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Refractive index and extinction coefficient for polar cubic crystals in the range of the single-phonon resonance

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Abstract. An expression for the complex dielectric constant of cubic polar crystals, valid in the frequency range of the transverse optical phonon mode Ω_{TO}, and including the local-field effects, is derived. The analysis of the reflectance data of GaP shows that in the range 27.20–27.40 μm the local-field effects introduce significative variations in the index of refraction (about 15–20%) and in the extinction coefficient (about 30–40%).

1. Introduction

In this paper we restrict our attention to the lattice-reflection spectra for III–V compounds in the frequency range of the phonon optical modes. It is common practice for those materials in the above frequency range to calculate the index of refraction $n(\omega)$ and the extinction coefficient $k(\omega)$ by using a classical harmonic oscillator model (Hass 1967). According to this model (Born and Huang 1954) the displacement difference $\boldsymbol{u} = \boldsymbol{u}_1 - \boldsymbol{u}_2$ ($\boldsymbol{u}_1$ and $\boldsymbol{u}_2$ are the displacements of the two ions in a unit cell) satisfies a classical harmonic oscillator equation of the eigenfrequency Ω_{TO} and damping constant γ, where Ω_{TO} is the frequency of the transverse optical modes at the $\boldsymbol{Q} = \boldsymbol{0}$ point in the Brillouin zone. The complex dielectric constant in the one-oscillator model as the form:

$$\varepsilon(\omega) = \mathrm{Re}\,\varepsilon(\omega) + \mathrm{i}\,\mathrm{Im}\,\varepsilon(\omega) = \varepsilon_\infty + \frac{(\varepsilon_0 - \varepsilon_\infty)\omega_0^2}{\omega_0^2 - \omega^2 - \mathrm{i}\gamma\omega} \tag{1}$$

where the frequency ω_0 is equal to the frequency Ω_{TO}. $\varepsilon_0 = \varepsilon(\omega = 0)$ is the static dielectric constant; ε_∞ is the dielectric constant at frequencies large compared to the optical phonon frequencies, which is included in calculations to account for the dielectric screening of the material due to the interband electronic transitions. The index of refraction, the extinction coefficient and the absorption coefficient $\alpha_f(\omega)$ for the fundamental resonance can be calculated by solving the following set of equations:

$$\mathrm{Re}\,\varepsilon(\omega) = n^2 - k^2 = \varepsilon_\infty + \frac{(\varepsilon_0 - \varepsilon_\infty)[1 - (\omega/\omega_0)^2]}{[1 - (\omega/\omega_0)^2]^2 + (\omega/\omega_0)^2(\gamma/\omega_0)^2} \tag{2a}$$

$$\mathrm{Im}\,\varepsilon(\omega) = 2nk = \frac{(\varepsilon_0 - \varepsilon_\infty)(\omega/\omega_0)(\gamma/\omega_0)}{[1 - (\omega/\omega_0)^2]^2 + (\omega/\omega_0)^2(\gamma/\omega_0)^2} \tag{2b}$$

$$a_f(\omega) = \frac{2\omega}{c}k(\omega). \tag{2c}$$

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The problem is reduced to a question of fitting the reflectivity data in terms of four parameters ε_0, ε_∞, ω_0 and γ to give the best fit of the calculated reflectivity curves $R(\omega)$ (of an infinitely thick plate at normal incidence)

$$R(\omega) = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2} \tag{2d}$$

to the experimental ones. According to the paper by Kleinman and Spitzer (1960) the best values of those parameters for GaP are:

$$\varepsilon_0 = 10.182 \qquad \varepsilon_\infty = 8.457 \qquad \omega_0 = 366.3 \text{ cm}^{-1} \ (\lambda_0 = 27.30 \ \mu\text{m})$$
$$\gamma = 0.003\omega_0. \tag{3}$$

The dielectric constant (1) does not take into account the rapidly varying part of the polarization of the crystal due to the atomic displacements, i.e. the local-field effects.

The purpose of this paper is to investigate the contributions of the local-field effects to the complex dielectric constant in the frequency range of the transverse optical modes. This theory is needed for more accurate interpretation of optical spectra in crystals, because the values of the index of refraction and the extinction coefficient are different depending on whether or not the local-field effects are taken into account.

