Oscillating electroreflectance of germanium with allowance for electron-hole Coulomb interaction

O. A. Makarov, N. N. Ovsyuk, and M. P. Sinyukov

Institute of Semiconductor Physics, Siberian Division, USSR Academy of Sciences

(Submitted 15 December 1982)
Zh. Eksp. Teor. Fiz. **84**, 2261–2267 (June 1983)

The nonparabolicity coefficients of the conduction band and of the light-hole band are determined for three directions in the vicinity of $\mathbf{k} = 0$ from the multi-oscillation electroreflectance spectra of germanium by using the theory of Aronov and Ioselevich [Sov. Phys. JETP **47**, 548 (1978); **54**, 181 (1981)]. It is shown that the use of the one-electron approximation over estimates the nonparabolicity coefficients by approximately three times compared with the exact exciton theory. An independent method of determining the band parameters of a semiconductor (the Luttinger parameters), which determine the dispersion law $E(\mathbf{k})$, is proposed. The obtained Luttinger parameters agree well with the values obtained from cyclotron resonance.

PACS numbers: 71.25.Rk, 71.25.Cx, 78.20.Jq

1. INTRODUCTION

The electroreflectance (ER) spectra of semiconductors in a strong electric field should contain an oscillating part connected, in accord with the Keldysh-Franz theory, with the modulation of the reduced density of states beyond the absorption edge. These oscillations are the result of interference between the wave functions of the electrons and holes, which appear in interband optical transitions. Their analysis can yield important information on certain parameters of the solid. Thus, the reduced effective masses were obtained in Ref. 1 for several critical points in the Brillouin zone of germanium, and the nonparabolicity coefficients of the germanium energy bands in the vicinity of the center of the Brillouin zone were determined in Ref. 2 for the [111] direction from the change of the period of the oscillations with changing photon energy. A shortcoming of these studies, however, is the use of the one-electron theory, although the period and amplitude of the oscillations should be influenced by the Coulomb interaction of the excited electron-hole pair. Recently Aronov and Ioselevich[3,4] obtained with the aid of a quasiclassical method the dielectric tensor for semiconductors with degenerate band and for the photon energies above the fundamental absorption edge in an electric field, with account taken of the exciton. This theory makes it possible to calculate the ER spectra more accurately than before.

The present paper is devoted to a determination of the nonparabolicity coefficients of the energy bands of germainium for three direction in the vicinity of $\mathbf{k} = 0$, using a theory that takes into account the Coulomb interaction of the electron and hole. Analytic expressions are obtained that connect the nonparabolicity coefficients with the Luttinger band parameters, and the values of these parameters are obtained.

2. EXPERIMENT

The ER spectra were recorded at room temperature for n-type 30 Ω·cm germanium samples for transitions at the Brillouin center, $\Gamma_8^+ - \Gamma_7^-(E_0)$ and $\Gamma_7^+ - \Gamma_7^-(E_0 + \Delta_0)$, at three orientations of the electric field, [100], [110], and [111]. The experimental procedure was described earlier in Ref. 2.

Figure 1 shows the experimental ER spectra for three electric-field directions. The ER spectrum in a semiconductor with a degenerate valence band is a linear combination of the spectra for the bands of the light and heavy holes. Since the two types of oscillation corresponding to the two types of hole have different periods, their sum contains "beats" in definite sections of the spectrum shown by the arrows in Fig. 1). Owing to degeneracy, the equal-energy surfaces are corrugated near the top of the valence band. The sections of these surfaces are given in many textbooks. Participating in the ER effect are simultaneously two bands between which

