ holds. Assuming $a \simeq \lambda_P$, we would thus need $\delta\epsilon = a/d$. Such values of $\delta\epsilon$ are possible under conditions (6). Accordingly, polaritons in thin films with frequencies near ω_0 may be localized over distances on the order of the original wavelength, even if the fluctuations in the film thickness are very small. Using the expression for the dielectric constant near an exciton transition,[4] we can show that conditions (6) hold for films with $d = 100$ Å under the conditions $\Gamma < 10^{13}$ $s^{-1} \simeq 10^{-2}\omega_0$.

Because of the excitation of such localized states by the incident light, the local field may increase significantly at the surface of the film. This effect should lead to a pronounced increase in the effective cross sections for the scattering of light by molecules adsorbed on the film, by analogy with the enhancement effect at irregular surfaces of metals and island films.[6]

We note in conclusion that we have considered only an adiabatic change in the resonant frequency of the film as its thickness is varied. This approach can be justified for only small and smooth variations of $d(\vec{\rho})$, over distances much greater than the average thickness. Incorporating effects of a localization of electrons in the film due to irregularities of the film boundaries in the case of sharper fluctuations in the thickness leads to a more complicated change in the spectrum of the film. The question of the fluctuations in ϵ in this case requires a separate study.

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