Spin relaxation in *n*-doped GaAs due to impurity and electron-electron Elliot-Yafet scattering

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We calculate the spin-relaxation time of conduction electrons in *n*-doped bulk GaAs. We consider the Elliot-Yafet spin-relaxation mechanism, driven by Coulombic-impurity and electron-electron scattering. We find that these two scattering mechanisms result in relaxation times of equal order of magnitude, but with dissimilar dependences on doping density and temperature. Our theoretical results are compared with experimentally measured spin-relaxation times in gallium arsenide.

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There is currently a great interest in the properties of semiconductors derived from the electronic spin. This interest stems largely from the foreseen growth of the scope of applications of the spin degree of freedom in electronics (spin tronics) and computer science (quantum computing).[1,2] Among the properties of interest in relation to potential applications, a central place is occupied by the spin-relaxation time, i.e., the characteristic time a spin-density imbalance lasts inside a given material or structure. Clearly, long relaxation times are generally desirable, and an important progress in the search of systems with long relaxation times has been made in a series of recent experiments.[3–5] In those experiments it was found, among other things, that the spin-relaxation time in semiconductors can be extended by more than two orders of magnitude with appropriate negative-type doping.

Several authors have recently analyzed this issue from a theoretical point of view. Song and Kim[6] reviewed the known spin-relaxation mechanisms and constructed a phase diagram to depict graphically which mechanism is dominant in each portion of parameter space. According to those authors, the D'yakonov-Perel (DP) (Ref. 7) mechanism dominates over the Elliot-Yafet (EY) one[8,9] (driven by electron-impurity scattering) except at low temperature in *n*-doped zincblende semiconductors. Wu and Ning[10] studied D'yakonov-Perel spin relaxation in *n*-doped GaAs. Their main finding, as far as explaining the above-mentioned experiments is concerned, is that the DP mechanism produces an increasing relaxation time as a function of applied magnetic field, in qualitative agreement with experiment at high density.[4] A related study, based on a kinetic equation formalism and including the k dependence of the conduction-band g factor, was done by Bronold *et al*.[11] These authors found a good agreement between their theory and experiment[4] for the relaxation time as a function of magnetic field at high doping density. While the high-doping-density case can be treated in terms of delocalized electrons—electrons at the bottom of the conduction band—the low-density regime calls for a description based on electrons localized at the impurity sites. Kavokin[12] and Gor'kov and Krotkov[13] studied this regime. The latter authors conclude that the Dzyaloshinskii-Moriya interaction between localized electrons does not explain the experimentally observed spin-relaxation times, as was claimed by Kavokin.

Here we report our calculations of the spin-relaxation time of electrons in the conduction band of *n*-type semiconductors caused by the EY scattering mechanism[8,9] mediated by both electron-impurity and electron-electron interactions. Our calculations extend those of Chazalviel[14] and Boguslawski[15] for electron-impurity and electron-electron scattering, respectively. Both of these authors considered only the case of almost equal up-spin and down-spin populations. Furthermore, Chazalviel studied only the zero-temperature limit while Boguslawski treated the high-temperature case by introducing Boltzmann rather than Fermi distributions. We generalize those calculations by considering arbitrary spin populations described by finite temperature Fermi distributions. Also, while those authors were mainly interested in indium antimonide, we concentrate here on GaAs, which is one of the preferred semiconductor alloys in recent studies of spin relaxation.

As it is clear that only an incomplete understanding of the experiments reported in Ref. 4 has emerged so far, our aim here is to provide a missing piece in the set of theoretical scenarios considered up to now. We point out that none of the recent theoretical studies of spin relaxation in *n*-type GaAs has taken into account the *electron-electron* EY mechanism, which had been studied by Boguslawski[15] in a different context.