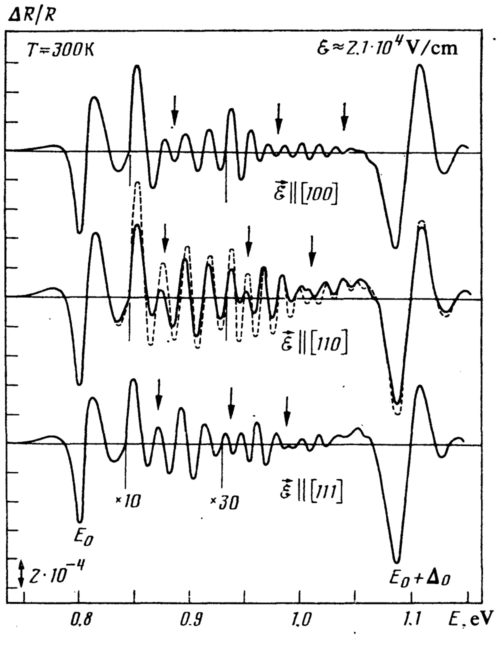

FIG. 1. ER spectra for three electric field directions. The solid line for $\mathscr{E}\|[110]$ shows the spectrum recorded with a light-wave polarization vector $\mathbf{e}\|[001]$, and the dashed—with $\mathbf{e}\|[1\bar{1}0]$. The two other spectra are independent of polarization. The arrows mark the positions of the "beats."

0038-5646/83/061318-04$04.00

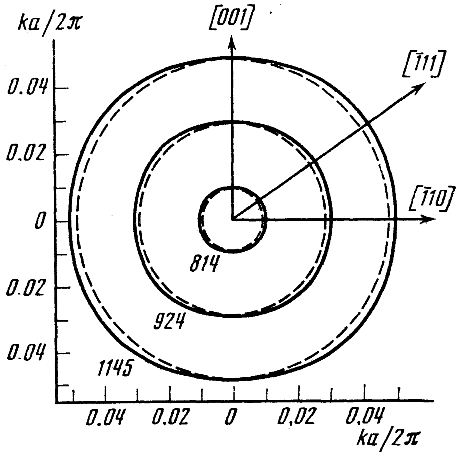

FIG. 2. Sections of equal-energy surfaces for interband optical transitions from the heavy-hole band to the conduction band of germanium. The arrows indicate the directions along which the electric field was applied. The dashed curves shown for comparison are circles. The numbers are the transition energies in MeV.

optical transitions take place. Since the conduction band is spherical and the electron mass is low, the corrugation of the equal-energy surface in the interband transitions becomes much smaller than that of one valence band. Figure 2 shows the intersections of the equal-energy surfaces with the (110) plane for transitions between the heavy-hole band and the conduction band. The distance from the center of the Brillouin band to an equal-energy line is proportional here to the product of the masses of the electron and the heavy hole. Despite the insignificant difference between the equal-energy surfaces in Fig. 2 and the spheres shown dashed in the figure, the ER spectra feel this difference. It can thus be seen from Fig. 1 that more frequent beats are observed for $\vec{\mathscr{E}}\parallel[111]$, for in this direction the ratio of the reduced masses of the heavy and light holes is a maximum. The noticeable approach of the beats at very weak anisotropy of the equal-energy surfaces for interband transitions, is evidence of the high sensitivity of the ER spectra to the values of the reduced effective masses.

3. COMPARISON WITH THEORY

To compare the oscillating part of the ER spectra with theory we used an asymptotic expression[5] that takes into account the band nonparabolicity and takes for the light and heavy holes the form

$$\frac{\Delta R}{R}(\omega, \vec{\mathscr{E}}) \sim (\hbar\omega)^{-2}(\hbar\omega-E_g)^{-1}\exp\left\{-\frac{2(\hbar\omega-E_g)^{1/2}\Gamma}{(\hbar\Theta)^{3/2}} + \frac{4C\mu^2\Gamma}{\hbar^4}\left(\frac{\hbar\omega-E_g}{\hbar\Theta}\right)^{3/2}\right\}\cos\left\{\varphi(\omega, \vec{\mathscr{E}}) + \frac{4}{3}\left(\frac{\hbar\omega-E_g}{\hbar\Theta}\right)^{3/2} \times\left[1-\frac{6}{5}\frac{C\mu^2}{\hbar^4}(\hbar\omega-E_g)\right]\right\}, \tag{1}$$

