

Spin relaxation of conduction electrons in p -type III-V compounds

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Spin relaxation of conduction electrons in p -type GaAs and GaSb crystals is investigated in detail in wide ranges of the acceptor density and of the temperature. It is established that the spin relaxation is due to the action of two mechanisms. The first is connected with exchange interaction of the electrons with equilibrium holes, and the other is due to spin splitting of the conduction band in crystals that have no inversion center. A detailed comparison of the experimental data with the results of theoretical calculations has made it possible to determine separately the role of each of the observed mechanisms, to study their behavior under various conditions, and find the conduction-band spin-splitting parameters and the exchange-interaction parameters that determine the effectiveness of these mechanisms.

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§1. INTRODUCTION

The method of optical orientation of photoexcited carriers makes it possible to measure short (down to 10^{-12} sec) spin-relaxation times of free electrons. This possibility is particularly valuable since the conditions for observing optical orientation differ substantially from ordinary conditions of observing electron paramagnetic resonance, and under these conditions there manifest themselves new spin-relaxation mechanisms whose investigation by traditional microwave-spectroscopy methods is quite difficult.

Even the first experiments^{1,2} on optical orientation have shown that the conduction electrons in p -type crystals can have very short spin-relaxation times, of the order of 10^{-10} – 10^{-11} sec. The strongest of the then known mechanisms of spin relaxation for free electrons, the Elliott-Yafet (EYa) mechanism,^{3,4} which takes into account the mixing of wave functions with different spins as a result of spin-orbit interaction, did not explain so rapid a relaxation of the spin. This stimulated a search for new possible mechanisms of conduction-electron spin relaxation. D'yakonov and Perel^{5,6} considered a mechanism due to spin splitting of the conduction band in crystals without inversion center (the DP mechanism). The distinguishing feature of experiments on optical orientation of electrons is the use of p -type crystals with relatively high hole density to ensure sufficiently intense luminescence. In this connection, Bir, Aronov, and Pikus⁸ considered a relaxation mechanism connected with exchange interaction of the electrons with the holes⁷ (the BAP mechanism).

Theoretical estimates carried out in Ref. 9 have shown that the BAP mechanism can play a dominant role in spin relaxation of thermalized electrons in a large number of III-V compounds in wide ranges of doping and temperatures. On the other hand, the DP and EYa mechanisms can predominate at relatively low doping levels and at high temperatures. Unfortunately, the earlier experimental investigations of the spin-relaxation processes in p -type compounds^{1,2,9–12} were insufficiently systematic and did not make it possible to identify reliably the mechanisms that dominate under some conditions or others. Only in Refs. 10 and 11 was a correct

conclusion drawn that the DP mechanism manifests itself in spin relaxation of electrons in moderately doped GaAlAs and GaAs crystals at high temperatures. The authors, however, were unable to explain the observed change in the time of the spin relaxation at low temperatures, and were therefore unable to single out and study the revealed mechanism. The dominant role of the DP mechanism was noted also in the case of spin relaxation of hot electrons in GaAs^{13,14} and GaSb crystals.¹⁵

We present in this paper the results of a systematic investigation of a spin relaxation of thermalized electrons in GaAs and GaSb, in wide ranges of the acceptor densities and temperatures. A detailed comparison of the experimental data with the results of theoretical calculations has made it possible to establish reliably the dominant relaxation mechanisms, to study their manifestations under various conditions, as well as to find the spin-orbit-splitting and exchange-interaction parameters that determine the effectiveness of the DP and BAP mechanisms.

Preliminary results were published earlier.^{16–18} Before we proceed to an exposition of the research results, we present briefly as summary of the principal conclusions of the theory.

§2. MECHANISMS OF SPIN RELAXATION OF CONDUCTION ELECTRON

a. The Elliott-Yafet mechanism

The spin orbit interaction leads to a mixing of the wave functions of the electrons in the conduction band with the wave functions of the valence band with opposite spin. This leads to nonzero values of the matrix elements of the transitions for electron scattering by impurities and by phonons, with simultaneous spin flip at a wave vector $\mathbf{k} \neq 0$. For cubic crystals with inversion center and with the band extrema position at the point Γ ($\mathbf{k} = 0$), these matrix elements $\propto k^2$, for example $\mathbf{k} \times \mathbf{k}'$, where \mathbf{k} and \mathbf{k}' are the final and initial values of the momentum. The corresponding spin relaxation time is given by

$$\frac{1}{\tau_s} = A \left(\frac{\Delta}{E_g + \Delta} \right)^2 \left(\frac{e_g}{E_g} \right)^2 \frac{1}{\tau_p}; \quad (1)$$

here τ_p is the momentum scattering time, ε_e is the kinetic energy of the electron, E_g is the width of the forbidden band, and Δ is the spin-orbit splitting of the valence band. According to the calculation results^{4,19,20} the numerical coefficient A in Eq. (1) depends on the dominant scattering mechanism and is close to unity in all the cases considered.

In crystals without an inversion center, in scattering by optical and acoustic phonons, the main contribution to $1/\tau_s$ is made by the deformation interaction. The corresponding matrix elements are proportional to $\mathbf{k} + \mathbf{k}'$ or $\mathbf{k} - \mathbf{k}'$, therefore

$$\frac{1}{\tau_s} \sim \left(\frac{\Delta}{E_g + \Delta} \right)^2 \frac{\varepsilon_e}{E_g} \frac{1}{\tau_p^{(1)}}, \quad (2)$$

where $1/\tau_p^{(1)}$ is the short-range contribution to the momentum relaxation time.²¹⁻²³ The quantity $1/\tau_s$ in (2) can also be expressed in terms of the momentum relaxation time $1/\tau_p$, which is determined mainly by the long-range interaction $1/\tau_p^{(2)}$. Since $\tau_p^{(2)}/\tau_p^{(1)} \propto \varepsilon_e$ (Ref. 24), in crystals without inversion center with zincblende structure the connection between the times τ_s and τ_p for the electrons is also determined by expression (1). However, owing to the lack of reliable data on the corresponding interband constants of the deformation potential, the constants A in (1) are more indeterminate in this case: $A \approx 1-10$. In scattering by charged impurities and in crystals without inversion center we have $A \approx 1$.

