

Spin relaxation of photoelectrons in *p*-type gallium arsenide

Guy Fishman

*Groupe de Physique des Solides\* de l'Ecole Normale Supérieure, Université Paris VII, 75221 Paris Cédex 05, France*

Georges Lampel

*Laboratoire de Physique de la Matière Condensée,† Ecole Polytechnique, 91128 Palaiseau Cédex, France*

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We have measured by optical-pumping methods the spin-relaxation time  $T_1$  of photocreated conduction electrons in *p*-type GaAs ( $N_A = 4 \times 10^{18} \text{ cm}^{-3}$ ) as a function of temperature. To analyze our results we present a detailed discussion of the possible relaxation mechanisms in *p*-type semiconductors. The electronic spin relaxation may originate from: (i) the splitting of the conduction band, (ii) the spin-orbit interaction, (iii) the hyperfine interaction with nuclei of the host crystal, and (iv) the exchange interaction between electrons and holes. The spin-relaxation time is given in each case as a function of experimentally attainable parameters and permits one to obtain a numerical result for all usual III-V compounds. It is established that in doped *p*-type GaAs the exchange interaction with the holes is the dominant relaxation mechanism at low temperatures, the other mechanisms being too weak by several orders of magnitude. For higher temperatures ( $T \geq 100^\circ\text{K}$ ), the relaxation due to the  $k^3$  splitting of the conduction band may become predominant for  $N_A \lesssim 10^{17} \text{ cm}^{-3}$ . The theory is compared with experimental data available in the literature and with our experiments. From our measurements we obtain in GaAs the value of the exchange splitting of  $1s$  exciton  $\Delta_{x,1s} \simeq 0.1 \text{ meV}$ . We also show that the observed spin depolarization at high kinetic energy (200–300 meV) can be explained by the splitting of the conduction band, taking into account the energy relaxation by optical-phonon scattering.

## I. INTRODUCTION

The investigation of many problems related to spin relaxation of electrons in semiconductors has been improved by the use of the optical-pumping techniques, i.e., the photoexcitation of carriers by circularly polarized light.<sup>1-3</sup> In particular this method is very well fitted to study the spin-relaxation of conduction electrons. Many experimental articles<sup>4-10</sup> have been published dealing with this specific subject and some recent theoretical papers<sup>11-13</sup> give a likely explanation to the results obtained by optical-pumping methods in *p*-type samples. However, up to now, the problem of the electronic spin relaxation in the conduction band was not completely solved: For example, the difference in efficiency of different spin-relaxation mechanisms with respect to different types of semiconductors was not clearly established.

In this paper our main concern is not optical pumping but rather spin relaxation. We focus our attention on some mechanisms which can explain the observed spin-relaxation time of conduction electrons in doped *p*-type gallium arsenide at low temperature ( $T < 100^\circ\text{K}$ ), and we apply the results of our calculations to other usual III-V compounds such as indium antimonide and gallium antimonide. At last, we compare the theory with available data and with our own experimental results obtained in GaAs. In contrast with this last material, the spin relaxation of conduction electrons has been widely studied in *n*-type InSb where the usual con-

duction-electron spin resonance technique can be conveniently applied.<sup>14</sup> Let us emphasize that gallium arsenide and indium antimonide represent two opposite cases. In the first one, the spin-orbit splitting is weak and the energy gap is large, whereas the contrary is true in InSb where it is well known that the conduction-electron spin relaxation originates from the spin-orbit interaction.<sup>14</sup>

We examine here in detail the most current mechanisms: (i) The lack of inversion symmetry in III-V compounds leads to a spin splitting of the conduction band.<sup>15</sup> D'yakonov and Perel<sup>11</sup> proposed a relaxation mechanism due to this splitting, which they analyze in terms of motional narrowing. We shall call this process the DP process. (ii) Taking into account the fact that the wave functions of the conduction band are nonpure spin states<sup>15</sup> due to the spin-orbit interaction, Elliott<sup>16</sup> and later Yafet<sup>17</sup> studied the spin relaxation by scattering on impurities or phonons. This mechanism will be called the EY process. (iii) Another possibility is the spin relaxation of conduction electrons by the hyperfine interaction with the nuclei of the host crystal, which was first studied by Overhauser<sup>18</sup> in metals. (iv) The exchange interaction mechanism between electrons and holes<sup>19</sup> can be very efficient in *p*-type semiconductors. This mechanism was studied in detail by Bir, Aronov, and Pikus,<sup>13</sup> and we shall call it the BAP process.

We do not take into account the spin-orbit part

associated with the impurity potential which is ineffective in III-V compounds.<sup>20</sup> As we deal with strongly doped *p*-type materials we suppose that the donor concentration is negligible. In these conditions we are concerned only by conduction electrons and we assume that there is no trapping on donors, so that the modulation of the hyperfine interaction in donor states will be neglected.<sup>21,22</sup>

In Sec. II, we recall the basic ideas concerning optical pumping in III-V compounds. In Sec. III, we briefly present a simple picture of the spin-relaxation theory, and give a presentation of the above-mentioned mechanisms. In each case the calculation is then carried out so that one can get a numerical result for usual III-V compounds. The relevance of our calculations for GaAs is discussed in connection with experimental results in the literature. Section IV is devoted to the discussion of our own experiments in GaAs. We show that (i) for thermalized electrons the exchange interaction mechanism accounts for our experimental results. This permits one to deduce the exchange splitting  $\Delta_{x,1s}$  of the exciton in the 1s state:  $\Delta_{x,1s} \approx 0.1$  meV; (ii) for electrons with high kinetic energy of the order of some hundreds of meV, the spin relaxation can be explained by the splitting of the conduction band after a reexamination of the conditions of validity of the DP process.

## II. SPIN POLARIZATION IN DIRECT-GAP III-V COMPOUNDS

In Sec. II A we recall briefly the basic ideas concerning optical pumping in III-V compounds. In order to study the spin-relaxation mechanisms of conduction electrons, we need the precise form of the wave functions of the conduction band. They are explicitly written in Sec. II B.

### A. Optical pumping and spin polarization

When a semiconductor is excited by circularly polarized light of energy  $h\nu$  equal to the band gap  $E_G$ , electrons are created in the conduction band and holes in the valence band. In steady-state regime the electronic polarization is given by<sup>1</sup>

$$P = \frac{n_{\uparrow} - n_{\downarrow}}{n_{\uparrow} + n_{\downarrow}} = P_i \frac{T_1}{T_1 + \tau}. \quad (1)$$

Here  $n_{\uparrow}$  ( $n_{\downarrow}$ ) is the steady-state concentration of photocreated electrons with spin up (down) quantized along the direction of propagation of the light,  $P_i$  is the initial polarization which is determined by the symmetry of the conduction and valence bands,  $T_1$  and  $\tau$  are, respectively, the spin-relaxation time and the lifetime of the electrons at the bottom of the conduction band. To obtain separately  $T_1$  and  $\tau$  one can use the Hanle effect

which is the depolarization produced by a transverse magnetic field  $B$ . It is given by<sup>2</sup>

$$P(B) = P_i \frac{T_1}{T_1 + \tau} \frac{1}{1 + [(B/\Delta B)]^2}, \quad (2)$$

where the Hanle linewidth is

$$\Delta B = \frac{\hbar}{|g^*| \mu_B} \left( \frac{1}{T_1} + \frac{1}{\tau} \right), \quad (3)$$

$\mu_B$  being the Bohr magneton and  $g^*$  the effective Landé factor of conduction electrons. In GaAs  $g^* = -0.44$ .<sup>23</sup>

