ABSTRACT

We present an investigation of electron-spin dynamics in p-doped bulk GaAs due to the electron-hole exchange interaction, aka the Bir-Aronov-Pikus mechanism. We discuss under which conditions a spin relaxation times for this mechanism is, in principle, accessible to experimental techniques, in particular to 2-photon photoemission, but also Faraday/Kerr effect measurements. We give numerical results for the spin relaxation time for a range of p-doping densities and temperatures. We then go beyond the relaxation time approximation and calculate numerically the spin-dependent electron dynamics by including the spin-flip electron-hole exchange scattering and spin-conserving carrier Coulomb scattering at the level of Boltzmann scattering integrals. We show that the electronic dynamics deviates from the simple spin-relaxation dynamics for electrons excited at high energies where the thermalization does not take place faster than the spin relaxation time. We also present a derivation of the influence of screening on the electron-hole exchange scattering and conclude that it can be neglected for the case of GaAs, but may become important for narrow-gap semiconductors.

1. INTRODUCTION

Much experimental and theoretical work in the area of spintronics has been focused on the control and manipulation of the electrons’ spin degree of freedom independently of charge. Oftentimes the ultimate goal is the realization of spintronics devices, in which electronic spins are the carriers of the information. A particularly appealing property of the manipulation of electronic spins in semiconductors that sets it apart from existing electronics in ferromagnetic materials is that the carrier densities can be varied over several orders of magnitude, and the existence of band gaps makes a controlled excitation and read-out with optical fields comparatively easy. The limiting factor for the usefulness of the information encoded in a spin polarization in a non-ferromagnetic semiconductor is the spin-polarization decay, which is caused by a variety of interaction mechanisms. A famous result on the physical limit of spin manipulation by relaxation phenomena is the measurement of a spin lifetime of about 1 nanosecond in n-doped GaAs at very low temperatures. However, for actual devices, one might be forced to use materials and excitation conditions that do not yield these extremely long spin relaxation-times for several reasons. First, one would likely prefer to work with undoped semiconductors, or even with dilute magnetic semiconductors, in which holes are generally the majority carriers. Second, carrier injection through metal-semiconductor interfaces or by optical fields may create nonequilibrium electronic distributions at energies far from the bottom of the bands. Third, a knowledge of the spin-dependent dynamics at moderate to high densities at room temperature is needed for most devices that interact with optical fields and may be used for switching of optical signals, such as amplifiers and lasers.

Much work has been done to characterize the spin-flip interaction mechanisms in semiconductors in terms of spin relaxation times. State-of-the-art ultrafast optical techniques such as time-resolved Faraday rotation and spin-resolved 2-photon photoemission make it possible to probe the spin and energy resolved carrier dynamics on short timescales. In this regime, one has to reexamine whether spin relaxation-times constitute a complete description of spin-dependent carrier dynamics, or whether one needs to resort to a microscopic description by including the relevant scattering processes that change the carrier momentum and their spin.

This paper is devoted to a theoretical study of the spin-dependent scattering processes due to the electron-hole exchange interaction in bulk GaAs. The spin-flip mechanism due to this interaction, which is the most important
one in semiconductors with high p-doping concentration, is the Bir, Aronov, and Pikus (BAP) mechanism. We first address the question about the conditions under which spin relaxation-times are appropriate to describe the spin-dependent dynamics of electrons in p-doped bulk GaAs. We therefore start our presentation in Section 2 with an analysis of how one can define a spin-relaxation time for the BAP process starting from the Boltzmann equation for electron-hole exchange scattering. We then show how this quantity is related to experimental results, and present numerical results for the BAP spin-relaxation time that are suitable for comparison with experiments over most of the experimentally interesting density and temperature range. In Section 3, we derive the dynamical Boltzmann equations for the electronic distributions including both exchange and direct scattering. This Boltzmann equation was already used in Section 2, but here we go back to the basic Hamiltonian that describes carriers in semiconductors interacting via the direct Coulomb and the electron-hole exchange interaction. Although these equations have been derived before, we include a discussion of screening effects in a fashion analogous with the usual random-phase approximation (RPA) treatment of screening and we pay special attention to the long-range and short-range contributions to the electron-hole exchange interaction. We then present numerical solutions of the Boltzmann equation including both the electron-hole exchange interaction and direct electron-hole Coulomb interaction for the case of the ultrafast optical excitation of a nonequilibrium spin-polarized distribution of electrons and hole. We compare the full energy-resolved spin polarization dynamics with the BAP spin-relaxation time.