b. The D'yakonov-Perel' mechanism

Spin-orbit interaction in crystals without inversion center leads to splitting of the spin states of the conduction band at $\mathbf{k} \neq 0$. In crystals with zincblende structure (of the GaAs type) this splitting is proportional near the point Γ to the cube of the electron momentum

$$\hbar\Omega_0(\mathbf{k}) = \alpha(2m_e^3 E_g)^{-1/2} \boldsymbol{\kappa}, \quad (3)$$

where $\kappa_x = p_x(p_y^2 - p_z^2)$, $\kappa_y = p_y(p_z^2 - p_x^2)$, $\kappa_z = p_z(p_x^2 - p_y^2)$, with p_x , p_y , and p_z the components of \mathbf{p} along the principal axes of the crystal, m_e is the effective mass of the electron, and α is the splitting parameter. According to Ref. 25,

$$\alpha = \frac{4\Delta}{[(E_g + \Delta)(3E_g + 2\Delta)]^{1/2}} \frac{m_e}{m_{ev}}, \quad (4)$$

where m_{ev} is a constant close in magnitude to the mass of the free electron.

The spin splitting of the conduction band is equivalent to a magnetic field acting on the spin, and the magnitude and direction of this field $\mathbf{B}_{\text{eff}} \propto \boldsymbol{\kappa}$ depends on the magnitude and direction of the electron momentum. It leads therefore to precession of the spin around the direction of $\boldsymbol{\kappa}$, with frequency Ω_0 . If the energy of the electron is so high that $\Omega_0\tau_p > 1$, each of the branches of the spectrum with definite spin direction must be considered separately; in this case $1/\tau_s \approx 1/\tau_p$.

For thermalized electrons, the inverse condition $\Omega_0\tau_p < 1$ is practically always satisfied. In this case, within the time between collisions that change the precession axis, the electron spin does not manage to deviate considerably from

the initial direction, and the spin relaxation is substantially slowed down. We then have $\tau_s^{-1} \sim \langle \Omega_0^2 \tau_p \rangle$, where the angle brackets denote averaging over the directions of the momentum and over the energy distribution of the electrons. For thermalized electrons we have in the nondegenerate case according to Ref. 6

$$\frac{1}{\tau_s} = q\alpha^2 \frac{T^3}{\hbar^2 E_g} \tau_p, \quad (5)$$

where T is the temperature in energy units. The numerical factor q reflects the change of the results of the averaging for different momentum relaxation mechanisms. In scattering of electrons by charged impurities we have $q = 1.5$; in scattering of acoustic phonons by a deformation potential $q = 3$.

c. The Bir-Aronov-Pikus mechanism

In p -type crystals the electrons can also be scattered by holes, with simultaneous spin flip. As shown in Ref. 8, the probability of such processes is determined by the exchange interaction of the electrons and holes. In cubic crystals with degenerate valence band Γ_8 , the exchange-interaction Hamiltonian can be written in the form⁷

$$H = D(\mathbf{J}\boldsymbol{\sigma})\delta(\mathbf{R})\delta(\mathbf{p}, \mathbf{p}'), \quad (6)$$

where \mathbf{J} is the hole angular-momentum operator, $\boldsymbol{\sigma}$ are Pauli matrices for the electrons, D is the exchange-interaction constant, \mathbf{R} is the difference of the coordinates of the electron and the hole, and \mathbf{p} is their total momentum.

The effectiveness of the exchange mechanism of the spin relaxation depends strongly on the state of the holes in the crystal and turns out to be different for free and bound holes. It changes also with the ratio of the electron and hole velocities v_e and v_h , and with the degree of Fermi degeneracy for the holes. This leads to a complicated dependence of τ_s on the hole density and on the temperature. We present the expressions obtained in Ref. 8 for $1/\tau_s$ in typical cases.

In scattering of electrons by holes bound to acceptors,

$$\frac{1}{2\tau_s} = \frac{1}{\tau_0} \frac{v_e}{v_B} (N_A a_B^3), \quad (7)$$

where

$$\frac{\hbar}{\tau_0} = \frac{\pi}{2} \frac{\overline{\langle D_s^2 \rangle}}{E_B}, \quad \overline{\langle D_s^2 \rangle} = \frac{5}{32} \Delta_{\text{exc}}^2, \quad (8)$$

here N_A is the acceptor density, $v_B = \hbar/\varepsilon_e a_B$, while $E_B = \hbar^2/2m_e a_B^2$, and a_B are respectively the Bohr energy and radius for the exciton, and $\overline{\langle D_s^2 \rangle}$ is the exchange constant expressed in terms of the exchange splitting of the exciton ground state Δ_{exc} . Equations (6) and (7), as well as the ones presented below, were derived under the assumption $m_e \ll m_h$, where m_h is the effective mass of the heavy holes.

When electrons relax on free nondegenerate holes we have

$$\frac{1}{2\tau_s} = \frac{1}{\tau_0} \frac{v_e}{v_B} |\psi(0)|^4 (N a_B^3), \quad \overline{\langle D_s^2 \rangle} = \frac{3}{32} \Delta_{\text{exc}}^2. \quad (9)$$

Here N is the density of the free holes. In the case of free electrons and holes, the Coulomb interaction of the carriers

leads to an increase of the scattering cross section as a result of the increase in the electron density $|\psi(0)|^2$ near the hole. According to Ref. 26, the Sommerfeld factor for an unscreened Coulomb potential is

$$|\psi(0)|^2 = \frac{2\pi}{\kappa} (1 - e^{-2\pi/\kappa})^{-1}, \quad \kappa = \left(\frac{\epsilon_e}{E_B}\right)^{1/2}. \quad (10)$$

In relaxation of electrons by degenerate holes in the case of "fast" electrons $v_e > v_F$ (v_F is the Fermi velocity of the holes), when the relative velocity of the particles is determined by the velocity of the electron,

$$\frac{1}{2\tau_s} = \frac{3}{2} \frac{1}{\tau_0} \frac{v_e}{v_B} \frac{T}{\epsilon_F} |\psi(0)|^4 (Na_B^3). \quad (11)$$