2. The complex constant beyond the one-oscillator model

In what follows we assume that the polarization of the crystal $\boldsymbol{P}(\boldsymbol{r}, t)$ at the point $\boldsymbol{r}$ is only due to the atomic displacements $u_\beta^{l\kappa}$:

$$P_\alpha(\boldsymbol{r}, t) = P_{\alpha\beta}^\kappa(\boldsymbol{r} - \boldsymbol{R}_l) u_\beta^{l\kappa}(t) \tag{4a}$$

where $l = 1, 2, \ldots, N$ numbers the unit cells in the crystal, $\kappa = 1, 2$ characterizes the two ions in the unit cell and $\boldsymbol{R}_l$ is the lattice vector of the lth unit cell. The phenomenological parameters $P_{\alpha\beta}^\kappa(\boldsymbol{r} - \boldsymbol{R}_l)$ which determine the polarization of the crystal have the following Fourier transform:

$$P_{\alpha\beta}^\kappa(\boldsymbol{r} - \boldsymbol{R}_l) = \frac{1}{\sqrt{V}} \sum_{\boldsymbol{Q}} \sum_{\boldsymbol{G}_n} \exp[\mathrm{i}(\boldsymbol{Q} + \boldsymbol{G}_n) \cdot (\boldsymbol{r} - \boldsymbol{R}_l)] P_{\alpha\beta}^\kappa(\boldsymbol{Q} + \boldsymbol{G}_n) \tag{4b}$$

where $\boldsymbol{G}_n$ is a reciprocal-lattice vector. The Fourier components $P_{\alpha\beta}^\kappa(\boldsymbol{Q} + \boldsymbol{G}_n)$ are connected with the rapidly varying part of the polarization of the crystal due to the atomic displacements, while $P_{\alpha\beta}^\kappa(\boldsymbol{Q})$ determines the slowly varying part of the polarization.

The Fourier coefficients $\varepsilon_{\alpha\beta}(\boldsymbol{Q} + \boldsymbol{G}_n; \boldsymbol{Q} + \boldsymbol{G}_m; \omega)$ of the dielectric tensor $\varepsilon_{\alpha\beta}(\boldsymbol{r}; \boldsymbol{r}'; t)$ can be calculated by means of the classical equation of the motion for the displacement $u_\beta^{l\kappa}$ of the (l, κ) ion (Born and Huang 1954) or by the Green function method (Glinskii and Koinov 1989). The final result for the dielectric tensor is:

$$\varepsilon_{\alpha\beta}(\boldsymbol{Q} + \boldsymbol{G}_n; \boldsymbol{Q} + \boldsymbol{G}_m; \omega) = \varepsilon_\infty \delta_{\alpha\beta} \delta_{\boldsymbol{G}_n \boldsymbol{G}_m} - \frac{4\pi}{M_0 \varepsilon_\infty} \sum_\lambda \frac{Z_\alpha(\lambda, \boldsymbol{Q} + \boldsymbol{G}_n) Z_\beta^*(\lambda, \boldsymbol{Q} + \boldsymbol{G}_m)}{\omega^2 - \Omega_\lambda^2(\boldsymbol{Q}) + \mathrm{i}\omega\gamma} \tag{5a}$$

where the phenomenological parameters $Z_\alpha(\lambda, \boldsymbol{Q} + \boldsymbol{G}_n)$ are defined as follows:

$$Z_\alpha(\lambda, \boldsymbol{Q} + \boldsymbol{G}_n) = P_{\alpha\beta}^\kappa(\boldsymbol{Q} + \boldsymbol{G}_n) e_\beta^\kappa(\lambda, \boldsymbol{Q}). \tag{5b}$$

Here $M_0 = \sum_\kappa M_\kappa$ is the total mass in the lth unit cell; $\Omega_\lambda(\boldsymbol{Q})$ are the 'bare' phonon frequencies with a wavevector $\boldsymbol{Q}$ in a Brillouin zone, λ is a branch index, and $\boldsymbol{e}^\kappa(\lambda, \boldsymbol{Q})$

denote the phonon eigenvectors. By using the fact that in the case of a cubic polar crystal with two atoms per cell in the $\boldsymbol{Q} \to \boldsymbol{0}$ limit one has:

$$\lim_{\boldsymbol{Q}\to\boldsymbol{0}} \sum_{\alpha} \frac{Z_{\alpha}(\lambda, \boldsymbol{Q})\boldsymbol{Q}_{\alpha}}{|\boldsymbol{Q}|} = Z\delta_{\lambda,LO} \tag{6a}$$

we can prove that the dielectric constant (1) is related to the $\boldsymbol{G}_n = \boldsymbol{G}_m = \boldsymbol{0}$ component of the longitudinal part of $\varepsilon_{\parallel}$ of the dielectric matrix $\varepsilon_{\alpha\beta}(\boldsymbol{Q}+\boldsymbol{G}_n; \boldsymbol{Q}+\boldsymbol{G}_m; \omega)$:

$$\varepsilon_{\parallel}(\boldsymbol{Q}+\boldsymbol{G}_n; \boldsymbol{Q}+\boldsymbol{G}_m; \omega) = \frac{(\boldsymbol{Q}+\boldsymbol{G}_n)_{\alpha}}{|\boldsymbol{Q}+\boldsymbol{G}_n|}\varepsilon_{\alpha\beta}(\boldsymbol{Q}+\boldsymbol{G}_n; \boldsymbol{Q}+\boldsymbol{G}_m; \omega)\frac{(\boldsymbol{Q}+\boldsymbol{G}_m)_{\beta}}{|\boldsymbol{Q}+\boldsymbol{G}_m|} \tag{6b}$$

$$\varepsilon(\omega) = \lim_{\boldsymbol{Q}\to\boldsymbol{0}} \varepsilon_{\parallel}(\boldsymbol{Q}+\boldsymbol{0}; \boldsymbol{Q}+\boldsymbol{0}; \omega). \tag{6c}$$

The phenomenological parameter Z is related to the transverse optical phonon frequency $\Omega_{TO} = \Omega_{TO}(\boldsymbol{Q} = \boldsymbol{0})$ as follows:

$$\frac{4\pi Z^2}{M_0\varepsilon_{\infty}(\varepsilon_0 - \varepsilon_{\infty})} = \Omega_{TO}^2. \tag{6d}$$

The main disadvantage of the relation (6*c*) is that the contributions of the local-field effects to the dielectric constant have not been taken into consideration. There are two methods that can be employed to overcome this problem. According to the first one, the correct definition of the dielectric constant, which considers the rapidly varying part of the polarization of the crystal due to the atomic displacements, is based on the matrix inversion procedure. In the case of a cubic polar crystal with two atoms per cell in the $\boldsymbol{Q} \to \boldsymbol{0}$ limit one has (Johnson 1975):

$$\varepsilon^{-1}(\omega) = \lim_{\boldsymbol{Q}\to\boldsymbol{0}} \varepsilon_{\parallel}^{-1}(\boldsymbol{Q}+\boldsymbol{G}_n; \boldsymbol{Q}+\boldsymbol{G}_n; \omega)|_{\boldsymbol{G}_n=\boldsymbol{G}_m=\boldsymbol{0}}. \tag{7}$$

The second way to calculate the dielectric constant without using the matrix-inversion procedure is to employ the so-called crystal optics approximation (Koinov and Glinskii 1988). According to this approximation the dielectric constant has the form:

$$\varepsilon(\boldsymbol{Q}, \omega) = \varepsilon_{\infty} - \frac{4\pi}{M_0\varepsilon_{\infty}} \sum_{\lambda,\lambda'} \frac{Z_{\alpha}(\lambda, \boldsymbol{Q})\boldsymbol{Q}_{\alpha}}{|\boldsymbol{Q}|} \tilde{S}^{(0)}_{\lambda\lambda'}(\boldsymbol{Q}, \omega) \frac{Z^*_{\beta}(\lambda', \boldsymbol{Q})\boldsymbol{Q}_{\beta}}{|\boldsymbol{Q}|}. \tag{8}$$

