Spin-lattice relaxation in p-type gallium arsenide single crystals

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An optical-pumping technique is used to measure the spin-relaxation time of photogenerated conduction electrons in several p-type GaAs single crystals doped with various amounts of acceptors in the 1.7-300 K temperature range. Our experimental results are compared with those of the literature and with the predictions of the existing theoretical calculations. From about 10 K, the Bir-Aronov-Pikus (BAP) mechanism is found to be relevant for moderately doped $(10^{17} - 10^{18})$ cm^{-3}), up to about 150 K, or degenerate (up to 300 K) semiconductors, using the electronic temperature, deduced from the luminescence spectra, rather than the sample temperature. The D'yakonov-Perel' (DP} process was found to be active above 200 K for moderately doped samples and from about 80 K to room temperature for samples doped in the $(1.6-6)\times10^{16}$ -cm⁻³ acceptor-concentration range. Our original results obtained at liquid-helium temperatures at whatever the doping level cannot be explained either by the DP mechanism or by the BAP process.

I. INTRODUCTION

The possibility of measuring the spin-relaxation time of photogenerated electrons in semiconductors from the value of the luminescence degree of circular polarization was recognized more than a decade ago.^{$1-3$} Let us recall that in optical-pumping conditions, that is, excitation of the semiconductor by circularly polarized light with convenient wavelength, the photogenerated electrons are spin polarized, the mean value of this polarization depending on the relative value of relaxation time T_1 and lifetime τ . Measuring the luminescence degree of circular polarization and its decrease in a transverse magnetic field (Hanle effect) allows us quite simply to obtain both T_1 and τ .

Optical-pumping studies of electronic spin-lattice relaxation were performed in several p-type semiconductors, but the most extensively studied compound has been GaAs, for which many experimental papers have been published. $4-10$ These results were compared with the various mechanisms proposed in the literature.

Firstly, the Elliot-Yafet process^{11,12} and the hyperfine interaction with the lattice nuclei were shown to play a negligible role for electron relaxation in gallium arsenide, since the calculated values of T_1 were found to be 2 or 3 orders of magnitude larger than the measured ones.

Two mechanisms were shown to be relevant, depending on temperature and acceptor concentration. The earlier results were interpreted correctly using the D'yakonov-Perel' (DP) mechanism.¹³ In 1976, Bir, Aronov, and Pikus¹⁴ (BAP) proposed a mechanism taking into account the strong efficiency of electron scattering

by holes with simultaneous exchange interaction. Fishman and Lampel,⁵ working on a degenerate semiconductor with $N_A = 4 \times 10^{18}$ cm⁻³, analyzed the various relaxation processes and found that the HAP mechanism explained the $T^{-1/2}$ dependence they observed for the spin-relaxation time. In 1983, Titkov and co-workers published interesting results on GaAs crystals doped published interesting results on GaAs crystals doped
with variable acceptor concentration.^{6,9,10} For moderately doped $(10^{17}-10^{18} \text{ cm}^{-3})$ and degenerat samples it was found that the HAP process was useful in explaining small parts of the $(1/T_1)(N_A)$ dependence both at liquid-nitrogen temperature (LNT) and room temperature (RT). A part or the totality of the hightemperature side (starting from 30 to 70 K according to the doping level) of the $(1/T₁)(T)$ variation was also accounted for by the same mechanism.⁹ Studying two samples with N_A =4 \times 10¹⁶ cm⁻³ and different degrees of compensation, Maruschak et al .¹⁰ explained the linear part of their log-log $(1/T_1)(T)$ curve as being due to the efficiency of the DP mechanism. Working at 6 K on a degenerate sample, Kleinman and co-workers^{7,8} found that $1/T_1$ is nearly independent of N_A for $N_A \ge 5 \times 10^{17}$ $cm⁻³$. They proposed a mechanism involving an electron-hole exchange via the creation of a resonant virtual photon which would dominate in the helium temperature range for a degenerate hole distribution.

In summary, the physics underlying the study of electron spin-lattice relaxation in p-type GaAs is very rich because at least two distinct mechanisms have been shown to be relevant. Moreover, the wealth of experimental results still does not allow us to obtain a complete physical picture of the system. In particular, there are discrepancies between the various reported results,

especially at low temperatures and for the purest samples. Therefore it seemed of interest to undertake a new systematic investigation as a function of acceptor concentration and of temperature including very low concentrations $(N_A = 1.6 \times 10^{16} \text{ cm}^{-3})$ and temperatures (1.7 and 4.2 K) to gain additional information. We shall see in the following that, with the exception of the lowest temperatures $(T < 10 K)$, the major part of our experimental results may be explained by means of the DP or BAP processes.