In direct-gap III-V compounds, the initial spin polarization  $P_i$  is equal to  $-0.5$  for  $h\nu = E_G$ .<sup>2,7,11</sup> If the energy  $h\nu$  is larger than the band gap, the initial polarization  $P_i$  remains approximately equal to  $-0.5$  as long as the wave functions at  $k \neq 0$  are not significantly different from the wave functions at  $k = 0$ .<sup>11</sup> In the general case, this holds only for  $(h\nu - E_G) \ll E_G$ . For GaAs this is well verified up to  $h\nu \sim E_G + \Delta$ , where  $\Delta$  is the spin-orbit splitting of the valence band. In GaAs (Refs. 24 and 25)  $E_G = 1.52$  eV and  $\Delta = 0.34$  eV. For  $h\nu > E_G + \Delta$ , the initial polarization decreases and becomes rapidly zero.<sup>11</sup>

### B. Symmetry and wave functions

The conduction band in direct-gap III-V compounds is of  $\Gamma_6$  symmetry<sup>15</sup> at  $\vec{k} = 0$ . The wave functions are  $|S\uparrow\rangle$  and  $|S\downarrow\rangle$ , where  $S$  is a  $\Gamma_1$ -type function and  $\uparrow$  and  $\downarrow$  stand for spin up and down. The valence band is of  $\Gamma_8$  symmetry<sup>15</sup> at  $\vec{k} = 0$ . The wave functions are described by  $|\frac{3}{2}, m\rangle$ , ( $m = \frac{3}{2}, \frac{1}{2}, -\frac{1}{2}, -\frac{3}{2}$ ), the spin being quantized along the  $z$  axis. The wave functions  $|\frac{3}{2}, m\rangle$  are linear combinations of spin up and down and of  $X, Y, Z$  functions which transform like  $p_x, p_y, p_z$  atomic wave functions in the tetrahedral group.

In the conduction band, at  $k \neq 0$ , the periodic parts  $u_{\vec{k}\pm}$  of the Bloch functions  $\Psi_{\vec{k}\pm} = (1/\sqrt{V}) e^{i\vec{k}\cdot\vec{r}} u_{\vec{k}\pm}$ , where  $V$  is the volume of the crystal, are given in the three bands approximation of the  $\vec{k}\cdot\vec{p}$  theory by<sup>26</sup>

$$\begin{aligned} u_{\vec{k}\uparrow} = & \left( iaS - \frac{b - c\sqrt{2}}{2} \frac{k^+}{k} R^- + \frac{b + c\sqrt{2}}{2} \frac{k^-}{k} R^+ + c \frac{k_z}{k} Z \right) \uparrow \\ & - b \left( \frac{k_x}{k} R^+ - \frac{k^+}{k} \frac{Z}{\sqrt{2}} \right) \downarrow, \\ u_{\vec{k}\downarrow} = & \left( iaS + \frac{b + c\sqrt{2}}{2} \frac{k^+}{k} R^- - \frac{b - c\sqrt{2}}{2} \frac{k^-}{k} R^+ + c \frac{k_z}{k} Z \right) \downarrow \\ & + b \left( \frac{k_x}{k} R^- - \frac{k^-}{k} \frac{Z}{\sqrt{2}} \right) \uparrow, \end{aligned} \quad (4)$$

where  $(k_x, k_y, k_z)$  are the components of  $\vec{k}$ ,  $k^{\pm} = k_x \pm ik_y$ ,  $R^{\pm} = (X \pm iY)/\sqrt{2}$ . At a given kinetic energy

$\epsilon$ , the coefficients  $a$ ,  $b$ ,  $c$  may be expressed as functions of  $\eta = \Delta/E_G$  and  $\epsilon/E_G$ . Neglecting higher-order terms in  $\epsilon/E_G$ , one gets

$$\begin{aligned} a &= 1 - \alpha\epsilon/E_G, \\ |b| &= \frac{\sqrt{2}}{3} \frac{\eta}{[(1+\eta)(1+\frac{2}{3}\eta)]^{1/2}} \left(1 - \frac{m^*}{m_0}\right)^{1/2} \left(\frac{\epsilon}{E_G}\right)^{1/2}, \\ |c| &= \left(\frac{1+\frac{2}{3}\eta}{1+\eta}\right)^{1/2} \left(1 - \frac{m^*}{m_0}\right)^{1/2} \left(\frac{\epsilon}{E_G}\right)^{1/2}, \end{aligned} \quad (5)$$

where

$$\alpha = \frac{1}{2} \left(1 + \frac{4}{3}\eta + \frac{2}{3}\eta^2\right) \left(1 + \frac{5}{3}\eta + \frac{2}{3}\eta^2\right)^{-1} \left(1 - m^*/m_0\right)$$

and where  $m^*/m_0$  is the ratio of the conduction-band effective mass to the free-electron mass.

The quantity  $a^2 + b^2 + c^2$  is the square of the modulus of the wave functions and is equal to  $1 + (\alpha\epsilon/E_G)^2$  so that the expressions are valid provided that  $(\alpha\epsilon/E_G)^2 \ll 1$ .

Numerically  $\alpha$  is nearly equal to  $\frac{1}{2}$  for all III-V compounds.

### III. SPIN-RELAXATION MECHANISMS

Let  $\tau_c$  be the correlation time of the interaction responsible for the relaxation process and  $\delta\phi$  the angle of rotation of the spin after  $\tau_c$ .<sup>27,28</sup> The correlation time is of the order of the momentum relaxation time  $\tau_p$ . When  $\delta\phi^2 \gg 1$ , the spin orientation is completely lost during one collision (strong collision case). When  $\delta\phi^2 \ll 1$ , the mean-square angular rotation after a time  $t \gg \tau_c$  is

$$\langle \Delta\phi^2 \rangle_t \sim \delta\phi^2 t / \tau_c \quad (6)$$

The spin-relaxation  $T_1$  is defined by  $\langle \Delta\phi^2 \rangle_{T_1} \sim 1$ .

It is convenient to distinguish between two descriptions, each one corresponding to two different physical situations. In the first case, the spin is constantly submitted to an interaction  $\hbar\vec{\omega} \cdot \vec{S}$ , where  $\vec{S}$  is the electronic spin operator and  $\vec{\omega}$  a precession vector which does not vary appreciably during a time  $\tau_c$ . Then

$$\delta\phi^2 \sim \langle \omega^2 \rangle \tau_c^2, \quad (7)$$

where  $\langle \omega^2 \rangle$  is an appropriate average of  $\omega^2$ . The spin-relaxation time becomes

$$1/T_1 \sim \langle \omega^2 \rangle \tau_c. \quad (8)$$

This is the well-known expression for the spin-relaxation time in the extreme narrowing case.<sup>27,28</sup>

In the second case, the scattering mechanism itself has a finite probability of reversing the spin and the angle of rotation  $\delta\phi$  is characteristic of the collision and independent of the correlation time  $\tau_c$

$$\frac{1}{T_1} \sim \delta\phi^2 \frac{1}{\tau_c}. \quad (9)$$

Examples of this calculation are given below: The DP process belongs to the first case, the three others to the second case.

#### A. Calculation of the relaxation processes

##### 1. D'yakonov-Perel' process

It is well known that the lack of inversion symmetry in III-V compounds produces a spin-dependent splitting of the conduction band, which can be described by the Hamiltonian  $\mathcal{H}(\vec{k})$  which adds to the isotropic effective-mass Hamiltonian<sup>15</sup>:

$$\mathcal{H}(\vec{k}) = \hbar\vec{\omega}(\vec{k}) \cdot \vec{S}, \quad (10)$$

where  $\vec{\omega}(\vec{k})$  can be interpreted as a precession vector corresponding to a  $\vec{k}$ -dependent effective magnetic field.

Due to the collisions which change the wave vector  $\vec{k}$ ,  $\vec{\omega}(\vec{k})$  undergoes random variations in both magnitude and direction: this results in a spin-relaxation mechanism.

When the scattering is quasielastic, one can define a relaxation rate  $1/T_1(\epsilon)$  at a given kinetic energy  $\epsilon$ , given by Eq. (8).