which is valid at $E \geqslant E_g + 2\hbar\Theta$, where E_g is the energy of the critical point, $\hbar\Theta = (e^2\mathscr{E}^2\hbar^2/2\mu_\parallel)^{1/3}$, $\mu_\parallel$ is the reduced mass of the electron and hole in the field direction, Γ is a phenomenological parameter that takes the scattering into account, $\varphi(\omega,\vec{\mathscr{E}}) = \varphi_{\text{opt}}(\omega) + \varphi_{\text{exc}}(\omega,\vec{\mathscr{E}})$ is a phase that depends on the optical constants of the system and on the electron-hole interaction force, and C is the interband nonparabolicity coefficient defined by the equation

$$E_c(\mathbf{k})-E_v(\mathbf{k})=E_g+(\hbar^2/2\mu)k^2-C(k^2)^2. \tag{2}$$

The phase $\varphi_{\text{opt}}(\omega)$, which depends on the optical constants of the semiconductor and of the surrounding medium, can be taken into account with the aid of the Seraphin coefficients α and β. An expression for the generalized Seraphin coefficients of an air-metal-semiconductor medium was obtained in Ref. 6:

$$\alpha-i\beta=K\exp[i\varphi_{\text{opt}}(\omega)]=-(2n_a n_s D)^{-1}, \tag{3}$$

$$D=\frac{\varepsilon_a-\varepsilon_s}{4\varepsilon_a}\left[1-\frac{4\pi i d_m n_s}{\lambda}\frac{\varepsilon_m-\varepsilon_a}{\varepsilon_s-\varepsilon_a}\right], \tag{4}$$

where ε_s, n_s, ε_a, and n_a are the dielectric constants and the refractive indices of the semiconductor and of the refracting medium respectively; ε_m and d_m are the dielectric constant and the thickness of the sputtered metallic layer. For example, for an aluminum film 40 Å thick and for $\hbar\omega = 0.8$ eV we have $K = 0.005$ and $\varphi_{\text{opt}}(\omega) = -0.3$.

The oscillations of the dielectric constant beyond the absorption edge are due to interference of the part of the electron and hole wave function that diverges from the center and the part reflected from the potential barrier produced by the electric field. It was found that in first-order approximation in the field only a small fraction of the electron-hole pairs that have spread out and then gathered again at a point contributes to the interference. In this case the phase of their wave function is the classical action calculated on the "returning" trajectory and is of the form

$$\Phi_{l,h}(\omega, \vec{\mathscr{E}}) = \frac{4}{3}\left(\frac{\hbar\omega-E_g}{\hbar\Theta_{l,h}}\right)^{3/2} + \frac{2R_{l,h}^{1/2}}{(\hbar\omega-E_g)^{1/2}}\ln 8\left(\frac{\hbar\omega-E_g}{\hbar\Theta_{l,h}}\right)^{3/2} + 2\delta_{l,h}(\hbar\omega-E_g), \tag{5}$$

where $R_{l,h} = \mu_{l,h}e^4/2\hbar\varepsilon_0^2$ is the exciton Rydberg. The first term in (5) is due entirely to the electric field and determines the phase of the oscillations in the one-electron approximation. The second term is due to the lowering of the potential energy of the electron-hole pair on account of the Coulomb interaction, and the last term is a correction that corresponds to the motion of an electron and hole at short distances from each other, where the Coulomb interaction cannot be described quasi-classically. In the absence of degeneracy we have

$$\delta(\hbar\omega-E_g)=\arg\Gamma(1-iR^{1/2}/(\hbar\omega-E_g)^{1/2}),$$

where $\Gamma(x)$ is the gamma function. It can be seen that the influence of the Coulomb interaction on the period of the oscillations manifests itself in the appearance of an additional phase parameter that depends on the energy:

$$\varphi_{\text{exc}\,l,h}(\omega, \vec{\mathscr{E}}) = \frac{2R_{l,h}^{1/2}}{(\hbar\omega-E_g)^{1/2}}\ln 8\left(\frac{\hbar\omega-E_g}{\hbar\Theta_{l,h}}\right)^{3/2} + 2\arg\Gamma\left(1-\frac{iR_{l,h}^{1/2}}{(\hbar\omega-E_g)^{1/2}}\right). \tag{6}$$

TABLE I.

