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ANISOTROPY OF INFRARED SPECTRAL ABSORPTION IN UNIAXIALLY-STRESSED *n*-GaP

A. E. BELYAEV,[1] O. P. GORODNICHII,[1] Z. A. DEMIDENKO[2] and P. M. TOMCHUK[2]
Institutes of [1]Semiconductors and [2]Physics, Ukrainian S.S.R. Academy of Sciences, Kiev, U.S.S.R.

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Abstract—A theoretical and experimental investigation of free-carrier absorption (FCA) in unstressed (optically isotropic) and uniaxially-stressed (optically anisotropic) *n*-GaP in the quantum frequency regions has been carried out. At a pressure $P = 10^4$ kgf cm^{-2}, parallel to the [100] direction, and at $T = 300$K, practically all the electrons previously uniformly distributed throughout all the equivalent valleys migrated into one lowered valley. This permitted the investigation of the components of the FCA coefficient at parallel ($\alpha_{\parallel}$) and perpendicular ($\alpha_{\perp}$) orientations of the field axis relative to the long axis of the mass tensor ellipsoid of the lowered valley. It is shown that the experimentally-observed singularities of FCA in *n*-GaP (varying frequency dependences α_0, $\alpha_{\parallel}$, $\alpha_{\perp}$, low value of the ratio $\alpha_{\perp}/\alpha_{\parallel}$ [(1)] and frequency dependence with the power less than 3/2) can be explained within the framework of FCA quantum theory only if the intermediate states in the upper conduction band Δ_2' and the intervalley scattering anisotropy are taken into account.

INTRODUCTION

In many-valley semiconductors (Ge, Si, GaP etc.) free electrons are uniformly distributed over the entire equivalent minima of the conduction band. This leads to integral isotropy of electrical and optical properties of such crystals, although the constant energy surfaces in each valley are stretched along definite crystallographic axes in the shape of ellipsoids. In the case of a uniaxial elastic deformation upsetting the equivalence of the extrema, a redistribution of charge carriers takes place between the valleys. In particular, when the stress axis coincides with the long axis of one of the isoenergetic ellipsoids, then, at a certain value of applied pressure $P \geqslant P_{sat}$, when the band splitting exceeds the average energy of current carriers, all free electrons migrate into the lowered valley, so that the crystal becomes quasi-one-valley and optically anisotropic.[(1)] By irradiating such a crystal with i.r. light, polarized along or across the long axis of the lowered ellipsoid, the frequency dependence of the longitudinal $\alpha_{\parallel}(\omega)$ or transverse $\alpha_{\perp}(\omega)$ components of the free-carrier absorption (FCA) coefficient (ω being the incident angular light frequency) can be measured.

As has been pointed out in Refs (2, 3), that the dependence $\alpha_0(\omega)$ obtained for unstressed crystals in non-polarized light in the quantum frequency region $\hbar\omega \gg kT$ should be described by the power law $\alpha \sim \omega^{-z}$ in which the value of power z is determined by the scattering mechanisms existing in the crystal being investigated ($z = 7/2$, $5/2$, $3/2$ for impurity, polar-optical and acoustic scatterings, respectively). Demidenko and Tomchuk[(4)] carried out quantum calculations for the values $\alpha_{\parallel}(\omega)$ and $\alpha_{\perp}(\omega)$, and it was shown that for the above intravalley scattering mechanisms these dependences are described by the same power laws as for $\alpha_0(\omega)$, while their ratio ($\alpha_{\perp}/\alpha_{\parallel}$) cannot be less than that of the effective masses $\beta = m_{\parallel}^*/m_{\perp}^*$, even in the quantum region [see Ref. (1)]. At the same time it was noted[(4)] that in the case of GaP it is necessary, when calculating $\alpha_{\parallel}(\omega)$ and $\alpha_{\perp}(\omega)$ to take into account the singularities of the FCA process resulting from the energetic structure of this material.

EXPERIMENTAL RESULTS

In our work we measured FCA spectra in unstressed GaP cystals and crystals uniaxially stressed in the direction [100], Te-doped ($N_D = 6 \cdot 10^{17}$ cm^{-3}, $N_A = 3.7 \cdot 10^{17}$ cm^{-3}), within the range of $0.3 \leqslant \hbar\omega \leqslant 0.08$ *eV*. Measurements were conducted at $T = 300$ K. At $P = 10^4$ kgf cm^{-2} the energy gap between the lower and upper valleys $\Delta_P = \Xi_u (S_{11} - S_{12})P$ attained 0.11 eV [$\Xi_u = 8.8$ eV being