The exact calculation has been carried out by D'yakonov and Perel'<sup>12</sup> who found

$$1/T_1(\epsilon) = \frac{2}{3} \omega^2(\epsilon) \tau_c(\epsilon), \quad (11)$$

where  $\omega^2(\epsilon)$  is the angular average of  $\omega^2(\vec{k})$  over all possible directions of  $\vec{k}$  at energy  $\epsilon$ :

$$\omega^2(\epsilon) = \int \frac{d\Omega_{\vec{k}}}{4\pi} \omega^2(\vec{k}). \quad (12)$$

The time  $\tau_c$  is related to the mean angle between the two directions of  $\vec{\omega}(\vec{k})$ , before and after the collision: This angle depends on the scattering process and is generally not equal to the mean angle of the directions of  $\vec{k}$  before and after collision. As a consequence the time  $\tau_c(\epsilon)$  which expresses the relaxation of the vector  $\vec{\omega}(\vec{k})$  is related, but not necessarily equal, to the usual relaxation time  $\tau_p(\epsilon)$  of the momentum. When the scattering process is isotropic (acoustical phonons) these two times are equal:  $\tau_c = \tau_p$ . On the contrary for an anisotropic scattering, this is no longer true. In particular, when small scattering angles are dominant<sup>12</sup>  $\tau_c = \tau_p/6$ .

Thus for the case of ionized impurity scattering, Eq. (12) becomes

$$1/T_1(\epsilon) = \frac{1}{9} \omega^2(\epsilon) \tau_{pi}, \quad (13)$$

where  $\tau_{pi}$  is the momentum relaxation time due to ionized impurity scattering.

Let us note that if the predominant scattering mechanism is inelastic, the spin-relaxation time  $T_1(\epsilon)$  at a given energy cannot be defined. Nevertheless, the interaction (10) produces a relative

loss of polarization of the order of  $\omega^2(\vec{k})\tau_p^2$  between each collision. We shall come back to this point in Sec. IV when discussing the optical-pumping experiments where polarized electrons are created above the bottom of the conduction band and lose their kinetic energy before recombining.

To evaluate the efficiency of the DP process in a given semiconductor, we have to obtain an explicit expression for  $\vec{\omega}(\vec{k})$ . From Ref. 15 it is easy to show that  $\vec{\omega}(\vec{k})$  is given by

$$\vec{\omega}(\vec{k}) = \sqrt{2} (ab/\hbar k) B' \vec{h}(\vec{k}), \quad (14)$$

where  $a$  and  $b$  are defined in Sec. II [Eq. (5)] and  $\vec{h}(\vec{k})$  is a vector whose components are  $h_x = k_x(k_x^2 - k_y^2)$ ,  $h_y = k_y(k_x^2 - k_y^2)$ , and  $h_z = k_z(k_x^2 - k_y^2)$ .

The parameter  $B'$  is defined as follows

$$B' = 2 \frac{\hbar^2}{m_0^2} \sum_u \frac{\langle S | p_x | u \rangle \langle u | p_y | Z \rangle}{E_c - E_u}. \quad (15)$$

The summation is made over all the states transforming like  $\Gamma_5$ ,<sup>29</sup> except for the upper valence band.  $E_c$  is the energy of conduction electrons at  $\vec{k} = 0$ .  $B'$  is estimated in the Appendix and ranges around  $10\hbar^2/2m_0$  in GaAs.

From Eq. (14), with  $a = 1$ , we obtain the value of  $\omega^2(\epsilon)$ :

$$\omega^2(\epsilon) = \frac{16}{315} \left( \frac{\Delta}{\hbar} \right)^2 \left( \frac{B'}{\hbar^2/2m_0} \right)^2 \left( 1 - \frac{m^*}{m_0} \right) \times \frac{1}{(1+\eta)(1+\frac{2}{3}\eta)} \left( \frac{m^*}{m_0} \right)^2 \left( \frac{\epsilon}{E_c} \right)^3, \quad (16)$$

which can be calculated numerically.

This equation shows that the occurrence of this relaxation process is due to both the spin-orbit interaction ( $\Delta \neq 0$ ) and the lack of inversion symmetry ( $B' \neq 0$ ); usually  $\tau_c(\epsilon)$  varies slowly with energy and the spin-relaxation rate increases when the kinetic energy increases.

## 2. Elliott-Yafet process

Due to the spin-orbit interaction, the Bloch functions  $\psi_{\vec{k}\pm}$  are generally nonpure spin states. For the  $\Gamma_1$  conduction band, the  $\vec{k} \cdot \vec{p}$  perturbation theory<sup>15</sup> shows that the admixture of the opposite spin component is proportional to  $k$  near the  $\Gamma_6$  point. Elliott<sup>16</sup> and Yafet<sup>17</sup> took this mixing into account to calculate the spin-relaxation time arising from the elastic scattering of the electrons by a perturbing potential  $U(\vec{r})$ .

The spin-flip and non-spin-flip transition probabilities are related to the matrix elements<sup>26</sup>

$$|M_{\vec{k}\pm, \vec{k}'\pm}^{\pm}|^2 = |\langle \psi_{\vec{k}'\pm} | U(\vec{r}) | \psi_{\vec{k}\pm} \rangle|^2.$$

More precisely  $1/T_1(\epsilon)$  is given by

$$\frac{1}{T_1(\epsilon)} = \frac{2\pi}{\hbar} \rho(\epsilon) \int \frac{d\Omega_{\vec{k}}}{4\pi} \int \frac{d\Omega_{\vec{k}'}}{4\pi} (|M_{\vec{k}\pm, \vec{k}'-}^{\pm}|^2 + |M_{\vec{k}\pm, \vec{k}'\pm}^{\pm}|^2), \quad (17)$$

while  $1/\tau_p(\epsilon)$  is given by

$$\frac{1}{\tau_p(\epsilon)} = \frac{2\pi}{\hbar} \rho(\epsilon) \int \frac{d\Omega_{\vec{k}}}{4\pi} \int \frac{d\Omega_{\vec{k}'}}{4\pi} (|M_{\vec{k}\pm, \vec{k}'\pm}^{\pm}|^2 + |M_{\vec{k}\pm, \vec{k}'-}^{\pm}|^2 (1 - \cos\theta')), \quad (18)$$

where  $\rho(\epsilon)$  is the density of state at energy  $\epsilon = \hbar^2 k^2/2m^*$  and  $\theta'$  is the angle between  $\vec{k}$  and  $\vec{k}'$ . For the usual scattering processes where  $\epsilon \ll E_c$

$$|M_{\vec{k}\pm, \vec{k}'-}^{\pm}|^2 \ll |M_{\vec{k}\pm, \vec{k}'\pm}^{\pm}|^2.$$

The ratio  $\tau_p(\epsilon)/T_1(\epsilon)$  is readily evaluated when the potential  $U(r)$  varies slowly on the scale of a unit cell since the Fourier transform of the potential can be factorized in the matrix elements. We get then

$$\frac{|M_{\vec{k}\pm, \vec{k}'-}^{\pm}|^2}{|M_{\vec{k}\pm, \vec{k}'\pm}^{\pm}|^2} \sim \frac{|\langle u_{\vec{k}\pm} | u_{\vec{k}'\pm} \rangle|^2}{|\langle u_{\vec{k}\pm} | u_{\vec{k}'\pm} \rangle|^2} \sim \left( \frac{\eta}{1+\eta} \right)^2 \left( \frac{\epsilon}{E_c} \right)^2. \quad (19)$$

Neglecting numerical factors of the order of unity arising from the angular average, we obtain<sup>7</sup>

$$\frac{1}{T_1(\epsilon)} \sim \left( \frac{\eta}{1+\eta} \right)^2 \left( \frac{\epsilon}{E_c} \right)^2 \frac{1}{\tau_p(\epsilon)}. \quad (20)$$