Here ϵ_F is the hole energy at the Fermi level. Equation (11) is valid at not too strong a degeneracy, when $v_e > v_F$, but $\epsilon_e/T < v_e/v_F$. For hot electrons with $\epsilon_e/T > v_e/v_F > 1$, i.e., at $\epsilon_e/T > m_h T/m_e \epsilon_F > 1$, it follows from Eq. (9) on Ref. 8 that

$$\frac{1}{2\tau_s} = \frac{1}{\tau_0} \frac{v_F}{v_B} \frac{\epsilon_e}{\epsilon_F} |\psi(0)|^4 (Na_B^3). \quad (12)$$

For degenerate holes, owing to the screening of the Coulomb potential at the Debye radius L_D , the factor $|\psi(0)|^4$ decreases drastically. However, as shown by model calculations,²⁷ total screening, i.e., equality of $|\psi(0)|^2$ to unity, is reached at carrier densities considerably exceeding the values corresponding to the onset of degeneracy.

Let us note the main differences that make it possible to distinguish between the indicated spin-relaxation mechanisms. The main difference between the DP and EYa mechanisms is their inverse dependence on τ_p ; according to (1) and (5) we have $\tau_{sEYa}^{-1} \propto \tau_p^{-1}$, whereas $\tau_{sDP}^{-1} \propto \tau_p$. Therefore τ_{sEYa} decreases with decreasing mobility, while τ_{sDP} increases. When scattering by the lattice vibrations predominates for both mechanisms, τ_s is independent of the hole density and decreases rapidly with increasing temperature. In the case of scattering by impurities whose density is equal to the hole density, $\tau_{sEYa}^{-1} \propto N_A$, and $\tau_{sDP}^{-1} \propto 1/N_A$. For the BPA mechanism, in the region of nondegenerate or bound holes, we also have $\tau_{sBAP}^{-1} \propto N_A$, and on going into the region of degeneracy the growth of τ_s^{-1} slows down: since $\epsilon_F \propto N^{2/3}$ we have according to (11) and (12) in this region $\tau_{sBAP}^{-1} \propto N^{1/3}$. The temperature dependence of τ_{sBAP} is determined primarily by the temperature dependence of $|\psi(0)|^4$, and in the region where there are both bound and free holes it is determined by the change of the relative densities with changing temperature. The change of the Sommerfeld factor with changing density as a result of screening can also influence strongly the dependence of τ_{sBAP} on N .

§3. EXPERIMENTAL PROCEDURE

The spin-relaxation times were measured with the aid of the optical orientation method.^{28,29} The method is based on the possibility of producing a nonequilibrium electron distribution over the spin states of the conduction band upon excitation of the crystal by circularly polarized light. The processes of spin relaxation lead, during the lifetime of the

electrons in the band, to partial equalization of the populations of the spin states. However, the residual disequilibrium of the population causes the appearance of circular polarization of the luminescence spectra. The degree P of polarization of the luminescence is proportional in this case to the degree of orientation of the electrons and is given by

$$P = P_0 (1 + \tau/\tau_s)^{-1}; \quad (13)$$

here τ is the electron lifetime and P_0 is the maximum possible degree of polarization, determined in the absence of spin relaxation by the selection rules for the interband transitions. For cubic crystals of III-V compounds, including GaAs and GaSb, we have $P_0 = 0.25$.

In a magnetic field \mathbf{B} transverse to the light propagation direction, depolarization of luminescence takes place on account of precession of the electron spin (the Hanle effect). The decrease of the degree of polarization of luminescence with increasing field is described by the expression

$$P(B) = P(0) (1 + \Omega_L^2 T_s^2)^{-1}, \quad (14)$$

where $\tau_s^{-1} = \tau^{-1} + \tau_s^{-1}$, $\Omega_L = g_e \mu_B B / \hbar$ is the Larmor precession frequency, g_e is the electron g -factor, and μ_B is the Bohr magneton. From (13) and (14) it follows that by measuring the degree $P(0)$ of polarization of the luminescence and its dependence on the magnetic field it is possible to determine independently the times τ and τ_s . Usually T_s is determined from the value of $B_{1/2}$ corresponding to $P(B_{1/2}) = 1/2P(0)$.

We investigated bulk GaAs and GaSb crystals doped with a shallow acceptor impurity, namely zinc, in the density interval $10^{17} < N_A < 5 \times 10^{19} \text{ cm}^{-3}$. For the characteristic state of the holes in the investigated density interval it is important to note that metallization of the shallow acceptors sets in GaAs at $N_{cr} = 4 \times 10^{18} \text{ cm}^{-3}$, and GaSb at $N_{cr} = 1.2 \times 10^{18} \text{ cm}^{-3}$ (see Ref. 30). Thus, the bound states of the holes on the acceptors are preserved only at $N_A < N_{cr}$. At higher doping levels, localization of the holes takes place, and the Fermi level for the holes penetrates according to Ref. 31 into the valence band to a depth $\epsilon_F = 0.6 \times 10^{-14} N^{2/3}$ in GaAs and $\epsilon_F = 1.0 \times 10^{-14} N^{2/3}$ in GaSb (ϵ_F is expressed here in eV, and N in cm^{-3}). It can be easily seen that at low temperatures $T \approx 10 \text{ K}$, the delocalized holes are degenerate at all $N > N_{cr}$. In the case of strong doping $N \geq 10^{19} \text{ cm}^{-3}$, the hole degeneracy is preserved also at temperatures close to room temperature.

The experiment was performed with the aid of the standard apparatus for investigations by optical orientation.^{1,2} The luminescence of the GaAs crystals was excited with a krypton laser at a wavelength 7525 \AA (1.647 eV). In the case of GaSb crystals, a helium-neon laser was used a wavelength $1.52 \mu\text{m}$ (0.814 eV). The excitation density in all the experiments did not exceed 10 W/cm^2 . At these excitation levels and at the electron lifetimes $10^{-9} - 10^{-10} \text{ sec}$ determined in the present study, the densities of the nonequilibrium carriers were always lower by several orders of magnitude than the density of the intrinsic holes in the samples.