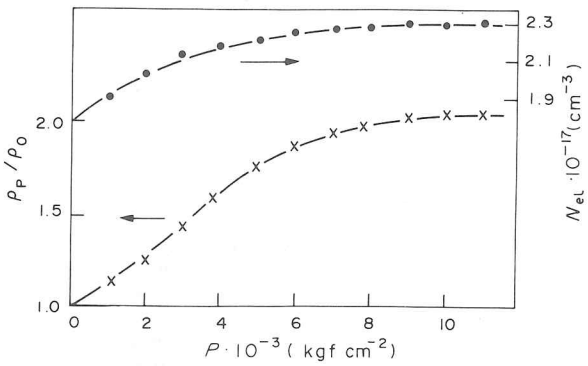

Fig. 1. Dependence of longitudinal piezoresistance ρ_P/ρ_0 and electron concentration N_{el} vs uniaxial compression $P\|[100]$ in n-GaP at $T = 300K$.

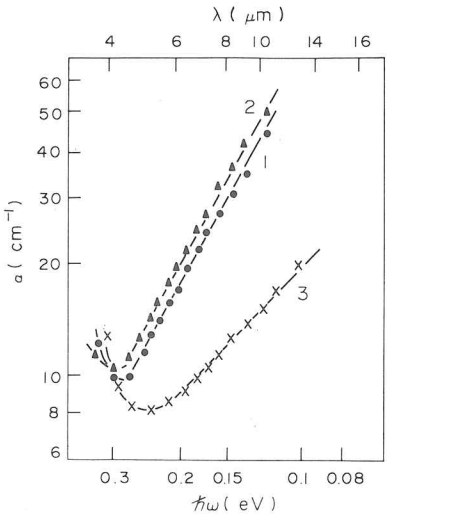

Fig. 2. Spectra of light absorption by free carriers in unstressed n-GaP(●) and in n-Gap uniaxially stressed(▲, ×) along [100]. 1, unpolarized light; 2, $l_\omega \perp [100]$; 3, $l_\omega \| [100]$.

the deformation potential shear constant of the conduction band;[5] $S_{11} = 9.539\cdot10^{-7}\,cm^2\,kg^{-1}$, $S_{12} = -2.931\cdot10^{-7}\,cm^2\,kg^{-1}$ being the compliance constants of the material under investigation[6]]. This led to a complete transmigration of free electrons into the lowered valley. This has been confirmed by saturation of the piezoresistance measured in the same crystals. Data on piezoresistance are given in Fig. 1. This figure shows also the dependence of the free-electron concentration on the uniaxial compression $P\|[100]$. Since at room temperature some of the donors in GaP prove to be unionized, then, due to the reduction of the energy of the ground impurity state during deformation [1] and ionization of donors, a rise of electron concentration is observed up to the value of $N_{el} = N_D - N_A$, signifying a complete ionization of the doping impurity at pressures $P \geqslant (8\text{–}9)\cdot10^3\,kgf\,cm^{-2}$. Thus, the contribution of the photoionization processes in $\alpha_\parallel(\omega)$ and $\alpha_\perp(\omega)$ is ruled out.

The optical absorption spectra were measured at normal light incidence on the (011) plane. The polarization vector was in this plane parallel or perpendicular to the direction [100]. The FCA spectra are shown in Fig. 2 as dependences of the absorption coefficient α upon the photon energy $\hbar\omega$. One can clearly see that:

(1) dependences $\alpha_\perp(\omega)$ and $\alpha_\parallel(\omega)$ differ greatly from one another ($\alpha_\perp \sim \omega^{-1.8}$, $\alpha_\parallel \sim \omega^{-1.0}$);

(2) the value of power z for $\alpha_\parallel(\omega)$ is notably lower than the minimum possible value $z = 1.5$ following from the theories of Rosenberg and Lax[2] and Visvanathan;[3]

(3) the $\alpha_\perp/\alpha_\parallel$ value is considerably lower than the ratio of the effective masses of electrons in GaP ($\beta = 10$).

THEORETICAL PROCEDURE

In calculating the FCA coefficients in GaP, along with the standard intravalley scattering mechanisms (impurities, polar optics and acoustics), we also took account of the intervalley scattering. In addition, we also took into consideration the intermediate state in the upper band Δ_2' of the conduction band, since in GaP the gap between the sub-band Δ_2' and Δ_1 at point X of the Brillouin zone is not large ($\Delta_X = X_3^c - X_1^c = 0.32\,eV$).[7] The transition $X_1^c \rightarrow X_3^c$ is dipole-permitted so it can be expected that the contribution of these components into $\alpha(\omega)$ will be appreciable.