This paper is organized as follows: in Sec. II we recall the predictions of the DP and BAP mechanisms; the experimental procedure and results are presented in Sec. III. These results are compared with the theory in Sec. IV.

II. OUTLINE OF THE PREDICTIONS OF THE DP AND BAP MECHANISMS

A. DP mechanism

In III-V compounds the lack of inversion symmetry leads to a spin splitting of the conduction band for $k\neq0$ (Ref. 15), which is described by the presence of a $k³$ term in the conduction-electron spin Hamiltonian. This splitting is equivalent to the presence in the crystal of an effective magnetic field inducing the precession of electron spins. In the case of thermalized electrons, it was shown¹³ that the spin-relaxation rate is given by

$$
\frac{1}{T} = Q\alpha^2 \frac{(k_B T)^3}{\hbar^2 E_g} \tau_P , \qquad (1)
$$

where τ_p is the electron momentum scattering time. The parameter Q depends on the operative scattering process of electrons and α characterizes the band structure in the $k³$ term of the conduction-electron spin Hamiltonian. From Eq. (1) it can be seen that the temperature dependence of the spin polarization rate is also sensitive to the variation of τ_p with temperature.

B. SAP mechanism

In p-type semiconductors, spin relaxation may result from electron scattering by holes with simultaneous exchange interaction. BAP (Ref. 14) proposed a mechanism taking into account the strong efficiency of such a process. They found that two terms, involving, respectively, free and bound holes, appear in the formula giving the spin-relaxation rate in the case of a nondegenerate semiconductor:

$$
\frac{1}{T_1} = \frac{2}{\tau_0} N_A a_B^3 \frac{v_e}{v_B} \left| \frac{N_P}{N_A} \right| \psi(0) \left| ^4 + \frac{5}{3} \left[1 - \frac{N_P}{N_A} \right] \right| , \quad (2)
$$

where $v_B = \hbar / \varepsilon_e a_B$ is the exciton Bohr velocity and $v_e = (3k_B T/m_e^*)^{1/2}$ for thermalized electrons. τ_0 is calculated from the exchange splitting Δ_{ex} of the exciton ground state, a_B is the exciton Bohr radius, N_P/N_A represents the acceptor degree of ionization, $|\psi(0)|^2$ is the Sommerfeld factor, and N_p is the density of free holes.

FIG. 1. Relative role of the BAP and DP mechanisms in GaAs after Aronov et al. (Ref. 9). The curve separates the concentration and temperature regions in which these mechanisms are expected to predominate.

For a given temperature N_P/N_A is constant and consequently it can be seen that $1/T_1$ is proportional to N_A . On the other hand, the variation of $1/T_1$ as a function of T is governed not only by the electron velocity but also by the relative importance of the two terms inside the large vertical bars of Eq. (2).

With *degenerate semiconductors*, the situation is still more complicated. Different formulas, each involving only one term, are obtained for thermalized or hot "fast" electrons [they satisfy the condition $\varepsilon_e > \varepsilon_F(m_e^*/m_h^*)$. Particularly, in the region of hole degeneracy, the theory formulated by

$$
\frac{1}{T_1} = \frac{3}{\tau_0} \frac{v_e}{v_B} \frac{k_B T}{\epsilon_F} | \psi(0) |^4 N_P a_B^3
$$
 (3)

for fast thermalized electrons predicts that $1/T_1$ varies as $N_p^{1/3}$. In Eq. (3) the hole Fermi velocity v_F is assumed to be lower than the electronic velocity v_e ; ε_F is the Fermi-level energy and $N_p = N_A$.

In summary, we have recalled in this section the predictions of the DP and BAP mechanisms. A straightforward calculation, in agreement with previous experimental results, allows us to predict the temperature and acceptor-concentration ranges for which each mechanism is dominant. This is shown in Fig. ¹ after Aronov et al ⁹. One sees that the DP mechanism is active at low acceptor concentration and high temperature, whereas the SAP process is more important at high acceptor concentration and low temperature.

III. EXPERIMENT

The method that we use to measure the spin-lattice relaxation time T_1 of the photoelectrons is now standard. Upon absorption of circularly polarized light of suitable wavelength, the steady-state spin polarization in zero magnetic field of the electrons is not zero; its value is given by

$$
P(0) = P_i \frac{T_1}{T_1 + \tau} \tag{4}
$$

where $P_i = \pm 0.5$ for σ^+ polarized light and τ is the lifetime of the free electrons. The decrease of P due to precession of the electronic spins in a magnetic field B perpendicular to the direction of light excitation is given by

$$
P(B) = P(0) \frac{(\Delta B)^2}{(\Delta B)^2 + B^2} , \qquad (5)
$$

where

$$
\Delta B = \frac{\hbar}{|g^*| \mu_B} \left[\frac{1}{T_1} + \frac{1}{\tau} \right]
$$
 (6)

and $g^* = -0.44$ (Ref. 16) is the absolute value of the free-electron Lande factor.