2. SPIN RELAXATION-TIME

2.1 Derivation from Boltzmann equation

In this Section, we discuss a derivation for the spin relaxation time due to the electron-hole exchange interaction in p-doped semiconductors (GaAs), i.e., the BAP spin relaxation mechanism. We start from the general Boltzmann equation for the electronic distribution functions. We assume in the following spherically symmetric energy dispersions so that the distribution functions depend only on the carrier energy instead of the vector momentum. We therefore neglect in this article other spin-relaxing mechanisms, such as the Dyakonov-Perel mechanism, which depends on anisotropies of the electronic band structure. We also assume that anisotropic electronic distributions, which are created by optical excitations are washed out in a few hundred femtoseconds by carrier-carrier scattering, so that for the timescales of interest for the electron-hole exchange interaction, we always have spherically symmetric electronic distributions. With these assumptions we can write the Boltzmann equation for the spin-dependent electronic distributions $n_s(E)$ in the form

$$\frac{\partial}{\partial t} n_s(E,t) = \int \left\{ -w_{\text{xc}}(E,E') [1 - n_{-s}(E')] n_s(E) + w_{\text{xc}}(E',E)n_{-s}(E') [1 - n_s(E)] \right\} D(E')dE'$$

$$+ \int \left\{ -w_{\text{dir}}(E,E') [1 - n_{-s}(E')] n_s(E) + w_{\text{dir}}(E',E)n_{-s}(E') [1 - n_s(E)] \right\} D(E')dE'$$

(1)

Here, $s = \uparrow, \downarrow$ denotes the electron spin projection quantum number along the quantization axis, which is chosen to be the $z$ axis. The details of the electron-hole exchange and direct scattering, such as direct carrier Coulomb scattering and/or carrier-phonon scattering, are contained in the scattering kernels which will be discussed in the next section. The exact form of the density of states is not important in the following, but for definiteness we consider parabolic electron bands with effective mass $m_e$, so that $D(E) = (2m_e/\hbar^2)^{3/2}/(2\pi)^2 \times \sqrt{E}$. This equation is derived and solved numerically in the following Section 3. Instead of a full numerical solution, we analyze first how a simplified description in terms of a spin-relaxation time is possible.

For the derivation of the spin-relaxation time, we focus on a simple pump-probe scenario. We assume that such an experiment is performed with a weak picosecond or sub-picosecond pump pulse that excites electrons close to the bottom of the band. Then a detailed microscopic description of the coherent carrier-creation process is not important for the electron spin dynamics because the time scale for the spin-dependent dynamics is much longer, namely on the order of several hundred picoseconds. The equilibration of the photoexcited electrons occurs via the direct scattering processes, i.e., via Coulomb scattering with equilibrated holes, which are present in the system due to the p-doping, or via electron-phonon scattering. For the present discussion, it is not important which mechanism is dominant, only that the electrons are thermalized after shortly after the pump pulse. If the
pump is weak enough, the thermalized electrons will be non-degenerate and described by a Maxwell-Boltzmann distribution

\[ n_s(E,t) = n_s(t)f_\beta(E) \]  
\[ f_\beta(E) = N \left( \frac{2\pi\hbar^2}{m_e\beta} \right)^{3/2} \exp(-\beta E). \]

