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Abstract. On the basis of the Kramers–Kronig relations and the dispersion equation the connection between the real and imaginary parts of the complex refractive indices for eigenwaves in weak spatial dispersion media is proposed. The theoretical description was applied to an experimental analysis of the amplitude–phase measurements of transmission spectra of semiconductor crystals near the exciton resonances. Two possibilities have been considered. The first is the detection of optical properties of normal waves diverted in space or in time. The second possibility refers to the analysis of the normal wave interference for plane-parallel crystals. It was shown that taking into account the interference of supplementary waves enables us to formulate the general concept and to derive the additional Kramers–Kronig dispersion relations and the dependence of the integral exciton absorption coefficient of the damping constant resulting from the influence of the spatial dispersion.

1. Introduction

It is known (Agranovich and Ginzburg 1979, Ginzburg and Meiman 1964) that the Kramers–Kronig dispersion relations (DRs) for the dielectric susceptibility tensor components $\varepsilon(\omega, k)$ (ω is the electromagnetic wave frequency and k is the wavevector) are valid in the absence of the spatial dispersion ($k = 0$). Nevertheless, since $\varepsilon(\omega, k)$ is an analytical function of the complex wavevector components, the same DRs are applicable for small k-values as for $k = 0$ (see, e.g., Kirzhnits 1976).

The question of the validity of these relations for the complex refractive indices is much more complicated. The breakdown of the DRs for 'normal' waves (eigenwaves) in the exciton spectrum region of the molecular and semiconductor crystals was pointed out by Brodin *et al* (1959) (see also Brodin and Strashnikova 1962) and was examined in detail by Ginzburg and Meiman (1964) and Davydov (1962). As was shown recently by Moskovskii and Solov'ev (1984), some progress has been achieved in the application of the DRs to the exciton reflection and absorption spectra of some semiconductor crystals.

2. The Kramers–Kronig dispersion relations in media with spatial dispersion

Under rather general assumptions about the properties of the equilibrium dielectric medium the DRs for the dielectric susceptibility tensor components $\varepsilon_{ij}(\omega, k) \equiv \varepsilon'_{ij}(\omega, k) + i\varepsilon''_{ij}(\omega, k)$ may be written as

$$\varepsilon'_{ij}(\omega, k) - \delta_{ij} = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega' \, \frac{\varepsilon''_{ij}(\omega', k)}{\omega' - \omega}$$

$$\varepsilon''_{ij}(\omega, k) = -\frac{1}{\pi} \int_{-\infty}^{\infty} d\omega' \, \frac{\varepsilon'_{ij}(\omega', k) - \delta_{ij}}{\omega' - \omega}.$$

(1)

From the experimental point of view it would be more interesting to derive similar relations containing real and imaginary parts of the complex refractive indices for normal waves such as $\bar{n}_l = n_l(\omega) + i\kappa_l(\omega)$. One may obtain the $\bar{n}_l^2$ values from the solution of the dispersion equation (see, e.g., Agranovich and Ginzburg 1979):

$$\varepsilon_{ij}s_is_j\bar{n}^4 - [(\varepsilon_{ij}s_is_j)\varepsilon_{ll} - \varepsilon_{il}\varepsilon_{lj}s_is_j]\bar{n}^2 - |\varepsilon_{ij}| = 0$$

(2)

where $s = k/|k|$.

If the spatial dispersion condition is valid, the roots of equation (2) will have singularities such as the branching points in the upper half-plane of $\bar{\omega}$. Because of this, relations similar to (1) cannot be obtained (Ginzburg and Meiman 1964).

Also, within the restrictions for the weak spatial dispersion case, i.e. taking into account only the low-order terms in the $\varepsilon_{ij}(k)$ expansion

$$\varepsilon_{ij}(\omega, k) = \varepsilon^0_{ij}(\omega) + i\gamma_{ijl}(\omega)k_l + \alpha_{ijlm}(\omega)k_lk_m + \ldots$$

(3)

one may use (1) to obtain certain relations between the set of n_l and κ_l values.