The value of electronic polarization is obtained from the measure of the degree of the circular polarization P of the luminescence, which is given by

$$
\mathcal{P} = P_r P(0) \tag{7}
$$

where, at least for free electrons, the quantity P_r is given by

$$
P_r = |P_i| = 0.5 \tag{8}
$$

Using Eqs. (4), (6), and (7), we finally obtain the value of $T₁$, which is given by

$$
T_1 = \frac{\hbar}{|g^*| \mu_B} \frac{1}{\Delta B} \left[\frac{P_i^2}{P_i^2 - P} \right].
$$
 (9)

The measurement of T_1 was performed as a function of temperature of 11 (111) p -type GaAs samples. They are doped either with cadmium or zinc, their acceptor concentrations being, respectively, 1.6×10^{16} , 6×10^{16} , 1.2×10^{17} , 2.6×10^{17} , and 10^{18} cm⁻³ (cadmium), and 2×10^{17} , 1.7×10^{18} , 3.2×10^{18} , 9.1×10^{18} , 2.8×10^{19} , and 1.1×10^{20} cm⁻³ (zinc). They originate from Meta Research (U.S.A.).

The exciting beam from a krypton-ion laser ($\lambda = 752.5$) nm, power ranging from ¹ to 100 mW) was focused on the sample, which can be introduced in two different cryostats. The first of them allows measurements at fixed temperatures $(1.7, 4.2,$ and 77 K). The second one, a gas-flow cryostat, permits measurements at any temperature between 8 and 300 K. The luminescence was analyzed and detected by an HRS 2 Jobin-Yvon monochromator and a photomultiplier with S_1 response. The luminescence degree of circular polarization was measured by a standard lock-in detection either with a rotating quarterwave plate or a photoelastic quartz modulator at 50 kHz.

The shape of the luminescence spectrum that we obtained, was, as expected, found to be strongly dependent on the acceptor concentration of the sample. For degenerate samples, the luminescence maximum was situated at an energy greater than the band gap.¹⁷ In the case of nondegenerate specimens, this maximum corresponds to the D^0 -h transition above liquid-nitrogen temperature, and to the donor-acceptor transition below.

In order to obtain the free-electron polarization, it would be natural to choose for P the degree of circular polarization of the band-to-band luminescence. However, we choose in all cases to measure the value of P at the maximum of the luminescence peak.¹⁸

The measure of the width ΔB of the depolarization curve in a transverse magnetic field was performed by modulation of the excitation light polarization, in order to avoid dynamic spin polarization of the lattice nuclei which might perturb the system behavior because of the large hyperfine nuclear field.^{19,20} In such a case, the electronic polarization is modulated and this allows dynamic polarization of the only nuclear spins with the spin-lattice relaxation time T_{1N} < 1/ Ω . By choosing the modulation frequency $\Omega/2\pi = 50$ kHz, and with the usual order of magnitude of T_{1N} , the nuclear hyperfine field cannot build up.

For $N_A \ge 2.6 \times 10^{17}$ cm⁻³, and for purer samples above a temperature of approximately 100 K, the decrease of P as a function of transverse magnetic field was found, as expected, to be Lorentzian, so that the determination of ΔB could be done straightforwardly. On the other hand, for purer samples at low temperatures, the depolarization curve was found to be strongly non-Lorentzian. This is shown in Fig. 2(a) in the case of $N_A = 6 \times 10^{16}$ cm⁻³ at $T = 1.7$ K.

Several possible interpretations can be given to this type of behavior. Firstly, nuclei with a very short relaxation time could exist in the sample, so that the nuclear magnetization and consequently the nuclear hyperfine field could follow the electronic magnetization. Although we cannot completely rule out this possibility, we do not believe in its relevance because this would imply unusually short nuclear relaxation times. Moreover, by a standard oblique magnetic field technique, 2^{1} we found a hyperfine field of 15 mT, which is likely to be negligible with respect to the external field. A possible explanation for the non-Lorentzian shape could involve

FIG. 2. Hanle curves recorded at 1.7 K with a sample containing 6×10^{16} acceptors per cubic centimeter. (a) Exciting power density $I_{\text{exc}}=60 \text{ W cm}^{-2}$. Resonant nuclear effects are indicated by arrows. (b) $I_{\text{exc}}=6 \text{ W cm}^{-2}$.

FIG. 3. $P(0)/P(B)$ plot as a function of the square of the magnetic field for the $N_A = 6 \times 10^{16}$ cm⁻³ sample. (a) $I_{\text{exc}} = 60$ $W \text{ cm}^{-2}$. We show here how we determine the width used in the calculation of T_1 . (b) $I_{\text{exc}}=6 \text{ W cm}^{-2}$.

