Spectrum and polarization of hot-electron photoluminescence in semiconductors

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Experimental and theoretical research on the recombination photoluminescence of free hot electrons in semiconductors (primarily GaAs) is reviewed. The polarization characteristics are discussed. These characteristics reflect, in particular, a momentum alignment of the electrons by linearly polarized light and an effect of a ripple in the constant-energy surfaces in the valence band. The dependence of the linear polarization on the spectrum is discussed in connection with various mechanisms for the energy relaxation of the hot electrons. The depolarization of the hot-electron photoluminescence in a magnetic field is discussed. A procedure is discussed for determining the energy relaxation times and the scale times for intervalley transitions through an analysis of depolarization curves. The energy distribution of the hot electrons is found from the hot-electron photoluminescence spectrum. The recombination luminescence of hot holes is discussed. These holes appear when the semiconductor is illuminated in the spin-split-off subband.

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CONTENTS

1.	Introduction	143
2.	Interband radiative transitions in semiconductors having a band structure of the	
	gallium arsenide type	146
3.	Linear polarization. Optical alignment of electron momenta	147
4.	Circular polarization. Optical alignment of electron spins	148
5.	Effect of the split-off subband	149
6.	Ripple effects	150
7.	Recombination of photoexcited electrons with holes localized at acceptors	153
8.	HEP spectrum and relaxation effects	153
9.	HEP depolarization in a magnetic field. Measurement of relaxation times	155
10	Phonon oscillations in the HEP spectrum	158
11	. Excitation through a side valley	16 0
12	. Reconstruction of the hot-electron-energy distribution from the HEP spectrum	161
13	. Hot-hole photoluminescence	161
14	. Conclusion	164
Ap	opendix. Role of elastic and inelastic collisions in the depolarization of hot-carrier	
ph	otoluminescence	164
Re	ferences	165

1. INTRODUCTION

Interband absorption of light in semiconductors gives rise to photoexcited electrons with an energy $\varepsilon_0 \approx \hbar \omega_{ex}$ $-E_{\rm g}$, where $\hbar \omega_{\rm ex}$ is the energy of the exciting photon, and E_{g} is the gap width. These electrons are then scattered by impurities, majority carriers, and lattice vibrations, losing their initial energy and momentum in the process. The electrons may also recombine with holes. The recombination time is usually several orders of magnitude longer than the scale time for energy and momentum relaxation. The overwhelming majority of the photoexcited electrons are thus thermalized in the conduction band before they undergo recombination. The electrons which are involved in the recombination luminescence are thus primarily thermalized electrons with an energy of the order of the thermal energy, ε $\sim kT$. These are the electrons that determine the greater part of the photoluminescence spectrum during interband transitions and during transitions from the conduction band to an impurity level.

the emitted photon) however, a faint high-frequency tail can be observed in the luminescence spectrum. This tail is produced by the recombination of hot electrons, i.e., electrons which recombine before their energy relaxation is completed. We will refer to this component of the recombination-photoluminescence spectrum as "hot-electron photoluminescence" (HEP). Studies of HEP have recently been carried out in crystals of GaAs, InP, and solid solutions of gallium arsenide.

The particular type of secondary emission with which we will be concerned in this review—the recombination luminescence of free hot electrons in semiconductors is a special case of a broader range of hot-carrier luminescence phenomena in crystals. Here we must not overlook the work by Rebane and Saari,¹ who were the first to observed the faint hot-carrier luminescence lines which correspond to the transitions of electrons in local centers from states which are not at equilibrium with respect to vibrational relaxation. The photoluminescence of hot excitons in semiconductors has been studied studied by Gross, Permogorov, *et al.*²

Under the condition $\hbar\omega - E_g \gg kT$, ($\hbar\omega$ is the energy of

Ivchenko, Pikus, and Takunov³ have derived a theory for the alignment and orientation of hot excitons in semiconductors, and the scattering of hot excitons by longitudinal optical phonons has been studied by Aristova et al.⁴ Ulbrich⁵ has studied the recombination luminescence of electrons heated by an electric field. We would also like to mention Betzler's detection of a very faint recombination luminescence of hot electrons which are produced during Auger processes in silicon.⁶ With regard to the spectrum of edge (interband) photoluminescence, we note that Shah and Leite⁷ and Meneses etal.⁸ have shown that the short-wave part of the spectrum in GaAs and CdS crystals can be described well by a dependence $\exp(-\hbar\omega/kT_{e})$, where the electron temperature $T_{\rm e}$ depends on the excitation intensity and is much higher than the temperature of the crystal lattice at high intensities. This means that in Refs. 7 and 8 a Maxwellian distribution was established for electrons and holes generally not at thermal equilibrium with the lattice. This conclusion applies to the bulk of the photo-excited carriers, which have already undergone energy relaxation and which are localized near the band edges, at distances $\sim kT_{e}$. The electron thermalization occurs through electron-electron collisions.

In the energy range with which we will be concerned here, which corresponds to hot electrons which have not undergone energy relaxation, the energy distribution is very non-Maxwellian. The HEP spectrum furnishes a sort of time sweep of the energy relaxation of the hot electrons, and a study of HEP can yield some interesting information about this relaxation. Research on HEP has revealed a momentum alignment of the electrons by linearly polarized light, which leads, in particular, to a linear polarization of the HEP. It turns out that the frequency dependence of the HEP polarization characteristics contains information about the energy and momentum relaxation.

From the HEP depolarization in an external magnetic field it is possible to determine the energy relaxation time of the hot electrons for various scattering mechanisms. In particular, this method has made it possible to measure the probability for the emission of an optical phonon by an electron in GaAs. The hot-electron distribution function can be constructed from the HEP spectrum. Our purpose in this review is to examine the results of this work.

Research on HEP was begun by Zakharchenya, Zemskii, and Mirlin,⁹ who studied p-GaAs crystals and crystals of solid solutions containing gallium arsenide (GaAs-AlAs and GaP-GaAs). The luminescence was excited with the beam from a He-Ne laser ($\hbar \omega_{ex} = 1.96$ eV), and the sample temperature was approximately 100 K. The experimental procedure in that and subsequent studies was similar to the procedure used to study the Raman effect in opaque crystals. The luminescence intensity spectrum $I(\hbar \omega)$ and the polarization characteristics were studied in Ref. 9. Measurements were carried out for several samples with various acceptor concentrations in the range $N_A \sim 10^{17} - 10^{19}$ cm⁻³. The HEP intensity was proportional to the excitation intensity over the entire spectrum; in p-type samples this result corresponds to the recombination of hot electrons with equilibrium holes in the valence band or at acceptor levels.

Under the experimental conditions of Ref. 9 the amount by which the excitation energy exceeded the gap width, $\hbar \omega_{ex} - E_{g}$, was 0.45 eV in the case of GaAs; correspondingly, the energy of the electrons at the time at which they were produced was quite high ($\varepsilon_0 \approx 0.4 \text{ eV}$). It was thus possible to follow the changes in the HEP characteristics over a rather large energy range. The primary results of Ref. 9, which stimulated further research on HEP, were the discovery of anomalously high degrees of circular polarization, $\rho_{\rm e}$ (with excitation by circularly polarized light), and the discovery of a linear polarization of the recombination emission in the hot region (with excitation by linearly polarized light). The latter result was unexpected, since a linear polarization of the recombination luminescence of free electrons in direct-gap cubic crystals (and a dichroism in interband absorption) had previously been observed only in an external electric field¹ (Refs. 10-12).

The spectrum and polarization characteristics shown for the recombination photoluminescence in Fig. 1 are typical of p-GaAs at $N_{\rm A} \sim 10^{18}$. There is a broad region of HEP on the high-frequency side of the peak corresponding to edge (thermalized) luminescence, above 1.6 eV. The HEP intensity is many orders of magnitude lower than the intensity at the peak of the edge band, even in the lowest-frequency part, next to the edge band. Also shown in Fig. 1 are the spectra of $\rho_c(\hbar \omega)$ and $\rho_i(\hbar \omega)$. We see that ρ_c is approximately equal to 0.45 at the time at which electrons are produced at the energy ε_0 . The appearance of circular polarization in the recombination-luminescence spectrum obtained with circularly polarized excitation is caused by an optical orientation of the electron spins by the circularly polarized light.¹⁴ The maximum possible theoretical value of ρ_c for the recombination luminescence of thermalized electrons in crystals with the GaAs band structure is 0.25, as follows from the selection rules for transitions between atomic levels with total angular momenta of 3/2 and 1/2 (Ref. 15). The same value can be found for interband recombination by taking an average over all the directions of the quasimomenta in the Brillouin zone.¹⁶ The values observed for ρ_c in Ref. 9 were thus anomalously high.

The frequency dependence of ρ_c is complicated, with two structural features: a minimum near 1.75 eV and a threshold near 1.6 eV. These features were attributed in Ref. 9 to an onset of transitions from a lighthole (lh) subband and a spin-split-off subband (the transition scheme is shown in Fig. 2). The reason why the

¹⁾In indirect-gap semiconductors, with several equivalent minima in the conduction band, the recombination emission may acquire a linear polarization because of a preferential filling of one of the minima of the conduction band by linearly polarized light. This effect has been observed in PbTe crystals, and a selective filling of valleys during indirect optical transitions has also been observed in other semiconductors.¹³



FIG. 1. The spectrum, $I(\hbar\omega)$, and the polarization characteristics of hot-electron photoluminescence. $\rho_{\rm c}$ —Degree of circular polarization (with circularly polarized excitation); $\rho_{\rm l}$ —degree of linear polarization (with linearly polarized excitation). p-GaAs sample, $N_{\rm A}^{\rm (Ge)} = 6 \cdot 10^{17} {\rm cm}^{-3}$; (100) plane of the sample; $T = 80 {\rm K}$.

structural features in the $\rho_{\rm c}(\hbar\omega)$ spectrum are of this nature is that upon excitation from these subbands electrons are produced with a spin orientation opposite to that which occurs when electrons are scattered out of a heavy-hole subband.^{16,17}

In the hot part of the spectrum, as we mentioned earlier, there is also a significant linear polarization in the case of linearly polarized exciting light (at the time of production, ρ_1 in Fig. 1 is approximately 0.25). As the relaxation of the electron energy proceeds, this linear polarization decreases. In the region of thermalized luminescence we find $\rho_1 = 0$ (in contrast with ρ_c).

As Dymnikov *et al.*¹⁷ have shown, linear polarization appears in the HEP spectrum, and the values of ρ_c are anomalously high, because the momentum distribution of the photoexcited electrons is anisotropic in the case of interband absorption of light in semiconductors having the GaAs band structure. For excitation from the heavy-hole subband, for example, the momenta of the



FIG. 2. The GaAs band structure near the center of the Brillouin zone; transition scheme. Arrows point upward—excitation into the conduction band (c) from the heavy-hole subband (hh), from the light-hole subband (Ih), and from the split-off subband (sh); arrows pointing downward—recombination to acceptor levels (A).

carriers which are produced are directed preferentially perpendicular to the electric vector (e), or polarization, of the exciting light. In excitation from the light-hole subband, in contrast, the momenta of the carriers which are produced are parallel or antiparallel to e. Since the selection rules in radiative recombination are the same as those for photoexcitation, the anisotropy of the momentum distribution causes a linear polarization of the photoluminescence. As was shown in Ref. 17, the appearance of an anisotropy in the momentum distribution also leads to anomalously high values of the degree of circular polarization of the HEP. As the electrons undergo energy relaxation, their momentum distribution tends to become isotropic. Correspondingly, linear polarization disappears from the recombinationluminescence spectrum, and circular polarization decreases.

Proceeding in the spirit of the correspondence principle, we will devote the first few sections (Secs. 2-5) of this review to interband radiative transitions in semiconductors of the gallium arsenide type. We will discuss the polarization characteristics, which reflect the optical momentum alignment of the electrons and the optical spin alignment. A ripple of the constant-energy surfaces in the heavy-hole subband has a strong influence on these effects (Sec. 6). At low temperatures the HEP results from band-acceptor transitions, and its polarization reflects the distribution among subbands and the quasimomentum distribution of the holes at the acceptors (Sec. 7). The variation of the linear polarization over the spectrum is determined by the relationship between the energy relaxation time and the momentum relaxation time. At a high hole concentration the hot electrons start losing most of their energy through collisions with heavy holes, accompanied by a conversion of the latter into light holes (Sec. 8). In moderately doped crystals $(N_A < 3 \cdot 10^{17} \text{ cm}^{-3})$ the hot electrons lose their energy primarily through the emission of LO phonons, as can be seen from the phonon oscillations in the HEP spectrum (Sec. 10). The imposition of a magnetic field results in a depolarization of the HEP, because the anisotropic part of the momentum distribution rotates. The scale time for the energy relaxation of the hot electrons can be determined from an analysis of the depolarization curves $\rho_1(H)$. This will be done in Sec. 9 for the case in which the energy loss results from collisions of hot electrons with neutral acceptors. In crystals which are doped more lightly, this approach yields the scale time for the emission of the LO phonons and that for the $\Gamma - L$ intervalley transition (Sec. 10). At high excitation frequencies, the photoelectrons from the central (Γ) minimum of the conduction band move to side valleys relatively rapidly, then, after reaching the bottom of the lowest side valley (L) as a result of several intervalley transitions, they end up back at the central minimum (Sec. 11). This pumping method (through the L band) has proved convenient for reconstructing the energy distribution of the hot electrons from the HEP spectrum and for analyzing this distribution (Sec. 12).

Finally, Sec. 13 deals with research on the photoluminescence of hot holes in n-GaAs crystals. The HEP spectrum in this case results from the radiative recombination of holes which are produced by the light in a split-off subband (Γ_7 ; see Fig. 2) with electrons from the conduction band, The HEP spectrum consists of a single band at $E_g + \Delta$ (1.85 eV). The hole lifetime in the Γ_7 band is short because of the relatively high probability for transitions between subbands of the valence band; as a result, the degree of circular polarization is approximately unity. In other words, there is a nearly total optical orientation of the holes in the case of circularly polarized excitation. The hole lifetime in the Γ_7 band can be determined directly from the depolarization of the luminescence in a transverse magnetic field.

In describing the HEP polarization, we denote the degree of linear polarization by ρ_1 and that of circular polarization by ρ_e . In addition, for a complete description of the polarization properties of the luminescence we will use the Stokes parameters ξ_1 , ξ_2 , ξ_3 , where $\xi_3 = \rho_1$ and $\xi_2 = \rho_e$ (see footnote 4 for more details).

Most of the experimental results which we will be discussing here were obtained with GaAs crystals. Figure 2 is a schematic reproduction of the GaAs band structure near the center of the Brillouin zone. The energy gaps are $E_{\rm g} \approx 1.5$ eV and $\Delta \approx 0.34$ eV. The effective masses of the electrons $(m_{\rm e})$ and the light holes $(m_{\rm 1h})$ and the average mass of the heavy holes $(m_{\rm hh})$ are approximately $0.07m_o$, $0.08m_o$, and $0.6m_o$, respectively.

In the analysis of the experimental data below we will use more accurate values for the band parameters, which reflect, in particular, the temperature dependence of $E_{\rm g}$, the ripple of the valence band, and the deviation from parabolic bands.

2. INTERBAND RADIATIVE TRANSITIONS IN SEMICONDUCTORS HAVING A BAND STRUCTURE OF THE GALLIUM ARSENIDE TYPE

The correspondence principle¹⁸ generates the most graphic description of direct interband transitions. In accordance with this principle, a classical dipole with a natural vibration frequency $\omega_{ab} = (E_a - E_b)/\hbar$ is asso-

TABLE I. Matrix elements of the dipole moment D_{vc}/D for various interband transitions $(\lambda, \mu, \text{ and } \nu \text{ are unit vectors}; \nu$ is directed along the momentum, while λ and μ are in the perpendicular plane).



ciated with each transition between two quantum states b and a. The amplitude of the dipole moment of this dipole is equal to the transition matrix element:

$$\mathbf{D}_{ab} = \langle a \mid \hat{\mathbf{D}} \mid b \rangle, \tag{1}$$

where \hat{D} is the dipole moment operator. The intensity and the polarization of the emission and absorption of light upon a quantum transition correspond completely (in the dipole approximation) to the intensity and polarization upon emission and absorption of light by the classical dipole in (1).