A feature of the luminescence spectra of the investigated crystals is the presence of luminescence lines correspond-

TABLE I. Momentum relaxation times for holes in the investigated GaAs and GaSb crystals.

GaAs			GaSb		
N_A, cm^{-3}	$\tau_{ph} \cdot 10^{13}, \text{sec}$		N_A, cm^{-3}	$\tau_{ph} \cdot 10^{13}, \text{sec}$	
	$T=77 \text{ K}$	$T=300 \text{ K}$		$T=77 \text{ K}$	$T=300 \text{ K}$
$2.2 \cdot 10^{17}$	3.3	1.1	$1.7 \cdot 10^{17}$	4.5	1.5
$1.2 \cdot 10^{18}$	0.4	0.5	$2.9 \cdot 10^{18}$	0.7	0.65
$4.1 \cdot 10^{19}$	0.3	0.2	$1.3 \cdot 10^{19}$	0.5	0.5

ing to recombination of free electrons with holes situated on acceptors or in the valence band.^{2,11,31} Direct study of the circular polarization of such bands has made it possible to obtain the times of the spin relaxation of the free electrons in the conduction band.

When determining the efficiency of different spin-relaxation mechanisms, it became necessary in this study to estimate the electron-momentum relaxation times. Since there are no direct measurements of electron mobilities in *p*-type crystals, while the theoretical calculations in the region where several mechanisms of scattering play a role are not reliable, we have assumed that the electron and hole relaxation times are of the same order. For nondegenerate electrons and holes, the ratio of the relaxation times does not exceed the square root of the ratio of their masses. In Table I are given the values of τ_p calculated for the holes from their mobilities.

Table II lists the parameters of the GaAs and GaSb crystals used in the study. The following notation is used: E_g is the width of the forbidden band, Δ is the energy of the spin-orbit splitting of the valence band, ϵ_0 is the dielectric constant, m_h is the effective mass of the heavy hole, m_e is the effective mass of the electron, m is the reduced mass of the exciton, a_B is the exciton radius, E_B is the exciton binding energy, v_B is the characteristic velocity, E_A is the ionization energy of single acceptor, and g_e is the *g* factor of the electron in the conduction band.

§4. EXPERIMENTAL RESULTS AND THEIR DISCUSSION
a. Dependence of the spin-relaxation times on the doping level

Figures 1 and 2 show the concentration dependences of the spin-relaxation times τ_s obtained at various temperatures for *p*-type GaAs and GaSb crystals. In the investigated doping and temperature intervals, the times τ_s vary in a wide range from 10^{-8} to 10^{-11} sec. Increasing the doping and

raising the temperature lead to a decrease of the times τ_s . It is of interest to note the rather weak dependence of the times τ_s on the doping level: increasing the acceptor density by three orders of magnitude decreases the times τ_s by only several times. Also noteworthy is the great similarity of the dependences of the times τ_s on N_A in both compounds.

It is easy to show that the observed changes of the times τ_s are not due to the action of the EYA mechanisms. Indeed, using expression (1), which characterizes the efficiency of the EYA mechanism, and substituting in it the known parameters of the investigated compounds (see Table II) as well as the values of the times τ_p determined from the carrier mobility, we obtain the following estimates for the time τ_s : as functions of the doping level they should vary at 77 K from 2×10^{-7} to 1.8×10^{-6} sec in GaAs and from 1.4×10^{-8} to 1.6×10^{-9} sec in GaSb, while at 300 K they should vary from 6×10^{-8} to 1.5×10^{-8} sec in GaAs. The estimates obtained exceed by more than two orders the corresponding experimental values. This discrepancy exceeds substantially the possible inaccuracy in the estimates of the times τ_s . Thus, at 77 K the scattering of the electrons in doped crystals is due mainly to charged impurities and the value of the coefficient *A* in expression (1) is close to unity. Therefore the inaccuracy of the estimates of τ_s can be due only to the inaccuracy in the determination of the times τ_p , which is patently smaller than two orders. The weak dependence of τ_s on N_A in the density range 10^{17} – 10^{18} cm^{-3} at high temperatures likewise does not agree with the severalfold decrease of the times τ_p in this range.

The observed decrease of times τ_s with increase in doping can likewise not be attributed to a manifestation of the DP mechanism, for when the doping is increased its effectiveness can only decrease. In the case when the DP mechanism predominates, the times τ_s should not decrease in the considered range of densities in accordance with expression (5), but, conversely, increase in inverse proportion to the times τ_p .

We consider now in greater detail the density dependences of the times τ_s in Figs. 1 and 2, paying first attention to the curves obtained at lower temperatures. At doping levels $N_A < N_{cr}$, the times τ_s decrease in inverse proportion to the acceptor density region N_A . In the density region $N_A \approx N_{cr}$, the decrease of the times τ_s stops, as is particularly clearly seen for the GaAs crystals. Further increase of the doping leads to a new decrease of the times τ_s , but now in proportion to $N_A^{-1/3}$.

TABLE II. Parameters of the GaAs and GaSb crystals.