More quantitatively for any slowly varying potential  $U(\vec{r})$  Eq. (20) can be written

$$\frac{1}{T_1(\epsilon)} = \beta_V \left( 1 - \frac{m^*}{m_0} \right)^2 \left( \frac{1 + \frac{1}{2}\eta}{1 + \frac{2}{3}\eta} \right)^2 \left( \frac{\eta}{1+\eta} \right)^2 \left( \frac{\epsilon}{E_c} \right)^2 \frac{1}{\tau_p(\epsilon)}, \quad (21)$$

where  $\beta_V$  is a dimensionless parameter which depends explicitly on the exact form of  $U(r)$ . When  $U(r)$  is due to screened ionized impurities and when the conditions of Born approximation are fulfilled, Chazalviel<sup>14</sup> obtains  $\beta_V = \frac{32}{27}$ . Let us remark that one could use for  $\tau_p = \tau_{pi}$  a simple version of the Brooks-Herring formula<sup>30</sup>

$$\frac{1}{\tau_{pi}} = N_I 2\pi (a_I^*)^2 \left( \frac{R_I^*}{\epsilon} \right)^2 v, \quad (22)$$

where  $N_I$ ,  $R_I^*$ , and  $a_I^*$  are, respectively, the concentration, the effective Rydberg and Bohr radius of the ionized impurities, and  $v$  the velocity of electrons at energy  $\epsilon$ . In this case, one could express the spin-relaxation rate as  $1/T_1(\epsilon) = N_I \sigma_{EY} v$ , where the spin-reversal cross section due to this process  $\sigma_{EY}$  does not depend on energy. This formulation would be very convenient to discuss the efficiency of this mechanism, but at low kinetic energy the Brooks-Herring formula is far from

being valid and for numerical evaluation we prefer to suppose that  $\tau_{p1}$  is approximately constant.

If the collisions are due to other causes for instance phonons, Eq. (20) is still a good approximation as shown by Zawadzki and Szymanska,<sup>26</sup> and the ratio of the momentum relaxation time to the spin-relaxation time is proportional to the square of the electronic kinetic energy.

### 3. Hyperfine coupling

For *s*-type electrons, the interaction between an electronic spin  $\vec{S}$  and a nuclear spin  $\vec{I}$  is the contact interaction  $\mathcal{H}_{\text{HFS}}$ <sup>28</sup>

$$\begin{aligned} \mathcal{H}_{\text{HFS}} &= (\mu_0/4\pi) \frac{2}{3} \pi g_0 \mu_B \gamma_n \hbar \vec{I} \cdot \vec{S} \delta(\vec{r}) \\ &= A_{\text{HFS}} V \vec{I} \cdot \vec{S} \delta(\vec{r}), \end{aligned} \quad (23)$$

where  $g_0 = 2$  is the Landé factor for free electrons,  $\gamma_n$  is the nuclear gyromagnetic ratio, and  $\vec{r}$  is the relative coordinate relative to the nucleus. The electronic relaxation time due to this interaction has been calculated in the case of metals<sup>18</sup> (Fermi statistics), and we give here a description valid for nondegenerate semiconductors where Boltzmann statistics are appropriate.<sup>21</sup>

To calculate the spin-flip transition probability one considers that the electrons are described by pure spin-state wave functions, i.e., one takes the functions  $u_{\vec{k}, \pm}$  defined by Eq. (4) for  $\vec{k} = 0$ . Thus the two functions of the conduction band for a given  $\vec{k}$  are

$$|\vec{k}\sigma\rangle = |\psi_{\vec{k}\sigma}(\vec{r})\rangle = |(1/\sqrt{V}) e^{i\vec{k}\cdot\vec{r}} S\rangle, \quad (24)$$

where  $\sigma = \uparrow$  or  $\downarrow$ . In this approximation  $\psi_{\vec{k}}(0) = \psi(0)$ , which is the value of the electron wave function at the nucleus site, is independent of the wave vector  $\vec{k}$ .

The transition probability from an initial electronic state  $|\vec{k}\uparrow\rangle$  of energy  $\epsilon$  and a nuclear state  $|I, \mu\rangle$  to a final electronic state  $|\vec{k}'\downarrow\rangle$  of energy  $\epsilon'$  and a nuclear state  $|I, \mu + 1\rangle$  is given by

$$\begin{aligned} w_{(\vec{k}\uparrow, \mu) \rightarrow (\vec{k}'\downarrow, \mu+1)} &= \frac{2\pi}{\hbar} [A_{\text{HFS}} V |\psi(0)|^2] \\ &\times \frac{1}{4} [I(I+1) - \mu(\mu+1)] \delta(\epsilon - \epsilon'). \end{aligned} \quad (25)$$

If the density of nuclei is  $N$ , the transition probability from an initial electronic state  $|\vec{k}\uparrow\rangle$  to a final state  $|\vec{k}'\downarrow\rangle$  is

$$w_{(\vec{k}\uparrow) \rightarrow (\vec{k}'\downarrow)} = \sum_{\mu} NV p_{\mu} w_{(\vec{k}\uparrow, \mu) \rightarrow (\vec{k}'\downarrow, \mu+1)}, \quad (26)$$

where  $p_{\mu}$  is the probability that a nucleus is in state  $|I, \mu\rangle$ .

The electronic spin-relaxation time for Boltz-

mann statistics and weak nuclear polarization ( $p_{\mu} \sim 1/2I + 1$ ) is

$$\begin{aligned} \frac{1}{T_1(\vec{k})} &= \frac{1}{T_1(\epsilon)} = \sum_{\vec{k}'} (w_{(\vec{k}\uparrow) \rightarrow (\vec{k}'\downarrow)} + w_{(\vec{k}\downarrow) \rightarrow (\vec{k}'\uparrow)}) \\ &= N(A_{\text{HFS}} V)^2 (|\psi(0)|^2 V)^2 \\ &\times \frac{2}{3} I(I+1) (2m^* \epsilon)^{1/2} / 2\pi \hbar^4 \end{aligned} \quad (27)$$

### 4. Bir-Aronov-Pikus process

We now consider the relaxing effect of the exchange interaction between electrons and holes.<sup>13</sup> This interaction is described by the exchange Hamiltonian<sup>19</sup>

$$\mathcal{H}_{\text{exch}} = A_{\text{exch}} V \vec{J} \cdot \vec{S} \delta(\vec{r}), \quad (28)$$

where  $\vec{J}$  is the angular-momentum operator of the hole and  $\vec{r}$  the position of the electron relative to the hole.

The constant  $A_{\text{exch}}$ , proportional to the exchange coupling between the periodic part of the conduction and valence wave functions, is given by

$$A_{\text{exch}} V = -\frac{2}{3} E_a V_c, \quad (29)$$

where  $e^{19,32}$

$$\begin{aligned} E_a &= -\frac{2}{3} \frac{1}{V_c} \int_{\text{cell}} S(\vec{r}_1) X(\vec{r}_2) \frac{e^2}{|\vec{r}_1 - \vec{r}_2|} \\ &\times X(\vec{r}_1) S(\vec{r}_2) d\vec{r}_1 d\vec{r}_2. \end{aligned} \quad (30)$$

Here  $V_c$  is the unit cell volume and the energy  $E_a$  is of the order of an atomic exchange energy.