The Coulomb interaction is independent of spin and therefore cannot cause spin-flip transitions between conduction-band states that are spin eigenstates. The possibility of a spin flip in the EY scattering mechanism arises from the fact that the conduction-band states of some semiconductors are not spin eigenstates, which in turn is due to the spin-orbit contribution to the crystal Hamiltonian. The conduction-band states of zincblende semiconductors are linear combinations of both spin eigenstates with coefficients that are functions of the crystal momentum $\mathbf{k}$. Usually one of the two components in these admixtures has a much larger amplitude than the other. Therefore, the mixed states retain a clear correspondence to the original pure-spin states, and are still referred to as the "spin-up" and "spin-down" states. We denote these states by $|\mathbf{k}\pm\rangle$. The matrix element of a spin-independent scattering potential (like that of ionized impurities and electron-electron interaction) between these states can thus be nonzero, thereby activating the spin-flip EY scattering mechanism.

Our general goal is to understand the spin-relaxation process in n-doped bulk semiconductors studied recently in ultrafast Faraday rotation experiments[4]. In this paper we do not investigate the initial stages of the relaxation process, when electron-hole scattering is predominant, but concentrate rather on the slower decay that takes place after the electron-hole recombination has been completed. Therefore, the starting point of our calculation is the assumption that there is a doped unpolarized electron gas of density n present in the host semiconductor, and an additional density n_{ex} of photoexcited electrons occupying conduction-band states of the type $|\mathbf{k}+\rangle$. Since we "look" at the system at least several hundreds of picoseconds after the pump pulse has created the photoexcited polarized electrons, we assume that the two spin species have reached thermal equilibrium and can be described by Fermi-Dirac distributions $f_{\pm}(\mathbf{k})$ with a common temperature T. We restrict our treatment to cases where $n_{ex}<n$.

We are interested in the relaxation of a spin-density difference $n_d \equiv n_+ - n_-$, where $n_{\pm}=(1/V)\,\Sigma_{\mathbf{k}} f_{\pm}(k)$ are the densities of spin-up and spin-down electrons in the conduction band. We assume that initially $n_d = n_{ex}$. The relaxation rate is defined by

$$\frac{1}{T_1} \equiv -\frac{\dot{n}_d}{n_d} = -\frac{\dot{n}_+ - \dot{n}_-}{n_d} = -\frac{2\dot{n}_+}{n_d}. \tag{1}$$

Here we have used that $\dot{n}_+ + \dot{n}_- = 0$ since no further excitation or recombination takes place in the considered time regime.

For electron-impurity scattering, the relaxation rate is calculated as

$$\begin{aligned}\frac{1}{T_1} &= -\frac{2n_i}{n_d}\sum_{\mathbf{k}}\sum_{\mathbf{k}'} f_-(k')[1-f_+(k)]T_{\mathbf{k}'-\to\mathbf{k}+} \\ &\quad - f_+(k)[1-f_-(k')]T_{\mathbf{k}+\to\mathbf{k}'-} \\ &= \frac{2n_i}{n_d}\sum_{\mathbf{k}}\sum_{\mathbf{k}'} [f_+(k)-f_-(k')]T_{\mathbf{k}+\to\mathbf{k}'-},\end{aligned} \tag{2}$$

where we have denoted the density of impurity scatterers by n_i, and used $T_{\mathbf{k}'-\to\mathbf{k}+} = T_{\mathbf{k}+\to\mathbf{k}'-}$. The spin-flip transition rate due to the impurity EY mechanism is

$$\begin{aligned}T_{\mathbf{k}+\to\mathbf{k}'-} &= \frac{2\pi}{\hbar}\delta(E_{\mathbf{k}'}-E_{\mathbf{k}})|\langle\mathbf{k}'-|V_i|\mathbf{k}+\rangle|^2 \\ &\approx \frac{2\pi}{\hbar}\delta(E_{\mathbf{k}'}-E_{\mathbf{k}})V_i(\mathbf{k}-\mathbf{k}')^2|\langle\mathbf{k}'-|\mathbf{k}+\rangle|^2.\end{aligned} \tag{3}$$