To what type of dipoles do the interband optical transitions in direct-gap cubic $A^{III}B^v$ semiconductors correspond? The bottom of the conduction band in these semiconductors is twofold degenerate and belongs to the Γ_6 representation. The valence band consists of two twofold-degenerate subbands²⁾ of heavy and light holes, and the corresponding wave functions at the center of the Brillouin zone transform by the Γ_3 representation. Below lies the so-called split-off subband (symmetry Γ_7 ; see Fig. 2).

The structural analysis of the states corresponding to the band edges¹⁹ can be simplifed substantially by using the so-called spherical approximation, in which the cubic symmetry of the crystal is replaced by a spherical symmetry. The effects of the real cubic symmetry are not small, and they will be discussed below (Sec. 6). but the spherical approximation is a useful starting point. In this approximation the conduction band can be characterized by orbital angular momentum l=0 and spin s = 1/2. The wave functions corresponding to the bottom of the band may be written

$$u_{1/2}^{(c)} = S \uparrow \quad u_{-1/2}^{(c)} = S \downarrow,$$
 (2)

where S is a function of the coordinates (in the spherical approximation, the boundary of the unit cell of the crystal is assumed to be a sphere, and the function S is spherically symmetric). The two spin directions are indicated by the arrows.

The valence band is characterized by orbital angular momentum l=1 and spin s=1/2, so that the total angular momentum has two possible values: J=1/2 and J=3/2. The value J=3/2 corresponds to the heavy-hole and light-hole subbands, while the value J=1/2 corresponds to the subband split off by the spin-orbit interaction. The corresponding wave functions for zero quasimomentum can be written easily by making use of the standard rules for combining angular momenta. For the subband of heavy holes (hh), which have angularmomentum projections $M_J = \pm 3/2$ onto the wave vector, these wave functions are

$$u_{3/2}^{(n)} = -\frac{1}{\sqrt{2}} (X + iY)^{\dagger}, \quad u_{-3/2}^{(n)} = \frac{1}{\sqrt{2}} (X - iY)^{\dagger}.$$
(3)

For the subband of light holes (lh), with $M_J = \pm 1/2$, we have

$$u_{1/2}^{(v)} = \frac{1}{\sqrt{3}} \left[-\frac{1}{\sqrt{2}} \left(X + iY \right) \downarrow + \sqrt{2}Z \uparrow \right],$$

²⁾Here and below, we are ignoring effects which result from the absence of a center of inversion in these semiconductors.

$$u_{-1/2}^{(v)} = \frac{1}{\sqrt{3}} \left[\frac{1}{\sqrt{2}} \left(X - iY \right) \dagger + \sqrt{2} Z_{\frac{1}{2}} \right].$$
 (4)

For the split-off subband (sh), the wave functions are

$$u_{1/2}^{(i)} = -\frac{1}{\sqrt{3}} \left[(X+iY) \downarrow + Z \uparrow \right], \quad u_{-1/2}^{(i)} = \frac{1}{\sqrt{3}} \left[Z \downarrow - (X-iY) \uparrow \right]; \quad (5)$$

where X, Y, Z are functions of the coordinates which transform as the coordinates x, y, z, respectively, under rotations. The subscripts on the wave functions are the projections of the angular momentum onto the momentum p. For the four degenerate states at the top of the valence band (p=0) the quantization axis can be chosen arbitrarily, but at nonzero momenta the degeneracy is lifted, and the quantization ax is (the z ax is) must be chosen along the momentum in order to construct the correct wave functions. We will thus assume that the zaxis is directed along the momentum, while the x and y axes lie in the plane perpendicular to the momentum (although this point is inconsequential for the conduction band and for the split-off subband, since for these bands the degeneracy at nonzero \mathbf{p} is the same as that at $\mathbf{p} = 0$). The wave functions in (2)-(5) are the Bloch amplitudes; the complete wave functions differ from these by a factor $\exp(i\mathbf{pr}/\hbar)$.

We can now write down the dipole moments which correspond to transitions between the different subbands of the valence band and the conduction band.²⁰ Here we should bear in mind that (1) the spin wave functions (indicated by the arrows) are orthogonal for opposite directions and are normalized, (2) momentum is conserved in the transition (this is the dipole approximation), and (3) the only nonvanishing matrix elements are

$$\langle S \mid \mathbf{D}_x \mid X \rangle = \langle S \mid \mathbf{D}_y \mid Y \rangle = \langle S \mid \mathbf{D}_z \mid Z \rangle = D.$$

Table I shows the matrix elements of the dipole moment vector constructed in this manner (see also Fig. 3). We can draw some conclusions from this table. The hh-c transitions correspond to two dipoles which are rotating clockwise and counterclockwise in the plane perpendicular to the momentum. The oscillations of these dipoles are 100% polarized perpendicular to the momentum. The lh-c transitions correspond to two dipoles which are oscillating parallel to the momentum and two which are rotating in the perpendicular plane. The resultant oscillation is 60% polarized parallel to the momentum. The sh-c transitions correspond to four dipoles (as do the lh-c transitions), but the resultant oscillation is not polarized.



FIG. 3. Dipoles corresponding to various interband transitions. The arrows representing the dipoles are labeled with the relative intensities of the transitions. The levels are labeled with the projections of the total angular momentum onto the quantization axis, which is directed along the momentum.

These results apply to those cases in which the momenta of the electrons participating in the transitions are not very high, specifically, when the kinetic energy of the holes is small in comparison with the spin-orbit splitting Δ . In the opposite case, the wave functions "get mixed up". The corresponding changes will be discussed below (Sec. 5); at the point we will assume that the momenta are small in the sense just defined.

3. LINEAR POLARIZATION. OPTICAL ALIGNMENT OF ELECTRON MOMENTA

It is clear from the discussion above that linearly polarized light excites charge carriers with an anisotropic momentum distribution. In hh-c transitions, for example, no electrons are excited with momenta parallel to e (the polarization unit vector of the light wave), since both oscillators corresponding to hh-c transitions are polarized perpendicular to the momentum. The momentum distribution of the photoexcited electrons, $F(\mathbf{p})$, at the time at which the electrons are produced is proportional to the sum of the squares of the projections onto the e direction for all the dipoles which correspond to the given type of transition. For hh-c transitions, for example, we find

$$F(\mathbf{p}) \sim 2 \cdot \frac{1}{2} \left[(\lambda \mathbf{e})^2 + (\mu \mathbf{e})^2 \right] = 1 - (\mathbf{v} \mathbf{e})^2 = \sin^2 \vartheta$$

where ϑ is the angle between **e** and **p**. In this manner we obtain results for all three transitions; these results may be written in the form¹⁷

$$F(\mathbf{p}) = F_0(\mathbf{p}) \left[1 + \alpha_0 P_2(\cos \vartheta) \right], \quad P_2(\mathbf{x}) = \frac{3x^2 - 1}{2} , \tag{6}$$

where $P_2(\cos \vartheta)$ is the second Legendre polynomial; $\alpha_0 = -1, +1, 0$ for hh-c, lh-c, sh-c transitions, respectively; and $F_0(p)$ is the symmetric part of the distribution function. Figure 4 shows the shape of the distribution function for the various transitions. We see that in the case of excitation from the heavy-hole subband the distribution of the photoexcited electrons is flattened along the polarization direction of the exciting light, while in the case of excitation from the light-hole subband the distribution is stretched out in this direction. This effect has been labeled the "optical alignment of electron





momenta." One of the most interesting manifestations of this effect is polarization of the HEP.

Let us examine the hot-carrier luminescence in ptype semiconductors. We may assume that at moderate excitation intensities the luminescence results from the recombination of photoexcited electrons with equilibrium holes. Since the holes are distributed isotropically, we may assume that a radiating dipole—one of the dipoles shown in Fig. 3—corresponds to each electron with momentum p. The relative intensity I_{\bullet} , (p) of the emission with polarization e' for a given momentum direction is expressed by an equation similar to (6):

$$I_{\mathbf{e}'}(\mathbf{p}) \sim 1 + \alpha_0 P_2(\cos \vartheta'), \tag{7}$$

where α_0 has the same value as in Eq. (6), and ϑ' is the angle between the electron momentum and the polarization vector e'. The total emission intensity with a given polarization e' is proportional to the product of (7) and the electron distribution $f(\mathbf{p})$, integrated over all angles of the vector \mathbf{p} :

$$I_{\mathbf{e}'} = \left\{ \left[1 + \alpha_0 P_2 \left(\cos \vartheta' \right) \right] f(\mathbf{p}) \, \mathrm{d}\Omega. \right.$$
(8)

The polarized component of the emission intensity thus contains information about the anisotropy of the distribution, which is described by the second Legendre polynomial.

In the case of excitation by linearly polarized light, the electrons, which had the distribution in (6) at the time they were produced and have "descended" to an energy ε , acquire the distribution

$$f = f_0(\varepsilon) \left[1 + \alpha P_2(\cos \vartheta) \right], \tag{9}$$

for which the anisotropy parameter is α (and depends on the energy ε). Substitution of (9) into (8) yields an expression for the intensity of the recombination radiation by the electrons with energy ε :

$$I_{e'} \sim 1 + \frac{\alpha a_0}{5} P_2(\cos \chi), \tag{10}$$

where χ is the angle between the polarization vector of the exciting radiation (e) and that of the luminescence (e').

We find the degree of polarization of the light emitted parallel or antiparallel to the exciting light to be

$$\rho_{1} = \frac{I_{1} - I_{\perp}}{I_{1} + I_{\perp}} = \frac{3\alpha\alpha_{0}}{20 + \alpha\alpha_{0}};$$
(11)

where I_{μ} is the intensity of the luminescence which has the same polarization as the exciting light ($\chi = 0$), and

TABLE II. Initial values of the linear polarization ρl of the recombination luminescence for various excitation and recombination pathways (spherical approximation). The numbers in parentheses are the polarization for the case $p^2 \gg m_{1h}\Delta$ (Sec. 5).

Recombination	c — hh	c – 1h	c — sh
Excitation hh-c lh-c sh-c	1/7 (1/7) 3/19 (1/7) 0 (1/3)	$\begin{array}{c} -3/19 (1/7) \\ 1/7 (1/7) \\ 0 (-1/3) \end{array}$	0 (-1/3) 0 (-1/3) 0 (1/2)



FIG. 5. Shape of the momentum distribution function of the photoexcited electrons for excitation by unpolarized (or circularly polarized) light.²¹ a—For hh-c transitions; b—for lh-c transitions. In both cases the distribution is symmetric with respect to the direction of the exciting beam (the vector n). The three-dimensional shape of the distribution can be found by rotating the distribution shown here around the vector n. The lengths of the vectors indicate the populations of the states having the corresponding momentum directions.

 I_{\perp} is the intensity of the luminescence which is polarized in the perpendicular direction $(\chi = \pi/2)$. In particular, for the luminescence which results from the recombination of electrons which have not yet undergone relaxation ($\alpha = \alpha_0$) we find the numbers shown in Table II for various types of excitation and recombination.

Under the experimental conditions corresponding to the results in Fig. 1, the high-frequency luminescence edge is shaped by transitions to acceptor levels. The selection rules for recombination to an acceptor are the same as those for c-hh transitions, since at high momenta the wave function of a hole at an acceptor is formed primarily from the heavy-hole states (Sec. 7). The initial value of ρ_1 found experimentally (about 0.25) is higher than that which follows from Table II for the hh-c-hh transitions (~0.14), because of a ripple of the constant-energy surfaces for the heavy holes (Sec. 6).

To conclude this section we note that for excitation by unpolarized or circularly polarized light the distribution function of the photoexcited electrons is also anisotropic (with a symmetry axis along the excitation direction). A momentum alignment (Fig. 5) occurs because of the transverse nature of the light wave. The distribution can be found by adding the two versions of function (6) corresponding to two mutually perpendicular directions of the polarization. We find

$$F = 2F_0 \left[1 - \frac{\alpha_0}{2} P_2 \left(\cos \theta \right) \right], \tag{12}$$

where θ is the angle between the momentum and the beam of exciting light. This distribution leads to polarization of the light which is emitted at an angle with respect to the exciting beam.

4. CIRCULAR POLARIZATION. OPTICAL ALIGNMENT OF ELECTRON SPINS

In the case of interband absorption of circularly polarized light, the photoexcited electrons have their spins aligned. It can be seen from Fig. 3 and Table I that the electrons whose momenta were directed along the exciting beam at the time of excitation turn out to have a 100% spin orientation for each type of excitation (the orientation is opposite to the photon angular mo-



FIG. 6. Relationship between the spin and momentum of an electron for excitation by circularly polarized light.²¹ a— For hh-c transitions; b—for lh-c transitions. The momenta are directed along the radii of a circle. The vectors which start at points on the circle indicate the average spins corresponding to the given momentum directions.

mentum in the case of hh-c transitions and along the photon angular momentum in the case of lh-c and sh-c transitions). If, on the other hand, the momentum of the electron makes some angle with the exciting beam, then it will have a different spin orientation. Figure 6 shows the mutual orientations of the momentum and the spin for the hh-c and lh-c transitions. In the case of the sh-c transitions, the electron spin is always directed along the photon angular momentum, and there is a 100% alignment. The following expression may be derived for the electron density matrix at the time at which the electrons are produced¹⁷:

$$\hat{F} = F_0 \left\{ \hat{I} \left[1 - \alpha_0 \frac{3 \left(\mathbf{n} \mathbf{v} \right)^2 - 1}{4} \right] + 2S_0 \sigma \mathbf{n} + \beta_0 \frac{3 \left(\sigma \mathbf{v} \right) \left(\mathbf{n} \mathbf{v} \right) - \left(\sigma \mathbf{n} \right)}{2} \right\}, \quad (13)$$

where \hat{I} is a 2×2 unit matrix, σ are the Pauli matrices, and n and ν are unit vectors along the direction of the photon angular momentum and along the momentum direction, respectively. The anisotropy parameters α_0 , β_0 , and S_0 are given in Table II. The parameter α_0 is a measure of the momentum alignment; S_0n gives the spin vector, averaged over all momentum directions; and β_0 is a measure of the correlation between the spin and the momentum. Relaxation leaves the density matrix in the same form as (13), but with anisotropy parameters α , β , and S which are smaller in magnitude than α_0 , β_0 , and S_0 . In particular, if the momentum distribution has become completely isotropic, while the spin has not undergone relaxation, we find a degree of orientation $2S_0 = -0.5$ for hh-c and lh-c transitions. This result was believed to be the limiting value for optical orientation involving these transitions.^{14,16} It leads to a maximum circular polarization of -0.25%. As a result of the correlation between the spin and the momentum, the circular polarization of the HEP is increased. From the expression for the density matrix in (13) with the anisotropy parameters α , β , and S, and from the selection rules in Table I, we find the

TABLE III. Initial values of the anisotropy parameters of the density matrix of the photoexcited electrons in the case of circularly polarized excitation.

Transition	αο	S_0	βa
$\frac{hh-c}{lh-c}$ $\frac{sh-c}{sh-c}$	-1	1/4	1
	1	1/4	1
	0	1/2	0

TABLE IV. Initial values of the degree of circular polarization, $\rho_{\rm c}$, of the recombination luminescence for various excitation and recombination pathways (spherical approximation). The numbers in parentheses are the polarizations for the case $p^2 \gg m_{\rm 1h}\Delta$ (Sec. 5).

