Manifestation of near-surface localized excitons in spectra of diffuse reflection of light

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The interaction of excitons with rough surfaces and its effect on light scattering spectra were investigated theoretically. We have employed a model for the excitonic surface potential based upon the generalized Morse potential, taking into account its random fluctuations, produced by the surface roughness. Applying first-order perturbation theory, we calculate the cross section of light scattering from a rough GaAs surface and analyze its frequency dependence in the presence of an extrinsic surface-potential well with excitonic bound states.

At present there is a great interest in the study of optical properties of spatially dispersive media such as excitonic crystals [1]. In these materials the interaction of excitons with the surface play an important role in determining the lineshape of their optical spectra (reflectivity, transmissivity, etc.). In the case of high-quality crystals the exciton-surface interaction is well described by a repulsive surface potential [1]. However, surface treatments (heating, intense illumination, electron and ion bombardment, doping, and electric-field application) modify the concentration of impurity ions in the transition layer and, consequently, the form of the surface potential. The extrinsic contribution to the surface potential may be attractive, unlike intrinsic potentials. The generation of excitonic bound state within an extrinsic potential well produces resonances (peaks and dips) in the optical spectra. Their resonance structure is very sensitive to the polarization of the incident light [2,3]. The resonances associated to transverse polaritonic modes appear in both *s*- and *p*-polarization geometries, whereas the longitudinal resonances, due to quantized polarization waves [3], are present only in the latter geometry.

In this paper we present theoretical results for spectra of light scattering from random rough surfaces of GaAs crystals, having near-surface localized excitons. We will show that there spectra give useful information about the behavior of excitons near treated surfaces.

We solved analytically the system of equations that describes the exciton-polariton fields inside the semiconductor. It is composed of the Maxwell equations coupled to the equation for the excitonic polarization [1]. In the calculation we considered a surface potential $U(\mathbf{r})$ given by

$$U(\mathbf{r}) = U_1 e^{-z/a} + U_2 e^{-2z/a} + \zeta(\mathbf{r}_\parallel)\big(U_1 e^{-z/a} + 2U_2 e^{-2z/a}\big)/a. \qquad (1)$$

This model holds when the surface is sufficiently smooth and its roughness is very small such that $\delta \ll a \ll L$ (δ is the mean-square deviation of the surface from its average plane $z = 0$, a is the characteristic size of the transition layer, L is the correlation length of the surface profile function $\zeta(\mathbf{r}_\parallel)$, which is assumed to be a stationary, Gaussian, stochastic process). Under these conditions, the exciton interacts, locally, with a flat surface and, therefore, the potential fluctuations are strongly correlated with the surface profile. The first two terms on the right-hand side of Eq. (1) represent the surface potential for an ideal flat surface, which is modeled here by the generalized Morze potential [3]. The terms proportional to $\zeta(\mathbf{r}_\parallel)$ in (1) describe the small (in $\delta/a \ll 1$) fluctuations of the surface potential.

The amplitudes of the electromagnetic fields both inside the semiconductor $(z > \zeta(\mathbf{r}_\parallel))$ and in the vacuum $(z < \zeta(\mathbf{r}_\parallel))$ are found by employing usual Maxwell boundary conditions together with the condition that the excitonic polarization vanishes at $z = \zeta(\mathbf{r}_\parallel)$. Applying first-order perturbation theory, we calculate the dimensionless cross section $d\sigma/d\Omega$, defined as the ratio of the intensity of light scattered into direction (θ, ϕ) (θ is the angle of scattering, measured from the normal of the average surface, and ϕ is the azimuthal angle) to the intensity of the incident light.

