Polaritons in thin semiconducting films

L. V. Keldysh
P.N. Lebedev Physics Institute, USSR Academy of Sciences

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It is shown that the oscillator strengths of the excitons in semiconducting films, whose thickness is less than the effective exciton radius and whose dielectric constant is much greater than that of the medium surrounding the film, increase sharply with decreasing thickness of the film. The reflectivity of the film increases in the neighborhood of the excitonic lines and conditions are established for propagation of the electromagnetic waves-polaritons that are localized near the film.

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It was shown in a recent letter[1] that in sufficiently thin semiconductor films whose dielectric constant ϵ greatly exceeds the dielectric constant of the substrate the Coulomb interaction between the charges will increase (compared with bulk samples of the same semiconductor) if the distance between them exceeds the thickness d of the film d. For $d \ll \alpha_0 \equiv \omega\hbar^2(me^2)^{-1}$ (e is the electron charge, $\hbar$ is the Planck constant, and m is the reduced mass of the electron and of the hole), this circumstance will increase the binding energy $\mathscr{E}_0(d)$ and decrease the effective radius α of the Vanier–Mott excitons

$$\mathscr{E}_0(d) = \frac{e^2}{\epsilon d}\left\{ \ln\left[\left(\frac{2\epsilon}{\epsilon_1+\epsilon_2}\right)^2 \frac{d}{a_0}\right] - 0.8\right\}, \tag{1}$$

$$a = \frac{1}{2}\sqrt{a_0 d}\,. \tag{2}$$

The subscripts 1 and 2 here and elsewhere denote quantities associated with the half-space on either side of the film. The excitons in this situation are almost two-dimensional.

The intensity of the optical transition for the dipole-active excitons examined below is determined by $|\phi(0)|^2$, where $\phi(r)$ is the wave function of the relative motion of the electron and hole.[2] It is clear that $|\phi(0)|^2$ is inversely proportional to the "exciton volume," i.e., because of Eq. (2) $|\phi(0)|^2 \sim a^{-2} \sim (a_0 d)^{-1}$. Thus, the binding energy of excitons and the oscillator strength corresponding to them increase with decreasing d. Taking this into account, the contribution of the excitonic transition to the electromagnetic response of the film to the field with the frequency ω and projection $\mathbf{k}$ of the wave vector on the plane of the film can be described by the induced current density $\mathbf{j}$:

$$j_\alpha(\mathbf{k}, \omega; z) = -i\omega\Lambda_{\alpha\beta}\,\chi_{\mathbf{k}\omega}\,\bar{E}_\beta 2\sin^2\frac{\pi z}{d}, \tag{3}$$

$$\chi_{\mathbf{k}\omega} = \frac{1}{\epsilon}\left(\frac{e^2}{dE_{\mathbf{k}}}\right)^2 m\,|V_{cv}|^2 \frac{2\mathcal{E}_{\mathbf{k}}}{\mathcal{E}_{\mathbf{k}}^2 - (\hbar\omega)^2)}, \tag{4}$$

$$\bar{\mathbf{E}} = 2d^{-1}\int_0^d \mathbf{E}_{\mathbf{k}\omega}(z)\sin^2\frac{\pi z}{d}\,dz. \tag{5}$$

Here $\mathbf{E}$ is the electric field, z is the coordinate normal to the plane of the film that occupies the band $0 \leqslant z \leqslant d$, V_{cv} is the matrix element of the velocity operator for a transition from the valence band to the conduction band, the functions $(2/d)^{1/2} \times \sin(\pi z/d)$describe the size-quantized transverse motion of the electrons and holes relative to the film, and $\mathcal{E}_k$ is the resonance energy of the excitonic transition. The dimensionless coefficients $\Lambda_{\alpha\beta} \sim 1$, which characterize the polarization properties of this transition, depend on the symmetry of the c and v bands. For simplicity, we assume that the materials of the film and of the substrate are optically isotropic. Thus, $\Lambda_{\alpha\beta}$ will contain only two different quantities: $\Lambda_\perp \equiv \Lambda_{zz}$ and $\Lambda_\parallel$. The matrix elements V_{cv} for typical semiconductors are associated with the masses and the forbidden bands. In the Kane model,[3] $m|V_{cv}|^2 = \mathcal{E}_k$ with an accuracy to a factor close to 1, which can be included in the definition of Λ. Thus,

$$\chi_{\mathbf{k}\omega} = \frac{2}{\epsilon}\left(\frac{e^2}{d}\right)^2 [\mathcal{E}_{\mathbf{k}}^2 - (\hbar\omega)^2]^{-1}. \tag{6}$$

The most characteristic properties of Eqs. (3)–(6) are the $\chi_{k\omega} \sim d^{-2}$ dependence and the coupling, nonlocal in z, of the induced current with the field.

Following the usual procedure for the optics of thin films,[4,5] we can solve with an accuracy to small terms $\sim [(\omega/c)d]^2$ and $(kd)^2$ the Maxwell equations together with Eqs. (3) and (5) and the equation of continuity in the $0 \leqslant z \leqslant d$ band, and use the resulting solution to match the solutions in the half-spaces 1 $(z<0)$ and 2 $(z>d)$. Henceforth the presence of a film will be taken into account only by these boundary conditions, which, if we allow for the nonlocalizability, have the form:

$$\mathbf{E}_2 - \mathbf{E}_1 = i\tilde{\epsilon}_\parallel\, \mathbf{n} d\left(\mathbf{k}\frac{\mathbf{E}_1 + \mathbf{E}_2}{2}\right) - i\frac{\omega}{c} d\left[\mathbf{n}\frac{\mathbf{H}_1 + \mathbf{H}_2}{2}\right] + i\frac{c\mathbf{k}}{\epsilon\omega} d\,\frac{\epsilon + 2\pi\Lambda_\perp\chi}{\epsilon + 6\pi\Lambda_\perp\chi}\left(\mathbf{k}\left[\mathbf{n}\frac{\mathbf{H}_1 + \mathbf{H}_2}{2}\right]\right), \tag{7}$$

$$\mathbf{H}_2 - \mathbf{H}_1 = -in\,d\left(\mathbf{k}\frac{\mathbf{H}_1+\mathbf{H}_2}{2}\right) + i\tilde{\epsilon}_{||}\frac{\omega}{2}d\left[\mathbf{n}\frac{\mathbf{E}_1+\mathbf{E}_2}{2}\right]. \tag{8}$$