First it should be noted that the DRs of type (1) are valid for all ε^0_{ij}, γ_{ijl}, α_{ijlm} tensor values. This must be taken into account for phenomenological determination of these parameters. Substituting (3) into (2), one may obtain the algebraic equation for $\bar{n}_l^2$. Its order is determined by the number of the terms in the representation (3). The coefficients of this equation are expressed by the series with positive terms for ε^0_{ij}, γ_{ijl}, α_{ijlm} or their products and are consequently analytical functions of ω, having no singularities at Im $\bar{\omega} \geq 0$. Because of this, a relation such as equation (1) may be written for every coefficient of equation (2).

If equation (2) is of the order N with respect to $\bar{n}^2$, N coefficients of it (as they are the complex functions of ω) will be determined by the real parts of $\bar{n}^2$. In this case, only N functions from $2N$ real functions n_l, κ_l will be independent. One may determine the others (as well as the coefficients in (3)) using the known relations between the roots of the algebraic equation and its coefficients.

The problem of the determination of the possible $\bar{n}_l^2$ values from the real and imaginary parts of the tensors ε^0, γ, α seems relatively simple and may be transformed into the problem of construction of the unknown parts of these functions using the Kramers–Kronig relations and equation (2). However, it is difficult to find out the coefficients in (3) from N given values of $n_l(\omega)$ and $\kappa_l(\omega)$ or their independent combinations.

For the simple case of an isotropic ($\varepsilon_{ij}^0 = \varepsilon^0 \delta_{ij}$) and non-gyrotropic ($\gamma_{ijl} = 0$) medium, the equation for the normal refractive indices $\bar{n}_l^2 \equiv X_l = X_l' + \mathrm{i} X_l''$ will take the form

$$X^2 - A(\varepsilon^0, \alpha)X + B(\varepsilon^0, \alpha) = 0 \tag{4}$$

where A and B are the known analytical functions of ε and α (Agranovich and Ginzburg 1979); the frequency dependence of ε and α are not determined. Also, the functions $X_1'(\omega)$ and $X_2'(\omega)$ are expressed as

$$
\begin{aligned}
X_1 + X_2 &= (X_1' + X_2') + \mathrm{i}(X_1'' + X_2'') = A \\
X_1 X_2 &= (X_1' X_2' - X_1'' X_2'') + \mathrm{i}(X_1' X_2'' + X_1'' X_2') = B.
\end{aligned}
\tag{5}
$$

Using the first equation of the Kramers–Kronig relations, one may find $X_1'' + X_2''$ and A.

The application of the Kramers–Kronig relations to the second equation leads to a non-linear integral equation of second order with a Coshy-type kernel. Its solution enables us to determine approximately the properties of the medium (i.e. ε^0 and α).

The increase in the order of equation (2) causes an increase in the degree of non-linearity of the equation and only an excess of data, as a rule, facilitates this problem. In particular, if in the above-mentioned case three functions X_1', X_2' and X_1'' are primarily determined, the construction of the X_2'' function may be carried out directly by the use of the Kramers–Kronig relations.

The same approach is valid for the analysis of the properties of medium, if equations (1) are written in terms of ε_{ij}^{-1}.

Thus, we proposed a new method for solution of the problem connected with the study of properties of media with spatial dispersion ($n_l(\omega)$ and $\kappa_l(\omega)$). This approach is supported by calculations performed for the isotropic non-gyrotropic medium model in the vicinity of the isolated absorption line (Agranovich and Ginzburg 1979). These calculations showed satisfactory stability of the solution with respect to the experimental errors of the extrapolated values of $n_l(\omega)$ and $\kappa_l(\omega)$ obtained in the region far from the absorption line.