	GaAs	GaSb		GaAs	GaSb	
E_g	$T=4,2 \text{ K}$ $T=77 \text{ K}$ $T=300 \text{ K}$	1.520 eV 1.509 eV 1.43 eV	0.814 eV 0.798 eV 0.69 eV	$m = m_e m_h / (m_e + m_h)$ $a_B = \hbar^2 \epsilon_0 / e^2 m$ $E_B = \hbar^2 / 2 m a_B^2$	0.06 m_0 114 Å 4.9 meV	0.037 m_0 210 Å 2.3 meV
Δ		0.35 eV	$v_B = \hbar / m a_B$	$1.7 \cdot 10^7 \text{ cm/sec}$	$1.4 \cdot 10^7 \text{ cm/sec}$	
ϵ_c		13.1	E_A	33 mēB	24 meV	
m_e		0.067 m_0	g_e	-0.44	-9.3	
m_h		0.48 m_0				

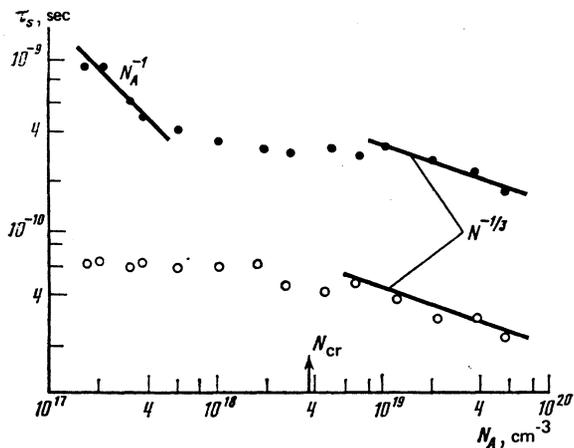


FIG. 1. Dependences of the times τ_s on the density of the shallow acceptors Zn in GaAs crystals at $T = 77$ K (●) and at $T = 300$ K (○).

The observed features of the density dependences of the times τ_s agree in detail with the predictions of the theory for the case of manifestation of the BAP mechanism. In the density region $N_A < N_{cr}$, the holes are distributed among the localized states on the acceptors and the states of the valence band. In this case the spin-relaxation rate of the electrons should be characterized by the sum of expressions (7) and (9), i.e.,

$$\frac{1}{2\tau_s} = \frac{1}{\tau_0} (N_A a_B^3) \left(\frac{N}{N_A} |\psi(0)|^4 + \frac{5}{3} \frac{N_A - N}{N_A} \right) \frac{v_e}{v_B}. \quad (15)$$

For the crystals GaAs at 77 K, the degree of ionization of the acceptors in the density range 10^{17} – 10^{18} cm^{-3} is approximately 30% and depends little on the doping level. This constancy is due to the fact that the increase of the density of states on the acceptors with increasing doping is offset by the accompanying decrease of the acceptor binding energy.³² Inasmuch as in this density region the screening by the holes is still insignificant, expression (15) leads to an experimentally observable dependence $\tau_s \propto N_A^{-1}$. A similar de-

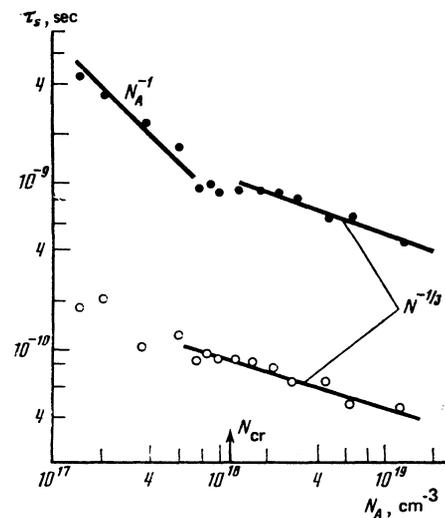


FIG. 2. Dependences of the times τ_s on the concentrations of the shallow acceptors Zn in GaSb crystals at $T = 15$ K (●) and $T = 77$ K (○).

pendence is obtained for GaSb crystals, if it is recognized that $T = 15$ K and $N_A < N_{cr}$ the degree of ionization of the acceptors for them is negligibly small.

The weak dependence of τ_s on N_A in the region $N_A \approx N_{cr}$ for GaAs can be explained, naturally, as being due to the decrease of the Sommerfeld factor $|\psi(0)|^2$, which cancels out almost completely the decrease of τ_s with increasing acceptor density. Thus for GaAs at 77 K and $N_A \approx 5 \times 10^{17}$ cm^{-3} , when there is no screening, $|\psi(0)|^4 = 20$. At densities $N \approx 10^{19}$ cm^{-3} we have $|\psi(0)|^4 \approx 1$, and consequently in this density region the product $N_A |\psi(0)|^4$ is practically constant.

In GaSb at $T = 15$ K and $N_A < 10^{18}$ cm^{-3} the electron scattering is primarily by bound holes, for which there is no factor $|\psi(0)|^4$. Therefore the decrease of the Sommerfeld factor for the free holes manifests itself in this case much more weakly than for GaAs.

In degenerate crystals, the value of the Sommerfeld factor is already small, and as shown by experimental investigations³³ and theoretical calculations,²⁷ at $N a_B^3 \gtrsim 10$, which corresponds to a density $N \approx 10^{18}$ cm^{-3} for GaSb and $N \approx 10^{19}$ cm^{-3} for GaAs, the values of $|\psi(0)|^2$ lie between 2 and 1. Further decrease of $|\psi(0)|^2$ with increasing doping is very slow because of the weak dependence of the screening radius on the hole density: $L_D \propto N^{-1/6}$. Without allowance for the dependence of $|\psi(0)|^4$ on N we have according to (11) $\tau_s \propto N^{-1/3}$, which agrees with the experimental data shown in Figs. 1 and 2.

To conclude the analysis of the concentration dependences of the times τ_s in GaAs and GaSb crystals, it remains to note that in the region of hole degeneracy a temperature rise does not change the character of the concentration dependence. On the other hand, at weaker doping $N < N_{cr}$ the dependence of the times τ_s on the acceptor density becomes noticeably weaker with rising temperature. The reason for this difference was made clear by separate studies of the temperature dependences of the times τ_s for the doping regions $N_A < N_{cr}$ and $N > N_{cr}$.

b. Temperature dependence of the spin-relaxation times in degenerate crystals ($N > N_{cr}$).

The temperature dependences of the times τ_s for several samples of degenerate crystals GaAs and GaSb are given in Fig. 3. The degree of doping of the samples was chosen such that the hole degeneracy was preserved in the entire investigated temperature interval. The curves shown in Fig. 3 reveal the same variation of the times τ_s with temperature for all the investigated samples: at low T the times τ_s vary little with temperature, and then begin to decrease rapidly in proportion to $T^{-3/2}$. This $\tau_s(T)$ dependence cannot be explained by the EYa and DP mechanisms when scattering by impurities predominates, when $\tau_p \sim T^{3/2}$, and according to (1) and (5) we have $\tau_{sEYa} \sim T^{-1/2}$ and $\tau_{sDP} \sim T^{-9/2}$.

