Spike in Spectra of Light Reflection by Crystals as a Criterion of Spatial Dispersion Appearance in Exciton Optical Properties

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Abstract

We report the results of modelling for the effects of spatial dispersion on the regularities of light reflection spectra in the region of polariton resonant excitation in crystal bulk with the plane parallel dispersion-free layer fastened on the crystal surface.

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Kew words: spatial dispersion, exciton, light reflection.

Introduction

After publication of a fundamental paper [1] on the theory of spatial dispersion (SD) in exciton region of spectrum, a considerable progress of further investigations has been conditioned by experimental observations of a "spike" (a narrow peak of the reflection coefficient) in the resonant reflection spectrum located in the region of longitudinal frequency ω_L and by explanation of its position in the SD exciton spectra [2][*)]. Out of any doubt, the mentioned paper has become a powerful impulse to understanding the core of the problem. However, the authors have not paid attention to some peculiarities of the exciton light reflection spectra formed in the presence of a plane parallel Fabry-Perot layer at the crystal surface[**)]. The urgency of such investigations is confirmed by the fact that studies of the light exciton effects and SD still remain topical [4-6]. Determination of the "spike" role as a criterion of SD manifestation in the spectra of resonant reflection represents the goal of the present paper. The calculations have been carried out for the "vacuum–dispersion-free layer–semi-infinite crystal" model.

[*)] The point of view [2] prevails also in all the other papers reviewed in the monograph [3].

Results and Discussion

It is known [2] that the amplitude of the resulting reflection coefficient for the case of multiple reflections of light wave in a plane parallel layer (see Figure 1) is equal to

$$\tilde{r} = \frac{r_{12} + \tilde{r}_{23} \exp\left(-i\,\tilde{\delta}\right)}{1 + r_{12}\,\tilde{r}_{23} \exp\left(-i\,\tilde{\delta}\right)}. \qquad (1)$$

Here r_{12} is the absolute Fresnel reflectance for the vacuum–dispersion-free layer interface (the index 12) with the refraction index n_0 and thickness d (the wave having the phase shift $\delta = \frac{4\pi\, n_0 d}{\lambda}$), and $\tilde{r}_{23}$ the absolute Fresnel reflectance for the interface of layer–semi-infinite crystal with the resonant type of absorption, which is modelled by one-oscillator function [2]

$$\tilde{\varepsilon}(\omega,\vec{k})=\varepsilon_0+\frac{4\pi\lambda\,\omega_0^2}{\omega_0^2-\omega^2+\dfrac{\hbar^2k^2}{2M^*}-i\omega\tau}\,. \qquad (2)$$

In Eq. (2), ε_0 denotes the background permittivity, $4\pi\lambda$ the oscillator strength of the resonant transition in the state with the frequency ω_0, τ the damping factor, and $\vec{k}$ the wave vector of the exciton with the effective mass M^*.

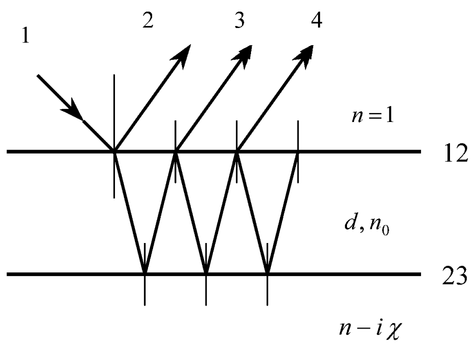

Fig. 1. Schematic diagram of multibeam Fabry-Perot interference.

According to Eq. (1), the energetic reflectance may be conveniently written with the envelope function of the interference minima as [7]

$$R=\frac{R_{\min}+b^2\sin^2\dfrac{\Delta}{2}}{1+b^2\sin^2\dfrac{\Delta}{2}}\,, \qquad (3)$$

where $\Delta=\phi_{23}-\delta$ and $b^2=\dfrac{4\sigma_{12}\sigma_{23}}{(1-\sigma_{12}\sigma_{23})^2}$, $R_{\min}=\left(\dfrac{\sigma_{12}-\sigma_{23}}{1-\sigma_{12}\sigma_{23}}\right)^2$ is the value of reflectance in the minimum of Fabry-Perot interference band, $\sigma_{12}=\dfrac{1-n_0}{1+n_0}$ the absolute value of the r_{12} amplitude, and $\sigma_{23}=\sqrt{\mathrm{Re}^2\,\tilde{r}_{23}+\mathrm{Im}^2\,\tilde{r}_{23}}$ the module of the $\tilde{r}_{23}=\sigma_{23}\exp(i\,\phi_{23})$ coefficient amplitude.