FIG. 4. Temperature dependence of the spin-relaxation time for two of our samples. (a) 1.6×10^{16} cm⁻³. The T^{-3} line was obtained from Eq. (1) with $\tau_p = 10^{-13}$ s and $\alpha = 0.07$. (b) 2×10^{17} cm⁻³. Our results (circles) are compared to those of Clark et al. (Ref. 4) (\times denotes the donor-acceptor transition, + the band-to-band transition) obtained on an epilayer with $N_A = 2 \times 10^{17}$ cm⁻³, and of Aronov et al. (Ref. 9) (triangles) obtained on a crystal with $N_A = 2.2 \times 10^{17}$ cm⁻³; the T^{-n} lines are given as a guide.

FIG. 5. $T_1(T)$ plot for two samples close to the limit of degeneracy. (a) $N_A = 10^{18}$ cm⁻³; (b) $N_A = 3.2 \times 10^{18}$ cm⁻³. We compare our results (circles) to those of Fishman and Lampel (Ref. 5) (crosses) obtained on a sample with $N_A=4\times10^{18}$ cm^{-3} , and of Aronov et al. (Ref. 9) (triangles) obtained on a crystal with $N_A = 5 \times 10^{18}$ cm⁻³. The $T^{-3/2}$ lines are shown as a guide.

resonant dynamic polarization of the nuclei in the frame rotating with the electronic polarization.^{19,22} In the same way, we cannot eliminate the possibility of inhomogeneity of the sample, which in the simplest case could lead to a Hanle depolarization consisting of the superposition of two Lorentzian components having very diferent widths.

At this point, we cannot make a conclusion as to the true reason for this complicated behavior. However, as

FIG. 6. Variation of T_1 as a function of T for two degenerate samples. (a) $N_A = 9 \times 10^{18}$ cm⁻³. (b) $N_A = 2.8 \times 10^{18}$ cm⁻³. The $T^{-3/2}$ lines are given as a guide

seen in Fig. 3(a), the high-field depolarization is indeed found to be Lorentzian. Furthermore, for a low excitation power density, as seen in Figs. 2(b) and 3(b), the curve becomes Lorentzian. Experiments carried out at low or high excitation power yield different values of Hanle width, as can be seen in Fig. 3. However, the values of T_1 calculated using Eq. (9) are the same in both cases, since the change of ΔB is essentially produced by a modification of the electronic lifetime. Thus, even for high-purity samples at low temperature, the measure of ΔB from the high-field Lorentzian depolarization curve gives a reliable value of electronic relaxation time T_1 .

The experimental results are summarized in Figs. 4-6, which show the measured dependences of spin-relaxation time as a function of temperature for the various samples. Most of these curves exhibit essentially the same overall features: there is a nearly constant plateau at temperatures below, typically, 77 K; above this temperature T_1 decreases as $T^{-3/2}$. However, in the particular case of the sample with $N_A = 2 \times 10^{17}$ cm⁻³, there exists an intermediate temperature range where the variatio of T_1 is as $T^{-1/2}$. Note also the exception of the pures sample $(N_A = 1.6 \times 10^{16} \text{ cm}^{-3})$ where the decrease is as T^{-3} .

The inffuence of acceptor concentration on the value of the relaxation time is shown in Figs. 7(a) and 7(b) for 1.7 and 77 K, respectively. For these two temperatures which correspond, respectively, to the plateau and to the

FIG. 7. Variation of T_1 as a function of the acceptor concentration at three different temperatures of the bath: (a) $T = 1.7$ K, (b) $T = 77$ K, and (c) $T = 300$ K. The line on the low concentration range in part (b) was obtained from Eq. (2) with $\Delta_{ex} = 0.15$ and taking into account the variation of N_P/N_A as a function of N_A . Other lines only join experimental points; some slopes are related to the theory [right-hand side of (a), (b), and (c), for instance]. The two other slopes [left-hand side and central part of (a)] are not related to the theory.

onset of the temperature decrease of $T₁$, the overall shape of these dependences is the same. Figure 7(c) shows the corresponding behavior at RT. In this case one finds a very weak temperature dependence at least up to a concentration of 3.2×10^{18} cm⁻³, and an $N_p^{-1/3}$ dependence for higher doping levels.

IV. DISCUSSIGN

In order to compare the experimental results, summarized in Figs. 4-7, with existing theories and with results of other workers, we shall first discuss the $T_1(N_A)$ dependence and we shall analyze the $T_1(T)$ variation in the second part of the present section.