Recombination	c – hh	c - 1h	c – sh
Excitation hh - c lh - c sh - c	5/7 (5/7) 5/19 (5/7) 1/2 (0)	5/19 (5/7) 5/7 (5/7) 1/2 (0)	-1/2 (0) -1/2 (0) 1 (0)

following expression¹⁷ for the degree of circular polarization, ρ_c :

$$\rho_c = \frac{80SS_0 + 10\beta\beta_0}{20 + \alpha\alpha_0}.$$
 (14)

In particular, for the recombination luminescence of electrons which have not yet undergone relaxation ($\alpha = \alpha_0, \beta = \beta_0, S = S_0$) we find the values of ρ_c shown in Table IV (a value $\rho_c > 0$ means that the angular momentum of a photon in the luminescence is directed preferentially in the same direction as in the exciting beam).

Table IV yields the value $\rho_c = 5/7 \approx 0.71$ for the initial degree of circular polarization for the case of hh-c-hh transitions (which determine the high-frequency HEP edge). The experimental value from Fig. 1, $\rho_c = 0.47$, is smaller (but much larger than the maximum value for the case of an isotropic electron momentum distribution: $\rho_c = 0.25$). At this point the reason for this discrepancy is not clear. One possibility is rapid spin relaxation of the hot electrons by the precession mechanism proposed by D'yakonov and Perel'.¹⁶

5. EFFECT OF THE SPLIT-OFF SUBBAND

If the energy of the photoexcited holes becomes comparable to the spin-orbit splitting Δ , the selection rules for interband transitions are changed. These changes are irrelevant for hh-c transitions, since the heavyhole subband is not coupled with the split-off subband.²² The energy spectrum and wave functions of the lighthole subband, however, undergo important changes at $\varepsilon_{1b} \ge \Delta$ (Fig. 2). The light-hole subband "gets heavier" and at $\varepsilon_{1h} \gg \Delta$ it runs parallel to the heavy-hole subband. In the limit of high hole energies, the role of the spin-orbit interaction lessens in importance. If it is ignored, then the merged subbands of heavy and "light" holes can be characterized by a projection ± 1 of the orbital angular momentum onto the momentum, while the split-off subband can be characterized by a projection 0.

We will now reproduce some equations from Ref. 23 which describe the anisotropy parameters of the density matrix (13) for the photoexcited electrons at the time of production. These equations incorporate the interaction of the light-hole subband with the split-off subband in the spherical approximation.

For hh-c transitions, the equations remain the same:

$$\alpha_0 = -1$$
, $S_0 = -\frac{1}{4}$, $\beta_0 = -1$.

For lh-c transitions, they are

$$\alpha_{0} = \frac{\varepsilon_{\rm sh} - (A+5B) p^{2}}{\varepsilon_{\rm sh} - \varepsilon_{\rm lh}}, \quad S_{0} = -\frac{1}{4} + \frac{3}{4} \frac{(A+B) p^{2} - \varepsilon_{\rm lh}}{\varepsilon_{\rm sh} - \varepsilon_{\rm lh}},$$
$$\beta_{0} = \frac{\varepsilon_{\rm sh} + (B-A) p^{2}}{\varepsilon_{\rm sh} - \varepsilon_{\rm lh}}. \quad (15)$$

If $\varepsilon_{1h} \gg \Delta$, then $\alpha_0 = -1$, $S_0 = 1/4$, and $\beta_0 = 1$. For sh-c transitions these equations are

$$\begin{aligned} \alpha_0 = \frac{(A+5B)\,p^2 - \varepsilon_{1h}}{\varepsilon_{sh} - \varepsilon_{1h}}, \quad S_0 = -\frac{1}{4} + \frac{3}{4}\,\frac{\varepsilon_{sh} - (A+B)\,p^2}{\varepsilon_{sh} - \varepsilon_{1h}}; \\ \beta_0 = \frac{(A-B)\,p^2 - \varepsilon_{1h}}{\varepsilon_{sh} - \varepsilon_{1h}}. \end{aligned}$$

If $\varepsilon_{1h} \gg \Delta$, then $\alpha_0 - 2$, $S_0 - 0$, and $\beta_0 - 0$.

The constants A and B in these equations determine the hole spectrum: $\varepsilon_{hh} = (A - B)p^2$,

$$\varepsilon_{\mathbf{l}\mathbf{b},\,\mathbf{s}\mathbf{b}} = \left(A + \frac{B}{2}\right)p^2 + \frac{\Delta}{2} \mp \frac{1}{2}\sqrt{9B^2p^4 - 2B\Delta p^2 + \Delta^2}.\tag{17}$$

The hole energies are assumed to be positive and are measured downward from the top of the valence band. The constants A and B are related to the hole masses in the limit $\mathbf{p} \rightarrow 0$ in the following way:

$$A = \frac{1}{4} \frac{m_{hh} + m_{hh}}{m_{hh} m_{hh}}, \quad B = \frac{1}{4} \frac{m_{hh} - m_{hh}}{m_{hh} m_{hh}}.$$
 (18)

As before, the degree of polarization of the recombination luminescence is given by Eqs. (11) and (14). Tables II and IV give (in parentheses) the degree of the initial linear and circular polarization in the limit of large quasimomenta, $\varepsilon_{1h} \gg \Delta$.

The weakening of the spin-orbit interaction causes the resultant degree of circular polarization to approach zero for the cases of hh-c and lh-c excitation. The reason is that in the absence of a spin-orbit interaction there is no optical orientation of the electrons, and the angular momentum of the photon is transferred entirely to a hole upon absorption.

The linear polarization is related to the momentum alignment of the electrons. With the spin-orbit interaction turned off, the excitation from the heavy- and lighthole subbands results in the same alignment ($\alpha_0 = -1$) and thus an identical linear polarization of the luminescence. In the case of excitation from the split-off subband, an alignment occurs [in the limit $\varepsilon_{1b} \gg \Delta$, we find $\alpha_0 = 2$ from (16)], since the split-off subband now corresponds to a dipole directed along the momentum. Figure 7 shows the interband transition scheme in the limit $\varepsilon_{1h} \gg \Delta$ along with a picture of the alignment corresponding to these transitions. In an analysis of experimental data it is important to take into account the deviation from a parabolic light-hole subband for a correct determination of the energy of the electrons excited from this subband; Eqs. (15) and the data in Tables II-IV are important for correctly finding the polarization caused by these electrons. For example, in the case of the excitation of HEP in gallium arsenide by photons with an energy $\hbar \omega_{ex} = 1.96$ eV, the electrons excited from the light-hole subband have an energy ε'_0 -0.26 eV and begin to contribute to the emission at 1.75 eV. Figure 1 reveals a structural feature at this energy on the curve for the circular polarization; specifically, $\rho_{\rm c}$ changes sign. The linear polarization, $\rho_{\rm l}$, on the other hand, has no such structural feature. This circumstance is puzzling from the standpoint of a theory which does not incorporate the effect of the split-off



FIG. 7. Dipoles corresponding to various interband transitions in the limit $\varepsilon_{lh} \gg 0$; shape of the momentum distribution of the photoexcited electrons for excitation by linearly polarized light. The levels are labeled with the projections of the orbital angular momentum onto the quantization axis, which is directed along the momentum.

subband. The structural feature on the $\rho_c(\hbar \omega)$ curve can be explained on the basis that for lh-c-hh transitions the sign of ρ_c is opposite to that for hh-c-hh transitions (Table IV), and the electrons which have been scattered out of the heavy-hole subband and which have descended to an energy ε'_0 have already undergone a partial momentum relaxation, and their contribution to the polarization decreases. The resultant polarization thus changes sign. The very same arguments would seem to apply to the linear polarization (Table II). However, when we take the split-off subband into account we find that the transitions lh-c-hh and hh-c-hh have opposite circular polarizations but linear polarizations of the same sign if the momenta of the recombining electrons are sufficiently high (see the numbers in parentheses in Tables II and IV). The same is true in the case of recombination to acceptor levels. Dymnikov et al.²³ calculated the polarization for lh-c-A transitions for the experimental conditions (Fig. 1). It turns out that while we have $\rho_{e} = -0.57$ for these transitions the linear polarization is positive and given by $\rho_1 = 0.05$; this value is approximately equal to the linear polarization of the electrons which have been excited from the heavy-hole subband and which have undergone energy relaxation. The incorporation of transitions from the light-hole subband thus has essentially no effect on the spectrum $\rho_1(\hbar\omega)$ (Fig. 1).

As another example we might cite the small value of ρ_c (0.08) which is observed in InP crystals near the high-frequency HEP edge²³ (1.9 eV). The value of Δ in InP is comparatively low (~0.1 eV), and at this energy we are essentially dealing with the limiting case $p^2 \gg m_{1h}\Delta$. In this case, the contributions of the transitions lh-c-hh and hh-c-hh to the recombination are apparently summed; the values of ρ_c corresponding to these two types of transitions are identical in magnitude but opposite in sign. This "cancellation" is responsible for the small resultant values of the circular polarization. The linear polarization has a magnitude of 0.14, in agreement with the theoretical value (Table II).

6. RIPPLE EFFECTS

What effect does the actual cubic symmetry of the crystal have on the HEP polarization?^{25, 26} This effect

results primarily from the ripple of the constant-energy surfaces of the valence band, i.e., from the dependence of the effective masses of the light and heavy holes on their quasimomentum direction. The ripple is greatest in the heavy-hole subband. In gallium arsenide, for example, the effective mass of the heavy holes in the [111] direction is $m_{\rm hh}^{[111]} = 0.8m_{\rm o}$, while that in the [100] direction is $m_{\rm hh}^{[100]} = 0.4m_{\rm o}$. Let us examine the consequences of this difference. The high-frequency edge of the HEP spectrum is shaped by transitions back into the heavy-hole subband of electrons which have been excited from this subband; after they return, they recombine with equilibrium holes. Holes with momentum directions along the {111} directions are predominant in the recombination under these conditions, since the population is greatest along these directions. As a measure of the preference for population of the {111} directions over the $\{100\}$ directions we may adopt the parameter $\Delta \varepsilon_{\rm wh}/kT$, where

$$\Delta \varepsilon_{\rm vh} = m_{\rm c} \left(\frac{1}{m_{\rm hh}^{(100]} + m_{\rm c}} - \frac{1}{m_{\rm hh}^{(111]} + m_{\rm c}} \right) (\hbar \omega_{\rm ex} - E_{\rm g}).$$
(19)

The meaning of $\Delta \varepsilon_{\rm vh}$ is clear from the transition scheme scheme in Fig. 8. At $\varepsilon \Delta_{\rm vh} \gg kT$, the recombination luminescence is produced primarily by carriers with momenta in the {111} directions. We will call these the "diagonal" directions. Because of the ripple, the electrons which appear in the conduction band upon monochromatic excitation as a result of hh-c transitions no longer have a monoenergetic distribution and are instead spread out over a band of width $\Delta \varepsilon_{\rm vh}$. In GaAs, this width is $\Delta \varepsilon_{\rm vh} \approx 0.07(\hbar \omega_{\rm ex} - E_{\rm g})$, so that at $\hbar \omega_{\rm ex} = 1.96$ eV we have $\Delta \varepsilon_{\rm vh} \approx 30$ MeV.

When the ripple is taken into account, the matrix elements for hh-c transitions are determined by equations more complicated than those given in Table I. It is important to note, on the other hand, that the equations in Table I remain valid for momenta directed along the $\{111\}$ directions. Accordingly, if it is assumed that the luminescence results exclusively from carriers with momenta along diagonal directions, then we can use Eqs. (8) and (9) to calculate the intensity I_{\bullet} , of the luminescence with a given polarization, except that the integration over angles is replaced by a summation over the four diagonal directions. If, for example, the exciting beam is directed along the [100] axis, and the polarization vector of the exciting light, e, and that of the luminescence, e', lie in the (100) plane, making angles φ and φ' , respectively, with the [001] direction, then the angles over which the summation is to be carried out can be determined from the expressions

$$\begin{aligned} \cos^2\vartheta_4 &= \cos^2\vartheta_2 = \frac{2}{3}\cos^2\left(\varphi - \frac{\pi}{4}\right),\\ \cos^2\vartheta_3 &= \cos^2\vartheta_4 = \frac{2}{3}\sin^2\left(\varphi - \frac{\pi}{4}\right). \end{aligned}$$

The angles ϑ'_i differ from ϑ_i in that φ is replaced by φ' . After the integration is replaced by the summation, Eqs. (8) and (9) yield the following intensity for c-hh transitions:



. .

FIG. 8. Diagram used to help explain the meaning of $\Delta \varepsilon_{vh}$. Shown here are the conduction band and the heavy-hole subband for two momentum directions, [100] and [111]. The length of the arrows corresponds to the excitation energy. At $\hbar\omega = 1.96$ eV, the value of $\Delta \varepsilon_{vh}$ in GaAs is about 30 meV.

$$I_{e'} = 1 - \frac{\alpha}{\lambda} \sin 2\varphi \cdot \sin 2\varphi'.$$
⁽²⁰⁾

It can be seen from Eq. (20) that with hh-c excitations ($\alpha < 0$) the maximum intensity corresponds to the luminescence polarized along [011] or [011], depending on whether sin 2φ is positive or negative, i.e., depending on which of these directions is closer to the vector e. It can also be seen that if $\varphi = 0$ or π , i.e., if the vector e is along [001] or [010], then the luminescence is unpolarized. The reason for this result can be seen in Fig. 9a. At these directions of e all the diagonals are equivalent and populated identically, just as if the distribution function were isotropic. The maximum degree of polarization of the luminescence should be observed at $\varphi = \pm \pi/4$ according to (20); i.e., it should be observed when e is directed along [011] or [011] (Fig. 9b).

If the polarization properties of the luminescence are characterized by the Stokes parameters⁴⁾ ξ_1 , ξ_2 , ξ_3 , then the following expressions²⁵ are found from (20) for this particular geometry (with the exciting beam along the [100] direction):

$$\xi_1 = -\frac{\alpha}{4}\sin 2q\cos 2\varphi, \quad \xi_3 = -\frac{\alpha}{4}\sin^2 2\varphi.$$
 (21)

Hence the maximum degree of polarization for a given value of φ is

$$l = \left| \frac{\alpha}{4} \sin 2\varphi \right|$$
 (22)

and it reaches its maximum value at $\varphi = \pm \pi/4$, when it is 0.25 for c-hh recombination of electrons which have not undergone relaxation ($\alpha = \alpha_0 = -1$). This is the geometry which corresponds to the data in Fig. 1. We

a.p

³⁾These values were calculated from the band parameters of GaAs found through a study of the cyclotron resonance.²⁴

⁴⁾We recall that the parameter ξ_3 gives the degree of linear polarization in the selected system of axes. In the present case, these axes are chosen along **e** and along the vector \mathbf{e}_1 , which is perpendicular to **e** and to the light beam so that we have $\xi_3 = \rho_1 = (I_n - I_1)/(I_n + I_1)$, where $I_{n,1}$ are the intensities of the luminescence polarized parallel and perpendicular, respectively, to the polarization of the exciting light, **e**. The Stokes parameter ξ_1 is the degree of linear polarization in a coordinate system rotated through $\pi/4$ with respect to the original system. The parameter ξ_2 is a measure of the degree of circular polarization $(\xi_2 = \rho_c)$. We have the relations²⁷ $\xi_1 = l \sin 2\psi$, $\xi_3 = l \cos 2\psi$, where l is the maximum polarization, and ψ is the angle between the corresponding direction and the vector **e**.