	$p_{1l,\,2l,\,3l}$, eV^{-1}	p_c, eV^{-1}
$\vec{\mathscr{E}} \parallel [100]$	590±20	740±80
$\vec{\mathscr{E}} \parallel [110]$	720±40	
$\vec{\mathscr{E}} \parallel [111]$	820±60	

Allowance for this phase, i.e., allowance for the Coulomb interaction of the electron and hole compared with experiment, leads to deformation of the oscillations and to a decrease of the obtained nonparabolicity coefficients by approximately three times compared with the "one-electron" case.

To determine the nonparabolicity coefficients, we compared the theoretical spectra obtained with the aid of (1) with the experimental ones for three electric-field directions. To decrease the number of independent adjustment parameters, all the spectra were obtained for several values of the field, and the spectra for $\vec{\mathscr{E}} \parallel [110]$, which depend on the polarization of light, were obtained also for different polarizations. A computer was then used to fit simultaneously the theoretical and experimental spectra for equal fields but different polarizations. All the parameters were varied until full coincidence was reached, except for the ratio of the contributions of the heavy- and light-hole bands. The resultant ratios of the contributions were then substituted in the spectra corresponding to the experimental spectra obtained at different fields but with like polarization, and a joint fitting again carried out. The nonparabolicity coefficients turned out to be the same in both cases, thus attesting to the uniqueness of the chosen parameters.

4. DISCUSSION OF RESULTS

The nonparabolicity coefficients obtained with coinciding theoretical and experimental spectra are given in Table I.The remaining parameters are listed in Table II. For all spectra, $E_g = 798$ meV and $\varphi_{\text{opt}} = -0.76$.

It can be seen that the broadening parameter for the light holes must be chosen larger than for the heavy ones. This indicates that the thermalization time is shorter for the light than for the heavy holes.

According to the published data, the nonparabolicity coefficients are most frequently determined with the aid of magneto-optical effects. For the nonparabolicity of the conduction band, magnetoreflectance yielded a value $p_c = (520 \pm 100)$ eV^{-1} (Ref. 7), and magneto-absorption yielded $p_c = (740 \pm 220)$ eV^{-1} (Ref. 8) or $p_c = 600$ eV^{-1} (Ref. 9). It is much more difficult to obtain by these methods information on the nonparabolicity of the light-hole band, since the number of maxima in the magnetoreflectance and magneto-absorption spectra, corresponding to transitions from the light-hole band, is as a rule small because of the large energy intervals between them. The only value of the light-hole-band nonparabolicity coefficient, obtained in a study of magneto-absorption[9] for the [110] direction, was $p_{2l} = 1400$ eV^{-1}. The light-hole-band nonparabolicity was recently measured[10] by another method, using the oscillations of the photoconductivity in a magnetic field under infrared illumination. The nonparabolicity coefficients for three directions, calculated from the presented plot of the mass of the light hole vs energy by using the formula

$$\frac{\partial^2 E_v(\mathbf{k})}{\partial k^2} = \frac{\hbar^2}{m_v^*(E)} \approx \frac{\hbar^2}{m_v^*(0)} - 6p_l\hbar^2 \frac{m_v^*(0)}{m_0^2}(E_v - E_g), \tag{7}$$

were found to be $p_{1l} = 280$ eV^{-1}, $p_{2l} = 420$ eV^{-1}, $p_{3l} = 470$ eV^{-1}. These are lower than calculated with the aid of Eqs. (A3), (A4), and (A5) using the known Luttinger parameters.[11] The nonparabolicity coefficients obtained from these equations were found to be $p_{1l} = (650 \pm 130)$ eV^{-1}, $p_{2l} = (710 \pm 160)$ eV^{-1}, $p_{3l} = (820 \pm 160)$ eV^{-1}. These are close to those obtained by us in experiment.