The above-mentioned considerations may be applied to the experimental results to determine the refractive indices and absorption coefficients of two normal waves diverted in space after light transmission through wedge-shaped crystals of CdS (Lebedev *et al* 1984) and the use of the laser pulse technique (Segawa *et al* 1978).

3. Additional Kramers–Kronig relations taking into account interference of supplementary waves

Other experimental possibilities appear when spatial and temporal diversion of supplementary waves are of no interest. This situation arises in the study of thin plane-parallel crystal transmission spectra near the exciton resonances.

Let us analyse the problem for hexagonal crystals of the CdS type with the optical axis lying in the plane of the plate. Expressions for the refractive indices for p-polarised waves for mixed excitons are as follows (Permagorov *et al* 1973):

$$\bar{n}_l^2 = \varepsilon_\parallel^0 + \sin^2 \varphi \, (1 - \varepsilon_\parallel^0/\varepsilon_\perp^0) + A \pm [A^2 + (\varepsilon_\parallel^0 \omega_{\mathrm{LT}} \sin^2 \varphi)/\varepsilon_\perp^0 \beta_\perp]^{1/2} \tag{6}$$

where $l = 1$ and $l = 2$ correspond, respectively, to $+$ and $-$ before the square root in (6);

$$
\begin{aligned}
A &= \{\omega - \omega_{\mathrm{L}} + \mathrm{i}\gamma - \beta_\perp [\varepsilon_\parallel^0 + \sin^2 \varphi \, (m_\perp^*/m_\parallel^* - \varepsilon_\parallel^0/\varepsilon_\perp^0)]\}(2\beta_\perp)^{-1} \\
\beta_\perp &= \hbar\omega^2/2m_\perp^* c^2.
\end{aligned}
\tag{7}
$$

$\varepsilon_\parallel^0, \varepsilon_\perp^0$ and $m_\parallel^*, m_\perp^*$ are the background dielectric susceptibilities and the effective masses of exciton for the light vector $k \parallel C_6$ and $k \perp C_6$; φ is the incident angle; ω_{LT} is the longitudinal–transverse splitting; ω_L is the longitudinal exciton frequency; c is the velocity of light.

For the isotropic case ($E \perp C_6; k \perp C_6$) the refractive indices will be expressed as

$$\tilde{n}_i^2 = \varepsilon_0 + B \pm (B^2 + \varepsilon_0 \omega_{LT}/\beta)^{1/2} \tag{8}$$

where

$$B = (\omega - \omega_T + i\gamma - \beta \varepsilon_0)(2\beta)^{-1}. \tag{9}$$

ω_T is the transverse exciton frequency; $\varepsilon_0 = n_0^2$ is the background dielectric susceptibility; n_0 is the background refraction index; γ is the exciton damping constant. Later for the transverse excitons the definitions $\beta \equiv \beta_\perp$ and $\varepsilon_0 \equiv \varepsilon_\perp^0$ will be used.

For the *total* wave which is the result of the light–exciton wave interference one has for the isotropic case

$$E(z) = E_1(z) + E_2(z) = E_{01}\{\exp[i(\omega/c)\tilde{n}_1 z] + q \exp[i(\omega/c)\tilde{n}_2 z]\} \tag{10}$$

where z is the thickness of the crystal; E_1, E_2 are the normal waves with the refractive indices $\tilde{n}_1$ and $\tilde{n}_2$, respectively; $\tilde{n}_{1,2} = n_{1,2} + i\kappa_{1,2}$; $E_{01} = E_1|_{z=0}$ is the wave amplitude E_1 on the crystal surface;

$$q(\omega) = |q(\omega)| \exp[i\phi(\omega)] = [E_2(\omega)/E_1(\omega)]|_{z=0}.$$

The $q(\omega)$-value represents the ratio of the complex amplitude of the $E_1(\omega)$ wave to that of the $E_2(\omega)$ wave on the first crystal surface and is determined by the supplementary boundary conditions; $\phi(\omega)$ is the phase of $q(\omega)$.