A. Dependence of $T₁$ on the doping level

The overall features of the $T_1(N_A)$ dependence are in agreement with the theoretical predictions. At high doping levels, where the SAP process should be dominant at all temperatures (see Fig. 1) we find an $N_P^{-1/3}$ dependence at 1.7 K, at 77 K, and at RT. This is in agreement with theoretical predictions for degenerate samples [see Eq. (3)]. Note that in the particular case of lower temperatures, the present results differ from those obtained by Miller and co-workers,^{7,8} who find that $1/T_1$ is independent of N_A for $N_A > 5 \times 10^{17}$ cm These authors were led to propose a relaxation mechanism involving an electron-hole exchange scattering process via a virtual photon in order to interpret their results.

At lower doping levels (see Fig. 1) one should expect the DP process to be efficient at room temperature, whereas the SAP one is relevant for lower temperatures. Indeed, at room temperature, $T₁$ does not depend on N_A , as expected for a DP mechanism provided the electronic collision time does not depend on temperature and concentration.

At liquid-nitrogen temperature, according to Eq. (2), a variation of T_1 as a function of N_A is expected from the BAP theory in the case of moderately doped samples. Due to the change of the ionization ratio N_P/N_A with concentration, there is no reason to find a T_1 dependence as N_A^{-1} if the acceptor ionization energy has a constant value, as observed from our luminescence spectra and from the cathodoluminescence results of Cusano.²³ Effectively the T_1 values calculated in these conditions in the (1.6×10^{16}) – (2.6×10^{17}) cm⁻³ concen tration range are found in the line shown in the left-hand side of Fig. 7(b), the slope of which is different from -1 . The agreement with experimental points is reasonable.

The weak increase of T_1 in the intermediate concentration range [Fig. 7(b)] may also be explained by the BAP process. Below $N_A = 7 \times 10^{17}$ cm⁻³ the acceptor ionization energy is independent of the Cd or Zn amount,²³ whereas the Sommerfeld factor $|\psi(0)|^2$ is decreasing due to the screening of the electron-hole interaction. When the hole-hole mean distance becomes lower than the Debye length, this factor is no longer given by $|\psi(0)|^2 = 2\pi /X[(1-e^{2\pi/X})]$ with

 $\chi = (\varepsilon_e / E_B)^{1/2}$, but by another more complicated expression given by Bir et al .¹⁴ which may yield values close to one.

On the other hand, at liquid-helium temperatures, the N_A^{-1} dependence predicted by the BAP process is not in agreement with the experimental results. Indeed, as seen in Fig. 7(a), we find an $N_A^{-2/3}$ dependence from 1.6×10^{16} cm⁻³. The inability of the BAP process to explain the results obtained at 1.7 K for the low doping levels will be further illustrated in the following section.

B. Dependence of $T₁$ on temperature

2. High-temperature range

A further verification of the ideas summarized in Fig. 1, and used for interpreting the dependence of T_1 on acceptor concentration, can be obtained by analyzing the rapid decrease of T_1 as a function of temperature for T larger than 100 K.

(i) For the purest samples $(N_A \le 6 \times 10^{16} \text{ cm}^{-3})$, the DP mechanism is largely dominant down to 77 K and a clear T^{-3} behavior is observed, as can be seen in Fig. 3(a), which corresponds to our 1.6×10^{16} cm⁻³ sample. Such a dependence may be explained using Eq. (1) with α =0.07 and a constant electronic momentum relaxation time $\tau_p = 10^{-13}$ s. We used these values, which were found by Maruschak et al.¹⁰ working on a 4×10^{16} cm specimen, because hole mobility measurements which permit one to obtain the values of a and τ_n were not realized in our case. The expected temperature decrease of τ_p above 80 K seems to be very weak.

(ii) The $T^{-3/2}$ dependence of T_1 observed in the case of degenerate samples with $N_A \ge 9 \times 10^{18}$ cm⁻³ [Figs. 6(a) and 6(b)] offers evidence of the predominant role of the BAP process. Such a dependence is deduced from Eq. (3) concerning degenerate holes in the case of "fast" thermalized electrons; the Sommerfeld factor $|\psi(0)|^2$ can be taken equal to unity, as shown by model calcula; $tion.²⁴$

Our expenmental results obtained for degenerate samples were fitted to Eq. (3) using a value of the exchange splitting $\Delta_{\rm ex}$ of the exciton ground state equal to 0.1 meV which is in relatively good agreement with its known estimate of (0.05 ± 0.05) meV (Ref. 25) and the value of 0.047 meV found by Aronov et al.