The electron-impurity scattering is caused by the Coulomb potential of the ionized silicon donors,[16] whose Fourier transform is given by $V_i(\mathbf{q})=4\pi e^2/\epsilon V(q^2+k_s^2)$, where ϵ is the lattice dielectric constant, V is the volume, and the screening wave vector in the quantum Debye-Hückel theory for a degenerate electron gas is $k_s=(6\pi e^2 n/\epsilon E_F)^{1/2}$. For n and E_F in this expression we take the values corresponding to the doped unpolarized electron gas. The spin-mixed conduction-band states of zincblende semiconductors can be calculated with the $\mathbf{k}\cdot\mathbf{p}$ perturbation theory.[17] The scalar product of opposite-spin states is given by[14,15]

$$\langle\mathbf{k}'-|\mathbf{k}+\rangle = \frac{\gamma\hbar^2}{4m^*E_G}(k_z k'_+ - k'_z k_+), \tag{4}$$

where $\gamma = 2\Delta(\Delta+2E_G)/(\Delta+E_G)(2\Delta+3E_G)$, $k_+ = k_x + ik_y$, m^* is the conduction-band effective mass, Δ is the valence-band spin-orbit splitting, and E_g is the band-gap energy. Substituting Eq. (4) into Eq. (3) yields

$$\begin{aligned}T_{\mathbf{k}+\to\mathbf{k}'-} &= \frac{2\pi}{\hbar}\delta(E_{\mathbf{k}'}-E_{\mathbf{k}})V_i(\mathbf{k}-\mathbf{k}')^2\left(\frac{\gamma\hbar^2}{4m^*E_G}\right)^2 \\ &\quad\times(k^2k_z'^2 + k'^2k_z^2 - 2k_zk'_z\mathbf{k}\cdot\mathbf{k}').\end{aligned} \tag{5}$$

Integrating over $\mathbf{k}'$ and the angular part of the integral over $\mathbf{k}$ one obtains

$$\begin{aligned}\frac{1}{T_1} &= \frac{n_i}{n_d}\frac{\hbar\gamma^2}{6\pi m^* E_G^2}\frac{e^4}{\epsilon^2}\int dk k^3[f_+(k)-f_-(k)] \\ &\quad\times\left[-2+(1+a)\ln\left(\frac{2+a}{a}\right)\right],\end{aligned} \tag{6}$$

where $a \equiv (k_s/k)^2/2$.

For electron-electron scattering, the relaxation rate is calculated from

$$\begin{aligned}\frac{dn_+}{dt} &= \sum_{\substack{\mathbf{k}\,\mathbf{k}'\\ \mathbf{q}\,\sigma}}\{f_-(\mathbf{k})f_\sigma(\mathbf{k}')[1-f_+(\mathbf{k}+\mathbf{q})][1-f_\sigma(\mathbf{k}'-\mathbf{q})]T_{\mathbf{k},-;\mathbf{k}',\sigma\to\mathbf{k}+\mathbf{q},+;\mathbf{k}'-\mathbf{q},\sigma} \\ &\quad - f_+(\mathbf{k})f_\sigma(\mathbf{k}')[1-f_-(\mathbf{k}+\mathbf{q})][1-f_\sigma(\mathbf{k}'-\mathbf{q})]T_{\mathbf{k},+;\mathbf{k}',\sigma\to\mathbf{k}+\mathbf{q},-;\mathbf{k}'-\mathbf{q},\sigma}\},\end{aligned} \tag{7}$$

where $T_{\mathbf{k}_1,\pm;\mathbf{k}'_1,\sigma\to\mathbf{k}_2,\mp;\mathbf{k}'_2\sigma}$ are the transition probabilities of scattering events where one of the electrons flips spin and the other does not. The likelyhood of double spin flips can be neglected. Taking into account the direct and exchange terms, the transition probability according to Fermi's golden rule is given by

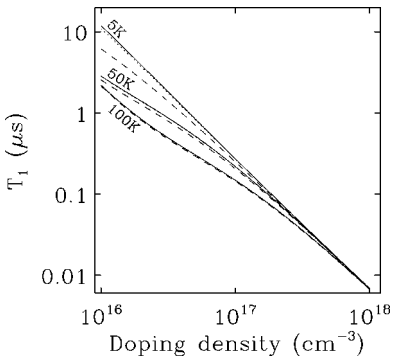

FIG. 1. Impurity EY spin-relaxation time of GaAs vs doping density at three different temperatures (5 K, 50 K, and 100 K) and three spin-density differences: $n_d=10^{14}$ cm^{-3} (solid lines), 10^{15} cm^{-3} (dotted lines), and 10^{16} cm^{-3} (dashed lines).

