Surface polaritons and Brewster waves in the exciton luminescence spectra

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We show that surface polaritons (SP), after scattering on phonons and surface irregularities, produce two peaks in the exciton luminescence spectra in the region of the longitudinal-transverse splitting. We also noticed a presence of the "narrow-neck" effect for SP and the angular distribution of low-temperature luminescence associated with the Brewster waves.

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The intensity of exciton luminescence at frequency ω, which is proportional to the integral $I \approx \int dz C(z) e^{-k(\omega)z}$, where $C(z)$ is the exciton concentration at a depth z and $k(\omega)$ is the coefficient of light absorption, is determined by the number of excitons in the surface layer of thickness $L \sim 1/k(\omega)$. In strongly absorbing crystals $L \sim 0.1 - 1\ \mu$. The surface polaritons have been experimentally observed in the surface layer at frequencies in the range of the longitudinal-transverse exciton splitting.[1,2] In the analysis of exciton luminescence spectra these states were not taken into account. At the vacuum boundary the dispersion law of surface polaritons

$$k_{\parallel}^2 = \frac{\omega^2}{c^2} \frac{\epsilon(\omega)}{1 + \epsilon(\omega)} \tag{1}$$

for $\epsilon(\omega) = \epsilon_\infty + S\omega_\perp^2/(\omega_\perp^2 - \omega^2)$ is shown in Fig. 1; the broken line represents depen-

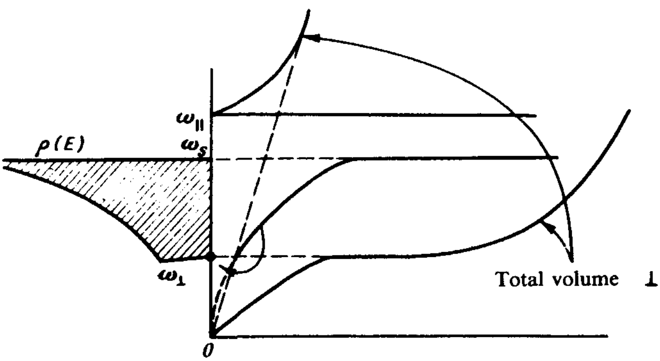

FIG. 1.

dence (1) corresponding to the Brewster waves.[3] The surface polaritons cannot be transformed into luminescence photons at the ideal plane boundary without taking into account the scattering processes. Allowance for the boundary irregularities and photon scattering, however, violates this constraint and leads to the appearance of the radiation width of the surface polaritons.

At relatively low temperatures, the excitons and surface polaritons with energy $E \geqslant \hbar\omega_\perp$ obey the Boltzmann distribution with $(E) = \rho(E)e^{-E/T}$. It can be seen in Eq. (1) that the density of states $\rho(E)$ for the surface polaritons per unit surface area

$$\rho(E) = \frac{\pi}{\hbar} \frac{(\omega_s^2 - \omega^2)^2 \epsilon_\infty (1 + \epsilon_\infty) + S\omega_s^2 \omega_\perp^2}{(\epsilon_\infty + 1)^2 (\omega_s^2 - \omega^2)^2}$$

has a sharp peak at the frequency $\omega = \omega_s$, $\epsilon(\omega_s) = -1$. Allowance for the spatial dispersion and finite lifetime of the surface polaritons smooths out the peak somewhat, without disturbing the overall picture of the $\rho(E)$ dependence. In the region $\omega = \omega_s$ the surface polaritons are practically elementary Coulomb excitations, since the contribution from the transverse field to their energy is small. On the other hand, the polaritons with frequencies $\omega \geqslant \omega_\perp$ are primarily photons, which interact weakly with the phonons but scatter strongly on the irregular surface. In this frequency range $|\epsilon| > 1$, the scattering on the irregular surface is accompanied primarily by a stripping process of the surface polariton, which becomes a vacuum photon.[4] Figure 2a shows schematically the density of states of the surface polaritons, Fig. 2b shows the probability of scattering by acoustical phonons, and Fig. 2c shows the probability of scat-

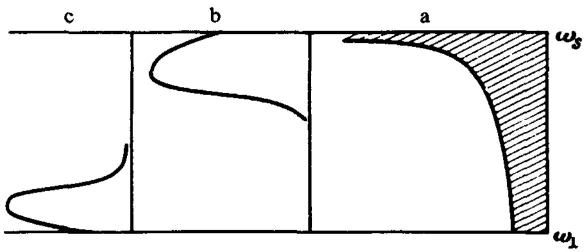

FIG. 2.

tering of the surface polaritons on the irregular surface.[1] Since practically all the states of the surface polaritons are populated at temperatures $T \geqslant \hbar|\omega_s - \omega_\perp|$, we can expect two peaks to appear in their luminescence spectrum in the region of longitudinal-transverse splitting; moreover, the intensity of the high-frequency peak should decrease with decreasing temperature. The interaction between the short-wave surface polaritons and the optical phonons of frequency Ω should produce vibrational repetitions in the luminescence spectra at frequencies $\omega_s - \Omega, \omega_s - 2\Omega, \ldots$, and at $\Omega > |\omega_s - \omega_\perp|$ these repetitions should be below the frequency $\omega_\perp$. As the frequency of the surface polariton ω approaches the frequency $\omega_\perp$, its interaction with the phonons and penetration depth of the field into the crystal decrease in the same way as for the bulk polaritons,[5] the "narrow-neck" effect. This unique situation differs from that discussed in Ref. 5 in that the surface polariton, after losing energy by going to a region below $\omega_\perp$ (this transition is indicated by an arrow in Fig. 1), becomes a Brewster wave which is no longer localized at the surface and which corresponds to a luminescence photon that escapes at the Brewster angle $\theta(\omega)$, $\tan\theta = [\epsilon(\omega)]^{1/2}$. The contribution of the Brewster waves to the exciton luminescence spectra can be determined experimentally from the angular dependence of the radiation intensity at fixed

ω. The Brewster waves can also be produced by scattering the bulk polaritons on phonons. Nonetheless, the contribution from the decay process of surface polaritons can be determined by studying the kinetics of luminescence at the frequencies of the Brewster waves. Since at frequencies $\omega \geqslant \omega_\perp$ the surface polaritons on partially irregular surfaces have relatively long lifetimes, the luminescence damping due to the generation of Brewster waves should also have a long lifetime. This time should correlate with the intensity of the lower peak. Since at present the spectra of polariton luminescence of crystals are widely investigated, the above discussion can be useful in explaining the obtained results.

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[1)] Detailed calculations will be published later. Figure 2b corresponds to the case $\omega_s - \omega_\perp > \omega_{\rm Debye}$, where $\omega_{\rm Debye}$ is the Debye frequency.

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