One may write the effective refractive index n^* as

$$E(z) = E_{01}(1 + q) \exp[i(\omega/c)\tilde{n}^* z] \qquad \tilde{n}^* = n^* + i\kappa^*. \tag{11}$$

The transmission function will take the form (Moskovskii and Solov'ev 1984)

$$\theta(\tilde{\omega}) = \exp[i(\omega_T/c)(\tilde{n}^* - n_0)z]. \tag{12}$$

In this case the effective refractive index depends on z. As was shown by Moskovskii and Solov'ev (1984), the transmission function as well as its logarithm are the analytical functions in the upper part of the complex plane $I_+(\tilde{\omega})$ except the zero points. In the classical case, as Nussenzweig (1972) pointed out, the transmission function cannot be zero because the refractive index is not z dependent.

So the additional DRs may be written as (Moskovskii and Solov'ev 1984)

$$n^*(\omega) - n_0 = \frac{2}{\pi} \int_0^\infty \frac{x\kappa^*(x)}{x^2 - \omega^2}\,\mathrm{d}x + \frac{2c}{\omega z}\sum_j \tan^{-1}\left(\frac{\gamma_{0j} - \gamma}{\omega_{0j} - \omega}\right) \tag{13}$$

where $\tilde{\omega}_{0j} = \omega_{0j} + i\gamma_{0j}$ corresponds to the zero values of the transmission function in $I_+(\tilde{\omega})$.

For practical use of the additional DRs (i.e. for the determination of the transmitted light phase using the absorption spectrum) the coordinates $\tilde{\omega}_{0j}$ must be calculated.

Neglecting the reflection from the second plane of the crystal, one may represent $\theta(\bar{\omega}) = 0$ by a system of two equations:

$$(\omega_T z/c)[n_1(\bar{\omega}) - n_2(\bar{\omega})] = \phi(\bar{\omega}) + (2j - 1)\pi$$
$$(\omega_T z/c)[\kappa_1(\bar{\omega}) - \kappa_2(\bar{\omega})] = -\ln|q(\bar{\omega})|$$

(14)

where $j = 1, 2, 3, \ldots, N$.

The number of the roots of the N-equation system and its values depend on the crystal thickness.

Taking into account the interference of many light beams does not lead to significant complication of the calculations. The substitution of the amplitude and phase of the functions $q^{(1)}(\bar{\omega})$ instead of $|q|$ and $\phi(\bar{\omega})$ gives

$$q^{(1)}(\bar{\omega}) = |q(\bar{\omega})|\bar{n}_2\{\exp[2i(\omega_T/c)\bar{n}_1 z] - 1\}/\bar{n}_1\{\exp[2i(\omega_T/c)\bar{n}_2 z] - 1\}.$$

(15)

The set of solutions for two-beam approximation may be taken as the zero approximation. The problem for the mixed exciton mode is solved in the same way using (6).

It should be emphasised that violation of the classical DRs arises not only because of the spatial dispersion. Therefore the analysis of experimental data, as we believe, may be performed using the dependence of the integral exciton absorption coefficient (IEAC) on the damping constant in a study of the amplitude–phase spectra. The latter problem has been solved in a number of theoretical and experimental studies (see, e.g., Nkoma 1980, Akhmediev 1980, Akhmediev *et al* 1983).

4. Integral exciton absorption coefficient

In the classical case for the absorption lines in gases, liquids and solid bodies the integral absorption coefficient $S(\gamma)$ is determined as the square under the spectral absorption coefficient curve and does not depend on γ. Nevertheless, experiment shows (Crescenzi *et al* 1979, Voight 1974) a considerable decrease in IEAC in the low-temperature region of the crystal which may be caused by the influence of spatial dispersion. The dependence of $S(\omega)$ is a possible reason for this effect (Agranovich and Ginzburg 1979). Because of this the observation of the IEAC dependence of γ cannot serve as proof of the spatial dispersion effect.