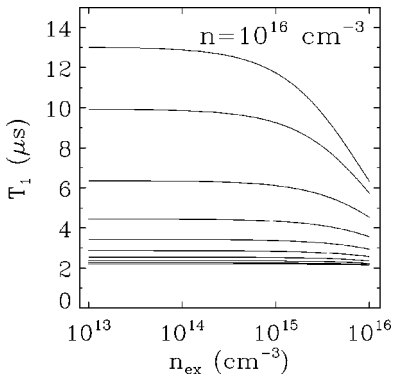

FIG. 2. Impurity EY spin-relaxation rate of GaAs vs spin-density difference ($n_d=n_{ex}$) for doping density $n=10^{16}$ cm^{-3} (equal to the impurity density n_i.) The various curves correspond, from top to bottom, to $T=1,10,20,\ldots,100$ K.

$$T_{\mathbf{k},-;\mathbf{k}',\sigma\to\mathbf{k}+\mathbf{q},+;\mathbf{k}'-\mathbf{q},\sigma}=\frac{2\pi}{\hbar}\delta(E_f-E_i)\cdot|\langle\mathbf{k}+\mathbf{q},+;\mathbf{k}'-\mathbf{q},\sigma|V|\mathbf{k},-;\mathbf{k}',\sigma\rangle-\langle\mathbf{k}'-\mathbf{q},\sigma;\mathbf{k}+\mathbf{q},+|V|\mathbf{k},-;\mathbf{k}',\sigma\rangle|^2. \quad (8)$$

The interaction potential here is again the screened Coulomb potential given above. Its matrix elements between two-particle states are given again by Eq. (4) after a few standard approximations are made.[15] The electron-electron EY relaxation time is obtained by solving Eq. (7) numerically.[18]

We first present results for the impurity EY mechanism obtained numerically from Eq. (6). This equation is valid for all zincblende semiconductor alloys, but in this paper we concentrate on GaAs, given the relative importance of this material in recent experimental studies. For a given material, the impurity EY spin-relaxation time depends on the dopant density n_i, the density of the excess electrons with spin up $n_d=n_{ex}$, and the temperature T. The density of donated electrons n, which also enters implicitly in the calculation, is taken to be $n=n_i$. We envision that the initial spin-density difference n_d comes from the photoexcitation caused by a circularly polarized pump laser pulse in a pump-and-probe experiment.

Our calculations show that for impurity EY scattering the dominant factors in the determination of T_1 [according to Eq. (6)] are the densities n and n_i. Therefore, we show first the time T_1 versus n for three different values of n_d and of T (Fig. 1). At low temperature and low spin-density imbalance, T_1 decreases by a factor of 1000 for a variation of a factor of 100 in n; thus, T_1 follows roughly a power law on n with an exponent of $-3/2$. This dependence comes from both n and n_i. The density of scattering centers n_i enters linearly in Eq. (6), while the density of donated electrons n affects the integral in that equation through the screening length $1/k_s$ and the Fermi functions.

The dependence of T_1 on T is strongest at low values of n, as could be expected from reasoning on the effect of temperature on Fermi functions for different densities. For $n=10^{18}$ cm^{-3} there is practically no temperature dependence up to the highest temperature considered here, $T=100$ K. The relaxation time also depends very weakly on n_d, except when n_d approaches n.

In Fig. 2 we present more detailed data on T_1 versus the spin-density imbalance n_d for doping density $n=10^{16}$ cm^{-3}. Temperatures from 1 K to 100 K are considered, as indicated in the figure caption. The main conclusion we can draw from Fig. 2 is that there is essentially no variation of T_1 with n_d for $n_d<n/10$. (This conclusion is also valid for $n=10^{17},10^{18}$ cm^{-3}, data not shown.) The variation of n between $n_d=n/10$ and $n_d=n$ is roughly of a factor 2 at low temperature, and almost none at high temperature. (The definition of low and high T depends on n, as mentioned above.)