Knowing the nonparabolicity coefficients for three directions in the Brillouin zone, we can obtain the Luttinger parameters that determine the dispersion $E(\mathbf{k})$ near $\mathbf{k} = 0$, using Eqs. (A3), (A4), and (A5). The obtained values are gathered in Table III. which gives for comparison the Luttinger parameters obtained from cyclotron resonance. The good agreement with the published data indicates that the high sensitivity of the ER oscillations to the values of the effective masses can be used to obtain the Luttinger band parameters by an independent method.

The authors thank V. N. Ovsyuk for a number of helpful remarks.

APPENDIX

We obtain equations that connect the nonparabolicity coefficients with the Luttinger band parameters, and make it possible to determine these parameters from the experimental nonparabolicity coefficients. We denote the nonparabolicity coefficients of the light-hole bands for the directions [100], [110], and [111] by p_{1l}, p_{2l}, and p_{3l}, respectively.

In the nonparabolic case the expansions of the energy in powers of k for the conduction and valence band are of the form

TABLE II.

	$\hbar\Theta_h$, meV	$h\Theta_l$, meV	Γ_h, meV	Γ_l, meV	B
$\vec{\mathscr{E}} \parallel [100]$	17.05	19.70	10	12	0.50
$\vec{\mathscr{E}} \parallel [110]$	16.58	19.90	10	14	0.65 for e ‖ [1$\bar{1}$0] 1.10 for e ‖ [001]
$\vec{\mathscr{E}} \parallel [111]$	16.23	19.77	10	16	0.50

TABLE III.

	Our data	[11]	[12]
γ_1	12.8±2.9	13.1±0.4	13.0±0.2
γ_2	4.0±0.3	4.2±0.6	4.4±0.1
γ_3	5.5±0.3	5.5±0.5	5.3±0.2

$$\begin{aligned} E_c(\mathbf{k}) &= E_g + \frac{\hbar^2 k^2}{2m_c} - p_c\left(\frac{\hbar^2}{2m_0}\right)^2 k^4, \\ E_{vl}(\mathbf{k}) &= -\frac{\hbar^2 k^2}{2m_l} + p_l\left(\frac{\hbar^2}{2m_0}\right)^2 k^4, \\ E_{vh}(\mathbf{k}) &= -\frac{\hbar^2 k^2}{2m_h} + p_h\left(\frac{\hbar^2}{2m_0}\right)^2 k^4. \end{aligned} \tag{A1}$$

The theoretical values of the nonparabolicity coefficients of the valence band for the three directions p_{1l}, p_{2l}, and p_{3l} can be obtained by solving the secular cubic equation that determines the electron energy spectrum $E(k)$.[12,13] This equation is expressed in terms of the parameters L, M, N, and λ, where $\lambda = E(k) - \hbar^2 k^2/2m_0$, and L, M, and N are constants, determined from cyclotron resonance, in terms of which one can express the sums of the products of the matrix elements for direct optical transitions between the valence bands. The band parameters L, M, and N are connected with the Luttinger parameters, which are more convenient for theoretical calculations, by the following relations:

$$\frac{\hbar^2}{2m_0}(\gamma_1 - 1) = \frac{1}{3}(L+2M), \qquad \frac{\hbar^2}{2m_0}\gamma_2 = \frac{1}{6}(L-M),$$

$$\frac{\hbar^2}{2m_0}\gamma_3 = \frac{N}{2}. \tag{A2}$$

The cubic equations for the three directions, and the nonparabolicity coefficients, take respectively the forms:

for the [100 direction]

$$(Lk_x^2-\lambda)(Mk_x^2-\lambda) - \frac{\Delta}{3}[2(Lk_x^2-\lambda)(Mk_x^2-\lambda)+(Mk_x^2-\lambda)^2]=0,$$

$$p_{1l} = \frac{1}{\Delta}\left[\frac{(\gamma_1-1)^2}{4} + (\gamma_1-1)2\gamma_2 + 3\gamma_2^2\right], \qquad p_{1h}=0; \tag{A3}$$

for the [110] direction

$$\{[(L-M)k_x^2-\lambda]^2-(Nk_x)^2\}\left\{(2Mk_x^2-\lambda)-\frac{\Delta}{3}\right\}$$

$$-\frac{2}{3}\Delta[(L+M)k_x^2-\lambda](2Mk_x^2-\lambda)=0,$$

$$p_{2l} = \frac{1}{\Delta}\left[\gamma_2^2+3\gamma_3^2+\frac{\gamma_2(9\gamma_3^2-\gamma_2^2)}{(3\gamma_3^2+\gamma_2^2)^{1/2}}\right], \tag{A4}$$

$$p_{2h} = \frac{1}{\Delta}\left[\gamma_2^2+3\gamma_3^2-\frac{\gamma_2(9\gamma_3^2-\gamma_2^2)}{(3\gamma_3^2+\gamma_2^2)^{1/2}}\right];$$

for the [111] direction

$$[(L+2M)k_x^2-\lambda]^3+2(Nk_x^2)^3-3[(L+2M)k_x^2-\lambda](Nk_x^2)^2$$

$$-\Delta\{[(L+2M)k_x^2-\lambda]^2-(Nk_x^2)^2\}=0,$$

$$p_{3l}=8\gamma_3^2/\Delta, \qquad p_{3h}=0. \tag{A5}$$

The heavy-hole-band nonparabolicity coefficient p_{2h} for the direction [110] is small and is neglected.

[1] D. E. Aspnes, Phys. Rev. Lett. **31**, 230 (1973).

[2] N. N. Ovsyuk and M. P. Sinyukov, Zh. Eksp. Teor. Fiz. **75**, 1075 (1978) [Sov. Phys. JETP **48**, 542 (1978)].

[3] A. G. Aronov and A. S. Ioselevich, Zh. Eksp. Teor. Fiz. **74**, 1043 (1978) [Sov. Phys. JETP **47**, 548 (1978)].

[4] A. G. Aronov and A. S. Ioselevich, Zh. Eksp. Teor. Fiz. **81**, 336 (1981) [Sov. Phys. JETP **54**, 181 (1981)].

[5] D. E. Aspnes, Phys. Rev. **B10**, 4228 (1974).

[6] D. E. Aspnes, J. Opt. Soc. Amer. **63**, 1380 (1973).

[7] R. L. Aggarwal, Phys. Rev. **B2**, 446 (1970).

[8] L. M. Both, B. Lax, and S. Zverdling, Phys. Rev. **114**, 90 (1959).

[9] A. V. Varfolomeev, B. P. Zakharchenya, A. Ya. Ryskin, R. P. Seisyan, and A. L. Éfros, Fiz. Tekh. Poluprov. **11**, 2301 (1977) [Sov. Phys. Semicond. **11**, 1353 (1977)].

[10] V. N. Zverev, Fiz. Tverd. Tela (Leningrad) **22**, 3282 (1980) [Sov. Phys. Solid State **22**, 1921 (1980)].

[11] R. N. Dexter, H. J. Zeiger, and B. Lax, Phys. Rev. **104**, 673 (1956).

[12] C. Dresselhaus, A. Kip, and C. Kittel, Phys. Rev. **98**, 368 (1955).

[13] E. O. Kane, J. Phys. Chem. Solids **1**, 249 (1957).

Translated by J. G. Adashko