It follows from Eq. (3) that an additional minimum appears at the frequency ω_m of phase compensation (see [8,9]), defined by the relation

$$\Delta=\phi_{23}-\delta=m\pi\,,\ m=0,1,2,...\,, \qquad (4)$$

in the contour of reflection with the amplitude $R_{\min}$ that tends to zero when the condition

$$\sigma_{12}=\sigma_{23} \qquad (5)$$

is satisfied. Irrespective of the character of frequency dispersion of the permittivity function, the frequency ω_m oscillates with changing layer phase thickness δ inside the region of longitudinal-transverse splitting ω_{L0} of the resonant state.

Let us now study the peculiarities of the minimum formed at the frequency ω_m and, hence, the formation of the additional maximum ("spike") with taking the SD effect into consideration. For this aim, we divide the function (2) into its real and imaginary parts:

$$\mathrm{Im}\,\tilde{\varepsilon}(\omega,\vec{k})=\frac{4\pi\lambda\left(\dfrac{\varepsilon_0}{G}-A_1-2\rho\sin\dfrac{\varphi}{2}\right)}{\left[\dfrac{\varepsilon_0}{G}-A_1-2\rho\sin\dfrac{\varphi}{2}\right]^2+\left[B_1+2\rho\cos\dfrac{\varphi}{2}\right]};$$

$$\mathrm{Re}\,\tilde{\varepsilon}(\omega,\vec{k})=\frac{4\pi\lambda\left(B_1+2\rho\cos\dfrac{\varphi}{2}\right)}{\left[\dfrac{\varepsilon_0}{G}-A_1-2\rho\sin\dfrac{\varphi}{2}\right]^2+\left[B_1+2\rho\cos\dfrac{\varphi}{2}\right]} \qquad (6)$$

and determine the refraction and absorption indices by solving the equation

$$\left[\mathrm{Re}\,\tilde{\varepsilon}(\omega,\vec{k})+i\,\mathrm{Im}\,\tilde{\varepsilon}(\omega,\vec{k})\right]=\left[n(\omega)-i\,\chi(\omega)\right]^2, \qquad (7)$$

where $A_1=1-(\omega/\omega_0)^2$, $B_1=\dfrac{\tau\omega}{\omega_0^2}$,

$A_2=\dfrac{A_1\varepsilon_0+4\pi\lambda}{G}$, $B_2=-\dfrac{B_1\varepsilon_0}{G}$, $G=\dfrac{M^*\omega_0c_0^2}{\omega^2}$,

$\rho^4 = A_2^2 + B_2^2$, and $\tan\varphi = -\dfrac{B_1}{A_1 + \dfrac{4\pi\lambda}{\varepsilon_0}}$.

As follows from Figure 2, the condition (5) of module equality in the ω_{L0} region is satisfied twice, quite independent of the δ value. The short-wave minimum of $R_{\min}$ contour with zero amplitude $R_{\min} = 0$ is formed at the resonant frequency ω_L, while the condition $n \cong n_0$ is satisfied at the ω_L in the approximation $\chi \to 0$. Therefore, the "spike" of reflection is localized in the region of frequency ω_L. The minimum of the reflection contour $R(\omega)$ appears at the frequency ω_m and has the amplitude $R_{\min}$.

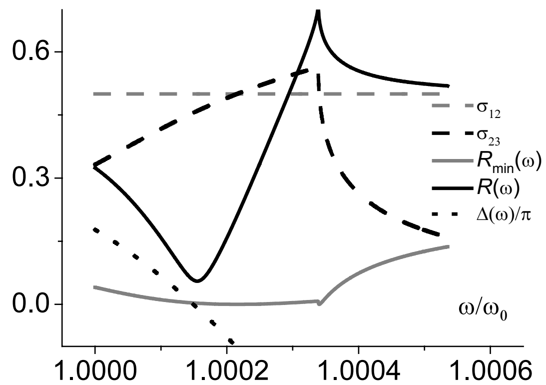

Fig. 2. Frequency curves for the modules $\sigma_{12,23}$ and the energetic reflectance $R_{\min}$ in the region of longitudinal-transverse splitting of resonant excitation. The initial parameters for calculations are $M^* = 0.87m_0$, $d = 18$nm and $\tau = 2\cdot 10^{-7}\ eV$.