We believe that correct data on the IEAC dependence of γ and the DRs would clarify the situation. Experiments of this type have been carried out recently only for quadrupole exciton line in CuO_2 crystals (Moskovskii and Solov'ev 1984). The connection between these phenomena becomes evident when we consider that, from the critical damping constant $\gamma_{cr} = 2(\varepsilon_0\omega_{LT}\beta)^{1/2}$ (for the transverse exciton) the IEAC becomes independent of γ. Here γ_{cr} corresponds to the exciton damping constant when the term in parentheses in (8) is equal to zero. Then the dispersive branches $\bar{n}_1(\omega)$ and $\bar{n}_2(\omega)$ cross at the point $\bar{\omega}_{cr} = \omega_{cr} + i\gamma_{cr}$ and the supplementary wave amplitude becomes very small. The IEAC attains the S_{cr}-value and for $\gamma > \gamma_{cr}$ it does not change. For the same γ_{cr} the classical Kramers–Kronig relations are valid. The explanation of this is that for $\gamma > \gamma_{cr}$ the role of the supplementary wave interference is neglected.
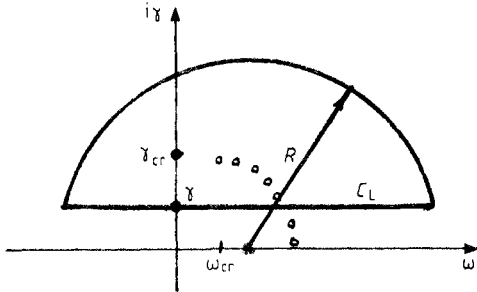
Figure 1. Integration contour and the positions (○) for the zeros $\tilde{\omega}_{0j}$.
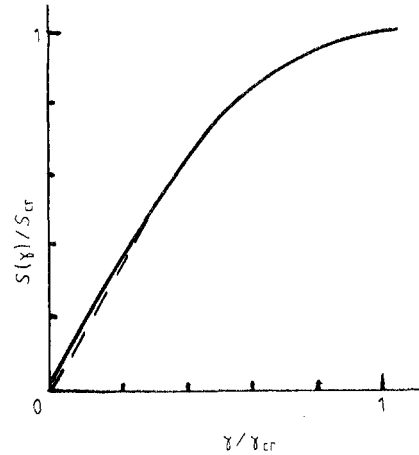

Figure 2. The IEAC for CdS crystals with $z = 0.3$ mm: ——, calculations from (22) for the transverse excitons ($E \perp C_6; k \perp C_6$); ---, calculations by Akhmediev (1980) using the parameters $\omega_T = 2.5527$ eV, $\omega_{LT} = 0.001\,86$ eV, $\varepsilon_\perp^0 = 8.3$ and $m_\pm^* = 0.9m_0$ (where m_0 is the electron mass).

The IEAC is expressed as (Akhmediev 1980)

$$S(\gamma, z) = -\frac{2}{z} \mathrm{Re}\left(\int_{-\infty}^{\infty} \ln \theta(\tilde{\omega}, z)\, d\omega\right) \tag{16}$$

where the absorption coefficient has the form $K(\omega) = (2\omega_T/c)\kappa(\omega)$. Thus we have

$$S(\gamma, z) = \frac{2}{z} \mathrm{Re}\left(\int_{-\infty}^{\infty} \omega \frac{\theta'}{\theta}\, d\omega\right). \tag{17}$$

This integral may be calculated from the analytical expansion of the function in $I_+(\tilde{\omega})$. Fixing the integration contour at the infinity we consider the integral

$$I = \int_C (\tilde{\omega} - i\gamma) \frac{\theta'}{\theta}\, d\tilde{\omega}.$$

The contour C consists of the linear part C_L for which Im $\tilde{\omega} = \gamma$ and semicircular part C_R in I_+ with a radius R and centre on the real axis (figure 1). Thus, we have $I = I_L + I_R$. For $R \to \infty$, $S = (2/z)\,\mathrm{Re}\,I_L = (2/z)\,\mathrm{Re}(I - I_R)$ and $I_R = 2\pi i \Sigma_j(\tilde{\omega}_{0j} - i\gamma_{0j})$, where $\tilde{\omega}_{0j} = \omega_{0j} + i\gamma_{0j}$ is simply the coordinates of the zeros of the transmission function determined above.