FIG. 9. Two versions of the orientation of the polarization vector \mathbf{e} of the exciting light with respect to the diagonal directions in a cubic lattice. $\mathbf{a} - \mathbf{e} \parallel [001]$. All the diagonals are oriented in equivalent manners with respect to \mathbf{e} and are therefore filled identically ($\rho_1 = 0$); $\mathbf{b} - \mathbf{e} \parallel [011]$. The vector \mathbf{e} lies in the plane which contains two of the diagonals and is perpendicular to the plane containing the two other diagonals. This, the most anisotropic orientation of \mathbf{e} , corresponds to the maximum linear polarization.

see that the initial value of the linear polarization is approximately equal to the theoretical value.

The Stokes parameters for other directions of the exciting beam can be found in a similar way. We will write out the results for the case in which the exciting beam is incident normally on the (111) plane (the "beam-along-[111] geometry"). In this case there is no anisotropy of the polarization. The Stokes parameters for c-hh transitions are

$$\xi_1 = 0, \quad \xi_3 = (-\alpha) \cdot \frac{2}{13}.$$
 (23)

(22)

These parameters differ only slightly from the corresponding values of the spherical approximation ($\xi_1 = 0$, $\xi_3 = -\alpha/7$). Figure 10 shows the angular characteristics of the degree of linear polarization (ξ_3) constructed from Eqs. (21) and (23).

Figure 11 shows the experimental dependence of the luminescence intensity I_{\bullet} , on the orientation of the polarization plane of the analyzer for the beam-along-[100] geometry at 100 K for three values of φ (Ref. 26). These results agree well with Eq. (20) with $\alpha = -1$.

As the temperature is raised, and kT becomes comparable to $\Delta \varepsilon_{vh}$, electrons and holes with all quasimomentum directions make some contribution to the recombination in the case of hh-c-hh transitions. In the calculation of the Stokes parameters, the summation over the diagonal directions must be replaced by an integration over all quasimomentum directions, with allowance for the populations corresponding to these directions. Figure 12 (the solid curves) shows results calculated for $\xi_a(\varphi)$ for 300 K from the equations de-



FIG. 10. Angular characteristics of the degree of linear polarization, $\xi_3(\varphi)$ [Eqs. (21) and (23)]. Here φ is the angle between the polarization vector of the exciting light, e, and the principal crystallographic axis which lies in the surface plane of the sample. a—Beam along [100], $\xi_3 = \frac{1}{4} \sin^2 2\varphi$; b—beam along [111], $\xi_3 = \text{const} = 0.154$.



FIG. 11. Experimental dependence of the luminescence intensity (I_{\bullet} .) on the orientation of the polarization plane of the analyzer for the beam-along-[100] geometry and for three values of the angle φ (Ref. 26). $1-\varphi = -5^{\circ}$; $2-15^{\circ}$; $3-45^{\circ}$ (Ref. 26). T=100 K, GaAs(Zn) sample, $N_{\rm A}-N_{\rm D}=2\cdot10^{19}$ cm⁻³, $\hbar\omega=1.93$ eV.

rived in Ref. 25, for the beam-along-[111] geometry. Also shown in this figure are experimental results for 300 K for $\hbar \omega = 1.93$ eV, in which case the HEP spectrum is shaped by interband transitions ($\hbar \omega_{ex} = 1.96 \text{ eV}$). The nature (the symmetry) of the experimental angular dependence corresponds to the theory, but, as can be seen from this figure, the measured values of $\xi_3(\varphi)$ for the (100) plane are more anisotropic than the calculated values and correspond better to low temperatures than to room temperature. Special measurements of the polarization anisotropy carried out by Mirlin and Reshina²⁶ have shown that it varies much more slowly with the temperature than would be expected on the basis of the theory of Ref. 25. The reasons for this discrepancy have yet to be explained. Interestingly, even under the assumption $T \rightarrow \infty$, i.e., even if the diagonal directions are not special directions in terms of the population, the polarization should remain substantially anisotropic (10.5% and 17% for $\varphi = 0$ and $\pi/4$, respectively), because of an anisotropy of the transition matrix elements. The ripple of the constant-energy surfaces in the gallium arsenide valence band thus makes the HEP linear polarization dependent on the orientation of the electric vector of the exciting light with respect to the crystallographic axes. To conclude this section, we note that the ripple should not have any important effect on the circular polarization of the HEP (with excitation by circularly polarized light).²⁵ For the beam-along-[100], -[110], and -[111] situations we would have ξ_2 = 3/4, 12/17, and 9/13, respectively (hh-c-hh transitions). We recall that the spherical model predicts ξ_2 = 5/7.



FIG. 12. Angular characteristics of the degree of linear polarization $\xi_3(\varphi)$, for T=300 K. a—Beam along [100]; b beam along [111]. Solid curves—calculated for T=300 K; points—experimental data for a GaAs(Zn) sample. $a-N_A-N_D$ $= 2 \cdot 10^{18}$ cm⁻³; $b-N_A-N_D=2 \cdot 10^{18}$ cm⁻³. T=300 K, $\hbar\omega=1.93$ eV.

7. RECOMBINATION OF PHOTOEXCITED ELECTRONS WITH HOLES LOCALIZED AT ACCEPTORS

At low temperatures in moderately doped p-type crystals, the equilibrium holes are localized at acceptors. In this case the HEP spectrum is shaped by transitions from the conduction band to acceptor levels (c-A transitions). Figure 13 shows the spectrum of the HEP intensity near the excitation line ($\hbar \omega_{ex} = 1.96 \text{ eV}$) for the same sample as in Fig. 1, at temperatures of 300 and 100 K. On the curve of I($\hbar \omega$) for T = 100 K there is a clearly defined dropoff, whose position (about 1.88 eV) corresponds to the recombination of the most energetic photoelectrons to acceptor levels.²⁸

It would seem that the anisotropy of the polarization which results from the ripple of the constant-energy surfaces of the heavy holes should be far less apparent in c-A transitions than in interband transitions: In transitions to acceptor levels, no role should be played by the special nature of the diagonal momentum directions (they are special because they are energetically preferred for holes; Sec. 6). However, an experiment carried out at 30 K, at which all the holes are clearly frozen at acceptors, showed²⁸ that the linear polarization remains highly anisotropic. The effect turns out to result from a pronounced anisotropy of the quasimomentum distribution of the holes at acceptors. The wave function (ψ_{a}) of a hole at a shallow acceptor is constructed from the free-hole wave functions $\psi_{an}(r)$:

$$\psi_{M}(\mathbf{r}) = \sum_{nn} C_{nM}(\mathbf{p}) \psi_{\mathbf{p}n}(\mathbf{r}); \qquad (24)$$

here $M = \pm 3/2$, $\pm 1/2$ specifies the four degenerate states of the hole at an acceptor. The index *n* takes on four values, which correspond to the twofold-degenerate subbands of heavy (hh) and light (lh) holes. The quantity

$$\mathfrak{D}_{n}\left(\mathbf{p}\right) = \sum_{\mathbf{M}} |C_{n\mathbf{M}}\left(\mathbf{p}\right)|^{2}$$
(25)

is a measure of the distribution among subbands and the quasimomentum distribution of the holes bound to acceptors.⁵⁾ Solution of the Schrödinger equation for the acceptor in the momentum representation, with allowance for the ripple, shows that at high quasimomenta, $p \gg \hbar/a$ (a is the acceptor radius) we have

$$\Phi_n(\mathbf{p}) \sim \frac{1}{p^{4} e_n^2(\mathbf{p})} = \frac{4m_n^2}{p^8},$$
(26)

where m_n is the mass in the corresponding subband, which depends on the direction of p. Equation (26) shows that at the high quasimomenta which are important for the radiative recombination of hot electrons the distribution of holes at acceptors is dominated by heavy holes with diagonal momentum directions, which correspond to the maximum mass. The special nature of the diagonal holes does not depend on the temperature in this case, however, since it is not related to the difference between the populations of different energy states.

At the highest-frequency HEP edge, the special posi-



FIG. 13. High-frequency edge of the HEP spectrum for excitation with $\hbar \omega_{ex} = 1.96 \text{ eV}$ (Ref. 28). $\mathbf{a} - T = 100 \text{ K}$. The slope change corresonds to recombination of the highest-energy photoelectrons to acceptor levels; $\mathbf{b} - T = 300 \text{ K}$. The HEP spectrum ranges up to $\hbar \omega_{ex}$. The GaAs(Ge) sample here was the same as in Fig. 1. The peak LO corresponds to Raman scattering of light by a longitudinal optical phonon.

tion of the diagonal directions and the associated anisotropy of the HEP polarization should become even more prominent, because (Sec. 6) the maximum energy ϵ_o is acquired during photoexcitation by those electrons which are scattered into the conduction band from diagonal directions in the valence band.

The measurements whose results are shown in Fig. 1 were carried out in the beam-along-[100] geometry and under conditions such that the HEP spectrum was dominated by transitions from acceptor levels (Fig. 13). The experimental values of the degree of linear polarization at the high-frequency edge of the spectrum turn out to be highest in the case $e \parallel [011]$ and approximately equal to the calculated value (0.25) (Fig. 1). As shown in Ref. 28, the minimum value is (in accordance with the calculations) approximately zero in the case $e \parallel [100]$.

8. HEP SPECTRUM AND RELAXATION EFFECTS

The energy relaxation of the photoexcited electrons may result from the emission of phonons and also from collisions with other electrons and with holes. In ptype crystals with a sufficiently high doping level, p>10¹⁸ cm⁻³, the primary energy-loss mechanism is that involving collisions of electrons with holes, either free or localized at acceptors.⁶ The energy loss due to the emission of optical phonons is of minor importance at such hole concentrations. The energy loss due to colli-

⁵⁾It is assumed that all four degenerate states of the acceptor are filled identically.

⁶⁾It is assumed that the excitation intensity and, correspondingly, the concentration of photoexcited electrons are not very high, so that the energy relaxation due to electronelectron collisions may be ignored. This simplification is valid for the papers considered in the present review.

sions of hot electrons with holes may be comparatively slight, and at any rate it may not occur in strictly fixed increments (in contrast with the energy loss resulting from the emission of optical phonons). The HEP spectrum in this case is therefore a continuous, structureless spectrum, as it is in Fig. 1, where it is shown for liquid nitrogen temperature. The spectrum is of the same nature at liquid-helium temperatures. The particular shape of the spectrum is determined by the distribution functions of the hot electrons and holes and also by the frequency dependence of the transition probability. Knowing this probability, we can reconstruct the hot-electron distribution function from the HEP spectrum, as we will see in Sec. 12. In moderately doped crystals $(N_A \sim 10^{17} \text{ cm}^{-3})$ at low temperatures (T < 30 K), there is a clearly defined structure in the HEP spectra: oscillations with a period equal to the energy of long-wave longitudinal optical (LO) phonons. The emission of LO phonons evidently becomes the primary mechanism for energy relaxation in this case. The spectra of this type will be described in more detail in Sec. 10.

As the photoexcited electrons undergo energy relaxation, they also undergo momentum and spin relaxation.⁷⁾ The result is a decrease in the degree of polarization with decreasing electron energy ε and, correspondingly, with decreasing photon energy in the HEP spectrum. The coefficient α , which is a measure of the degree of anisotropy of the momentum distribution in Eq. (9) and thus a measure of the linear polarization [Eq. (11)], decreases with decreasing energy. The dependence of α on the energy ε can be determined from the simple equations¹⁷

$$\frac{d\alpha}{dt} = -\frac{1}{\tau_{\mathbf{p}_{\mathbf{i}}}} \alpha, \quad \frac{d\varepsilon}{dt} = -\frac{1}{\tau_{\varepsilon}} \varepsilon, \qquad (27)$$

where τ_c is the energy relaxation time, and τ_{p_2} is the relaxation time for the anisotropy associated with the second Legendre polynomial. For small-angle scattering, such as that in the case of the scattering of holes or impurities by a Coulomb potential, τ_{p_2} can easily be expressed in terms of the ordinary momentum relaxation time τ_{p} :

$$\frac{\tau_p}{\tau_{p_3}} = \lim_{\vartheta \to 0} \frac{1 - P_{\vartheta}(\cos \vartheta)}{1 - \cos \vartheta} = 3.$$

From (27) we find⁸⁾

$$\alpha = \alpha_0 \exp\Big(-\int_{\varepsilon}^{\varepsilon_0} \frac{\tau_e}{\tau_{p_s}} \frac{\mathrm{d}\epsilon}{\varepsilon}\Big).$$
 (28)

In the case of high acceptor concentrations, in which the relaxation is dominated by an interaction with holes or charged centers, the ratio τ_c/τ_{p_2} is independent of the energy with logarithmic accuracy. From (28) we then find

$$\frac{\alpha}{\alpha_0} = \left(\frac{\varepsilon}{\varepsilon_0}\right)^{r_c/r_{p_2}}.$$
(29)

The spectrum of the degree of linear polarization of the HEP is essentially always identical to the energy dependence of α [see Eq. (11)], since the energy of the photon which is emitted in direct interband transitions is unambiguously related to the energy of the recombining electron. For C-hh transitions, for example, we have

$$\hbar\omega = E_{g} + \varepsilon \left(1 + \frac{m_{\rm C}}{m_{\rm hh}}\right).$$

In a collision of a light particle (an electron with mass $m_{\rm c}$) with heavy particles (heavy holes with mass $m_{\rm hh}$) the electron loses energy more slowly than it loses momentum,⁹⁾ by a factor of $m_{\rm hh}/m_{\rm C}$. Since we have $m_{\rm C} = 0.07m_0$ in GaAs, and the average mass of the heavy holes is $m_{\rm hh} = 0.6m_0$, we conclude that $\tau_{\rm c}$ should exceed $\tau_{\rm p_2}$ by about an order of magnitude. The ratio $\tau_{\rm c}/\tau_{\rm p_2}$ turns out to be even larger (about 20) if we take into account the fact that the time $\tau_{\rm p_2}$ (but not $\tau_{\rm c}$) is reduced further by collisions with impurities. Such a large value of $\tau_{\rm c}/\tau_{\rm p_2}$ would correspond to a far more rapid polarization decay than is actually observed. The experimental results for $\rho_1(\hbar\omega)$ in this case can be approximated well by Eq. (29) with $\tau_{\rm c}/\tau_{\rm p_2} \approx 5$.

This question was taken up in Ref. 30, where an effective mechanism was proposed for the energy relaxation of hot electrons in p-type semiconductors. It was shown in that paper than the primary mechanism for energy transfer from fast electrons to holes is that of collisions of an electron with heavy holes, accompanied by the conversion of the latter into light holes (i.e., by the scattering of heavy holes into the light subband). The electron energy relaxation which results from this mechanism turns out to be the same as that which would occur in the case of ordinary Coulomb collisions with particles having the mass of the light hole (m_{1b}) and a concentration equal to half that of the heavy holes. This mechanism is effective for electrons with an energy ε $>(m_{\rm hb}/m_{\rm lb})kT$. Because of this mechanism, the energy relaxation time of the fast electrons turns out to be comparable in magnitude to the momentum relaxation time. The picture is similar when the holes are localized at acceptors, i.e., when electrons collide with neutral acceptors. If the electron momentum satisfies $p \gg \sqrt{m_{\rm hh} \varepsilon_{\rm A}}$, where $\varepsilon_{\rm A}$ is the acceptor ionization energy, the hole bound at the acceptor may be treated as free.

According to the estimate in Ref. 30, the proposed mechanism for the electron energy relaxation becomes more effective than the mechanism involving the emission of optical phonons at a hole concentration exceeding $2 \cdot 10^{17}$ cm⁻³ in gallium arsenide. Taking into account only those collisions which are accompanied by interband transitions of holes, we find $\tau_c/\tau_{p_2} \approx 1.5$. When we also take into account ordinary collisions with

⁷⁾Here and below we are ignoring absolutely elastic collisions. This question will be taken up in the Appendix.