It is seen from Figure 3 that the amplitude of the principal maximum of reflection in the region of resonant frequency ω_0. This has been observed in [2]. However, the amplitude of the "spike" also increases with increasing exciton effective mass M^*, the fact that has not been reported yet. Consequently, the appearance of the additional maximum of reflection is conditioned by the additional minimum at the frequency ω_m caused by the phase compensation. Its intensity is determined by the

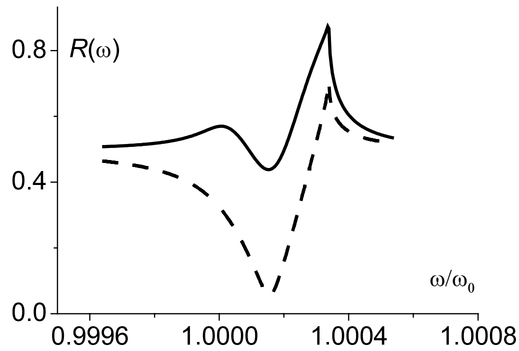

Fig. 3. Frequency curves for the energetic reflectance $R_{\min}$ determined for the two values of the exciton effective mass: $M^* = 0.87m_0$ (dash curve) and $M^* = 8.7m_0$ (solid curve).

difference between ω_m and the frequency, for which the condition of equality of Fresnel coefficient modules for the opposite boundaries of plane parallel layers at the crystal surface occurs.

It is worthwhile that the observation of the "spike" in the exciton reflection spectra enables one to evaluate the phase thickness of the layer.

According to the condition (4), at the frequency ω_m we have $\tan\phi_{23} = \tan\delta$, where $\tan\phi_{23} = \dfrac{\operatorname{Im}\tilde{r}_{23}}{\operatorname{Re}\tilde{r}_{23}}$ from the above definitions. Therefore the tangent of the phase change at the interface 23 for the reflection model "vacuum–dispersion-free layer–semi-infinite crystal" is equal to

$$\tan\phi_{23} = \frac{2\chi n_0}{\varepsilon_0 - n^2 - \chi^2}. \quad (8)$$

Since the SD effects are significant for small levels of damping, solving Eq. (8) and taking Eqs. (2) and (7) into account in the limit of $\gamma = 0$ gives

$$\omega_m\big|_{\gamma=0} = \frac{\omega_m\big|_{M^*\to\infty}}{\sqrt{\left(1 + \dfrac{\hbar\omega_0\varepsilon_0}{M^* c_0^2}\tan^2\dfrac{\delta}{2}\right)}}, \quad (9)$$

where $\omega_m\big|_{M^*\to\infty}$ determines the frequency of the phase compensation without the SD consideration. According to [9], we have

$$\omega_m\Big|_{M^*\to\infty} = \omega_0\sqrt{1+\frac{\gamma^2}{2\omega_0^2 tg\delta}+\frac{2\pi\alpha}{\varepsilon_0}-\sqrt{\left(\frac{2\pi\alpha}{\varepsilon_0}+\frac{\gamma^2}{2\omega_0^2 tg\delta}\right)^2+\frac{\gamma^2}{\omega_0^2 tg^2\delta}-\frac{(4\pi\alpha)^2\sin^2\delta}{4\varepsilon_0^2}}}\ , \qquad (10)$$

or, neglecting the dependence on γ,

$$\omega_m \approx \omega_0\sqrt{1+\frac{4\pi\alpha}{2\varepsilon_0}\sin^2\delta}\ .$$

In that way, when the “spike” width is defined as $\Delta\omega \approx \omega_L - \omega_m$, it is approximately equal to

$$\Delta\omega \approx \omega_{L0}\sin^2\frac{\delta}{2}\ . \qquad (11)$$

Conclusions

The non-dispersion layer thickness may be evaluated with measuring experimentally the “spike” width. As a result, the fact of the “spike” existence only is not sufficient for revealing unambiguously that the SD alone is responsible for the peculiarities of spectral contour of exciton reflection.

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