We shall now move on to the results for the electron-electron EY scattering. We define the relative density n_{exr} as the ratio between the density of photoexcited electrons and

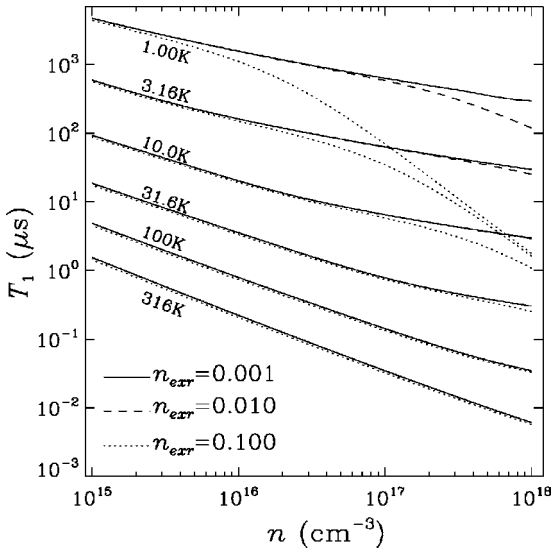

FIG. 3. Electron-electron EY relaxation time vs doping density n for several values of the temperature and the relative density of photoexcited electrons n_{exr}.

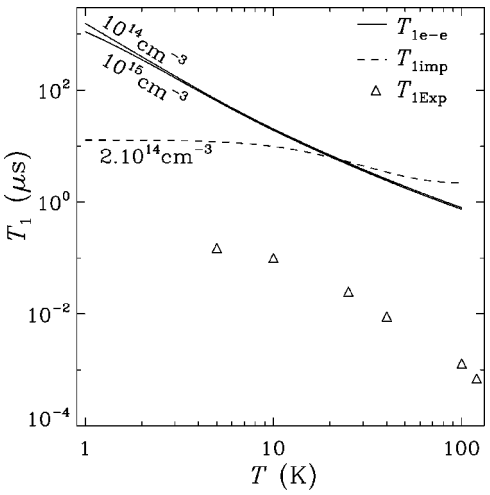

FIG. 4. Spin-relaxation time vs temperature. Comparison between experimental data (Ref. 4) and results of the electron-electron EY scattering and impurity scattering calculations. The theoretical curves are for $n=10^{16}\,\mathrm{cm}^{-3}$ and the values of n_{ex} are indicated on the graph.

the doping density, $n_{exr}\equiv n_{ex}/n$. Figure 3 shows the spin relaxation time T_1, obtained by solving Eq. (7) and using Eq. (1), as a function of n for $n_{exr}=0.001,0.01,0.1$, and various temperatures between 1 K and 315 K. Once again we obtain that T_1 decreases rapidly with doping density much as it does in the case of impurity EY scattering. On the other hand, Fig. 3 shows also a strong temperature dependence, not present for impurity scattering.

In Fig. 3 we can identify different regimes that can be broadly classified according to the doping density and the temperature. A high-temperature regime appears for $T>10$ K and $n\lesssim 10^{16}\,\mathrm{cm}^{-3}$, or $T>T_F$, where T_F is the Fermi energy of the doped electron gas. The low-temperature regime is located on the ''opposite corner (upper-right)'' of the plot, and is given roughly by $T<3.2$ K and $n\gtrsim 10^{17}\,\mathrm{cm}^{-3}$ $(T<T_F)$. Other combinations of n and T correspond to $T\approx T_F$. The following main conclusions can be drawn from Fig. 3: (i) At high temperature $(T_F<T)$, T_1 does not depend significantly on n_{exr}; (ii) at low temperature $(T<T_F)$, as n increases T_1 decreases and this decrease is sensibly greater the higher the value of n_{exr}; (iii) at low temperature (see curves for $T=1$ K, 3.16 K), high n_{exr} (see $n_{exr}=0.1$), and high density $(n\approx 10^{18}\,\mathrm{cm}^{-3})$, the temperature dependence of T_1 disappears. We have analyzed in detail the dependence of T_1 on n, T, and n_{exr}, but at this point we shall just refer the interested reader to Ref. 18.