To calculate the spin-flip transition probability, it is necessary to have an explicit expression of the wave function describing an electron-hole pair. In the Wannier approximation, which is valid in III-V compounds,<sup>33</sup> this function can be written as<sup>32,34</sup>

$$|\Psi_{\sigma, m}^{\vec{k}, \xi}(\vec{R}, \vec{r})\rangle = (e^{i\vec{k}\cdot\vec{R}} / \sqrt{V}) \Phi_{\xi}(\vec{r}) \left| \frac{1}{2}, \sigma \right\rangle \left| \frac{3}{2}, m \right\rangle; \quad (31)$$

$\vec{r}$  is the position of the electron relative to the hole,  $\vec{R}$  is the center-of-mass coordinate,  $\vec{K}$  is the center-of-mass wave vector,  $|\frac{1}{2}, \sigma\rangle$  and  $|\frac{3}{2}, m\rangle$  stand, respectively, for  $\Gamma_6$  and  $\Gamma_8$  functions of conduction and valence bands.  $\xi$  stands for the quantum numbers of the bound states and labels the continuum for unbound states. If the Coulomb interaction between the electrons and the holes is not screened the envelope function  $\Phi_{\xi}(\vec{r})$  is hydrogenlike, if  $\xi$  corresponds to a bound state  $\Psi_{\sigma, m}^{\vec{k}, \xi}$  describes an exciton, otherwise  $\Psi_{\sigma, m}^{\vec{k}, \xi}$  describes a free electron-hole pair submitted to a Coulomb interaction. Let us remark that  $A_{\text{exch}}$  is related to the exchange splitting  $\Delta_{x,1s}$  which is the difference in energy between triplet and quintuplet states of an exciton in its 1s state

$$A_{\text{exch}}V = -\frac{1}{2}\Delta_{x,1s}/|\Phi_{1s}(0)|^2. \quad (32)$$

If the Coulomb interaction is screened,  $\Phi_{\frac{1}{2}}$  is no more a hydrogenlike function. An important case is that where the screening length  $\lambda_s$  is much smaller than the first Bohr radius  $a_{x,1s}$  of the exciton,<sup>35</sup> then  $|\Phi_{\frac{1}{2}}(0)|^2 \approx 1/V$ .

When the holes obey Boltzmann statistics like the electrons (the conditions where this is valid are discussed in Sec. IV), one obtains the electronic spin-relaxation time due to the exchange interaction by following the same steps as for the case of the hyperfine interaction, the holes replacing the nuclei.

As the effective mass of the holes is much larger than that of the electrons in III-V compounds, one may consider that the scattering of an electron on a hole is elastic so that Eq. (25) is valid with some changes in notation:  $A_{\text{HFS}}V \rightarrow A_{\text{exch}}V$ ,  $|I, \mu\rangle \rightarrow |\frac{3}{2}, m\rangle$ ,  $|\psi(0)|^2 \rightarrow |\Phi_{\frac{1}{2}}(0)|^2$ ,  $N \rightarrow N_h$  which is the concentration of holes which contributes to the electronic-spin relaxation. Because the Pauli principle does not play any part for holes, one can use Eq. (26) where  $p_\mu$  must be replaced by  $p_m$  which is the probability for a hole to be in a state  $|\frac{3}{2}, m\rangle$ . The holes being nonpolarized,  $p_m = \frac{1}{4}$  and the relation (27) is obtained with the changes of notation stated above. With the hypothesis of strong screening by free carriers,  $\lambda_s \ll a_{x,1s}$  and  $|\Phi_{\frac{1}{2}}(0)|^2 = 1/V$ : the spin-relaxation time due to exchange interaction between electrons and holes can be expressed as a function of parameters which can be experimentally measured

$$\frac{1}{T_1(\epsilon)} = N_h \left( \frac{(5\pi)^{1/2}}{8} \frac{\Delta_{x,1s}}{E_{x,1s}} a_{x,1s} \right)^2 \left( \frac{2\epsilon}{m^*} \right)^{1/2}. \quad (33)$$

$E_{x,1s} = \hbar^2/2m_x a_{x,1s}^2$  is the binding energy of an exciton in the 1s state,  $m_x$  is the reduced mass of the exciton and we have taken  $m_x = m^*$ .

The formula (33) can be written as  $1/T_1(\epsilon) = N_h \sigma_1 \nu$ , where  $\sigma_1$  can be interpreted as a spin-flip cross section. In GaAs  $\sigma_1 \sim 10^{-16}$  cm<sup>2</sup> (see numerical data in following part).

### B. Discussion

In this part we compare numerically the efficiency of the above relaxation mechanisms and discuss some results obtained by optical-pumping techniques. The comparison with our experiments will be made in the next section.

In most of the optical-pumping experiments, the densities of photoelectrons are so low that they obey Boltzmann statistics. The relaxation time for a statistical distribution of electrons at temperature  $T$  is then given by

$$\frac{1}{T_1(T)} = \int \frac{1}{T_1(\epsilon)} \rho(\epsilon) e^{-\epsilon/k_B T} d\epsilon / \int \rho(\epsilon) e^{-\epsilon/k_B T} d\epsilon, \quad (34)$$

where  $\rho(\epsilon)$  is the density of states and  $k_B$  the Boltzmann constant. For the spin-relaxation mechanisms due to the hyperfine and exchange interaction one only needs to calculate the average of the electronic velocity  $v = (2\epsilon/m^*)^{1/2}$  which gives  $(4\pi)^{1/2}(2k_B T/m^*)^{1/2}$ . But when the spin-relaxation time depends explicitly on the momentum relaxation  $\tau_p$ , the average is not so simple because the variation of  $\tau_p$  as a function of  $\epsilon$  is not well known. In order to compare the efficiency of the different mechanisms, we simply replace in the formulas  $\epsilon$  by  $k_B T$  since the energy average given by the expression (34) does not change the orders of magnitude.

Numerical results are detailed in Table I for GaAs, GaSb, InP, InAs, and InSb. For the DP and EY process one needs to know  $\tau_p$ . In the following

TABLE I. Numerical evaluation of the relaxation rate  $1/T_1$  for different processes.  $1/T_1$  is given in sec<sup>-1</sup>.  $\epsilon$  is given in meV. For the DP and EY processes  $\tau_p$  has been taken constant and equal to  $10^{-13}$  sec. For the DP process,  $B' = 10\hbar^2/2m_0$  in GaAs, GaSb, and InSb,  $B' = 3\hbar^2/2m_0$  in InAs and InP. For the exchange interaction mechanism we suppose that the hole concentration is  $N_h = 4 \times 10^{18}$  cm<sup>-3</sup> and that the atomic exchange energy is the same for the five semiconductors:  $E_a = 14$  eV.

$1/T_1$	GaAs	GaSb	InP	InAs	InSb
DP process: Eqs. (13) and (16)	$1 \times 10^4 \epsilon^3$	$6 \times 10^4 \epsilon^3$	$2 \times 10^3 \epsilon^3$	$4 \times 10^3 \epsilon^3$	$7 \times 10^4 \epsilon^3$
EY process: Eq. (20)	$1 \times 10^5 \epsilon^2$	$3 \times 10^6 \epsilon^2$	$2 \times 10^4 \epsilon^2$	$1 \times 10^6 \epsilon^2$	$1 \times 10^8 \epsilon^2$
Hyperfine interaction: Eq. (27)	$6 \times 10^4 \epsilon^{1/2}$	$2 \times 10^5 \epsilon^{1/2}$	$5 \times 10^5 \epsilon^{1/2}$	$9 \times 10^4 \epsilon^{1/2}$	$9 \times 10^4 \epsilon^{1/2}$
Exchange interaction: Eqs. (33) and (35)	$1 \times 10^{10} \epsilon^{1/2}$	$8 \times 10^9 \epsilon^{1/2}$	$2 \times 10^{10} \epsilon^{1/2}$	$3 \times 10^9 \epsilon^{1/2}$	$2 \times 10^9 \epsilon^{1/2}$

we shall take  $\tau_p = 10^{-13}$  sec, which is a representative value of the momentum relaxation time in strongly doped materials. For the BAP mechanism we have performed the calculation assuming that  $N_h = 4 \times 10^{18} \text{ cm}^{-3}$  for all these compounds; this concentration permits a direct comparison with our experiments (see Sec. IV).

For the DP mechanism, the important factor is  $\omega^2(\epsilon)$ . From Eq. (16) one calculates that  $\omega^2(\epsilon)$  is equal to  $10^{18}\epsilon^3$  in GaAs ( $\omega$  in  $\text{rad sec}^{-1}$  and  $\epsilon$  in meV). For scattering on ionized impurities, Eq. (13) is valid and  $1/T_1(\epsilon) \sim 10^{17}\epsilon^3\tau_p(\epsilon)$ .

To obtain an order of magnitude valid as well for impurity scattering as for phonon scattering, Eq. (20) is used for the EY mechanism.