⁸⁾A more rigorous analysis, based on the solution of the kinetic equation,²⁹ shows that τ_{ε} in (27) and (28) should be replaced by the time τ'_{ε} , which incorporates the correlation between the change in the energy and that in the momentum during the collision. For the case of interest, however, of small-angle Coulomb scattering, we have $\tau_{\varepsilon} = \tau'_{\varepsilon}$.

⁹⁾Collisions of electrons with light holes may be ignored since their concentration is lower by a factor of $(m_{\rm hh}/m_{\rm lh})^{3/2}$ than that of heavy holes.



FIG. 14. Dependence of the degree of linear polarization of the HEP on the energy of the recombining electron. Circles experimental data, for the p-GaAs sample of Fig. 1; curve calculated from Eq. (19) from Ref. 30.

holes and with ionized acceptors, we find

$$\frac{\tau_{\rm e}}{\tau_{\rm p_{\rm f}}} = 6 \frac{m_{\rm h}(m_{\rm h})}{m_{\rm C}(m_{\rm h}) + m_{\rm h}}$$

from which we find $\tau_c/\tau_{p_2} \approx 6$ for GaAs (for $m_c = 0.07m_0$, $m_{1h} = 0.085m_0$, and $m_{hh} = 0.6m_0$). This value must be reduced slightly because of the deviation from a parabolic conduction band, since the increase in the mass of the electrons along with the increase in their energy leads to a decrease in τ_c . The final theoretical dependence in Fig. 14 (the solid curve) agrees quite well with the experimental data found from the linear-polarization spectrum in Fig. 1.

9. HEP DEPOLARIZATION IN A MAGNETIC FIELD. MEASUREMENT OF RELAXATION TIMES

In a magnetic field H the Lorentz force rotates the momenta of the photoexcited electrons. The HEP polarization is accordingly changed, unless the field is directed along the symmetry axis of the electron momentum distribution. The simplest case is that in which the magnetic field is directed along the light beam. In this case the symmetry axis of the distribution, which is directed along the polarization vector **e** of the exciting light at the time of excitation, is rotated around H, remaining perpendicular to it at all times. The angular rotation frequency is the cyclotron frequency:

$$\omega_c = \frac{eH}{m_C^c} \,. \tag{30}$$

Let us assume that the HEP photon energy corresponds to the recombination of electrons with an energy ε . The momentum-direction distribution for these electrons is rotated (with respect to e) through an angle $\psi_H(\varepsilon, \varepsilon_0) = \omega_c \tau(\varepsilon, \varepsilon_0)$, where $\tau(\varepsilon, \varepsilon_0)$ is the time over which the electrons descend from their original energy ε_0 to the energy ε . This rotation may be observed if the angle ψ_H is not too small. If $\tau(\varepsilon, \varepsilon_0) \sim 10^{-12} - 10^{-13}$ s, then the condition $\omega_c \tau \sim 1$ with $m_c \sim 0.1 m_0$ holds in fields H $\sim 10^4 - 10^5$ Oe. Experimentally, the effect of a magnetic field on the HEP polarization becomes reliably detectable at $\omega_c \tau \sim 0.3$.

Let us express the Stokes parameters of the HEP in terms of the rotation angle³¹ $\omega_{c}\tau(\varepsilon,\varepsilon_{0})$. In the beamalong-[100] geometry for C-hh transitions it is a good approximation to use Eq. (20), in which the replacement $\varphi - \varphi + \omega_{c}\tau(\varepsilon,\varepsilon_{0})$ should be made. We then find the following expression for the intensity of the luminescence with polarization e':

$$I_{e'} \sim 1 + \left(-\frac{\alpha}{4}\right) \sin\left(2\varphi + 2\omega_c \tau\left(\varepsilon, \varepsilon_0\right)\right) \cdot \sin 2\varphi', \tag{31}$$

where α is the anisotropy parameter of the distribution function (this parameter, we recall, is negative for hh-C excitation), and φ and φ' are the angles made by the vectors e and e' with the [001] axis of the crystal.

In the coordinate system tied to the polarization vector of the exciting beam, we find from (31) the following expressions for the Stokes parameters¹⁰⁾:

$$\xi_1 = l \cos 2\varphi, \quad \xi_3 = l \sin 2\varphi,$$
 (32)

$$l = \left(-\frac{\alpha}{4}\right) \sin\left[2\varphi + 2\omega_{\rm c}\tau\left(\varepsilon, \varepsilon_{\rm 0}\right)\right]. \tag{33}$$

It follows from these equations that again in a magnetic field, despite the rotation of the distribution function, the direction of maximum polarization is along the [011] or $[0\overline{11}]$ axis, as would be expected for this geometry according to Sec. 6. The degree of maximum polar-ization (*l*), on the other hand, oscillates with the magnetic field, reaching a maximum whenever the symmetry axis of the distribution crosses the [011] or $[0\overline{11}]$ direction.

For the beam-along-[111] geometry (for C-hh transitions), the Stokes parameters are³¹

$$\begin{aligned} \xi_1 (H) &= \xi_3 (0) \sin 2\omega_c \tau (\varepsilon, \varepsilon_0), \\ \xi_3 (H) &= \xi_3 (0) \cos 2\omega_c \tau (\varepsilon, \varepsilon_0), \end{aligned} \tag{34}$$

where $\xi_3(0) = -2\alpha/13$. [The Stokes parameters are of the same form in the spherical approximation, except that $\xi_0(0) = -\alpha/7$. We again see that the ripple has almost no effect in the beam-along-[111] geometry.] In this case, on the other hand, the direction of the maximum polarization rotates around the magnetic field along with the distribution function, while the magnitude of the maximum polarization remains constant.

Equations (32)-(35) hold only when the descent time $\tau(\varepsilon, \varepsilon_0)$ is a strictly determined quantity for all the electrons. This is approximately the situation when an electron loses energy in small portions, and the interval between ε_0 and ε contains a large number (N) of such portions. Although the emission time for each portion fluctuates, the fluctuations $\Delta \tau$ in the total time $\tau(\varepsilon, \varepsilon_0)$ are small ($\Delta \tau/\tau \sim 1\sqrt{N}$). Under these conditions, the descent time can be expressed in terms of the energy relaxation time τ_{ε} by making use of the second equation in (27):

$$\tau(\varepsilon, \varepsilon_0) = \int_{\varepsilon}^{\varepsilon_0} \tau_{\varepsilon'} \frac{d\varepsilon'}{\varepsilon'}.$$
 (36)

Even if $\Delta \tau / \tau$ is small, the phase coherence of the oscillations should be lost in strong magnetic fields, such that $\omega_c \Delta \tau \gtrsim 1$, so that these oscillations will be damped. This question has been studied in detail elsewhere.³¹

Near the excitation line, when the difference $\varepsilon_0 - \varepsilon$ is smaller than or comparable to the characteristic energy lost in a collision, the fluctuations in the descent

¹⁰⁾See the definition of the Stokes parameters in footnote 4).

time are large, and the nature of the magnetic depolarization changes. In this case, $\tau(\varepsilon, \varepsilon_0)$ represents the time spent by an electron in the interval between ε and ε_0 , and in the limit $\varepsilon - \varepsilon_0$ it becomes simply the time spent by the electron in its initial state, τ .

The probability that the time au' lies in the interval d au' is

$$W(\tau') d\tau' = e^{-\tau'/\tau} \frac{d\tau'}{\tau}, \qquad (37)$$

where τ is the average time spent by an electron in the state with initial energy ε_0 (the time required for it to leave its initial state). An expression describing the behavior of the magnetic depolarization in this case can be derived by averaging Eqs. (32)-(35) over the various realizations of the time $\tau(\varepsilon, \varepsilon_0) = \tau'$ with the probability in (37). We then find the following expression for the maximum degree of polarization, to replace (33), in the beam-along-[100] geometry:

$$l = \left(-\frac{\alpha}{4}\right) \frac{\sin 2\varphi + 2\omega_{\rm c}\tau \cos 2\varphi}{1 + 4\omega_{\rm c}^{\rm c}\tau^{\rm s}}.$$
(38)

In the beam-along-[111] geometry (and also for the spherical model), we find instead of Eqs. (34) and (35) the following expressions for the Stokes parameters:

$$\xi_1(H) = \xi_3(0) \frac{2\omega_c \tau}{1 + 4\omega_c^2 \tau^2},$$
(39)

$$\xi_3(H) = \xi_3(0) \frac{1}{1 + 4\omega_2^2 \tau^2}.$$
(40)

Equations (39) and (40) are completely analogous to the equations describing the magnetic depolarization of the atoms of a gas¹⁵ which results from the rotation of the atomic moment in a magnetic field (the Hanle effect). It follows from these equations that the maximum rotation of the polarization plane (as $\omega_c \tau \rightarrow \infty$) is 45°. The effect of a magnetic field on the HEP polarization was first studied experimentally in Ref. 32. Detailed experiments on the depolarization of the HEP in magnetic fields up to 70 kOe were carried out in Ref. 33. The experimental arrangement is shown in Fig. 15. The sample (1) and a spherical mirror (2) are placed inside a superconducting solenoid (3). The beam from the exciting laser is directed along the axis of the solenoid (this is the Faraday geometry) and is focused onto the surface of the sample by a long-focal-length objective (not shown in Fig. 15). The light emitted from the sample is collected by the spherical mirror (2), directed



FIG. 15. Diagram of the liquid-helium cryostat with a superconducting solenoid for measurements of the magnetic depolarization.³³ The sample (1) and the spherical mirror (2) which collects the luminescence are inside the solenoid (3). The arrows show ray directions.



FIG. 16. Dependence of the Stokes parameters ξ_1 and ξ_3 on the magnetic field.³³ a—Beam along [100], $\mathbf{e} \parallel [011]$, GaAs(Zn) sample, $N_{\mathbf{a}} - N_{\mathbf{D}} = 2 \cdot 10^{18} \text{ cm}^{-3}$; b—beam along [111], GaAs(Zn), $N_{\mathbf{A}} - N_{\mathbf{D}} = 0.9 \cdot 10^{17} \text{ cm}^{-3}$.

out of the cryostat, and focused onto the entrance slit of a DFS-24 spectrometer. This arrangement, with a short-focal-length collecting mirror (2), was chosen because of the low intensity of hot-carrier luminescence. Despite the large aperture ratio of mirror (2) (1:1.4), the maximum deflection of the luminescence rays inside the sample is only about 6°, because of the high refractive index of GaAs. Accordingly, the experimental results may be compared without any appreciable error with a theory in which the directions of the excitation and luminescence rays are assumed to be parallel. In the measurements, the sample was in liquid helium with pumping (T = 1.6 K), or in helium vapor (in this case the crystal temperature was about 30 K).

Figure 16 compares the results measured for the Stokes parameters for the beam-along-[100] geometry for $\varphi = \pi/4$ and for the beam-along-[111] geometry. These measurements were carried out with excitation by a He-Ne laser ($\hbar \omega_{ex} = 1.96 \text{ eV}$) at the high-frequency edge of the luminescence spectrum ($\hbar \omega = 1.88$ eV; the luminescence resulted from transitions between the conduction band and an acceptor level). It follows from this figure that for the beam-along-[100] geometry the parameter ξ_1 remains essentially equal to zero, while ξ_3 decreases with increasing magnetic field. These results are in agreement with Eqs. (32) and (38) for $\varphi = \pi/2$ 4 and indicate that there is no rotation of the polarization plane. For the beam-along-[111] geometry the decrease in ξ_3 is accompanied by an increase and then a decrease in ξ_1 , implying a rotation of the polarization plane and in agreement with Eqs. (39) and (40). [The



FIG. 17. Spectrum of the degree of linear polarization, ξ_3 , without a magnetic field and in a magnetic field of 70 kOe. GaAs(Zn) sample, $N_A - N_D = 1.2 \cdot 10^{18}$ cm⁻³, T = 1.6 (the upper curve corresponds to H=0).



FIG. 18. Curves of the depolarization in a magnetic field for various energies $\hbar \omega$ in the HEP spectrum.³² 1-1.893 eV; 2-1.85 eV; 3-1.81 eV; 4-1.77 eV (Ref. 32). The sample is the same as in Fig. 17. T=1.6 K, $\hbar \omega_{ex}=1.96$ eV, beamalong-[111] geometry.

rotation angle in the field H = 45 kOe in Fig. 16b is $\psi = (1/2) \arctan(\xi_1/\xi_3) = 13^\circ$.] When H is reversed, the rotation also reverses.

The measurements described below were carried out in the beam-along-[111] geometry, for which the orientation of the electric vector of the exciting light with respect to the crystal axes is inconsequential.¹¹

Figure 17 compares the ξ_3 spectrum in a magnetic field H = 70 kOe with the corresponding spectrum in zero field. In addition to the depolarization of the luminescence—the decrease in ξ_3 —caused by the tendency for the distribution function to become isotropic, there is a further decrease in ξ_3 in a magnetic field because of the rotation of the HEP polarization plane. As a result of this effect, the polarization disappears far more sharply in a strong magnetic field.

Figure 18 shows a series of depolarization curves $\xi_{\omega}(H)$ for various energies $\hbar \omega$ in the luminescence spectrum. The smaller energies $\hbar \omega$ correspond to a longer descent time and thus larger rotation angles in a given field. As a result, the dependence $\xi_3(H)$ becomes sharper as $\hbar \omega$ decreases. The shape of the curves should be described by Eq. (40) if $\hbar \omega$ is near the highfrequency luminescence edge, or by Eq. (35) far from the edge, where the recombining electrons lose many portions of energy. Actually, an intermediate situation is the most probable. From Eqs. (40) and (35), however, we find very nearly equal values for the angle $2\omega_{\tau}$, at which the degree of depolarization decreases by a factor of two $[\pi/3 \text{ from Eq. } (35) \text{ and } 1 \text{ from Eq.}$ (40)]. Working from the value of the magnetic field $(H_{1/2})$ which corresponds to a halving of $\xi_3(H)$ we can therefore determine τ , using

$$2\omega_{c1/2}\tau = 1.$$
 (41)



FIG. 19. Dependence of the descent time τ on the energy of the recombining electron, ε . The times τ are determined from the half-widths of the depolarization curves in Fig. 18. For $\varepsilon \rightarrow \varepsilon_0$ we have $\tau \approx 0.5 \cdot 10^{-13}$ s, and in this case this time is the time taken to leave the initial state.

In one limiting case, this value of τ will represent the average time taken for an electron to leave the state with the initial energy, while in the other limit it corresponds to the descent time. Values of τ determined in this manner¹²⁾ from the curves in Fig. 18 are shown in Fig. 19, as a function of the energy of the recombining electrons, τ .

In the limit $\varepsilon - \varepsilon_0$ we find the departure time $\tau = 0.5 \cdot 10^{-13}$ s. Figure 20 shows this time for several p-GaAs samples doped with Zn and Cd, with uncompensated-acceptor concentrations $N_{\rm A} - N_{\rm D}$ ranging from $0.9 \cdot 10^{17}$ cm⁻³ to $1.4 \cdot 10^{18}$ cm⁻³ [for the high-frequency edge of the luminescence spectrum, the experimental curves of $\xi_3(H)$ can be described well by Eq. (40)]. The measured results on τ as a function of $N_{\rm A} - N_{\rm D}$ are approximated by a straight line

$$\frac{1}{\tau} = \frac{1}{\tau} + C (N_{\rm A} - N_{\rm D}), \tag{42}$$

where $C = (0.9 \pm 0.3) \cdot 10^{-5}$ cm³/s and $1/\tau_1 = 1.4 \cdot 10^{13}$ s⁻¹. In samples having a high acceptor concentration, the



FIG. 20. Dependence of the departure time on $N_{\rm A} - N_{\rm D}$ for several p-GaAs samples (Ref. 33). The experimental dependence is approximated by the expression $1/\tau = (1/\tau_1) + C(N_{\rm A} - N_{\rm D})$, where $C = (0.9 \pm 0.3) \cdot 10^{-5}$ cm³/s and $1/\tau_1 = 1.4 \cdot 10^{13}$ s⁻¹.

$$m(\mathbf{s}) = \mathbf{m}(0) \left(\mathbf{1} + \frac{2\varepsilon}{E_{p}}\right),$$

¹¹¹¹In the interpretation of the results on the depolarization of the HEP in transitions to acceptors, the splitting of the fourfold-degenerate state of the acceptor was ignored, since this splitting is smaller than the spread of the acceptor levels in doped crystals. As a result, the degree of circular polarization of the HEP in the case of linearly polarized excitation does not exceed a few percent.