(iii) For intermediate acceptor concentrations, we expect the two relaxation processes to have comparable efficiencies. Above 150 K the variation of T_1 is then determined by the DP mechanism which becomes active. Effectively Eq. (1) shows that $1/T_1 \sim \tau_P T^3$. As the mobility, ²⁶ and consequently τ_p , is likely to be proportion to $T^{-3/2}$ one expects that $1/T_1$ varies as $T^{3/2}$. Our $T_1(T)$ variation above 160 K is in good agreement with this prediction.

For $N_A = 10^{18}$ and 3.2×10^{18} cm⁻³, this competition is clearly evidenced by the presence of a shoulder between 150 and 200 K. A detailed analysis of the results between 80 K and RT allows us to distinguish three different behaviors

Firstly, the $T^{-3/2}$ dependence of T_1 from 80 to 150 K corresponds to a degenerate behavior (as observed by Aronov et al.⁹ using a $N_A = 5 \times 10^{18}$ cm⁻³ sample) which can be explained by means of the BAP mechanism [Eq. (3)]. Secondly, above 200 K, the DP process is dominant and a good description of the experimental data may be obtained.

Finally, to describe the beginning of saturation observed between 150 and 200 K, which corresponds to a nondegenerate behavior, it is mandatory to use Eq. (2) characterizing the BAP process for electron scattering by both free holes and holes bound to acceptors. However, in this concentration range, it is known that the acceptor ionization energy depends on the doping level^{23,27} so that the relative concentrations of free and bound holes may change with both temperature and doping level. As numerous adjustable parameters must be introduced in the two BAP expressions, the detailed analysis of our results was not undertaken.

2. Lou-temperature range

In order to interpret the temperature dependence of electronic relaxation time $T₁$ for temperatures lower than 100 K, we point out that special attention must be paid to the fact that the temperature T_e of the electron gas is likely to be larger than the lattice temperature. This is due to the existence of a finite rate of exchange of energy between the electron gas and the lattice. The value of T_e can be measured quite simply from the slope of the high-energy side of the luminescence line. This value is given in Fig. 8, as a function of temperature for $N_A = 2 \times 10^{17}$ cm⁻³. One sees that for $T > 77$ K, one has $T_e = T$. On the other hand, for $T < 77$ K, the electronic temperature is almost independent of lattice temperature and equal to 60 K for the excitation density that we use.

In the case of degenerate samples, using Eq. (3), we obtain a good fit with the experimental results using the value of electronic temperature instead of the lattice temperature. This is shown in Fig. 9 for $N_A = 2.8 \times 10^{19}$ $cm⁻³$. The theoretical variation (solid line) is obtained using Eq. (3) and $\Delta_{ex} = 0.1$ meV. Nevertheless, and in accordance with the $T_1(N_A)$ variation, it can be seen that at liquid-helium temperatures, the experimental relaxation velocity is greater than that deduced from the SAP mechanism.

FIG. 8. Dependence of the electronic temperature T_e on the bath temperature T for the $N_A = 2 \times 10^{17}$ cm⁻³ sample. I_{exc} = 60 W cm⁻².

FIG. 9. Theoretical fit of our results on the 2.8×10^{19} cm⁻³ sample obtained using Eq. (3) with $\Delta_{ex} = 0.1$ and where the temperature is that of the electron gas T_e (T_e is greater than T below LNT).

In the case of samples with intermediate doping levels, we are able to predict the overall variation of the $T_1(T)$ dependence using the BAP model. This is shown in Fig. 10 for the $N_A = 2 \times 10^{17}$ cm⁻³ sample. The dashed line is obtained from Eq. (2) using T_e instead of T with Δ_{ex} = 0.15 meV. This value is comparable with (but not equal to) that of 0.¹ meV used for degenerate samples. The experimental data are satisfactorily interpreted up to 150 K where the DP process becomes active. Note that the solid line obtained using the lattice temperature is unable to interpret the data, with the exception of the small 10-30-K temperature interval.

In the case of weakly doped samples $(N_A \le 6 \times 10^{16}$ cm^{-3}), where (see Fig. 1) we also expect the BAP process to be relevant at low temperature, we have previously seen that the $T_1(N_A)$ dependence is not in agreement with the predictions of the BAP model. The same type of feature is revealed by the $T_1(T)$ dependence. Even if we take the value of the electron gas temperature, we do not predict the experimental results satisfactorily. This is seen in Fig. 11, concerning the $N_A = 1.6 \times 10^{16}$ cm⁻³ sample, where the curve a is obtained using Eq. (2) with Δ_{ex} = 0.15 meV. On the same figure it can be seen that the DP process has too great an efficiency below 100 K.