The efficiency of the hyperfine interaction mechanism is calculated from Eq. (27):  $A_{\text{HFS}}V$  is known and the values of  $|\psi(0)|^2V$  are easily calculated.<sup>36</sup>

For the BAP mechanism, the numerical value of the spin-relaxation time cannot be obtained precisely since the value of the exchange integral  $E_a$  is generally not known. However, if we suppose that the screening length is small enough so that the relation  $|\phi_x(0)|^2V = 1$  holds, Eq. (33) is valid. So we can calculate  $T_1$  in GaAs:  $E_{x,1s}$ ,  $a_{x,1s}$ , and  $m^*$  are known<sup>23</sup> and the value  $\Delta_{x,1s} = 0.1$  meV is deduced from our experiments. A discussion of the validity of Eq. (33) and of the value of  $\Delta_{x,1s}$  is given in Sec. IV. Let us remark that if  $|\phi_x(0)|^2$  is constant, the dependence of  $T_1$  on band parameters in semiconductors arises from

$$\left(\frac{\Delta_{x,1s} a_{x,1s}}{E_{x,1s}}\right)^2 \frac{1}{\sqrt{m^*}} = \left(\frac{8}{3\pi\hbar^2}\right)^2 E_a^2 V_c^2 (m^*)^{3/2}. \quad (35)$$

In the absence of any other indication about  $E_a$  we have assumed that it is constant and that the only variation from one semiconductor to another arises from  $V_c^2(m^*)^{3/2}$ . From the above equation we can deduce  $E_a$  in GaAs:  $E_a = 14$  eV.

Figure 1 shows the energy dependence of the expressions of Table I for GaAs.

Before beginning the discussion of the experimental results, we want to point out that the temperature dependence alone is not conclusive for the interpretation of the experimental data, and one should first look for an order of magnitude agreement. First  $\tau_p$  being not very well known, the temperature dependence of the DP and EY processes are given only as an indicative trend; second, the exchange interaction gives a  $T^{1/2}$  temperature dependence only under certain conditions. Bir, Aronov, and Pikus showed that the temperature dependence could vary according to experimental conditions.<sup>13</sup> On the contrary, the orders of magnitude of the different processes are so different that they constitute the only valuable test.

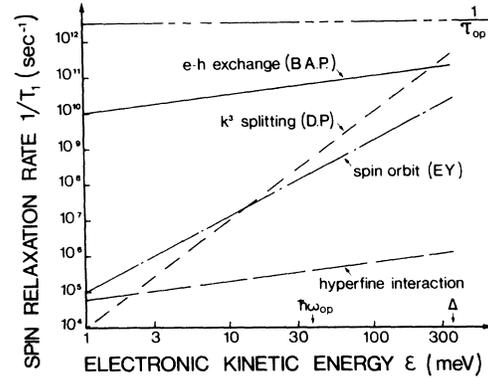


FIG. 1. Theoretical variation of the electron spin-relaxation rates as a function of kinetic energy of conduction electrons in  $p$ -doped GaAs,  $N_A = 4 \times 10^{18} \text{ cm}^{-3}$ . The momentum relaxation time  $\tau_p$  is  $10^{-13}$  sec. The exchange splitting is 0.1 meV.  $\tau_{\text{op}} = 3 \times 10^{-13}$  sec is the energy relaxation time by optical-phonon emission, and is taken constant for simplicity;  $\hbar\omega_{\text{op}}$  is the energy of the optical phonon, and  $\Delta = 0.34$  eV the spin-orbit splitting of the valence band.

If the orders of magnitude of two processes are the same, a more accurate theoretical study of their temperature dependence must be made and compared with the experiments.

We want now to compare briefly our evaluations with some published data. The results corresponding to a temperature below  $100^\circ\text{K}$  in  $p$ -doped GaAs or  $\text{Ga}_{1-x}\text{Al}_x\text{As}$  by several authors<sup>4-7,9</sup> give  $T_1 \sim 10^{-10}$ – $10^{-9}$  sec depending on the doping. This order of magnitude is well accounted for by the exchange interaction mechanism. Above  $100^\circ\text{K}$ , other mechanisms can be effective and the problem is not always clear. In Ref. 4, the authors find an experimental law  $1/T_1 \propto T^{5/2}$  in  $\text{Ga}_{1-x}\text{Al}_x\text{As}$  with  $T_1 \sim 10^{-10}$  sec at  $T \sim 100^\circ\text{K}$  and attribute the spin relaxation to acoustical phonon scattering following a theoretical calculation by Pavlov.<sup>37</sup> In GaAs the numerical calculation from Pavlov's paper gives  $T_1 \sim 10^{-7}$  sec at  $T \sim 100^\circ\text{K}$ , so that this process is too weak by three orders of magnitude and should be disregarded. This process is like an EY mechanism and the rough formula of Table I gives the same order of magnitude. More recently, Clark *et al.*<sup>10</sup> studied experimentally the temperature dependence of the spin-relaxation time in  $p$ -doped GaAs ( $N_A \sim 10^{17} \text{ cm}^{-3}$ ). Below  $50^\circ\text{K}$  their experimental results can be fitted by the law  $1/T_1 \sim 10^8 T^{1/2}$  which seems to indicate that the relaxation is due to the BAP process. Above  $100^\circ\text{K}$  they attribute the spin relaxation to the DP process: Table I shows that for  $N_A \sim 10^{17} \text{ cm}^{-3}$  and for acoustical-phonon scattering the right order of magnitude is effectively obtained.

## IV. EXPERIMENTAL RESULTS AND DISCUSSION

## A. Experimental results

Our experiments are performed on *p*-type GaAs doped with  $N_A \sim 4 \times 10^{18} \text{ cm}^{-3}$  acceptors. The experimental setup is a classical optical-pumping one which has been described elsewhere.<sup>2,7</sup> The measurement of the electronic polarization has been performed on the band-acceptor line at 1.49 eV. When the concentration of free holes is large enough, the electron-hole collisions are very efficient to thermalize the electronic gas below the optical-phonon energy. The conduction electrons which recombine on the acceptors are then at the lattice temperature. On the other hand, the steady-state spin polarization of the holes is zero because their spin-relaxation time is usually very small as compared to the lifetime of electron-hole pairs and furthermore in these *p*-type samples the concentration of equilibrium holes is much larger than that of the photoexcited ones. In this case, the degree of circular polarization of the luminescence is proportional to the electronic polarization and for band-to-band or band-to-acceptor recombination the proportionality factor is  $P_t = -0.5$ .<sup>3</sup> Thus the polarization measurement on the acceptor luminescence line yields a direct measurement of the polarization of the thermalized electrons. Figure 2 shows the polarization versus the exciting photon energy  $h\nu$  at different temperatures. One sees that the polarization of the electrons hardly varies up to  $h\nu \sim 1.75$  eV. In this case, it has been shown<sup>38</sup> that the Hanle effect yields the spin-relaxation time and the lifetime of electrons at the bottom of the band. Figure 3 shows the experimental results for  $h\nu \sim 1.6$  eV. These data yield the  $T_1$  and  $\tau$  values as a function of temperature (Fig. 4) and we obtain the following experimental variation

$$\frac{1}{T_1} \sim 3 \times 10^9 T^{1/2} \sim 10^{10} \epsilon^{1/2},$$

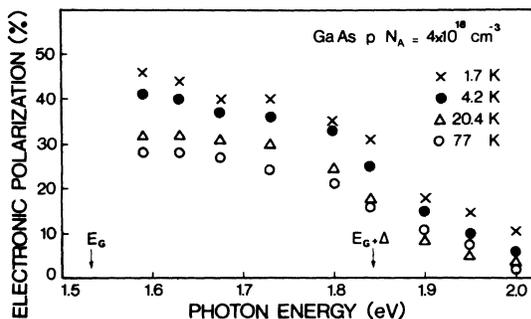


FIG. 2. Experimental variation of the electronic polarization as a function of exciting photon energy.