The use of the dimensionless variables η where $\eta = (\omega - \omega_T - \beta\varepsilon_0)(2\beta)^{-1}$ helps us to calculate the integral I_R (Akhmediev 1980). In this case, part of integration contour C_R will be transferred in the semicircular curve C_{R_1} in $I_+(\eta)$ with a radius $R_1 = R/2\beta$ and a centre point $\eta = 0$; so we have

$$I_R = \int_{C_{R_1}} (2\beta\eta + \omega_T + \beta\varepsilon_0 - i\gamma) \frac{\theta'}{\theta}\, d\eta. \tag{18}$$

In the limiting case, $R_1 \to \infty$ integration may be performed along all the semi-circular

contour neglecting the part between the real axis and the linear part of the integration path. Transforming the variable $\eta = R_1 \exp(i\xi)$, we have

$$I_R = iR_1 \int_0^\pi [2\beta R_1 \exp(i\xi) + \omega_T + \beta\varepsilon_0 - i\gamma) \frac{\theta'}{\theta} \, d\eta. \tag{19}$$

In the $R_1 \to \infty$ limit all the terms in equation (19) may be neglected except the first. Sufficient accuracy for θ'/θ is of order R_1^{-2}. Thus, for the θ'/θ-value, one obtains

$$\theta'/\theta \simeq i\alpha\omega_T z/4cn_0 R_1^2 \exp(2i\xi) \tag{20}$$

where

$$\alpha \equiv \varepsilon_0 \omega_{LT}/\beta.$$

Finally, I_R can be expressed as

$$I_R = -\pi\alpha\beta\omega_T z/2cn_0 \tag{21}$$

and is in accordance with the results obtained by Akhmediev (1980). This result is evident because of the small influence of the supplementary waves in the region far from the exciton resonance.

As a result, we have

$$S(\gamma, z) = \pi\gamma_{cr}\left(\frac{\omega_T \alpha^{1/2}}{2cn_0} - \frac{4}{z}\sum_j \frac{\gamma_{0j} - \gamma}{\gamma_{cr}}\right). \tag{22}$$

It is clear that, to obtain the IEAC dependence on γ, we should determine only the imaginary coordinates of the transmission zeros. This formula enables us to calculate accurate values for the IEAC for arbitrary values of z (figure 2, full curve). The calculations carried out by Akhmediev (1980) were made using the approximation of one-wave full absorption (see figure 2, broken curve). Also, the calculations accomplished in this work show that, for $\gamma = 0$, $S(\gamma, z) \to S_{cr}$ when $z \to 0$ but, according to Akhmediev (1980), $S(\gamma) = 0$ for every z. For large z-values the $S(\gamma, z)$-dependences calculated using (22) and by Akhmediev (1980) coincide with rather good accuracy.