The decrease of the times $\tau_s \sim T^{-3/2}$, which was reliably observed in a wide temperature interval, agrees well with the theoretical relation (11) for the case when the electrons that relax on the holes are fast, i.e., $v_e > v_F$. In GaAs and GaSb, even in the case of strong degeneracy on account of the large

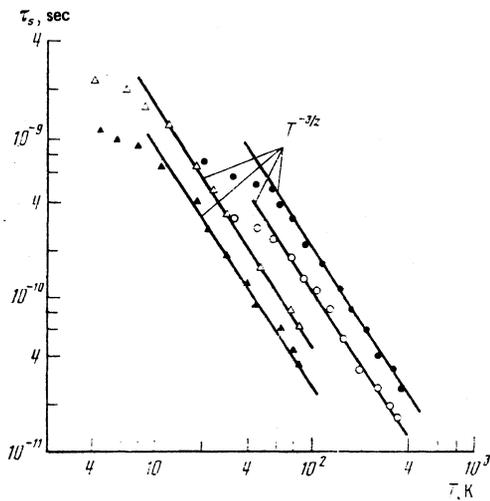


FIG. 3. Temperature dependences of the times τ_s in degenerate GaAs and GaSb crystals: \blacktriangle —GaSb, $N_A = 6.5 \times 10^{18} \text{ cm}^{-3}$; \triangle —GaSb, $N_A = 1.8 \times 10^{18} \text{ cm}^{-3}$; \circ —GaAs, $N_A = 5.5 \times 10^{19} \text{ cm}^{-3}$; \bullet —GaAs, $N_A = 7.8 \times 10^{18} \text{ cm}^{-3}$.

difference between the effective masses of the electrons of the heavy holes, the condition $v_e > v_F$ is satisfied even at relatively low temperatures: starting with 10–20 K for GaSb and with 20–35 K for GaAs.

The dependences of the times τ_s on the temperature and on the doping level, obtained for degenerate crystals, offer evidence of the predominant role of the BAP mechanism. In the strong-doping region, the dependences of the times τ_s on the hole density and on the temperature are well described by the following expressions [(16a) for GaAs and (16b) for GaSb]:

$$\tau_s^{-1} = 2.3 \cdot N^{1/2} T^{3/2}, \quad (16a)$$

$$\tau_s^{-1} = 14.1 \cdot N^{1/2} T^{3/2}, \quad (16b)$$

where τ_s is in seconds, N is in cm^{-3} and T is in $^\circ\text{K}$.

From a comparison of expressions (16a) and (16b) with (11) we obtained the values of the parameter τ_0 and of the energy of the exchange splitting of the ground state of the exciton Δ_{exc} in the investigated crystals: for GaAs we have $\tau_0 = 1.0 \times 10^{-8} \text{ sec}$ and $\Delta_{\text{exc}} = 4.7 \times 10^{-5} \text{ eV}$, while for GaSb we have $\tau_0 = 1.8 \times 10^{-8} \text{ sec}$ and $\Delta_{\text{exc}} = 2.4 \times 10^{-5} \text{ eV}$. The value of Δ_{exc} obtained for GaAs agrees well with its known estimates.^{34,35} The value of Δ_{exc} for GaSb was obtained here for the first time. The values of Δ_{exc} were chosen for best agreement between the theory and the experimental data also for nondegenerate crystals (see below). For strongly degenerate crystals it was necessary to set the factor $|\psi(0)|^2$ equal to 1.5 for GaAs and to 1.7 for GaSb, values that do not contradict the estimates above.

Let us discuss the possible causes of the saturation of the $\tau_s(T)$ dependences with decreasing temperature. As can be seen from Fig. 4, in GaAs crystals this saturation begins already at rather high temperatures $T \approx 40\text{--}60 \text{ K}$. In Ref. 9, a weak dependence of the times $\tau_s \sim T^{-1/2}$ in GaAs was observed up to 100 K. We note immediately that the observed saturation of times τ_s with decrease in temperature cannot

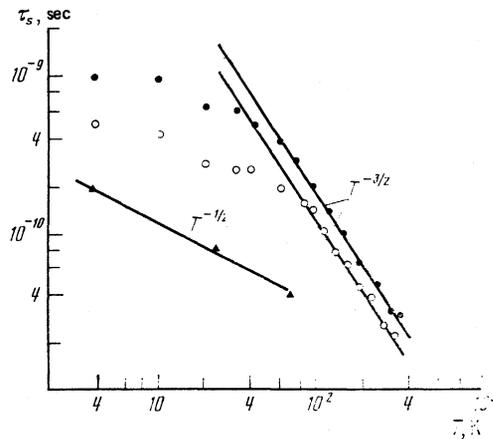


FIG. 4. Temperature dependences of the times τ_s in degenerate GaAs crystals with different doping levels and different photoelectron lifetimes: \bullet — $N_A = 4 \times 10^{18} \text{ cm}^{-3}$, $\tau = 0.6 \times 10^{-3} \text{ sec}$ (from the data of Ref. 9), \circ — $N_A = 5 \times 10^{18} \text{ cm}^{-3}$, $\tau = 2.8 \times 10^{-10} \text{ sec}$; \circ — $N_A = 4 \times 10^{19} \text{ cm}^{-3}$, $\tau = 2.4 \times 10^{-10} \text{ sec}$.

be attributed to a transition to the case of slow electrons, inasmuch as in accordance with the theory⁸ one should expect in this case an even stronger dependence, $\tau_s \sim T^{-2}$.