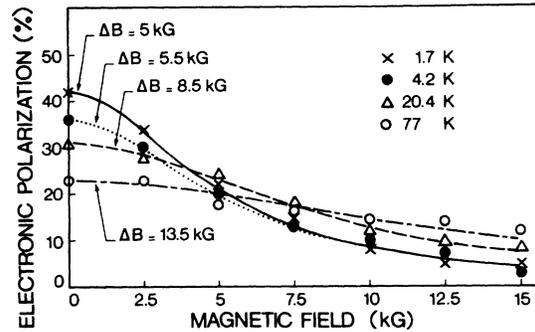


FIG. 3. Transverse magnetic-field depolarization in *p*-type GaAs. ( $N_A = 4 \times 10^{18} \text{ cm}^{-3}$ ) for an excitation energy  $h\nu \sim 1.6$  eV. The continuous curves are the Lorentzian fit (half-width  $\Delta B$ ) to the experimental points.

$T_1$  in sec,  $T$  in  $^\circ\text{K}$ ,  $\epsilon$  in meV.

For the discussion we distinguish two types of results: (i) the spin-relaxation rate of thermalized electrons; (ii) the variation of the polarization of thermalized electrons as a function of exciting photon energy.

## B. Discussion

In semiconductors when the doping is such that the wave functions of the shallow impurity states begin to overlap, the description of these states is a difficult problem related to the formation of an impurity band.<sup>39</sup> In GaAs the screening length  $\lambda_s$  of the holes is of the order of magnitude of the Bohr radius  $a_A$  of acceptors for a concentration  $N_0 \sim 2 \times 10^{18} \text{ cm}^{-3}$ , so that for a concentration  $N_A \geq N_0$ , the acceptors are ionized. On the other hand, the critical density  $N_m$  for metallic conduc-

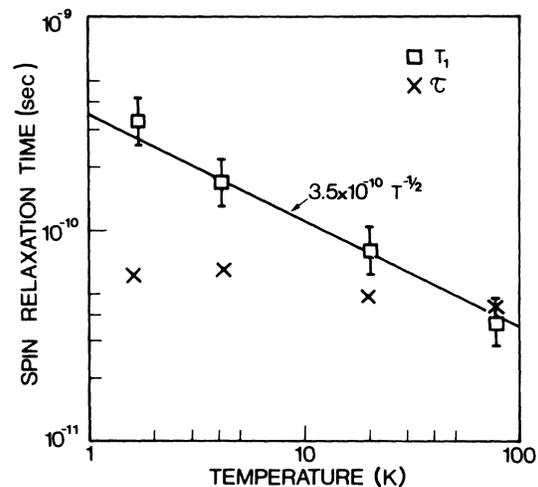


FIG. 4. Variation of the spin-relaxation time  $T_1$  and of the lifetime  $\tau$  for thermalized electrons as a function of temperature. The continuous line is the  $T^{-1/2}$  fit.

tion defined as  $\frac{4}{3}\pi a_A^3 N_m \sim 1$  ranges around  $N_m \sim 4 \times 10^{19} \text{ cm}^{-3}$ . With  $N_A \sim 4 \times 10^{18} \text{ cm}^{-3}$  we are in the intermediate range, and we make the assumption that all the holes are delocalized in an impurity band and contribute to the electronic relaxation without limitation due to the Pauli principle<sup>40</sup> so that  $N_A = N_h$ , where  $N_h$  is the number of holes appearing in Eq. (33). Moreover since  $N_A > N_0$ , the Coulomb interaction is screened and we take  $|\Phi_i(0)|^2 = 1/V$ . The expression given in Table I for the spin-relaxation rate due to the electron-hole exchange interaction has been obtained with these hypotheses and must be modified if they do not apply.

### 1. Thermalized electrons

One sees from Table I that for energies in the meV range, the first three mechanisms are much too ineffective to explain our results. For all reasonable values of  $\Delta_{x,1s}$ , the exchange interaction mechanism gives the right order of magnitude. The  $T^{1/2}$  law predicted for the spin-relaxation rate  $1/T_1$  is well verified from 1.7 to 77 °K. From our results we determine  $\Delta_{x,1s} \approx 0.1 \text{ meV}$  which compares favorably with the value  $\Delta_{x,1s} = 0.05 \text{ meV} \pm 0.05 \text{ meV}$  given in Ref. 24.

### 2. Depolarization during the thermalization process

When the excitation energy is larger than the band gap, the photocreated electrons must lose their excess energy before they reach the bottom of the conduction band where they recombine. It is necessary to consider how the electronic spin is affected by this thermalization process. This process may be divided into two steps; the first

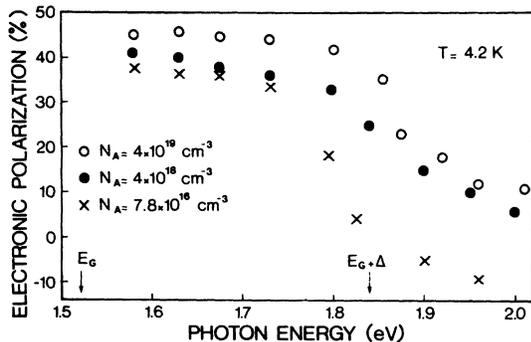


FIG. 5. Variation of the electronic polarization for various doping levels in *p*-doped GaAs. The experimental data corresponding to samples  $N_A = 4 \times 10^{19} \text{ cm}^{-3}$  and  $N_A = 7.8 \times 10^{16} \text{ cm}^{-3}$  have been obtained by Ekimov and Safarov (Ref. 5). The fact that in less-doped samples the electronic polarization becomes negative for  $\epsilon > \Delta$ , due to the split-off valence band, is explained in Ref. 5 and is well accounted for by a critical energy  $\epsilon_c < \Delta$ .

one is the decrease in energy by emission of optical phonons from the energy of creation to states near the bottom of the conduction band within an optical-phonon energy  $\hbar\omega_{op}$ . The second step is the thermalization from these states to thermalized states at the bottom of the conduction band. The depolarization during this last step has been studied elsewhere: It has been shown<sup>38</sup> that in GaAs, due to the shortness of the energy relaxation time in doped samples, there is no spin depolarization during this thermalization. It remains to study the loss of polarization which arises for larger energies, where emission of optical phonons occurs.

Figure 5 shows together the results obtained by Ekimov and Safarov<sup>5</sup> and by us in order to exhibit the experimental variation of the spin depolarization as a function of doping level. Following an analysis given first by D'yakonov and Perel<sup>11</sup> these results were interpreted by introducing a critical energy  $\epsilon_c$  depending on the doping: only for kinetic energies higher than  $\epsilon_c$  a significant depolarization occurs in the high-energy states. This analysis supposes that a spin-relaxation time can be defined at each energy and neglects the fact that the energy relaxation occurs by finite quanta of the order of several tens of meV. Here we consider the case where the energy relaxation is due to emission of optical phonons in a time  $\tau_{op}$ . In GaAs  $\tau_{op} \sim 3 \times 10^{-13} \text{ sec}$  and  $\hbar\omega_{op} = 36 \text{ meV}$ .<sup>41</sup> Figure 1 shows that none of the calculated relaxation rates is effective enough to destroy the spin orientation during a time of the order of  $\tau_{op}(\tau_{op}/T_1 \ll 1)$ . Even the relaxation time due to the modulation of the  $\hbar^3$  term given by Eq. (13) is too long to explain the observed depolarization at high kinetic energies of the order of 200 meV. At this point we must wonder whether or not the usual theories of spin relaxation are valid on a time scale as short as  $\tau_{op}$ . In fact we are going to see that we deal here with a special case where it is not always possible to define a spin-relaxation time, but it is still possible to evaluate a relative loss of polarization during a time  $\tau_{op}$ . The formula which gives  $T_1$  in the motional narrowing case was established with the two following hypothesis: (i) a time average during a time  $t \gg \tau_c$  (ii) a mean rotation given by  $\delta\phi^2 \sim \omega^2(\epsilon)\tau_c^2 \ll 1$  where  $\omega^2(\epsilon)$  is given by Eq. (16). For thermalized electrons  $t$  is the lifetime which is of the order of  $10^{-10} \text{ sec}$  and  $\omega^2(\epsilon)\tau_c^2 \ll 1$ , so that these conditions are fulfilled. But for an energy  $\epsilon > \hbar\omega_{op}$  the time  $t$  is equal to  $\tau_{op}$ . If the momentum relaxation time due to impurity scattering  $\tau_{p1}$  is larger than  $\tau_{op}$ , the correlation time is equal to  $\tau_{op}$  and it is not possible to define a spin-relaxation time  $T_1(\epsilon)$  at energy  $\epsilon$ . The mean angular rotation  $\delta\phi^2$ , that is the loss of

TABLE II. Spin depolarization at high kinetic energy.