¹²⁾Here [and also below, in a calculation of the cyclotron frequency, which appears in (41) and (47)] we are taking the nonparabolic shape of the conduction band into account. The cyclotron mass $m(\varepsilon)$ is found from the approximate expression for a Kane band,

where $m(0) = 0.067m_0$, and $E_g = 1.52 \text{ eV}$. For $\varepsilon_0 = 0.38 \text{ eV}$ (this is the initial energy of the electrons in the case of excitation by a He-Ne laser, with $\hbar\omega_{ex} = 1.96 \text{ eV}$), the cyclotron mass is $m = 0.1m_0$.

primary mechanism for energy relaxation is apparently the collision of electrons with neutral acceptors, accompanied by the excitation or ionization of the latter. A theoretical estimate of the total cross section (σ) for the excitation and ionization of neutral hydrogen-like atoms by fast electrons³⁵ in a medium with the dielectric permittivity of GaAs yields $\sigma v \approx 0.5 \cdot 10^{-5}$ cm³/s (at $v \approx 10^8$ cm/s), in satisfactory agreement with the value given above for C.

At low impurity concentrations, the processes primarily responsible for the energy relaxation of photoexcited electrons are the emission of long-wave LO phonons by these electrons and intervalley scattering¹³; these processes also determine the time τ_1 in Eq. (42), as can be seen from the appearance of phonon oscillations in the HEP spectrum (Sec. 10).

Let us examine Fig. 19, which corresponds to a relatively high impurity concentration $(N_A - N_D \approx 1.2 \cdot 10^{18} \text{ cm}^{-3})$. The data shown for τ in this figure for the largest values of $\varepsilon_0 - \varepsilon$ represent the descent time $\tau(\varepsilon, \varepsilon_0)$. The energy dependence of τ in this region can be approximated by a straight line drawn through the origin (the dashed line). This line corresponds to Eq. (36), which takes the following form in the case $\varepsilon_0 - \varepsilon \ll \varepsilon_0$:

$$\tau(\varepsilon, \varepsilon_0) = \frac{\tau_{\varepsilon_0}}{\varepsilon_0} (\varepsilon_0 - \varepsilon). \tag{43}$$

The energy relaxation time determined from the slope of the dashed line, $\tau_{c_0} = 5.6 \cdot 10^{-13}$ s at $\tau_{c_0} = 0.38$ eV, agrees satisfactorily with the theoretical result for the total energy loss of fast electrons through the excitation and ionization of neutral acceptors⁵¹:

$$\frac{1}{\tau_{\varepsilon_{\bullet}}} = \sqrt{2m}_{\mathrm{C}} (m_{\mathrm{h}\mathrm{h}} + m_{\mathrm{h}\mathrm{h}}) (m_{\mathrm{h}\mathrm{h}} m_{\mathrm{h}\mathrm{h}})^{-1} \frac{\pi e^{4} (N_{\mathrm{A}} - N_{\mathrm{D}})}{\kappa^{2} \varepsilon_{0}^{3/2}} \ln \frac{4\varepsilon_{0}}{I}, \qquad (44)$$

where \varkappa is the dielectric permittivity, and *I* is a quantity of the order of the acceptor ionization energy. Adopting the values $\varepsilon_0 = 0.38 \text{ eV}$, I = 0.03 eV, and $N_A - N_D = 1.2 \cdot 10^{18} \text{ cm}^{-3}$, we find the value $\tau_{\varepsilon_0} \approx 6 \cdot 10^{-13} \text{ s}$ from (44).

10. PHONON OSCILLATIONS IN THE HEP SPECTRUM

At an acceptor concentration $N_A < 5 \cdot 10^{17}$ cm⁻³ and at temperatures below ~50 K, oscillations appear in the HEP spectrum with a period of about 37 meV, which is equal to the energy of long-wave LO phonons in gallium arsenide. Figure 21 shows the high-frequency regions of the HEP spectra obtained in a sample with $N_A - N_D$ = 0.9 $\cdot 10^{17}$ cm⁻³ excited by beams from He-Ne and Kr⁺ lasers ($\hbar \omega_{ex}$ = 1.96, 1.83, 1.65 eV). The first high-frequency maximum on each of the spectra I, II, III in Fig. 21 results from transitions to acceptor levels of electrons photo-excited from the heavy-hole subband which



FIG. 21. Hot-carrier photoluminescence spectra of GaAs(Zn) (Refs. 33 and 34). $N_A - N_D = 0.9 \cdot 10^{17}$ cm⁻³, for three different excitation lines: I-1.96 eV; II-1.83 eV; III-1.65 eV. The arrows show the positions of the exciting lines. The TO peaks correspond to Raman scattering of the light by TO phonons (scattering by LO phonons is forbidden in this experimental geometry). The inset shows the transition scheme. The excitation is from the heavy-hole (hh) and light-hole (lh) sub-bands. Peaks 1, 3, and 2 in the luminescence spectrum correspond to excitation from the hasubband and recombination to acceptor (A) levels of electrons from the production point and after the emission of one and two phonons, respectively.

have not undergone energy relaxation. The second and third maxima correspond to the recombination of electrons which have emitted one and two optical phonons in succession; the corresponding transition scheme is shown in the inset in Fig. 21. The lower-frequency part of spectra I and II is complicated by the onset of recombination transitions of electrons excited out of the light-hole subband. The oscillations in the HEP spectrum are quite clearly defined at $N_{\rm A} \sim 3 \cdot 10^{17}$ cm⁻³, and their amplitude does not change as $N_{\rm A}$ is reduced further. This result is evidence that emission of LO phonons becomes the primary mechanism for energy relaxation of hot electrons at these impurity concentrations.

The width of the peaks in the spectra is determined by several factors, in particular, (a) the energy spread of the final states (the acceptor levels) which results from the spatial fluctuations caused by the impurity – impurity interaction and (b) the broadening of the initial electron energy distribution by the ripple of the valence band (Sec. 6). The broadening caused by the ripple decreases with decreasing excitation energy, as is observed experimentally. Furthermore, we do not rule out the possibility that the peaks are broadened somewhat by the proximity of the crystal surface and that this factor becomes less important with decreasing energy because of the decrease in the absorption coefficient.

Polarization measurements have revealed that the degree of linear polarization decreases from peak 1 to peak 2 and also from peak 2 to peak 3, by a factor of about 1.5 in each step. This decrease results from the tendency toward an isotropic electron distribution function accompanying the emission of an optical phonon. It is not difficult to calculate the behavior of the anisotropic part of the distribution function. The prob-

¹³^{The} contribution of scattering by acoustic phonons may be ignored. The scale time for the emission of an acoustic phonon in GaAs at an energy of 0.4 eV is ~5 $\cdot 10^{-12}$ s, while the energy relaxation time is $\tau_{\rm c} \sim 10^{-9}$ s. Electron-electron collisions also make only a minor contribution to the energy relaxation. Estimates show that at the concentration of photo excited electrons under experimental conditions, ~10¹⁵ cm⁻³, we have $\tau_{\rm ee} \sim 10^{-10}$ s.

ability per unit time for a transition due to the emission of an optical phonon, $w(\mathbf{p}, \mathbf{p}')$, is proportional to $|\mathbf{p} - \mathbf{p}'|^{-2}$, where p and p' are the initial and final momenta of the electron. If the distribution function before the transition, $f_n(\mathbf{p})$, is of the form in (9), then the distribution function after the transition will be of the same form, with

$$\frac{\alpha_{n+1}}{\alpha_n} = \int \frac{P_2(\cos\vartheta_{\mathbf{pp}'})}{|\mathbf{p}-\mathbf{p}'|^2} \, \mathrm{d}\Omega \Big/ \int \frac{\mathrm{d}\Omega}{|\mathbf{p}-\mathbf{p}'|^2} \, .$$
(45)

where $\vartheta_{\mathtt{p}\mathtt{p}'}$ is the angle between p and p'. A calculation yields

$$\frac{\alpha_{n+1}}{\alpha_n} = \frac{3\varepsilon_n^2 + 3\varepsilon_{n+1}^2 + 2\varepsilon_n\varepsilon_{n+1}}{8\varepsilon_n\varepsilon_{n+1}} - \frac{3}{4} \frac{\varepsilon_1 + \varepsilon_{n+1}}{\sqrt{\varepsilon_n\varepsilon_{n+1}}} \left[\ln \frac{\varepsilon_n + \varepsilon_{n+1} + 2\sqrt{\varepsilon_n\varepsilon_{n+1}}}{\hbar\omega_{1,0}} \right]^{-1},$$
(46)

where ε_n and ε_{n+1} are the energies of the electron before and after the emission of the optical phonon. With ε_n = 0.4 eV we find for GaAs ($\hbar \omega_{\rm LO} = 37$ meV) the value $\alpha_n / \alpha_{n+1} \approx 1.6$ from this expression. This is close to the value found experimentally.

The dependence of the Stokes parameter ξ_3 (the degree of linear polarization) on the magnetic field has been studied at the frequencies of the first, second, and third peaks in the spectrum in Fig. 21. The corresponding experimental data are shown in Fig. 22. Shown in the same figure are theoretical curves plotted from the equations derived in Ref. 33 for cascade transitions¹⁴):

$$\begin{aligned} \xi_{3}^{(1)}(H) &= \frac{1}{1 + 4\omega_{0}^{2} \tau_{1}^{2}} \xi_{3}^{(1)}(0), \\ \xi_{3}^{(2)}(H) &= \frac{1 - 4\omega_{0}^{2} \tau_{1} \tau_{2}}{(1 + 4\omega_{0}^{2} \tau_{1}^{2}) \xi_{3}^{2}(0)}, \\ \xi_{3}^{(3)}(H) &= \frac{1 - 4\omega_{0}^{2} (\tau_{1} \tau_{2} + \tau_{1} \tau_{3} + \tau_{2} \tau_{3})}{(1 + 4\omega_{0}^{2} \tau_{1}^{2}) (1 + 4\omega_{0}^{2} \tau_{3}^{2}) (1 + 4\omega_{0}^{2} \tau_{3}^{2})} \xi_{3}^{2}(0); \end{aligned}$$

$$\begin{aligned} (47) \\ \end{aligned}$$

here τ_1 , τ_2 , and τ_3 are the effective times at which electrons leave the states with the energies ε_0 , ε_0 $-\hbar\omega_{\rm LO}$, and $\varepsilon_0 - 2\hbar\omega_{\rm LO}$, respectively, as the result of any energy-relaxation process. In Fig. 22 the theoretical curves correspond to the times

$$\tau_1 = 0.7 \cdot 10^{-13} \text{ s}, \quad \tau_2 = 0.8 \cdot 10^{-13} \text{ s}, \quad \tau_3 = 1 \cdot 10^{-13} \text{ s}.$$

The time τ_1 corresponds to the time at which electrons leave the point at which they are produced ($\varepsilon_0 = 0.38 \text{ eV}$); τ_2 corresponds to the time at which electrons leave the point $\varepsilon_0 - \hbar \omega_{\text{LO}}$, and τ_3 corresponds to the time at which electrons leave the point $\varepsilon_0 - 2\hbar \omega_{\text{LO}} \approx 0.31 \text{ eV}$. The difference between τ_1 and τ_3 (as we will see below) results from the additional contribution of intervalley transitions to the energy relaxation from the state with energy ε_0 .

The GaAs conduction band contains, in addition to its central Γ minimum, some side minima, at the points L and X of the Brillouin zone. According to the present understanding, the lowest of these side minima are the L minima (valleys), which lie $\Delta E_{\Gamma L} \approx 0.31$ eV above the central minimum (Sec. 11). Since the electrons in the experiment under discussion here have an energy $\varepsilon_0 = 0.38$ eV at their point of production, i.e., since they



FIG. 22. Depolarization of the hot-carrier photoluminescence in a magnetic field.³³ a, 2, 3—Measurements at peaks 1, 2, and 3 in spectrum I in Fig. 21 (excitation energy of 1.96 eV); symbols—experimental results. The sample is the same as in Fig. 21. Solid curves—approximation of the experimental results by Eqs. (47).

lie above the bottom of the *L* valleys, they may be scattered not only in events which leave them in the central minimum but also in events which cause them to move to one of the *L* valleys, emitting an "intervalley" phonon with $q \sim \pi/a$ in the process. The probability for the scattering of an electron from the point of production can thus be written

$$\tau_{1}^{-1} = \tau_{\rm phO}^{-1} \tau_{\rm FL}^{-1}, \tag{48}$$

where τ_{ph0}^{-1} is the probability for the emission of a longwave LO phonon, and $\tau_{\Gamma L}^{-1}$ is the probability for scattering into the *L* valleys, accompanied by emission of a short-wave phonon. For a determination of τ_{ph0} the electron lifetime was measured at the point of production for conditions such that transitions to the *L* valleys are impossible, i.e., for $\varepsilon_0 < \Delta E_{\Gamma L}$. With this aim in view, measurements of $\xi_3(H)$ were carried out during excitation by the line of a Kr⁺ laser with $\hbar \omega_{er} = 1.83$ eV ($\varepsilon_0 = 0.26$ eV) (Fig. 23). The result is $\tau_1 = \tau_{ph0} = (1.0 \pm 0.05) \cdot 10^{-13}$ s.

This value can be compared with the calculated value. For GaAs at T = 0 K and $\varepsilon = 0.3$ eV, we have τ_{phO} = 2.10⁻¹³ s, and this value depends only slightly on the



FIG. 23. Depolarization of the hot-carrier photoluminescence in a magnetic field at excitation energies of 1.96 eV and 1.83 eV (Refs. 33 and 34). The measurements were taken at the peaks 1 on spectra I and II in Fig. 21; the sample is the same as that in Fig. 21.

¹⁴⁾Equations (14) are completely analogous to the equations which describe the Hanle curves for cascade transitions in atoms.

energy.³⁶ When the deviation from a parabolic conduction band is taken into account in the calculations, the value for τ_{phO} turns out to be slightly (15–20%) smaller.^{37,38} Comparison of the value $\tau_{phO} = 1 \cdot 10^{-13}$ s with the lifetime $\tau_1 = (0.7 \pm 0.05) \cdot 10^{-13}$ s measured during excitation with a He-Ne laser ($\varepsilon_0 = 0.38$ eV) with the help of Eq. (48) yields the intervalley-scattering time $\tau_{\Gamma L}$ referred to the value $\varepsilon = 0.38$ eV: $\tau_{\Gamma L} = (2.5 \pm 0.8) \cdot 10^{-13}$ s. This value corresponds to the interaction constant³⁷ D_{ΓL} = 0.8 \cdot 10⁹ eV/cm for scattering from the Γ minimum into the L valley.