It seems that the cause of the observed behavior of the times may be the incomplete thermalization of the electrons in the conduction band during the lifetime. The degenerate crystals investigated by us are characterized by rather short lifetimes τ : $(2\text{--}3) \times 10^{-10} \text{ sec}$ in GaAs and $(4\text{--}5) \times 10^{-10} \text{ sec}$ in GaSb. These times are governed by the radiative transitions and depend little on the doping level and on the temperature.^{31,36}

Let us compare these values of τ with the energy relaxation times τ_s of nonequilibrium electrons in the passive energy region, where the energy relaxation of the electrons is by scattering from acoustic phonons or else from holes with energies close to the Fermi level. According to Ref. 37, the times of energy relaxation of the electrons due to scattering by acoustic phonons, τ_{eac} , lie for the investigated crystals in the range $10^{-9}\text{--}10^{-8} \text{ sec}$. These values exceed the lifetimes substantially.

Calculation shows that the rates of the energy relaxation in scattering by degenerate holes is determined by the expression

$$\tau_{e,h}^{-1} = \frac{4\pi}{105\hbar} \frac{m_e}{m_h} \frac{e_e^2}{\epsilon_F} F(\gamma); \quad (17)$$

here $\gamma = v_e/v_F$ and $F(\gamma > 1) = 1$, while $F(\gamma < 1) = 1 - (1 - \gamma)^5 [1 + 5\gamma + 15\gamma^2]$. The times $\tau_{e,h}$ can vary, depending on the electron energy and on the degree of degeneracy of the holes, in a rather wide range, from 10^{-12} to 10^{-8} sec . In the region of the lowest temperatures $T \lesssim 10 \text{ K}$ we have according to (17) $\tau_{e,h} > 0.8 \times 10^{-8} \text{ sec}$ at $N \approx 5 \times 10^{18} \text{ cm}^{-3}$ and $\tau_{e,h} > 2 \times 10^{-8} \text{ sec}$ at $N \approx 3.8 \times 10^{19} \text{ cm}^{-3}$. In both cases these times exceed considerably the lifetimes, which are respectively 2.8×10^{-10} and $2.4 \times 10^{-10} \text{ sec}$ for these samples. Therefore under these conditions the electron temperature certainly cannot manage to reach that of the lattice.

If the saturation of the temperature dependences of the times τ_s is indeed due to heating of the electrons, their average temperature T_e can be obtained with the aid of expression (12). If it is assumed that the factor $|\psi(0)|^4$ is independent of τ_e in the strong-screening region, it follows from (11) and (12) that

$$T_e = T \frac{\tau_s(T)}{\tau_s(T_e)} \left(\frac{3 m_h T}{2 m_e \epsilon_F} \right)^{1/2}, \quad (18)$$

where $\tau_s(T)$ is the spin-relaxation time for thermalized electrons (i.e., according to Fig. 4 at $T > 50$ K), and $\tau_s(T_e)$ is the time of the spin relaxation of the hot electrons. An estimate based on Eq. (18) yields $T_e \approx 40$ K for a sample with $N = 3.8 \times 10^{19} \text{ cm}^{-3}$, and $T_e \approx 29$ K for a sample with $N = 5 \times 10^{18} \text{ cm}^{-3}$. For electrons with such an energy the times τ_{eh} are of the order of 10^{-10} sec.

The temperature dependence of the times τ_s , obtained in Ref. 9 for a GaAs crystal with approximately the same doping level, differ strongly from those observed by us. This difference cannot be attributed to heating of the electrons, even if account is taken of the very short lifetime 6×10^{-11} sec of the electrons in the investigated crystal. To this end it would be necessary to assume that the electrons become thermalized only above room temperature.

At the same time, such short lifetimes offer evidence of the presence in the investigated crystal of additional recombination centers, which usually have paramagnetic properties in III-V compounds. Scattering by such centers could explain also the observed $\tau_s \propto T^{-1/2}$ dependence.

c. Temperature dependence of the times of spin relaxation in nondegenerate crystals ($N_A < N_c$)

In this doping region, in the case of the action of the BAP mechanism, the rate of the spin relaxation of the electrons should be described by expression (15). The temperature dependence of the times τ_s is connected here not only with electron velocity but also with the temperature-dependence of the degree of the acceptor ionization and with the decrease of the Sommerfeld factor $|\psi(0)|^2$. A characteristic feature of the dependence is its nonmonotonicity: the acceptor depletion region the fast decrease of the Sommerfeld factor should lead to a slowing down of the spin relaxation. In this case the time τ_s is proportional to v_e and should increase with increasing temperature $\propto T^{1/2}$.

The temperature dependences of the times τ_s in nondegenerate crystals GaAs and GaSb are shown in Figs. 5a and 5b and in Fig. 6a, respectively. The solid lines in the figures show the theoretical curves calculated with the aid of expression (15) at the values indicated above, Δ_{exc} (GaAs) = 4.7×10^{-5} eV and Δ_{exc} (GaSb) = 2.4×10^{-5} eV. With allowance for the small role of the screening in the investigated samples, the values of $|\psi(0)|^2$ were obtained from (10). The degree of ionization of the acceptors was calculated from the known formulas³⁸ with account taken of the dependence of the acceptor ionization energy E_A on the doping level.³² It can be seen that the obtained theoretical dependences describe well the experimental data also in the case of nondegenerate crystals. The theory explains also the

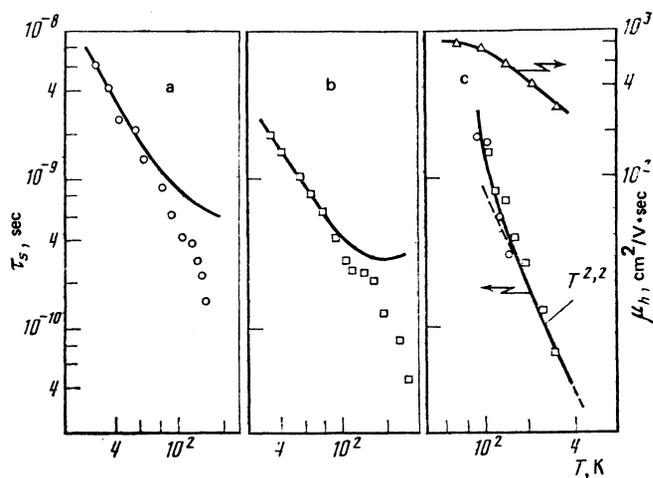


FIG. 5. Temperature dependences of the times τ_s in nondegenerate GaAs crystals: a, b) \circ, \square —experimental data; solid lines—theoretical dependences for the BAP mechanism according to (15); a — $N_A = 2.2 \times 10^{17} \text{ cm}^{-3}$, b — $N_A = 3.5 \times 10^{17} \text{ cm}^{-3}$, c) \circ, \square —times $(1/\tau_s) = (1/\tau_s)_{\text{expt}} - (1/\tau_s)_{\text{BAP}}$, solid line—theoretical dependence for the DP mechanism according to (5), Δ —temperature dependence of the Hall mobility μ_h .

saturation of the $\tau_s(T)$ dependences which sets in GaAs at $T \approx 100$ – 150 K.