The above approach for transverse excitons is also valid for mixed excitons. In this case the critical damping constant γ_{cr} and the critical frequency ω_{cr} may be calculated from (6) and (7). These values correspond to the crossing point of the dispersive branches for the waves $\tilde{n}_1(\omega)$ and $\tilde{n}_2(\omega)$ and are expressed by

$$\begin{aligned}\gamma_{cr} &= 4\beta_\perp \omega_{LT}(\varepsilon_\parallel^0/\varepsilon_\perp^0)\sin^2\varphi \\ \omega_{cr} &= \omega_L + \beta_\perp\varepsilon_\parallel^0 + \sin^2\varphi\,(\beta_\parallel - \beta_\perp\varepsilon_\parallel^0/\varepsilon_\perp^0)\end{aligned} \tag{23}$$

where

$$\omega_L = \omega_T + \omega_{LT} \qquad \beta_\parallel = \hbar\omega^2/2m_\parallel^* c^2 \qquad \beta_\perp = \hbar\omega^2/2m_\perp^* c^2.$$

The dependences of γ_{cr} (curve A) and ω_{cr} (curve B) for CdS crystals for mixed excitons are presented in figure 3.
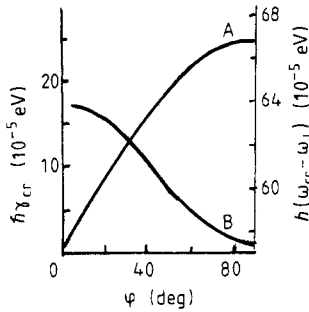
Figure 3. The angular dependences of the critical damping constant γ_{cr} (curve A) and the critical frequency ω_{cr} (curve B) in CdS crystals for mixed excitons with the parameters $\omega_T = 2.5527$ eV, $\omega_{LT} = 0.001\,86$ eV, $\varepsilon_\perp^0 = 8.3$, $\varepsilon_\parallel^0 = 9.0$, $m_\perp^0 = 0.9m_0$ and $m_\parallel^* = 5m_0$ (where m_0 is the electron mass).
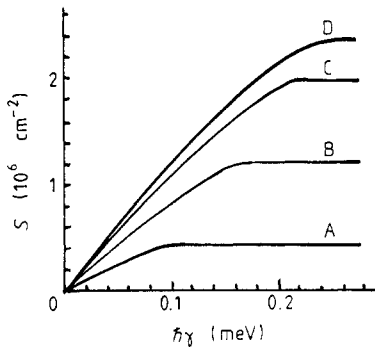

Figure 4. The γ-dependence of the IEAC in CdS crystals for mixed excitons: curve A, $\varphi = 25°$; curve B, $\varphi = 45°$; curve C, $\varphi = 65°$; curve D, $\varphi = 80°$. The calculations were performed using the same parameters as in figure 3.

For mixed excitons, equation (22) takes the form

$$S(\gamma, z, \varphi) = \pi\gamma_{cr}\left(\frac{\omega_L\alpha^{1/2}}{2c\varepsilon^{1/2}} - \frac{4}{z}\sum_j \frac{\gamma_{0j} - \gamma}{\gamma_{cr}}\right) \tag{24}$$

where

$$\alpha = \varepsilon_0\omega_{LT}\sin^2\varphi/\beta_\perp\varepsilon_\perp^0 \qquad \varepsilon = \varepsilon_\parallel^0 + \sin^2\varphi\,(1 - \varepsilon_\parallel^0/\varepsilon_\perp^0).$$

The angular dependences of $S(\gamma)$ for CdS crystals for mixed excitons are represented in figure 4. These numerical results from (24) are in rather good agreement with the experimental data obtained by Novikov *et al* (1986).

5. Conclusions

In conclusion, the application of the Kramers–Kronig relations to media with spatial dispersion has been studied. The advanced theory may be applied to the experimental analysis of the amplitude–phase measurements of semiconductor crystal transmission spectra near the exciton resonances. Two possibilities have been considered. The first is the detection of optical properties of normal waves diverted in space or in time (i.e. in wedge-shaped crystals or for a pulsed technique). The second possibility is the analysis of the normal wave interference for plane-parallel crystals. In this case on the basis of an analysis of the analytical properties of the transmission function it is possible to derive the additional Kramers–Kronig relations accounting for the supplementary wave interference. On the contrary, using the same theoretical description the dependence of the IEAC on the damping constant may be explained.

Acknowledgments

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