FIG. 10. Theoretical fit of our results on the 2×10^{17} cm⁻³ sample, obtained using Eq. (2) with $v_e = (3k_B T/m_e^*)^{1/2}$, $\Delta_{\text{ex}} = 0.24 \text{ meV (curve } a); v_e = (3k_B T_e/m_e^*)^{1/2} \text{ below LNT, and}$ $v_e = (3k_B T/m_e^*)^{1/2}$ above LNT and $\Delta_{ex} = 0.15$ meV (curve b).

FIG. 11. Theoretical curves $T_1(T)$ obtained in the case of the $N_A = 1.6 \times 10^{16}$ cm⁻³ sample using (a) Eq. (2) with $\Delta_{ex} = 0.15$ meV and $v_e = [(3k_BT_e)/(m_e^*)]^{1/2}$ (BAP process); and (b) Eq. (1) with $T = T_e$ below 80 K (DP process). The circles are the experimental points.

Curve b was obtained using Eq. (1) with $Q = 1.5$ (diffusion by ionized impurities), $\tau_{pe} = 10^{-13}$ s, and T_e (rather than T). The use of the electronic temperature in the equations describing the two processes is able to explain the existence of the low-temperature plateau. Nevertheless the addition of the two processes leads to T_1 values still lower than the experimental ones.

To conclude this subsection we shall say that our results are not well understood by the light of the existing theories. Eftectively, according to Fig. 1, the relevance of the BAP process was expected in the whole concentration range. We have seen that this is not the case, particularly below $N_A = 2.6 \times 10^{17}$ cm⁻³ (variation as $N_A^{-2/3}$ rather than N_A^{-1}). If an $N_p^{-1/3}$ slope, as predicted by the SAP theory, was obtained for degenerate samples, the calculated T_1 values are always longer than those we measured. On the other hand, we recall that for the purest samples $(N_A \le 6 \times 10^{16} \text{ cm}^{-3})$ the DP mechanism was found to have, in total contradiction to Fig. 1, too strong an efficiency below 100 K. Therefore, at liquid-helium temperatures, we are led to assume the existence of an unknown relaxation process which would explain the experimental results whatever the concentration.

C. Comparison with results obtained by others

Most of the results presented here are in agreement with the theoretical predictions. However, significant discrepancies appear, particularly between our data and the experimental results obtained by other groups. There is first the case of degenerate samples at low temperature which we have already discussed. We point out now that discrepancies also exist in the case of $N_A = 2 \times 10^{17}$ cm⁻³. As seen in Fig. 4(b), the present results differ from the results of Clark et $al.$,⁴ as well as from these of Aronov et $al.^9$ In the preceding subsection the overall shape of our $T_1(T)$ variation is interpreted by the HAP model using the value of the electronic temperature rather than that of the sample. Qn the other hand, Aronov et al .⁹ interpret their results by supposing a dependence of the acceptor ionization energy on the doping level, which does not seem to agree with the cathodoluminescence results of $Cusano²³$ and with our experimental observation for $N_A \leq 7 \times 10^{17}$ cm

In fact, several uncontrolled parameters are likely to influence the spin-lattice relaxation, so that the results might be in some cases sample dependent. There is first the possible presence of paramagnetic impurities of unknown origin which might create other relaxation processes. Internal stresses might also play a role in the 'value of T_1 .¹⁹ Finally, shallow acceptor levels are strongly sensitive to internal stresses which could modify the value of P , in the case of donor-acceptor recombina $tion.²⁸$

V. CONCLUSION

In this work we have investigated, by means of the optical-pumping technique, the variation of spinrelaxation time T_1 of the photogenerated electrons in ptype GaAs specimens with an increasing amount of acceptor concentration N_A . The T_A dependences as a function of N_A or as a function of the observation temperature T were compared to the predictions of the various theories dealing with relaxation processes in zincblende-type semiconductors.

Concerning the variation of T_1 as a function of N_A we have found that either the HAP mechanism or the DP process are able to explain our experimental results down to liquid-nitrogen temperature. At liquid-helium temperatures, it was found that none of the known relaxation processes could explain our results, in the whole concentration range. Nevertheless, if we except the purest samples $(N_A \le 6 \times 10^{16} \text{ cm}^{-3})$, the use of the value of the electronic temperature T_e rather than the sample temperature T in the BAP formulas dealing with moderately doped or degenerate samples leads to a good fit of our experimental results from about 150 down to 10 K, assuming a value of the exchange splitting $\Delta_{\rm av}$ of the exciton ground state equal to (0.12 ± 0.03) meV.