The light scattering spectra in Figs. 1 and 2 were calculated for GaAs near exciton resonance frequency $\hbar\omega_T = 1.515$ eV. The rough-surface parameters, used here, are $\delta = 15$ Å, $L = 5000$ Å. Figs. 1 *a* and 2 *a* correspond to the case of a high-quality GaAs crystal with an intrinsic (exponential) potential ($U_1 = 0.5$ meV, $U_2 = 0$, $a = 130$ Å). The curves (*2*) in Figs. 1 and 2 were obtained for a surface potential well ($U_1 = -2.718$ meV, $U_2 = 3.694$ meV, $a = 550$ Å), having three excitonic bound states at the frequencies $\hbar\omega_{T1} = 1.51465$ eV, $\hbar\omega_{T2} = 1.51485$ eV, $\hbar\omega_{T3} = 1.51497$ eV. The near-surface localized excitons produce transverse resonances (broad peaks) in the frequency dependence of $d\sigma/d\Omega$ close to their eigenvalues ω_{Tn} ($n = 1, 2, 3$). These peaks coalesce and form a huge maximum below the exciton resonance frequency ω_T in the spectrum $d\sigma(\omega)/d\Omega$ (see Figs. 1 *b* and 2 *b*). In the case of *p*-polarized incident light, besides transverse modes, quantized polarization waves [3] are excited at frequencies $\omega_{Ln} = \omega_{Tn} + \omega_{LT}$ (ω_{LT} is the frequency value of the longitudinal-transverse splitting). These longitudinal modes give rise to new resonances in the spectrum $d\sigma(\omega)/d\Omega$ that interfere with the transverse ones. As in seen Fig. 2 *b*, the longitudinal resonances manifest themselves as dips in the

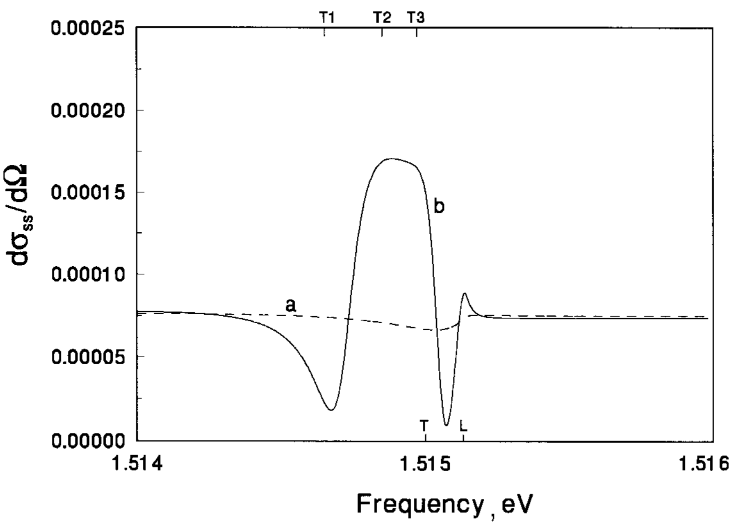

Figure 1. Frequency dependence of the dimensionless cross section of light scattering from a rough GaAs surface. The incident light is s-polarized with an angle of incidence $\theta_0 = 40^\circ$, and the scattered light is s-polarized in the plane of incidence ($\phi = 0$) with $\theta = 10^\circ$. Curves (a) and (b) were calculated for an exponential potential and an extrinsic surface potential well, respectively.

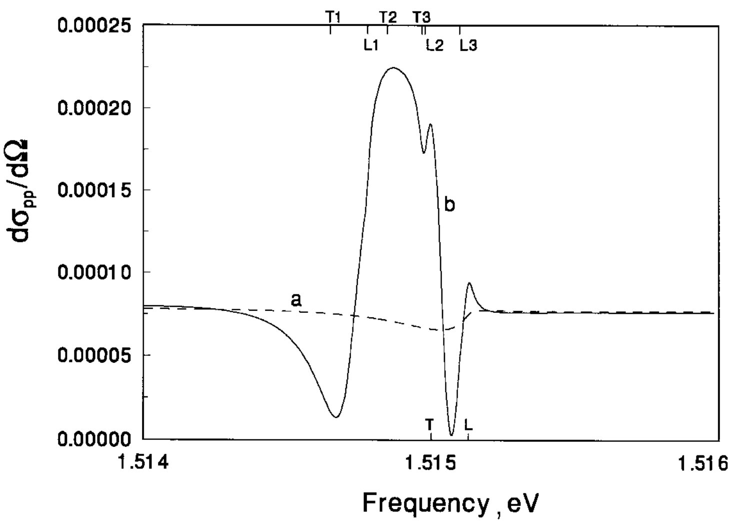

Figure 2. The same as in Fig. 1, but both the incident light and the scattered light are p-polalized.

spectrum of light scattering for GaAs. It is noteworthy that this manifestation is similar to that observed in reflectivity spectra [3]. This fact can be explained as a consequence of the assumed strong correlation between the potential fluctuations and the surface roughness.

In conclusion, the spectra of diffuse reflection can be useful to characterize the state of the surface transition layer and defect near-surface localized excitons.

The present work was partially supported by the Consejo Nacional de Ciencia y Tecnología.

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