We now compare our theoretical results with the corresponding experimental findings of Ref. 4. The longest spin-relaxation times reported in Ref. 4 were obtained at low magnetic field and with a doping density of $n=10^{16}\,\mathrm{cm}^{-3}$. In Fig. 4 we plot the relaxation time versus temperature for that situation. The two solid curves are results from our electron-electron (e-e) EY scattering calculation and the triangles are experimental data from Ref. 4. The result for impurity EY scattering is plotted as a dashed line. Although the experimental points have a similar qualitative behavior as the minimum between our e-e and impurity curves, there is a disagreement of roughly two orders of magnitude between the experimental points and the calculated curves. The fact that both types of EY relaxation times are much longer than the ones measured experimentally could indicate that another, more efficient, spin-flip mechanism dominates the spin relaxation in this system. A partial agreement between experiment and theory has been found in terms of the D'yakonov-Perel spin-flip mechanism in a calculation that includes a detailed treatment of the semiconductor band structure and experimentally measured mobilities as input parameters.[19] That treatment seems to fit the experimental relaxation times at a temperature of around 100 K, but is off by two orders of magnitude at low temperature.

In Ref. 4 it is mentioned that the e-e EY mechanism could help understand the temperature dependence of the relaxation time at low temperature (below 30 K). However, as noted by the authors, the estimates based on the treatment of Boguslawski,[15] which assumes classical statistics, yield a relaxation rate that is actually too fast compared to experiment. Our calculation of the e-e EY mechanism shows that this mechanism, when it is evaluated using the appropriate Fermi-Dirac distribution, yields relaxation times that are much longer than those measured experimentally. That is to say that the Fermi distribution and Pauli exclusion radically reduce the effectiveness of the e-e EY relaxation mechanism, and therefore Boguslawski's results cannot be used to interpret experiments at low temperature.

A partial agreement between impurity EY scattering and experiment was reported in Ref. 4 for a temperature of about 20 K, at zero magnetic field. This agreement contrasts with the disagreement between theory and experiment that we report here for the impurity EY scattering relaxation. The explanation for these conflicting estimates of the impurity EY scattering rate must be sought in the fact that the fit in Fig. 4(b) of Ref. 4 is obtained using experimentally measured mobilities as input parameters in the expression of the momentum-scattering rate (which is proportional to the spin scattering rate in the theory of Chazalviel). This fact, together with the partial agreement obtained by Lau *et al*. mentioned earlier,[19] which also relies on experimentally measured mobilities as input parameters, indicates that the probable cause of disagreement between our theory and experiment is not specific to spin-flip scattering, but that it would also be reflected in comparisons of momentum-scattering rates.

Therefore, we cannot rule out that a more refined version of the EY calculation presented here could help explain the measured relaxation times in *n*-doped GaAs, particularly at low temperature. The refinement in the calculation should probably be made on the expression of the spin-mixed conduction-band states, the treatment of screening of the Coulomb potential, and also the Born approximation to scattering rates used here and in Refs. 14 and 15. Furthermore, at low density the possibility of electron localization at impurity sites due to the metal-insulator transition has to be taken into account.[12,13]

There is an interesting magnetic-field dependence in the spin-relaxation times which has not been addressed in this

paper. Notably, the experiments show a strong reduction of T_2^* with applied magnetic field at a doping density of $n = 10^{16}\ \mathrm{cm}^{-3}$ (Ref. 4). At higher doping densities, the magnetic-field dependence is less strong and it is well described by a combination of the DP mechanism and the variation of the g factor with momentum.[10,11] Given that EY scattering is possibly a relevant spin-flip mechanism at densities around $n = 10^{16}\ \mathrm{cm}^{-3}$ and low temperatures (roughly below 30 K), it is worth extending our calculations to the case of finite applied magnetic field and to confront the results with the available experimental data.[4] We plan to undertake that study in the future.

In conclusion, we have generalized the treatments of Chazalviel[14] and Boguslawski[15] of impurity and electron-electron EY spin-flip scattering in *n*-doped zincblende semiconductors. Our treatment of spin relaxation takes into account the dependence of T_1 on doping density, spin-density imbalance, and temperature. We apply this theory to GaAs and analyze the main features of the dependence of T_1 on those system variables. This theory should mainly be considered as a starting point for more refined treatments of the Elliot-Yafet mechanism, as a complete understanding of recent experiments[3,4] based on the known spin-flip mechanisms in semiconductors has not emerged so far.

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