With the above-mentioned results, and taking those of Ref. (4) into consideration, according to Demidenko and Tomchuk[4] dependences $\alpha_{0,\perp,\parallel}(\omega)$ can be written as follows

$$\alpha_{\parallel}(\omega) = B\frac{1}{\beta}\sum_j [A_j^2(\omega)\, L_j^{\parallel}(\beta) + A_{ji}^2(\omega)\, L_{ji}^{\parallel}(\beta)] \tag{1}$$

$$\alpha_{\perp}(\omega) = B\sum_j [A_j^2(\omega)\, L_j^{\perp}(\beta) + A_{ji}^2(\omega)\, L_{ji}^{\perp}(\beta)] \tag{2}$$

$$\alpha_0(\omega) = \left(\frac{\alpha_{\parallel} + 2\alpha_{\perp}}{3}\right)_{P=0}, \tag{3}$$

where

$$B = \frac{4\sqrt{2m_{\parallel}}\, e^2}{Cn\hbar^2}\cdot\frac{N_{\mathrm{el}}}{(\hbar\omega)^{3/2}}, \tag{4}$$

j numbers the scattering mechanisms (ji accounts for the intermediate states in zone Δ_2').

It can be seen from equations (1)–(4) that the scattering intensity $A_j^2(\omega)$ determines the frequency dependence of each of the j-scattering mechanisms while the multipliers $L_j(\beta)$ are its anisotropy. Such presentation is convenient, since it permits the use of the analogy with the transfer phenomena to introduce for each scattering mechanism a ratio $(L_j^{\perp}/L_j^{\parallel})$ characterizing the degree of scattering anisotropy K_j (in a low-frequency process, i.e. at $\hbar\omega \ll kT$ it is characterized by the ratio of relaxation times $\tau_j^{\parallel}/\tau_j^{\perp}$):

$$\alpha_j^{\perp}/\alpha_j^{\parallel} = \beta(L_j^{\perp}/L_j^{\parallel}) = \beta K_j. \tag{5}$$

Forms $A_j^2(\omega)$ and $L_j(\beta)$ for intraband scattering mechanisms are given in Table 1.

Let us consider in more detail the second components in equations (1)–(3) which appear as a result of accounting for the intermediate states in the next conduction band. These formulae show that such consideration leads formally to the appearance of a certain additional mechanism which we call ji-scattering. For each of the j-mechanisms of the intraband scattering the intensity of the ji-scattering has the following form

$$A_{ji}^2(\omega) = A_j^2(\omega)\,\eta_i(\omega) \tag{6}$$

Table 1

	Acoustic	Optical	Impurity	Intervalley
$A_j^2(\omega)$	$\dfrac{\Xi_u^2 kT}{\rho V_{\parallel}^2}$	$\dfrac{3\pi e^2\hbar^2}{\varepsilon_0 m_{\perp}}\left(\dfrac{\varepsilon_0}{\varepsilon_\infty}-1\right)\dfrac{\omega_0}{\omega}\chi_0$	$3\left(\dfrac{\pi e^2\hbar^2}{\varepsilon_0 m_{\perp}}\right)^2\dfrac{N_{\mathrm{imp}}}{(\hbar\omega)^2}$	$\dfrac{\mathscr{D}_{\mathrm{inter}}^2\hbar}{\rho\omega_{\mathrm{inter}}}\chi_{\mathrm{inter}}$
$L_j^{\parallel}$	$y^2+1.79y+1.026$	$2(1-\delta)(\beta-1)^{-1}$	$2(\beta\delta-1)[\beta(\beta-1)]^{-1}$	2β
$L_j^{\perp}$	$y^2+1.052y+0.78$	$(\beta\delta-1)(\beta-1)^{-1}$	$[\delta(\beta-2)+1](\beta-1)^{-1}$	$(\beta+1)\beta^{-1}$
γ_j	$\frac{3}{2}[y^2+1.252y+0.86]$	δ	$(\beta\delta+1)\beta^{-1}$	$\frac{3}{2}$

$y = \Xi_d/\Xi_u$ is the ratio of deformation potential constants (for numerical estimations $y = -1.77$[8]).

$\delta = \dfrac{\mathrm{arctg}\sqrt{\beta-1}}{\sqrt{\beta-1}}$, $V_{\parallel}$ is the longitudinal sound velocity.

$\mathscr{D}_{\mathrm{inter}}$ is the electron–phonon coupling constant.

ω_0, ω_{inter} are the phonon frequencies.