		$N_A < N_c \sim 10^{18} \text{ cm}^{-3}$ $\tau_{pi} > \tau_{op} \sim \tau_c$ no averaging $\epsilon_c \sim 200 \text{ meV}$	$N_A > N_c \sim 10^{18} \text{ cm}^{-3}$ $\tau_{op} > \tau_{pi} \sim \tau_c$ $N_A', \tau_{pi}', \epsilon_c'$ $\epsilon_c' > 200 \text{ meV}$
$\epsilon > \hbar\omega_{op}$	$\epsilon < \epsilon_c$	no $T_1(\epsilon)$	$\frac{1}{T_1(\epsilon)} = \frac{1}{9} \omega^2(\epsilon) \tau_{pi}$
		$\omega^2(\epsilon) \tau_c^2 \sim \delta\phi^2 \ll 1$	$\frac{1}{T_1} \ll \frac{1}{\tau_{op}}$
		no spin depolarization	no spin depolarization
	$\epsilon > \epsilon_c$	$\omega^2(\epsilon) \tau_c^2 \sim \delta\phi^2 > 1$	$\frac{1}{T_1} > \frac{1}{\tau_{op}}$
	spin depolarization	spin depolarization	

polarization due to the emission of an optical phonon, is of the order of  $\omega^2(\epsilon)\tau_{op}^2$ . When  $\tau_{pi}$  is shorter than  $\tau_{op}$  the correlation time is again of the order of  $\tau_{pi}$  and the loss of polarization during a time  $\tau_{op}$  is approximatively  $\omega^2(\epsilon)\tau_{pi}\tau_{op}$ .

In first approximation one can then conclude that the critical energy  $\epsilon_c$  is given by  $\omega^2(\epsilon)\tau_c\tau_{op} \sim 1$  where  $\tau_c \sim \tau_{op}$  for pure enough samples ( $\tau_{pi} > \tau_{op}$ ) and  $\tau_c \sim \tau_{pi}$  for more doped samples ( $\tau_{pi} < \tau_{op}$ ). Using Brooks-Herring formula, given by Eq. (22), which is valid for high kinetic energies, one calculates that in *p*-doped samples with concentration lower than  $N_c \sim 10^{18} \text{ cm}^{-3}$ ,  $\tau_{pi}$  is larger than  $\tau_{op}$  and  $\epsilon_c \sim 200 \text{ meV}$ . On the contrary for  $N_A > N_c$ ,  $\tau_{pi} < \tau_{op}$  and  $\omega^2(\epsilon)\tau_{pi}\tau_{op}$  becomes of the order of unity for an energy  $\epsilon_c$  which is greater than 200 meV: As  $\tau_{pi} \propto N_A^{-1}$ , the higher the doping  $N_A$  ( $N_A > N_c$ ), the shorter the momentum relaxation time  $\tau_{pi}$  and thus the higher the critical energy  $\epsilon_c$ . These conclusions, which are in agreement with the results of Fig. 5 are summarized in Table II.

At last, let us remark that these conclusions on the depolarization at high energy cannot be applied directly in other III-V compounds. In GaSb the critical energy  $\epsilon_c$  is below the first optical-phonon energy and the problem is completely different.<sup>8</sup> In InSb the EY process is very efficient and the corresponding depolarization due to optical phonons is very effective.

## V. CONCLUSION

We have shown that the exchange interaction mechanism between electrons and holes, first proposed by Bir, Aronov, and Pikus, is the only process which can explain the spin relaxation of thermalized conduction electrons in strongly doped *p*-type GaAs at low temperature ( $T \lesssim 100 \text{ K}$ ). The other studied mechanisms are too weak by several

orders of magnitude. Moreover this mechanism is also very efficient for other strongly *p*-doped III-V compounds. From our measurement as a function of temperature of the spin-relaxation time of conduction electrons, we deduce the exchange splitting  $\Delta_{x,1s} \sim 0.1 \text{ meV}$ . For electrons with high kinetic energy ( $\epsilon \gtrsim \hbar\omega_{op}$ ), none of the calculated  $T_1$  can explain the observed results in GaAs, due to the shortness of the energy relaxation time. However, a careful examination of the relaxation mechanism due to the spin-splitting of the conduction band accounts for the experimental results in strongly doped *p*-type GaAs. But this last conclusion is probably far less general among III-V compounds where the importance of the spin-orbit splitting and the smallness of the energy gap causes the EY mechanism to be very efficient for high kinetic energy.

## ACKNOWLEDGMENTS

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## APPENDIX: EVALUATION OF THE BAND-SPLITTING PARAMETER $B'$

If we restrict ourselves to the  $\Gamma_5$  symmetry band which is the closest to the conduction band (except for the upper valence band), one can write

$$B' \sim 2 \frac{\hbar^2}{2m_0 m_0} \frac{2 \langle S | p_x | \Gamma_5 \rangle \langle \Gamma_5 | p_y | Z \rangle}{E_c - E(\Gamma_5)}.$$

$E_c$  is the energy of the conduction band at  $\vec{k}=0$  and  $\Gamma_5$  is nothing but the second conduction band. It is possible to get  $(2/m_0) |\langle \Gamma_5 | p_y | Z \rangle|^2$  using La-

waetz' results<sup>42</sup>; one finds  $2/m_0 |\langle \Gamma_5 | p_x | Z \rangle|^2$  close to 18 eV for GaAs, GaSb, InSb, and InAs and close to 15 eV for InP. Furthermore, Hermann and Weisbuch<sup>43</sup> have shown that  $P'^2 = (2/m_0) \times |\langle S | p_x | \Gamma_5 \rangle|^2$  is ranging between 6 and 9 eV for GaAs, 9 and 11 eV for GaSb, 10 and 13 eV for InSb. For GaSb if we take  $P'^2 = 10$  eV we find  $B' \sim 9\hbar^2/2m_0$ . This result is to be compared to the experimental data of Seiler *et al.*<sup>44</sup> who find  $B' \sim 10\hbar^2/2m_0$ . For InAs and InP the value of  $P'^2$  is not determined so accurately. In Ref. 43 it is shown that  $P'^2$  is probably ranging between 0.2 and 4 eV for InAs and between 2 and 6 eV for InP. Finally as we only look for a reasonable order of

magnitude we shall take  $B' \sim 10\hbar^2/2m_0$  for GaAs, GaSb, and InSb and  $B' \sim 3\hbar^2/2m_0$  for InAs and InP. With the above values for  $B'$  and using Eq. (16) we find

$$\begin{aligned} \omega^2(\epsilon) &\sim 1 \times 10^{18} \epsilon^3 \text{ for GaAs,} \\ &\sim 6 \times 10^{18} \epsilon^3 \text{ for GaSb,} \\ &\sim 7 \times 10^{18} \epsilon^3 \text{ for InSb,} \\ &\sim 4 \times 10^{17} \epsilon^3 \text{ for InAs,} \\ &\sim 2 \times 10^{16} \epsilon^3 \text{ for InP;} \end{aligned}$$

$\omega$  is in rad sec<sup>-1</sup> and  $\epsilon$  in meV.

\*Laboratoire associé au Centre National de la Recherche Scientifique.

†Equipe de Recherche du Centre National de la Recherche Scientifique.

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