Here, \( \beta = \frac{1}{k_B T} \), \( k_B \) is Boltzmann’s constant, and \( N \) is the total density of electrons. With this normalization, \( n_s(E) \) is in the range of 0 to 1. For this low density of nondegenerate electrons, the effect of the direct scattering is to keep the energy dependence of the electronic distributions constant. This is already built into the electronic distribution function, Eq. (2), and its time development can therefore be calculated by inserting Eq. (2) in Eq. (1), and keeping only the exchange contribution, i.e., the first line, in Eq. (1). In addition, for low density distributions as assumed here, \( 1 - n_s(E,t) \approx 1 \). Integrating the resulting equation over \( \int_0^\infty D(E)dE \) and using the normalization \( \int_0^\infty dE D(E)f_\beta(E) = N \) one obtains from Eq. (1) a dynamical equation for the density of electrons with spin \( s \),

\[ \frac{\partial}{\partial t} n_s(t) = -\langle \gamma_{xc} \rangle \beta \left[ n_s(t) - n_{-s}(t) \right] \]

The energy-dependent out-scattering rate \( \gamma_{xc} \), defined by

\[ \gamma_{xc}(E) = \int_0^\infty w(E,E')D(E')dE' \]

enters Eq. (5) only in an energy-averaged sense. [See Appendix B for the numerical solution of Eq. (6)]. The average over the electronic Maxwell-Boltzmann distribution is defined by

\[ \langle \gamma_{xc} \rangle \beta = \frac{1}{N} \int_0^\infty \gamma_{xc}(E)f_\beta(E)D(E)dE. \]

From Eq. (5) and Eq. (2), the spin relaxation time, i.e., the decay time of the spin polarization, defined by

\[ P(E,t) = \frac{n_\uparrow(E,t) - n_\downarrow(E,t)}{n_\uparrow(E,t) + n_\downarrow(E,t)} \]

can be deduced. The energy resolved spin polarization is experimentally accessible, e.g., by spin resolved 2-photon photoemission techniques\(^\text{12}\) However, for low electronic excitation conditions, the energy dependence of the non-degenerate electronic distributions of the form (2) cancels in Eq. (8), so that the polarization \( P(E) \) is energy independent and equal to the total spin polarization \( P(E,t) = P(t) \equiv [N_\uparrow(t) - N_\downarrow(t)]/[N_\uparrow(t) + N_\downarrow(t)] \). From (5) it then follows that

\[ \frac{d}{dt} P(t) = -\frac{1}{\tau_{\text{spin}}} P(t) \]

where the spin relaxation-time is defined by

\[ \frac{1}{\tau_{\text{spin}}} = 2 \langle \gamma_{xc} \rangle \beta . \]

For a low density of electrons in an energy range, in which the direct scattering leads to an effective equilibration on timescales shorter than the spin relaxation-time, one therefore obtains an energy independent spin relaxation time. Only this quantity is experimentally accessible, even with experimental methods that possess an energy resolution that is better than the spread in energy of the Maxwell-Boltzmann distribution \( f(E) \), which is used to
calculate the average in Eq. (7). Moreover, it is impossible to measure an energy resolved spin-relaxation time, which is sometimes defined by

\[
\frac{1}{\tau_{\text{spin}}(E)} = 2\gamma^x_c(E)
\]  

(11)

Thus the only meaningful spin relaxation time is the one originally introduced by Bir, Aronov, and Pikus, but without the energy dependence, which has been discussed in the literature. For the theoretical description of experimental results that do not meet the conditions for the derivation of a single, energy-independent spin relaxation-time, one needs to solve the full Boltzmann equation numerically to obtain the energy or momentum-resolved electron spin dynamics. If the form of the electronic distribution functions changes appreciably over the time of the spin relaxation, one should not expect a single electron spin-relaxation time, but an energy-dependent one. However, from the energy-dependent dynamics one can determine the time evolution accessible by the experiment and try to fit the spin-polarization dynamics by an exponential decay, if possible. Another experimental consequence of this analysis is that a measurement of the electronic spin dynamics by different techniques, such as Faraday rotation, 2-photon photoemission or differential transmission, may yield different results, and one may extract different spin relaxation-times from these measurements, as has been shown for the case of hole spin-relaxation.