χ_0, χ_{inter} are the multipliers accounting for the absorption and emission of the corresponding phonons

$$\chi_0 = \tfrac{1}{2}\left[n_{\mathrm{opt}}\left(1+\frac{\omega_o}{\omega}\right)^{1/2} + (n_{\mathrm{opt}}+1)\left(1-\frac{\omega_0}{\omega}\right)^{1/2}\right.$$

$$\left.\chi_{\mathrm{inter}} = \tfrac{1}{2}\left[n_{\mathrm{inter}}\left(1+\frac{\omega_{\mathrm{inter}}}{\omega}\right)^{3/2}\left(1-\frac{\Delta_P}{\hbar(\omega+\omega_{\mathrm{inter}})}\right)^{3/2} + (n_{\mathrm{inter}}+1)\left(1-\frac{\omega_{\mathrm{inter}}}{\omega}\right)^{3/2}\left(1-\frac{\Delta_P}{\hbar(\omega-\omega_{\mathrm{inter}})}\right)^{3/2}\right].\right.$$

For ji-scattering all powers are lesser by one unit.

n_j is the phonon distribution function; ρ is the crystal density.

ε_0, ε_∞ are the low- and high-frequency dielectric constants.

where the multiplier $\eta_i(\omega)$ is the same for all scattering mechanisms and at $\hbar\omega < \Delta_X$

$$\eta_i(\omega) = \frac{\hbar\omega}{\Delta_X}\left(1 - \frac{\hbar\omega}{\Delta_X}\right)^{-2}, \tag{7}$$

i.e. in the distance from the band corresponding to the transition $X_1^c \to X_3^c$, $\eta_i(\omega) \sim \omega$ for all scattering mechanisms. Therefore, the presence of this multiplier, reducing by one unit the absolute value of the exponent z for intraband scattering mechanisms, leads to the appearance of non-standard dependence $\alpha(\omega)$. The anisotropy factors L_{ji} for ji-scattering, as follows from the calculations, are characterized by the presence of L_i multipliers identical for all scattering mechanisms

$$L_{ji} = \tfrac{1}{2}\frac{m_\perp}{m}\gamma_j(\beta)\, L_i(\beta, f_X), \tag{8}$$

where

$$L_i^{\parallel} = \beta f_X^{\parallel}, \quad L_i^{\perp} = f_X^{\perp}, \quad f_X = \frac{2\mathscr{P}_X^2}{m\Delta_X}, \tag{9}$$

f_X is the oscillator strength of transition $X_1^c \to X_3^c$; $\mathscr{P}_X$ is the matrix element of transition $X_1^c \to X_3^c$. Numerical estimations of the oscillator strength were carried out on the basis of data from Ref. (7). Form $\gamma_j(\beta)$ is given in Table 1.

From comparison of equations (6)–(9) with Table 1 it is evident that the second components in equations (1)–(3), in contrast to the first ones, have no analogue in the low-frequency processes (Demidenko and Tomchuk[4] show that for the intraband scattering mechanisms in high-frequency and low-frequency cases such an analogy exists and it is so complete that the degree of anisotropy K_j simply coincides). It follows, from equations (8) and (9) that for ji-components $K_{ji} = L_i^{\perp}/L_i^{\parallel}$, i.e. all scattering mechanisms have the same degree of anisotropy. It is precisely this fact which permits the statement that ji-processes are additional scattering mechanisms.

DISCUSSIONS AND CONCLUSIONS

The results of accounting for the non-standard scattering mechanisms mentioned above can be conveniently illustrated by an example of the degree of anisotropy K_j. The calculation of K_j by the formulae given in Table 1 produces $K_j = 2.1$, 2.7 and 6.7 for the acoustic, optical and impurity scattering, respectively; $K_j = 0.055$ and 0.042 for the intervalley and ji-scattering, i.e. for the first three mechanisms $K_j > 1$, and for the latter two $K_j < 1$. It can be seen that the transitions which were accounted for are characterized not only by other frequency dependences, but also by their anisotropy which differs from the standard scattering mechanisms. Therefore, in accordance with equation (5) it follows that the first three mechanisms contribute mainly to $\alpha_\perp$, and the remaining ones to $\alpha_\parallel$, this results in the above-mentioned singularities in dependences $\alpha_0(\omega)$, $\alpha_\parallel(\omega)$ and $\alpha_\perp(\omega)$, thus permitting the calculation to comply with the experiment.

Thus, the experimentally-observed singularities in the process of light absorption by free carriers in n-GaP (differing frequency dependences $\alpha_\parallel$ and $\alpha_\perp$, low value of $\alpha_\perp/\alpha_\parallel$ ratio and unusually low exponent value in dependences $\alpha_\parallel(\omega) \sim \omega^{-z}$, as well as a comparatively low frequency dependence of the absorption coefficient $\alpha_0 \sim \omega^{-(1.4-1.8)}$) can be explained correctly within the framework of the quantum theory of FCA with the intervalley scattering anisotropy and the intermediate states in band Δ_2' taken into account. From this work it also follows that information on the scattering mechanisms obtained from investigations of temperature and field dependences of the kinetic coefficients in a general case can differ from the information obtained from the studies of the spectral dependences $\alpha_{0,\parallel,\perp}(\omega)$ in the quantum frequency region.

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