11. EXCITATION THROUGH A SIDE VALLEY

The HEP spectra which we have been discussing were locked in frequency with the exciting line. For example, a shift of $\hbar \omega_{ex}$ from 1.96 eV to 1.83 eV in Fig. 21 and then to 1.65 eV shifted the entire system of oscillations in a corresponding way. A different picture is observed when the excitation energy lies in the range 2.4 -2.8 eV, i.e., when the electrons in the conduction band are produced with a significantly higher energy.^{34,39,40} In this case the HEP is very faint near the excitation line. At the same time we observe a much more intense luminescence with a high-frequency edge near 1.8 eV and with a spectrum which does not depend on the excitation energy $\hbar\,\omega_{\rm ex}$ in the range specified (Fig. 24). Identical spectra were found with excitation by the lines of an Ar^{*} laser (2.41 and 2.54 eV) and a He-Cd laser (2.81 eV). The initial electron energies ε_0 corresponding to these values of $\hbar \omega_{ex}$ are 0.75, 0.85, and 1.07 eV (we mean average values here, with allowance for the ripple, of the energy ε_0 of the electrons excited from the heavy-hole subband). In the spectra of moderately doped samples, as during excitation by the lines of He-Ne and Kr⁺ lasers, we observe oscillations with a period of about 37 meV. An interpretation of these results which was proposed in Refs. 34 and 39 is based on the existence in the GaAs conduction band of side minima (valleys) in addition to the central Γ minimum at the point k=0. These side valleys are the points $L(\pi/a, \pi/a, \pi/a)$ and $X(2\pi/a, 0, 0)$ in the Brillouin zone. The bottoms of these side (L and X) valleys are raised above the level of the edge of the conduction band



FIG. 24. Photoluminescence spectrum $I(\hbar\omega)$ of a p-GaAs(Zn) sample.³⁹ $N_A \sim N_D = 0.9 \cdot 10^{17}$ cm⁻³, T = 1.6 K. $1 - \hbar\omega_{ex} = 2.54$ eV; $2 - \hbar\omega_{ex} - 2.41$ eV; $3 - dI/d\omega_e$. Shown in the inset at the bottom left is the scheme for the transition $L \rightarrow \Gamma \rightarrow A$ (to an acceptor level).

[the point $\Gamma(0,0,0)$] by about 0.3 and 0.5 eV, respectively.43 Since these energies are noticeably lower than the values of ε_0 cited above, and the state density in the side bands is quite high,¹⁵) the photoexcited electrons undergo a relatively rapid transition to these valleys. Accordingly, with sufficiently high-frequency excitation, the HEP intensity is also low near $\hbar \omega_{ex}$. Through successive intervalley transitions, the photoexcited electrons slide down to the bottom of the lowest side valley (L); then, emitting an "intervalley" phonon with $q(\pi/a, \pi/a, \pi/a)$, they return to the central Γ minimum of the conduction band (see the transition scheme in Fig. 24a). The L valley thus becomes a secondary source of monoenergetic electrons. As before, the HEP results from the recombination of electrons at the Γ minimum with holes at acceptor levels, and the oscillations in the spectra result from the successive emission of LO phonons in the course of the energy relaxation. The first high-frequency maximum in the spectrum, at $\hbar \omega = 1.78$ eV, is, in light of the assumptions above, due to the recombination of electrons from the state at the Γ minimum, which they reach after an $L \rightarrow \Gamma$ intervalley transition accompanied by the emission of an LO phonon $(\hbar \omega_{\rm ph} = 29 \text{ meV})$ or an LA phonon $(\hbar \omega_{\rm ph} = 26 \text{ meV})$ with q $=(\pi/a)$ (1, 1, 1). These are precisely the types of transitions which are allowed by the selection rules in GaAs crystals. The energies of the LO and LA phonons are approximately equal, and the corresponding peaks in the spectrum are accordingly not resolved. On the high-frequency slope of the spectrum, however, we can see a knee at about 1.8 eV; this knee is clearly visible in the differential spectrum (curve 3 in Fig. 24). This structural feature may be attributed to an $L \rightarrow \Gamma$ transition which is accompanied by the emission of a TA phonon ($\hbar \omega_{\rm ph} = 8$ meV). Such a transition may be partially allowed because the electron wave vector k at the Γ valley is not small after an intervalley transition (k $\approx 10^7$ cm⁻¹). The position of the L valley with respect to that of the Γ valley is found from the positions of the peaks on curve 3 in Fig. 24 to be 310 ± 10 meV. This result agrees well with results obtained by other methods: 330 ± 40 meV (Ref. 57), 310 meV (Ref. 58), and 297 ± 10 meV (Ref. 59). The high-frequency tail in the spectrum is possibly a consequence of some energy spread of the electrons coming from the L valley, caused by the accumulation near the bottom of the L valley of electrons with energies below the energy of an optical phonon. In the case of excitation through a side valley, the emission turns out to be unpolarized if the pump is linearly polarized. (This is a natural result, since the initial anisotropy of the electron distribution is lost upon intervalley transitions.)

This effect gives us a method for directly measuring the energy difference ΔE between the bottom of the conduction band and the bottom of the lowest-lying side valley in the case of a direct-gap semiconductor. In Ref. 39, for example, the value $\Delta E = 210 \pm 10$ meV was found for the mixed crystal Ga_{0.8}Al_{0.2}As.

¹⁵⁾Estimates in Ref. 43 put the state-density masses in the L and X valleys at $0.56m_0$ and $0.85m_0$, respectively.

A photoluminescence spectrum in p-GaAs identical to that shown in Fig. 24 was recently described by Olego and Cardona.⁴¹ Their interpretation is completely different from that offered above: They attribute this spectrum to photoluminescence in a band which arises upon transitions from the conduction band to the splitoff subband (a band at $E_{\rm b} + \Delta$; see Sec. 13) and phonon repetitions of this band. We do not find their interpretation convincing. The photoluminescence spectrum in the region $E_{g} + \Delta$ in n-GaAs crystals, which has been studied over a broad concentration range, consists of a single band at about 1.85 eV with a rather sharp lowfrequency threshold.⁴² We find it difficult to accept that it would be in p-type crystals that phonon repetitions of the $E_{g} + \Delta$ band should arise. Furthermore, in the model proposed by Olego and Cardona⁴¹ we would expect a quadratic dependence of the luminescence on the excitation intensity (the recombination of photoexcited electrons with photoexcited holes). Experimentally, however, a linear dependence is observed¹⁶) for the spectrum in Fig. 24.

12. RECONSTRUCTION OF THE HOT-ELECTRON ENERGY DISTRIBUTION FROM THE HEP SPECTRUM

The energy distribution of the free carriers, $f(\varepsilon)$, can be reconstructed from the spectrum of the bandimpurity radiative recombination, provided that the probability for the corresponding transition is known. This probability is proportional to the square of the modulus of the carrier wave function at the center in the momentum representation, $\Phi(\mathbf{p})$. For a hydrogenlike center with an isotropic effective mass we would have

$$\Phi(\mathbf{p}) \sim \frac{1}{(1+x)^4},
x = \frac{p^2}{2m_A e_A},$$
(49)

where ε_A is the binding energy of the center, and m_A is the effective mass. Equation (49) does give the corresponding transition probability in this very simple case.⁴⁴ For a shallow acceptor associated with a degenerate band, Eq. (49) would presumably also give a reasonable approximation, but it is not clear just what mass should be used in it. Since the wave functions of holes at acceptors consist primarily of the wave functions of the heavy-hole subband at sufficiently high quasimomenta (Sec. 7), m_a was set equal to the average mass of the heavy holes $(0.6m_0)$ in using Eq. (49) in Ref. 39 to reconstruct $f(\varepsilon)$ from the HEP spectrum. The results of Ref. 39 were recalculated in Ref. 45 with the function $\Phi(\mathbf{p})$ found through a direct numerical calculation by the method of Ref. 46. It turns out that for electron energies $\varepsilon > \varepsilon_A$ the wave function $\Phi(p)$ can be approximated well by (49) with $m_{\rm A} = 0.66m_0$ (it is customary to assign ε_{A} the value 30 meV, which is the ionization energy of Zn in GaAs). Hot electrons were produced in Ref. 39 by the beam from an Ar⁺ laser (2.54 eV). According to Fig. 24, the high-frequency





edge of the HEP spectrum and, correspondingly, the maximum electron energy (about 0.3 eV) are determined by the position of the L valley above the bottom of the conduction band.

Figure 25 shows the results calculated for $n(\varepsilon)$ = $f(\varepsilon)g(\varepsilon)$ from the data of Fig. 24 [$g(\varepsilon)$ is the state density]. The oscillations on the curve of $n(\varepsilon)$ are, as before, caused by the sequential emission of LO phonons in the course of energy relaxation. We see that the average value of $n(\varepsilon)$ over the interval 100-300 meV depends slightly on the energy. In this particular sample, scattering by optical phonons was evidently predominant, and the average value was⁴⁷ $\overline{n(\varepsilon)} \sim G\tau_{\rm phO}(\varepsilon)$, where G is the pump intensity and $\tau_{\rm phO}(\varepsilon)$ is the emission time of the optical phonon. The weak dependence $n(\varepsilon)$ in this energy range corresponds to a weak energy dependence³⁸ $\tau_{\rm phO}(\varepsilon)$. At $\varepsilon < 37$ meV the emission of optical phonons becomes impossible, and $n(\varepsilon)$ correspondingly increases sharply (Fig. 25a).

13. HOT-HOLE PHOTOLUMINESCENCE

Up to this point we have been dealing with the hotcarrier photoluminescence caused in p-type samples by the recombination of nonequilibrium hot electrons with equilibrium holes, either free or localized at acceptor levels. In principle, we could invert the situation and examine the luminescence resulting from the recombination of hot holes. In this section we will ex-



FIG. 26. Photoluminescence spectrum of an n-GaAs sample.⁴² Solid curve— $N_D = 3.4 \cdot 10^{17}$ cm⁻³, T = 82 K; dashed curve—T = 200 K; plus signs—degree of circular polarization ξ_2 at 82 K. $\hbar \omega_{ex} = 1.96$ eV.

¹⁶)See Fig. 30b in Section 13, where the dependence on the pump intensity for the $E_g + \Delta$ band is approximately quadratic even for lightly doped n-type samples.

amine some results found on the luminescence caused in n-GaAs samples by the recombination of holes which are nonequilibirum holes and, as we will see, hot (unthermalized). They appear in the Γ_7 subband of the valence band and recombine with electrons of the Γ_6 conduction band.

The photoluminescence spectrum of n-GaAs crystals contains, along with an intense edge-luminescence band at about 1.51 eV, a significantly weaker band at about 1.85 eV (Fig. 26). It has been shown^{42,48,49} that this band results from transitions between the conduction band and the Γ_7 split-off subband, i.e., from the recombination of photoproduced holes in the Γ_7 band with electrons from the conduction band. The position of this band is determined by the sum of E_g (1.51 eV at 80 K) and Δ (0.34 eV; Ref. 50); the corresponding transition scheme is shown in Fig. 27.

The intensity of the band at $E_{g} + \Delta$ in Fig. 26 is five orders of magnitude lower than that of the band at E_{a} . Since the probabilities for the excitation of electrons from the Γ_7 and Γ_8 subbands are identical in order of magnitude, such a pronounced difference in intensity is a consequence of a difference between the lifetimes of the holes in the degenerate Γ_8 band and the Γ_7 split-off subband. In the latter case the lifetime, even in crystals with a relatively heavy doping, is apparently determined by $\Gamma_7 - \Gamma_8$ transitions, accompanied by the emission of an optical phonon (Fig. 27), which occur with a rather high probability. An estimate based on the partial $\Gamma_8 \rightarrow \Gamma_6$ and $\Gamma_7 \rightarrow \Gamma_6$ absorption coefficients,¹⁶ the emission probabilities, and corrections for selfabsorption leads to the following ratio of band intensities⁵¹:

$$\frac{I(E_g)}{I(E_g+\Delta)} = a \frac{\tau}{\tau_0},$$

where τ and τ_0 are the hole lifetimes in the Γ_8 and Γ_7 bands, respectively. The coefficient *a* depends on the particular form of the hole distribution function $f_h(\varepsilon_h)$, having a value of approximately 30 for the function $f_h(\varepsilon_h)$ shown in Fig. 29. Adopting $I(E_g)/I(E_g + \Delta) = 10^5$ and $\tau = 10^{-9}$ s (this is a typical value for a moderately doped sample) for an estimate, we find $\tau_0 = 3 \cdot 10^{-13}$ s. The results of a direct measurement of τ_0 will be reported below.

Figure 28 shows the photoluminescence spectrum in the vicinity of the $E_g + \Delta$ band for one of some compar-



FIG. 27. Transitions scheme for excitation of radiative recombination between the conduction band Γ_6 and the split-off subband Γ_7 . Upward arrow—excitation; downward arrow recombination with an energy $E_g + \Delta$; oblique arrows—hole transitions accompanied by the emission of an optical phonon.



FIG. 28. Photoluminescence spectrum of an n-GaAs sample in the region $E_g + \Delta$ (Ref. 42). $N_D = 7.6 \cdot 10^{15} \text{ cm}^{-3}$, T = 100 K. Solid curve—experimental; dashed—theoretical, for direct interband transitions.

atively lightly doped n-GaAs samples. Shown along with the experimental spectrum is a theoretical spectrum calculated for direct interband $\Gamma_6 - \Gamma_7$ transitions.¹⁷) In the calculation, the electron distribution function was assumed to be an equilibrium (Fermi) function, while the hole distribution function was assumed to be a Boltzmann function, $f_{\rm b}(\varepsilon_{\rm b}) \sim \exp(-\varepsilon_{\rm b}/kT)$. We see from the figure that the peaks of the theoretical and experimental curves coincide well. The tendency for the spectrum to stretch out on the low-frequency side is customary for doped crystals (because of the formation of tails on the state densities in the band gap and transitions involving impurity levels). The discrepancy between the theoretical and experimental spectra at high frequencies is probably due to the nonequilibrium nature of the distribution function of the holes participating in the radiative recombination. Figure 29 shows the hole energy distribution $f_{\rm h}(\varepsilon_{\rm h})$ found from an analysis of the experimental luminescence spectrum in Fig. 28. We can see that the function $f_{\rm h}(\varepsilon_{\rm h})$ increases with the energy. This tendency of $f_{\rm h}(\varepsilon_{\rm h})$ is explained on the basis that in the course of the energy relaxation of the holes there is some "leakage" of holes due to transitions between subbands of the valence band, so that a significant fraction of the photoproduced holes do not manage to reach the top of the valence band. Zakharchenya et al.42 have derived the following expression for the hole distribution function at $\varepsilon_h \gg kT$ for this particular case under the assumption of a quasielastic scattering mechanism:

$$f_{\rm h}(\varepsilon_{\rm h}) \sim \frac{1}{\epsilon_{\rm s}^{\frac{3}{2}-\nu}} \exp\left(\frac{1}{\nu} \frac{\tau_{\rm s}}{\tau_{\rm o}}\right),\tag{50}$$

where ν is the exponent in the energy dependence of the



FIG. 29. Hole energy distribution function found from an analysis of the spectrum in Fig. 28 (Ref. 42).

¹⁷⁾Zakharchenya *et al.*⁴² have shown that the band observed at $E_g + \Delta$ at liquid-nitrogen temperatures cannot be explained on the basis of donor-band transitions and is instead a consequence of $\Gamma_6 \rightarrow \Gamma_7$ interband transitions.

intraband relaxation time $(\tau_{\rm c} \sim \tau^{\nu})$. It can be seen from (50) that the behavior of $f_{\rm h}$ is substantially determined by the relationship between $\tau_{\rm c}$ and $\tau_{\rm o}$. In the case in which the holes lose energy in the $\Gamma_{\rm \gamma}$ band in collisions with free electrons,¹⁸⁾ we can set $\tau_{\rm c} \sim n^{-1} \varepsilon_{\rm h}^{-3/2}$, where *n* is the concentration of free electrons. We then have

$$f_{\rm h}\left(\varepsilon_{\rm h}\right) \sim \exp\left(\alpha n^{-1}\varepsilon_{\rm h}^{3/2}\right),$$
 (51)

where α is a coefficient which is independent of the energy. With increasing *n*, the rate of the intraband energy relaxation of the holes increases, and at a sufficiently high value of *n* the function f_h becomes a weak function of the energy according to (51), in agreement with the behavior observed in Ref. 42. In lightly doped samples, as long as the time τ_c for holes with an energy $\varepsilon_h \sim kT$ is long in comparison with τ_0 , the function $f_h(f_h)$ will have no Maxwellian region, even at very low energies.