Here $\tilde{S}^{(0)}_{\lambda\lambda'}(\boldsymbol{Q}, \omega)$ is the Fourier transform of the 'bare' phonon Green function which takes into account the rapidly varying part of the polarization of the crystal due to the atomic displacements. $\tilde{S}^{(0)}_{\lambda\lambda'}(\boldsymbol{Q}, \omega)$ satisfies the following set of equations:

$$\sum_{\lambda''} \left\{ \delta_{\lambda\lambda''}[\omega^2 - \Omega^2_{\lambda}(\boldsymbol{Q})] - \frac{\omega^2}{M_0c^2} \sum_{\boldsymbol{G}_n\neq\boldsymbol{0}} Z_{\alpha}(\lambda, \boldsymbol{Q}+\boldsymbol{G}_n) D^{(\infty)}_{\alpha\beta}(\boldsymbol{Q}+\boldsymbol{G}_n, \omega) Z^*_{\beta}(\lambda'', \boldsymbol{Q}+\boldsymbol{G}_n) \right\}$$
$$\times \tilde{S}^{(0)}_{\lambda''\lambda'}(\boldsymbol{Q}, \omega) = \delta_{\lambda\lambda'}. \tag{9a}$$

Here $D^{(\infty)}_{\alpha\beta}(\boldsymbol{Q}+\boldsymbol{G}_n, \omega)$ is the Fourier coefficient of the phonon propagator, screened (due to the interband electron transitions) by the dielectric constant ε_{∞}:

$$D^{(\infty)}_{\alpha\beta}(\boldsymbol{K}, \omega) = \frac{4\pi c^2}{\varepsilon_{\infty}\omega^2 - c^2K^2 + \mathrm{i}0^+} \left[\delta_{\alpha\beta} - \frac{c^2\boldsymbol{K}_{\alpha}\boldsymbol{K}_{\beta}}{\varepsilon_{\infty}\omega^2 + \mathrm{i}0^+} \right].$$

The above propagator can be written as a sum of a longitudinal (instantaneous) part and a transverse (retardation) part. The most simple way to obtain the exact expression for

$\tilde{S}^{(0)}_{\lambda\lambda'}(\boldsymbol{Q},\omega)$ is to neglect the retardation part of the photon Green function in comparison to its instantaneous part (Koinov and Glinskii 1988):

$$D^{(\infty)}_{\alpha\beta}(\boldsymbol{Q}+\boldsymbol{G}_n,\omega)\approx\frac{4\pi c^2}{\varepsilon_\infty\omega^2+\mathrm{i}0^+}\frac{(\boldsymbol{Q}+\boldsymbol{G}_n)_\alpha(\boldsymbol{Q}+\boldsymbol{G}_n)_\beta}{|\boldsymbol{Q}+\boldsymbol{G}_n|^2}.$$

The instantaneous approximation simplifies the left-and side of (9*a*) which assumes the following form:

$$\sum_{\lambda''}\left\{\delta_{\lambda\lambda''}[\omega^2(\boldsymbol{Q})-\Omega^2_\lambda(\boldsymbol{Q})]-\frac{4\pi}{M_0\varepsilon_\infty}\sum_{\boldsymbol{G}_n\neq\boldsymbol{0}}\frac{Z_\alpha(\lambda\boldsymbol{Q}+\boldsymbol{G}_n)(\boldsymbol{Q}+\boldsymbol{G}_n)_\alpha}{|\boldsymbol{Q}+\boldsymbol{G}_n|}\right.$$
$$\left.\times\frac{Z^*_\beta(\lambda'',\boldsymbol{Q}+\boldsymbol{G}_n)(\boldsymbol{Q}+\boldsymbol{G}_n)_\beta}{|\boldsymbol{Q}+\boldsymbol{G}_n|}\right\}\tilde{S}^{(0)}_{\lambda''\lambda'}(\boldsymbol{Q},\omega)=\delta_{\lambda\lambda'}. \tag{9b}$$

The solution of the above equations can be written in the form:

$$\tilde{S}^{(0)}_{\lambda\lambda'}(\boldsymbol{Q},\omega)=\sum_\mu\frac{A_{\lambda\mu}(\boldsymbol{Q})A^*_{\mu\lambda'}(\boldsymbol{Q})}{\omega^2-\tilde{\Omega}^2_\mu(\boldsymbol{Q})} \tag{9c}$$

where the unknown quantities $\tilde{\Omega}_\mu$ and $A_{\lambda\mu}(\boldsymbol{Q})$ have to be determined by solving the following set of linear homogeneous algebraic equations:

$$\sum_{\lambda'}\left\{\delta_{\lambda\lambda'}[\tilde{\Omega}^2_\mu(\boldsymbol{Q})-\Omega^2_\lambda(\boldsymbol{Q})]-\frac{4\pi}{M_0\varepsilon_\infty}\sum_{\boldsymbol{G}_n\neq\boldsymbol{0}}\frac{Z_\alpha(\lambda,\boldsymbol{Q}+\boldsymbol{G}_n)(\boldsymbol{Q}+\boldsymbol{G}_n)_\alpha}{|\boldsymbol{Q}+\boldsymbol{G}_n|}\right.$$
$$\left.\times\frac{Z^*_\beta(\lambda',\boldsymbol{Q}+\boldsymbol{G}_n)(\boldsymbol{Q}+\boldsymbol{G}_n)_\beta}{|\boldsymbol{Q}+\boldsymbol{G}_n|}\right\}A_{\lambda'\mu}(\boldsymbol{Q})=0. \tag{9d}$$

The solutions $A_{\lambda\mu}(\boldsymbol{Q})$ can be interpreted as components of an unitary matrix which transforms $\tilde{S}^{(0)}_{\lambda\lambda'}(\boldsymbol{Q},\omega)$ into a diagonal form.

In the instantaneous approximation the dielectric constant (8) has the form:

$$\varepsilon(\boldsymbol{Q},\omega)=\varepsilon_\infty-\frac{4\pi}{M_0\varepsilon_\infty}\sum_\mu\frac{1}{\omega^2-\tilde{\Omega}^2_\mu(\boldsymbol{Q})+\mathrm{i}\omega\gamma}\left|\frac{\tilde{Z}_\alpha(\mu,\boldsymbol{Q})Q_\alpha}{|\boldsymbol{Q}|}\right|^2 \tag{10a}$$

where

$$\tilde{Z}_\alpha(\mu,\boldsymbol{Q})=\sum_\lambda A_{\lambda\mu}(\boldsymbol{Q})Z_\alpha(\lambda,\boldsymbol{Q}).$$

In a cubic polar crystal with two atoms per cell the relation (6*a*) holds and the dielectric constant assumes the form:

$$\varepsilon(\omega)=\varepsilon_\infty-\frac{4\pi Z^2}{M_0\varepsilon_\infty}\sum_\mu\frac{|A_{LO,\mu}|^2}{\omega^2-\tilde{\Omega}^2_\mu+\mathrm{i}\omega\lambda}. \tag{10b}$$

The frequencies $\tilde{\Omega}_\mu=\tilde{\Omega}_\mu(\boldsymbol{Q}=\boldsymbol{0})$ and the quantities $A_{\lambda\mu}=A_{\lambda\mu}(\boldsymbol{Q}=\boldsymbol{0})$ are determined by the solutions of the following set of equations:

$$\begin{pmatrix}\tilde{\Omega}^2_\mu-\Omega^2_{TO}-\Delta_{TO,TO} & -\Delta_{TO,TO} & -\Delta_{TO,LO}\\ -\Delta_{TO,TO} & \tilde{\Omega}^2_\mu-\Omega^2_{TO}-\Delta_{TO,TO} & -\Delta_{TO,LO}\\ -\Delta_{LO,TO} & -\Delta_{LO,TO} & \tilde{\Omega}^2_\mu-\Omega^2_{TO}-\Delta_{LO,LO}\end{pmatrix}$$
$$\times\begin{pmatrix}A_{TO,\mu}\\ A_{TO,\mu}\\ A_{LO,\mu}\end{pmatrix}=0. \tag{11}$$

The quantities $\Delta_{\lambda,\lambda'}$ ($\lambda, \lambda' = TO, LO$) depend on the rapidly varying part of the polarization of the crystal due to the atomic displacements:

$$\Delta_{\lambda,\lambda'} = \frac{4\pi}{M_0\varepsilon_\infty} \sum_{\boldsymbol{G}_n \neq \boldsymbol{0}} \frac{Z_\alpha(\lambda, \boldsymbol{G}_n)(\boldsymbol{G}_n)_\alpha}{|\boldsymbol{G}_n|} \frac{Z^*_\beta(\lambda', \boldsymbol{G}_n)(\boldsymbol{G}_n)_\beta}{|\boldsymbol{G}_n|}. \tag{12a}$$