Here $\mathbf{E}_{1,2}$ and $\mathbf{H}_{1,2}$ are the boundary values of the electronic and magnetic fields in the half-spaces 1 and 2, c is the velocity of light, $\mathbf{n}$ is the unit vector of the normal to the film, and $\tilde{\epsilon}_{||} = \epsilon + 4\pi\Lambda_{||}\chi$. Henceforth, we drop the small terms $\sim(\omega/c)d$ and kd [the second term in Eq. (7) and the first term in Eq. (8)] but retain the terms $\sim\epsilon(\omega/c)d$ and ϵkd. The third term in Eq. (7) should be taken into account only in the narrow frequency range in which $|1 + 6\pi\Lambda_{\perp}\chi\epsilon^{-1}| \ll \epsilon^{-1}$.

The coefficient of reflection of light from the structure under consideration at normal incidence

$$R = \frac{(\sqrt{\epsilon_1} + \sqrt{\epsilon_2})^2 + \left(\frac{\omega}{c}d\tilde{\epsilon}_{||}\right)^2}{(\sqrt{\epsilon_1} + \sqrt{\epsilon_2})^2 + \left(\frac{\omega}{c}d\tilde{\epsilon}_{||}\right)^2} \tag{9}$$

has a maximum near $\omega = \omega_0 = \hbar^{-1}\mathscr{E}_{k=0}$, which increases as d^{-2} due to Eqs. (4) and (6). For oblique incidence when the electric vector lies in the incident plane a second maximum R appears near the frequency

$$\omega_l = \left[\omega_0^2 + 12\pi\Lambda_{\perp}\left(\frac{e^2}{\epsilon\hbar d}\right)^2\frac{m|V_{cv}|^2}{\hbar\omega_0}\right]^{1/2}. \tag{10}$$

If the film is sufficiently perfect and homogeneous in thickness, then a fast increase of $\chi_{\mathbf{k}\omega}$ with decreasing d will lead to the appearance of pronounced poles (maxima) and zeros in the quantities $\epsilon_{||}$ and $\epsilon + 6\pi\Lambda_{\perp}\chi$, which are necessary for manifestation of the polariton effects. Of course, we can discuss only the modes such as $E_{1,2}(\mathbf{r}) \sim \exp[i\mathbf{kr} - \kappa_{1,2}|z|]$, which are localized near the film,[5-8] because the film determines only their spectrum. The dispersion of these waves is characterized by the quantities Δ_E and Δ_M

$$\Delta_E = 4\pi\frac{e^2}{\hbar c}\,\frac{e^2}{\epsilon d}\,\frac{m|V_{cv}|^2}{\hbar\omega_0}, \qquad \Delta_M = \frac{4\pi}{\epsilon}\left(\frac{e^2}{\epsilon d\hbar\omega_0}\right)^2 m|V_{cv}|^2. \tag{11}$$

I. Transverse electric polaritons. $\mathbf{E}||[\mathbf{kn}]$. These exists in the frequency range $2\Lambda_{||}\hbar^{-1}\Delta_E/\sqrt{|\epsilon_1 - \epsilon_2|} \geqslant \omega_0 - \omega > 0$. The dispersion law is

$$\omega = \omega_0 - \frac{2\Lambda_{||}\Delta_E}{\hbar\sqrt{|\epsilon_1 - \epsilon_2|}}\left\{\frac{c^2k^2}{\omega_0^2} - \left[\left(\frac{c^2k^2}{\omega_0^2} - \epsilon_1\right)\left(\frac{c^2k^2}{\omega_0^2} - \epsilon_2\right)\right]^{1/2} - \frac{\epsilon_1 + \epsilon_2}{2}\right\}^{1/2}, \tag{12}$$

$$\kappa_{1,2} = \frac{\omega_0}{2c}\left(\frac{\Lambda_\parallel \Delta_E}{\hbar\omega_0 - \hbar\omega} \pm \frac{\epsilon_2 - \epsilon_1}{2}\right). \tag{13}$$

II. Transverse magnetic polaritons $\mathbf{H}\|[\mathbf{kn}]$. There are two branches of such waves near the frequencies ω_0 and ω_l in frequency intervals of width $\sim\hbar^{-1}\Delta_M$. In the simplest case, $\epsilon_2 = \epsilon_1$, their dispersion laws are

$$\hbar\omega = \hbar\omega_0 + \Lambda_\parallel \Delta_M \frac{d\left(k^2 - \epsilon_1\frac{\omega_0^2}{c^2}\right)^{1/2}}{2\frac{\epsilon_1}{\epsilon} + d\left(k^2 - \epsilon_1\frac{\omega_0^2}{c^2}\right)^{1/2}}, \tag{14}$$

$$\hbar\omega = \hbar\omega_l + \frac{\epsilon_1}{2}\Lambda_\perp\Delta_M \frac{k^2 d}{\left[k^2 - \epsilon_1\frac{\omega_l^2}{c^2}\right]^{1/2}}. \tag{15}$$

If $\Lambda_\perp = 0$, then the branch (15), as well as the reflection peak near ω_l, will be missing.

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