At $T > 150$ K, however, for GaAs and $T > 40$ K for GaSb, instead of saturation of the $\tau_s(T)$ dependence expected in accordance with the theory of the BPA mechanism one observed in experiment a further decrease of the times τ_s . This decrease is obviously evidence that one more mechanism of spin relaxation comes into play. In Fig. 5a and 6b are shown the values of the times $(1/\tau_s) = (1/\tau_s)_{\text{expt}} - (1/\tau_s)_{\text{BAP}}$, determined from the experimental values in Figs. 5a and b and Fig. 6a, and corresponding to the demonstrated mechanism. The same figures show the temperature dependences of the hole mobilities in the investigated crystals; this gives an idea of the temperature dependence of the times τ_p .

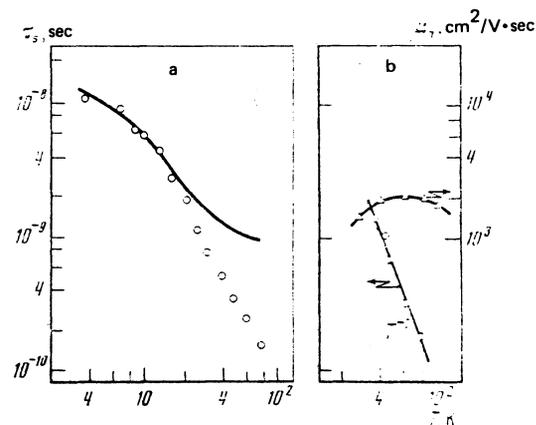


FIG. 6. Temperature dependence of the times τ_s in nondegenerate GaSb crystals: a) \circ —experimental data, solid line—theoretical dependence for the BPA mechanism according to (15), $N_A = 2.8 \times 10^{17} \text{ cm}^{-3}$; b) \circ —times $(1/\tau_s) = (1/\tau_s)_{\text{expt}} - (1/\tau_s)_{\text{BAP}}$, solid line—theoretical dependence for the DP mechanism according to (5), Δ —temperature dependence of the hole mobility μ_h .

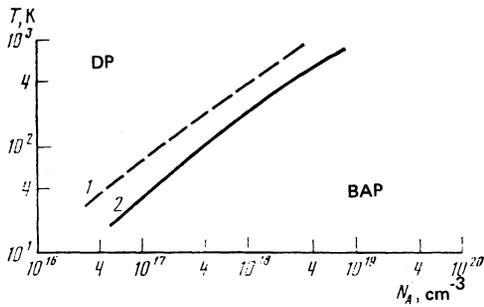


FIG. 7. Boundaries between the temperature regions and the hole-density regions in which the BAP and DP mechanism predominate, for crystals GaAs (curve 1) and GaSb (curve 2).

The temperature dependences of the times τ_s obtained in Fig. 5c and 6b cannot be attributed to a manifestation of the EYa mechanism. It was already noted earlier that the times τ_s corresponding to the EYa mechanism are substantially longer than the measured values. It is also impossible to explain within the framework of the EYa mechanism the character of the obtained $\tau_s(T)$ dependences. Thus, for GaSb, according to (1) and when account is taken of the constancy of the times τ_p in the considered temperature interval, the times τ_s should have changed in proportion to T^{-2} , and in GaAs and $T > 100$ K one should expect a relation $\tau_s \propto T^{-3}$, inasmuch as the times τ_p decrease here approximately in proportion to $T^{-0.8}$.

The observed $\tau_s(T)$ dependences found a natural explanation within the framework of the DP mechanism. The constancy of the times τ_p in EYa leads, in accordance with (5), to an experimental relation $\tau_s \propto T^{-3}$, and the shortening of the times $\tau_p \propto T^{-0.8}$ in GaAs leads to a relation $\tau_s \propto T^{-2.2}$ at $T > 100$ K. The increase of the slope of the plot of $\tau_s(T)$ at $T > 100$ K in GaAs agrees with the slowing down of $\tau_p(T)$ in this case. The quantitative agreement between the experiment and the theory was reached by choosing the parameter α in (5) equal to 0.05 for GaAs and 0.1 for GaSb. The obtained values of α agree well with their theoretical estimate (4). Thus, our data have confirmed the conclusion drawn in Ref. 11 that the DP mechanism comes into play in moderately doped GaAs crystals.

The experimental investigations and the theoretical analysis of the obtained data have made it possible to establish reliably the mechanisms of the spin relaxation which predominate under conditions of optical orientation in the crystals GaAs and GaSb. Figure 7, plotted on the basis of the data presented above, show the boundaries of the temperature regions and of the hole-density regions within which the BAP and DP mechanisms predominate. On the boundaries, the contributions of both mechanisms become equalized. According to the obtained estimate, the contribution of the EYa mechanism in the entire range of values of N_A and T indicated in Fig. 7 is smaller than the contributions of the BAP and DP mechanisms.

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Note added to proof (26 January 1983). Later measurements of spin relaxation in pure GaAs crystals in a magnetic field, the results of which will be published separately, have made it possible to measure τ_p directly and by the same token refined the value of α , which was found to equal 0.06 (in place of the value $\alpha = 0.05$ indicated in the text for GaAs).

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