The solutions of the equations (11) are:

$$\tilde{\Omega}_1^2 = \Omega_{TO}^2 \rightarrow A_{TO,1} = -A_{TO,1};\ A_{LO,1} = 0 \tag{12b}$$

$$\tilde{\Omega}_2^2 = \omega_0^2 + \Delta\omega_0^2 \rightarrow A_{TO,2} = A_{TO,2} = \frac{\Delta_{TO,TO} - \Delta_{LO,LO}/2 + D}{2\Delta_{TO,LO}} A_{LO,2} \tag{12c}$$

$$\tilde{\Omega}_3^2 = \omega_0^2 - \Delta\omega_0^2 \rightarrow A_{TO,3} = A_{TO,3} = \frac{\Delta_{TO,TO} - \Delta_{LO,LO}/2 - D}{2\Delta_{TO,LO}} A_{LO,3} \tag{12d}$$

where

$$D = \sqrt{\left(\Delta_{TO,TO} - \frac{\Delta_{LO,LO}}{2}\right)^2 + 2|\Delta_{LO,TO}|^2}. \tag{12e}$$

Here the frequencies ω_0 and $\Delta\omega_0$ are defined as follows:

$$\omega_0^2 = \Omega_{TO}^2 + \Delta_{TO,TO} + \tfrac{1}{2}\Delta_{LO,LO} \tag{13a}$$

$$(\Delta\omega_0) = 4\sqrt{(\Delta_{TO,TO} - \tfrac{1}{2}\Delta_{LO,LO})^2 + 2|\Delta_{TO,LO}|^2}. \tag{13b}$$

Due to the rapidly varying part of the polarization (i.e. the local-field effects) the threefold degenerate phonon energy Ω_{TO} at the centre of the Brillouin zone is split into three energies $\tilde{\Omega}_\mu$; $\mu = 1, 2, 3$. Using the solutions (12) and (13) one can rewrite the dielectric constant (10*b*) in the following form:

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{(\varepsilon_0 - \varepsilon_\infty)\omega_0^2}{(1+C)C_1} \left[\frac{1}{\omega_0^2 + (\Delta\omega_0)^2 - \omega^2 - \mathrm{i}\omega\gamma} + \frac{C}{\omega_0^2 - (\Delta\omega_0)^2 - \omega^2 - \mathrm{i}\omega\gamma}\right] \tag{14}$$

where

$$C = \frac{|A_{LO,3}|^2}{|A_{LO,2}|^2} \qquad C_1 = \frac{1 - [(1-C)/(1+C)]\Delta\omega_0^2/\omega_0^2}{1 - \Delta\omega_0^4/\omega_0^4}. \tag{15}$$

The dielectric constant (14) depends on the six parameters: ε_0, ε_∞, ω_0, γ, $\Delta\omega_0$ and C. Three of them ε_0, ε_∞, γ have a clear physical meaning, low and high dielectric constants and the damping constant. The frequency ω_0 is very close to Ω_{TO}, because a characteristic feature of the polar crystals is the predominance of the slowly varying part of the polarization as compared with its rapidly varying part. Actually, ω_0 and $\Delta\omega_0$ determine the renormalization (due to the local-field effects) of the three degenerate 'bare' phonon frequencies Ω_{TO}. By rewriting the dielectric constant (14) in the form:

$$\varepsilon(\omega) = \varepsilon_\infty + \sum_{\mu=2,3} \frac{F_\mu}{\tilde{\Omega}_\mu^2 - \omega^2 - \mathrm{i}\omega\gamma}$$

one sees that the constant C is related to the oscillator strength F_μ ($C = F_3/F_2$).

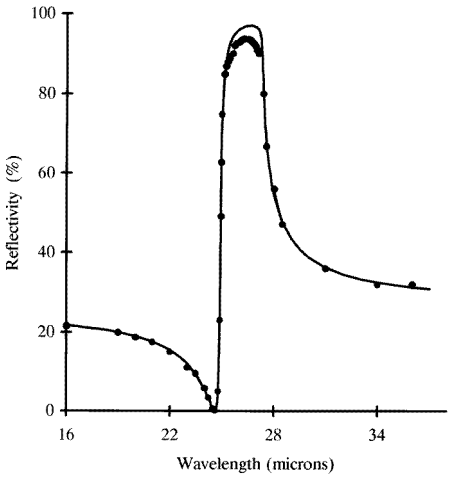

Figure 1. Lattice reflection spectrum of GaP. The experimental data (Kleinman and Spitzer 1960) are shown in circles, and the calculated fit by using six parameters by the solid line.