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- ¹G. Lampel, Phys. Rev. Lett. 20, 491 (1968).
- ${}^{2}G$. Lampel, in Proceedings of the 12th International Conference on the Physics of Semiconductors, Stuttgart, 1974, edited by M. H. Pilkuhn (Teubner, Stuttgart, 1974), p. 743.
- ³C. Hermann, G. Lampel, and V. I. Safarov, Ann. Phys. (Paris) 10, 313 {1985).
- ⁴A. H. Clark, R. D. Burnham, D. J. Chadi, and R. M. White, Solid State Commun. 2Q, 385 (1976).
- 5G. Fishman and G. Lampel, Phys. Rev. B 16, 820 (1977).
- 6V. I. Safarov and A. N. Titkov, in Proceedings of the 15th International Conference on the Physics of Semiconductors, Kyoto, 1980 [Suppl. A, J. Phys. Soc.Jpn. 49, 623 (1980)].
- ⁷D. A. Kleinman and R. C. Miller, Phys. Rev. Lett. 46, 68 $(1981).$
- ⁸R. C. Miller, D. A. Kleinman, W. A. Nordland, Jr., and R. A. Logan, Phys. Rev. B 23, 4399 (1981).
- ⁹A. G. Aronov, G. E. Pikus, and A. N. Titkov, Zh. Eksp. Teor. Fiz. 84, 1170 (1983) [Sov. Phys. -JETP 57, 680 (1983)].
- ¹⁰V. A. Maruschak, M. N. Stepanova, and A. N. Titkov, Fiz. Tverd. Tela (Leningrad) 25, ³⁵³⁷ (1983) [Sov. Phys.—Solid State 25, 2035 (1983)].
- ¹¹R. J. Elliott, Phys. Rev. 96, 266 (1954).
- 12 Y. Yafet, in Solid State Physics, edited by F. Seitz and D. Turnbull {Academic, New York, 1963), Vol. 14, pp. ¹—98.
- ¹³M. I. D'yakonov and V. I. Perel', Zh. Eksp. Teor. Fiz. 60, 1954 (1971) [Sov. Phys.—JETP 33, 1053 (1971)].
- ¹⁴G. L. Bir, A. G. Aronov, and G. E. Pikus, Zh. Eksp. Teor. Fiz. 69, ¹³⁸² (1975) [Sov. Phys.—JETP 42, ⁷⁰⁵ (1976)].
- ¹⁵E. O. Kane, J. Phys. Chem. Solids 1, 249 (1957).
- ¹⁶C. Weisbuch and C. Hermann, Phys. Rev. B 15, 816 (1977).
- ¹⁷D. Olego and M. Cardona, Phys. Rev. B 22, 886 (1980). corresponds, according to the temperature, either to the D^0 -
- 18 For nondegenerate samples, the maximum luminescence peak h or to the D^0 - A^0 transition. In this last case the D^0 -h transition appears as a small shoulder on the high-energy side of the luminescence spectrum. The value of P is, within the experimental error, constant between the shoulder and the D^0 - $A⁰$ peak. In the case of degenerate samples for which all the peaks appear at energies greater than the gap one, the values of P measured at various points of the luminescence spectrum are the same within the experimental uncertainty.
- ¹⁹Optical Orientation, Modern Problems in Condensed Matte. Science, edited by F. Meier and B. P. Zakharchenya (North-Holland, Amsterdam, 1984), Vol. 8.
- 20D. Paget, G. Lampel, B. Sapoval, and V. I. Safarov, Phys. Rev. B 15, 5780 (1977).
- 2'M. I. D'yakonov, V. I. Perel', V. L. Berkovits, and V. I. Safarov, Zh. Eksp. Teor. Fiz. 67, ¹⁹¹² (1974) [Sov. Phys.— JETP 40, ⁹⁵⁰ (1975)].
- 22V. K. Kalevitch, V. D. Kul'kov, and V. G. Fleisher, Fiz. Tverd. Tela 23, ¹⁵²⁴ (1981) [Sov. Phys.—Solid State 23, ⁸⁹² (1981}].
- ²³D. A. Cusano, Solid State Commun. 2, 353 (1964).
- 24G. L. Bir, G. E. Pikus, and A. S. Skal, Fiz. Tekh. Poluprovodn. 8, 1096 (1974) [Sov. Phys. - Semicond. 8, 715 (1974)].
- 25D. D. Sell, S. E. Stokowski, R. Dingle, and J. V. Di Lorenzo, Phys. Rev. B 7, 4568 {1973).
- ²⁶P. Kireev, La Physique des Semiconducteurs (Mir, Moscou, 1975), p. 461.
- ²⁷Sh. M. Gasanli, O. V. Emel'yanenko, V. K. Ergakov, F. P. Kesmanly, T. S. Lagunova, and D. N. Nasledov, Fiz. Tekh. Poluprovodn. 5, 1888 (1971) [Sov. Phys.—Semicond. 5, 1641] (1972)].
- ²⁸G. Bacquet, J. Bandet, F. Fabre, J. Frandon, and D. Paget, J. Appl. Phys. 58, 3541 (1985).