A "leakage" of the holes in the course of their energy relaxation in the Γ_{γ} band can also explain the strong dependence of the intensity of the band at $E_{g} + \Delta$ on the excitation energy $\hbar \omega_{ex}$ (and thus on the initial hole energy, ϵ_{h}^{0} ; Ref. 51). As $\hbar \omega_{ex}$ is increased from 1.96 to 2.33 eV, for example (this increase corresponds to an increase in ϵ_{h}^{0} from 35 to 200 meV), the intensity at the peak of the $E_{g} + \Delta$ band decreases by a factor of about five.

The luminescence intensity in the $E_g + \Delta$ band turns out to be linear in the excitation intensity in moderately and lightly doped crystals; i.e., it corresponds to the recombination of photoproduced holes with equilibrium electrons. If, however, the concentration of the equilibrium electrons (in lightly doped crystals) is comparable to the photoelectron concentration, then the luminescence intensity increases more rapidly with the pumping (compare the data for the two samples in Fig. 30).

The short value of τ_0 makes the luminescence in the $E_g + \Delta$ band very convenient for observing optical orientation of free holes. As a rule, experiments on the optical orientation of free carriers in semiconductors have dealt with only the spin orientation of the free



FIG. 30. Dependence of the luminescence intensity in the band $E_g + \Delta$ on the excitation intensity for two doping levels (a and b).

163



FIG. 31. Depolarization of the photoluminescence in the band $E_g + \Delta$ in a transverse magnetic field.⁵³ n-GaAs sample $(N_D = 1.4 \cdot 10^{18} \text{ cm}^3)$. Points—experimental; curve—theoretical, with a lifetime 1.3 $\cdot 10^{-13}$ s.

electrons—not the holes in the valence band.¹⁴ The reason lies in the rapid spin relaxation of the holes in the Γ_8 band, which is degenerate at k = 0. In this case the spin relaxation time of the holes, τ_8 , is comparable to the momentum relaxation time τ_p and much shorter than the hole lifetime. For this reason, the degree of optical orientation of the holes, which is proportional to the quantity $\tau_8(\tau_8 + \tau_0)^{-1}$ is small.¹⁹

There is a different situation in the simple (nondegenerate) Γ_{τ} band. Here the spin relaxation time τ_{s} is not rigidly tied to the value of τ_p and may be significantly longer than the latter; the lifetime au_0 , as noted earlier, is determined by the relatively high probability for Γ_7 - Γ_8 transitions. Consequently, the holes produced near the edge of the Γ_7 band do not have time, during the lifetime τ_0 , to lose their initial spin orientation, created upon photoexcitation by circularly polarized light. If the spin relaxation is ignored, then the degree of circular polarization of the luminescence, ξ_2 , for Γ_6 - Γ_7 transitions is equal to the degree of spin orientation of the holes, P_{sh} . The theoretical value of P_{sh} upon excitation by circularly polarized light with an energy $\hbar \omega_{ex} = E_g + \Delta$ is equal to unity^{16,49}; for $\hbar \omega_{ex} = 1.96$ eV, we have $P_{sh} = 0.99$ (Ref. 49). The large values of ξ_2 in the $E_{e} + \Delta$ band (of the order of 0.8-0.9) are in fact observed upon excitation by circularly polarized light with $\hbar \omega_{ex}$ = 1.96 eV (Refs. 42 and 49). This means that the lifetime of the nonequilibrium holes in the Γ_7 band is significantly shorter than their spin relaxation time.

The large values of ξ_2 have made it a simpler matter to design an experiment for a direct determination of τ_0 (Ref. 53). The lifetime τ_0 has been determined by exploiting the depolarization of the luminescence in a transverse magnetic field upon excitation by circularly polarized light. We know that this effect is caused by the precession of the magnetic moments (in this case, the magnetic moments of holes) around the magnetic field direction. Figure 31 shows the dependence $\xi_2(H)$. Over this particular range of magnetic fields, the experimental data in Fig. 31 can be described satisfactor-

¹⁸)For excitation by a He-Ne laser (1.96 eV) the hole energy at the time of production in the Γ_7 band is of the order of 30-35 meV, i.e., smaller than the energy of the optical phonon. In this case we may therefore ignore energy relaxation accompanied by emission of optical phonons.

¹³⁾The optical orientation of the holes in the Γ_8 band is intensified substantially upon a deformation which lifts the degeneracy of the light- and heavy-hole subbands, with the result that the spin relaxation is retarded. At a load of 3 kgf/mm², the degree of circular polarization of the recombination luminescence caused in GaAs by the spin orientation of holes reaches 10% (Ref. 52).

ily by the Lorentz formula

$$\frac{\xi_2(H)}{\xi_2(0)} = (1 + \omega_L^2 T^2)^{-1},$$

where T is the spin lifetime $(T^{-1} = \tau_0^{-1} + \tau_8^{-1}); \omega_L = g\mu_B H/\hbar$, where μ_B is the Bohr magneton; and g is the g-factor, set equal to four in accordance with the results of Refs. 50 and 54. The solid curve in Fig. 31 corresponds to $T = 1.3 \cdot 10^{-13}$ s. Ignoring τ_s^{-1} in comparison with τ_0^{-1} on the basis of the above discussion, we may identify T as the hole lifetime at the top of the split-off subband.²⁰⁾

The value found for τ_0 was compared in Ref. 53 with a value calculated there for τ_{pbO} , the time for the emission of an optical phonon in the Γ_7 transition. The corresponding expression under the condition $\Delta - 3\hbar\omega_{\rm LO}$ $\gg \sqrt{m_{eb}} m_{mb}\Delta$ is

$$\frac{1}{\tau_{\rm p}} = \frac{4}{3} \alpha_{\rm hh} \omega_{\rm LO} \sqrt{\frac{\hbar \omega_{\rm LO}}{(\Delta/3) - \hbar \omega_{\rm LO}}}; \qquad (52)$$

where α_{bb} is the constant of the interaction of heavy holes with optical phonons of frequency $\omega_{
m LO}$, and $m_{
m hh}$ and m_{sk} are the effective masses in the heavy-hole subband and in the spin-split-off band, respectively. Expression (52) corresponds to hole transitions from Γ_{τ} to the light-hole subband (transitions to the heavy-hole subband from a state with k=0 are forbidden by the selection rules), and the circumstance that the interaction constant α_{hh} appears in (52) is a result of a pronounced deviation of the light-hole subband from a parabolic shape near energies of order Δ . For GaAs, Eq. (52) yields $\tau_{\rm pbO} = 1.1 \cdot 10^{-13}$ s. The condition written above for the applicability of (52) in GaAs is satisfied poorly. However, an exact expression derived for τ_{phO} without any assumptions regarding the relationship between masses leads to the value $\tau_{\rm pb0} = 2 \cdot 10^{-13}$ s, which is not greatly different. Comparison of the theoretical and experimental results for τ_{pb0} and τ_0 and estimates of the probabilities for other scattering channels show that the hole lifetime τ_0 in this situation is in fact determined primarily by the probability for the emission of longitudinal optical phonons in transitions between the subbands of the valence band.

In lightly doped p-GaAs crystals $(N_A < 10^{17} \text{ cm}^{-3})$, as long as the hot-electron photoluminescence (described in the preceding sections) is faint, the band $E_g + \Delta$ is also observed at 1.85 eV (Ref. 9). In this case, this band is caused by a recombination of photoexcited electrons and of photoexcited holes, and it also exhibits a pronounced circular polarization. A band near 1.78 eV was recently observed (at 80 K) in Ref. 56 in the photoluminescence spectrum of heavily doped p-GaAs crystals (at concentrations N_A in the range $10^{17}-10^{20}$ cm⁻³). Olego and Cardona⁵⁶ also attributed this band to $E_g + \Delta$ transitions. They attributed the 70-meV frequency shift from the position of the band in *n*-type samples to an effect of the dopant (Zn) on the value of Δ in GaAs.

164 Sov. Phys. Usp. 25(3), March 1982

This interpretation, does not, however, explain why this shift does not depend on $N_{\mathbf{A}}$.

The recombination luminescence which arises in the $E_{g} + \Delta$ band in GaSb and GaAs crystals as a result of Auger processes was studied in Ref. 55. Holes were scattered into the Γ_{7} band from the Γ_{8} band by the energy E_{g} liberated upon the recombination $\Gamma_{6} - \Gamma_{7}$. This process naturally occurs more efficiently in GaAs, where $E_{g} \approx \Delta$.

14. CONCLUSION

It is fair to say that the basic behavior of hot-carrier photoluminescence in GaAs crystals has now been established. In other crystals, however, there may be some important differences, stemming from the symmetry, the band structure, the nature of the impurity centers, etc. Incidentally, even in GaAs all that has been studied up to this point in addition to the interband recombination luminescence is the hot-carrier photoluminescence which involves transitions to shallow acceptors. The role played by the surface in hot-carrier photoluminescence has not been revealed. The surface may prove to be quite important, since the hot electrons undergo their energy relaxation at only a small depth in the crystal.

A remarkable property of the hot-carrier photoluminescence is the relationship between its characteristics and the hot-electron relaxation kinetics. Because of this relationship, it becomes possible to directly determine the energy relaxation times and the hot-electron distribution functions in a variety of situations. We should expect further research developments along this line.

The optical momentum alignment of electrons by polarized light which was discovered in the course of research on hot-carrier photoluminescence may undoubtedly be reflected in other phenomena. An example might be the polarization photogalvanic effect which was recently discovered in GaAs crystals.⁶⁰

APPENDIX, ROLE OF ELASTIC AND INELASTIC COLLISIONS IN THE DEPOLARIZATION OF HOT-CARRIER PHOTOLUMINESCENCE

Let us examine a simplified model in which there are collisions of two types:

1) Absolutely elastic collisions, characterized by the time t_{p2} (this is the relaxation time for the anisotropy described by the second Legendre polynomial).

2. Inelastic collisions, which cause the electron to "slide down" the energy steps $\varepsilon_1, \varepsilon_2 = \varepsilon_1 - \delta \varepsilon, \ \varepsilon_3 = \varepsilon_2 - \delta \varepsilon$, etc. We denote by ν a unit vector along the electron momentum direction, and we introduce $n_k(\nu)$, the distribution of electrons in the momentum direction at the k-th step. We set $n_k(\nu) = a_k + b_k P_2(\cos \vartheta)$, where ϑ is the angle which the momentum makes with some direction which we have singled out (with the polarization vector of the exciting light). For the coefficients a_k and b_k we can write the simple balance equations

²⁰⁾The measured value of τ_0 is two orders of magnitude smaller than the value which was adopted as an *a priori* assumption in Ref. 55 in an evaluation of the efficiency of Auger processes in GaAs.

$$\frac{1}{\tau_{h}} a_{h} = \frac{1}{\tau_{h-1}'} a_{h-1}, \quad \left(\frac{1}{\iota_{p_{2}}} + \frac{1}{\tau_{h}}\right) b_{h} = \frac{1}{\tau_{h-1}'} \gamma_{h-1} b_{h-1};$$
(A1)

here τ_k is the total time required for an electron to leave the k-th step, and τ'_{k-1} is the time required to leave the (k-1)-th step as a result of a transition to the k-th step. If there are no other exit processes (e.g., an exit to a different valley), then $\tau'_k = \tau_k$. We ignore transitions upward along the energy scale. In the second equation in (A1), $1 - \gamma_k$ is the relative change in the anisotropy of the distribution described by the second Legendre polynomial in a transition from the (k - 1)-th step to the k-th step:

$$\gamma_{k-1} = \frac{\int W_{k-1, k}(\vartheta) P_2(\cos \vartheta) \sin \vartheta \, \mathrm{d}\vartheta}{\int W_{k-1, k}(\vartheta) \sin \vartheta \, \mathrm{d}\vartheta}$$

where $W_{k-1,k}(\vartheta)$ is the probability for a transition from the state ν at the (k-1)-th step to the state ν' at the kth step. If we introduce the anisotropy parameter α_k = b_k/a_k , we find from (A1)

$$\alpha_{k} = \frac{l_{p_{2}}}{\tau_{k} + l_{p_{2}}} \gamma_{k-1} \alpha_{k-1}.$$
 (A2)

This equation can also be applied to the first step, if we use $\gamma_0 = 1$ and take α_0 to be the anisotropy parameter at the time of photoexcitation; i.e., $\alpha_1 = \alpha_0 t_{p2} (\tau_1 + t_{p2})^{-1}$. Since the initial degree of polarization found experimentally is approximately equal to the theoretical value, we have $\alpha_1 \approx \alpha_0$ and thus $t_{p2} \gg \tau_1$; i.e., the absolutely elastic collisions are inconsequential in the cases which have been studied.

If the primary mechanism for the energy relaxation is the emission of longitudinal optical phonons, then we have $\gamma_{k-1} = 1/1.6$, as mentioned earlier, and this quantity is essentially independent of the energy (i.e., independent of k). The measured ratio α_k / α_{k-1} is approximately equal to this value, which also indicates that the condition $t_{p2} \gg \tau_k$ holds under the experimental conditions (τ_k in this case is the scale time for the emission of a single optical phonon).

In crystals which are doped relatively heavily, and for which the spectrum of the hot-carrier photoluminescence is structureless, energy relaxation occurs upon the excitation or ionization of an acceptor, or it occurs with free holes. Again in this case, the measurements of the initial polarization reveal $\alpha_1 \approx \alpha_0$, i.e., $t_{p2} \gg \tau_1$. This result may be explained on the basis that the scattering of the fast electrons is through small angles. Under these conditions we should have $1 - \gamma_k \ll 1$, $t_{p2} \gg \tau_k$ (provided only that the concentration of charged centers does not exceed the concentration of acceptors or of free holes by too large a margin).

In this case, Eq. (A2) can be converted into a differential equation,

$$\frac{d\alpha}{l\varepsilon} = \frac{\tau_{\varepsilon}}{\varepsilon \tau_{p_2}} \alpha, \tag{A3}$$

if we set $\tau_{\varepsilon} = \tau_{k} \varepsilon / \delta \varepsilon$ and

$$\frac{1}{\tau_{p_2}} = \frac{1}{t_{p_2}} + (1 - \gamma_{k-1}) \frac{1}{\tau_k} \, .$$

Equation (A3) gives the result in (29).

We turn now to the depolarization of the hot-carrier

photoluminescence in a magnetic field. Calculations similar to those of Ref. 33 lead to the dependence

$$\rho_{ln}(H) = \rho_{ln}(0) \operatorname{Re} \prod_{k=1}^{n} (1 + 2i\omega_{c}T_{k})^{-1},$$
 (A4)

where ρ_{ln} is the degree of linear polarization of the emission which appears upon the recombination of electrons of the *n*-th step with equilibrium holes, and T_k is the duration of the momentum alignment at the *k*-th step, given by

$$\frac{1}{T_h} = \frac{1}{\tau_h} + \frac{1}{t_{p_2}}.$$

With k = 1, 2, 3, Eq. (A4) leads to Eqs. (47), in which τ_k should be replaced by the times T_k . If $t_{p2} \gg \tau_k$, however, the times τ_k and T_k are essentially the same. At large values of k and at $t_{p2} \gg \tau_k$, this equation leads to (35), if we make use of the following equality, which holds under the condition $\omega_c \tau_k \ll 1$:

$$\prod_{k=1}^{n} (1+2i\omega_c \tau_k)^{-1} = \exp\left(-2i\omega_c \sum_{k=1}^{n} \tau_k\right).$$

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