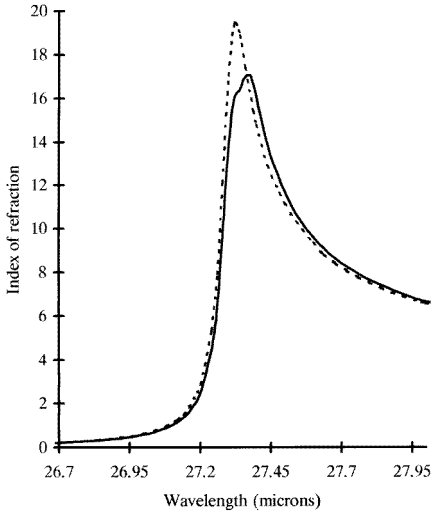

Figure 2. Index of refraction of GaP, calculated by using a one-oscillator model (- - - -), and by taking into account the local-field effects (solid line).

3. Analysis of the reflectance data of GaP

It is physically reasonable to expect that the adjustable values of the low and high dielectric constants and the value of the damping constant should slightly depend on the rapidly varying part of the polarization. The major result of the local-field corrections is that the triple degeneracy of the optical phonon bands at the centre of the Brillouin zone is removed by the local-field effects arising from the rapidly varying part of the polarization.

The problem now is to fit the reflectance data of GaP by means of the dielectric constant (14), i.e. in terms of six parameters ε_0, ε_∞, ω_0, γ, $\Delta\omega_0$ and C. Our numerical calculations show that the best values of ε_0, ε_∞, ω_0, γ, $\Delta\omega_0$ and C, obtained by adjusting those six

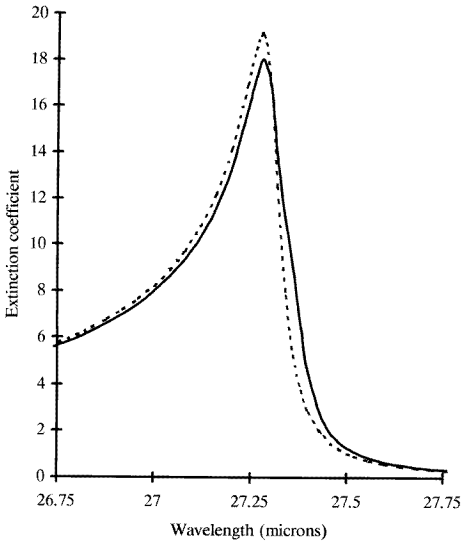

Figure 3. Extinction coefficient of GaP, calculated by using a one-oscillator model (- - - -), and by taking into account the local-field effects (solid line).

parameters to give the best fit of the calculated reflectivity curve to the experimental one (Kleinman and Spitzer 1960), are

$$\varepsilon_0 = 10.182 \qquad \varepsilon_\infty = 8.457 \qquad \gamma/\omega_0 = 0.003 \qquad \omega_0 = 365.9\ \text{cm}^{-1}\ (\lambda_0 = 27.33\ \mu\text{m})$$
$$\Delta\omega_0/\omega_0 = 0.045 \qquad C = 0.419. \tag{16}$$

The reflectivity $R(\lambda)$, calculated by means of the set of parameters (16) is shown in figure 1. The position of the reflectivity minimum is at 24.60 μm. The experimental data by Kleinman and Spitzer (1960), which were given in graphical form, were read out directly and plotted in figure 1. The small discrepancy in the range of the reflectivity maximum between the experimental data and calculated curve is due to the surface effects (Kleinman and Spitzer 1960). The index of refraction $n(\lambda)$ and the extinction coefficient $k(\lambda)$, plotted against wavelength in micrometres obtained from the reflectivity calculations by using parameters (3) and (16), are presented in figures 2 and 3.

In conclusion it is worth noting that the quality of the fit achieved with six adjustable parameters is almost the same as that achieved by Kleinman and Spitzer. The percentage correction to the reflectivity calculated using the dielectric constant (1) with the set of parameters (3) and that shown in figure 1 is less than 1% in the range 16–40 μm. However, the percentage corrections due to the local-field effects to the index of refraction and extinction coefficient near resonance (27.20–27.40 μm) have been estimated as being significant (about 15–20% for the index of refraction and about 30–40% for the extinction coefficient).

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