2.2 Numerical results for spin relaxation-times

As discussed above, a spin relaxation-time for the BAP process, Eq. (10), can be rigorously defined for experiments probing a low density of thermalized (non-degenerate) electrons. For this case, we numerically evaluate the expression (10) using also Eqs. (7) and (6); see Appendix B and the set of parameters shown in Table 1 in that Appendix. The results obtained in this Section can therefore be compared to the spin relaxation-times measured by optical pump-probe techniques, such as 2-photon photoemission or Faraday-rotation measurements, as long as the experiments only probes equilibrated electrons. In this case, as stressed above, the spin relaxation time is energy independent.

An important characteristic of a spin dependent scattering process is its temperature dependence because different mechanisms differ strongly in their respective temperature dependences and a measurement of this

![Figure 1. Temperature dependence of \( \tau_{\text{spin}} \) for different p-doping concentrations.](attachment:image.png)
quantity can sometimes be used to identify the dominant scattering process for the material and doping concentration under study. Fig. 1 shows the computed spin relaxation-time $\tau_{\text{spin}}$ from 25 K up to room temperature. It confirms the well-known general trend that the BAP mechanism becomes important for “high” temperatures, i.e., around room temperature, where the spin relaxation-time can reach values below 100 ps for typical doping densities of $N = 5 \times 10^{18} \text{cm}^{-3}$ and beyond.

Another important “fingerprint” of the BAP process is its dependence on the density of holes introduced by the p-doping. The evaluation of Eq. (7) allows one to obtain the BAP spin relaxation-time for arbitrary hole concentrations, as shown in Fig. 2. The doping-density dependence is obtained for the whole density range without having to patch together analytical results, which can be obtained for the limiting cases of low and high doping densities. Since both the analytical results are not valid around the densities, for which the holes become degenerate, a discontinuity results if one plots the standard analytical results in one figure over the whole doping density range. One can compare the results of Fig. 2 with recent evaluations of the contribution of the Dyakonov-Perel effect or with measurements for undoped GaAs, i.e., hole concentrations of well below $10^{17} \text{cm}^{-3}$. For these low densities, even around room temperature, the BAP process is not the dominant relaxation process. This result shows that there is no “rule of thumb” that (for intrinsic GaAs) the Dyakonov-Perel effect is dominant at low temperatures and the BAP becomes important for “higher” temperatures. The BAP vs. Dyakonov-Perel problem, including the effects of carrier heating, has been explored in more detail recently.

In Fig. 3 we examine the importance of different contributions to the spin-relaxation time due to the electron-hole exchange interaction. The long-range contribution to the electron-hole exchange interaction is clearly the dominant one, as can be seen from comparing the top and bottom solid lines in Fig. 3. If one leaves out the short-range contribution one gets a good approximation to the true spin-relaxation time especially for higher p-doping concentrations approaching or exceeding $N = 10^{20} \text{cm}^{-3}$. It is even a good approximation to only include the contribution of the heavy-hole to heavy-hole transitions to the electron spin relaxation-time. Since the results of Fig. 3 again cover the p-doping density range around hole degeneracy where the approximate results are not valid, using just the long-range contribution due to heavy-hole to heavy-hole transitions will give a much better estimate of the true spin relaxation-time than using any one of the approximate expressions.

Figure 2. Dependence of $\tau_{\text{spin}}$ on the p-doping concentration for different temperatures.
Figure 3. Dependence of $\tau_{\text{spin}}$ on the p-doping concentration for $T = 300$ K. (a) The bottom solid line is the result including short and long range contributions to the electron-hole exchange scattering and contributions from hh-hh, hh-lh, and lh-lh transitions. (b) The long-dashed line is the result including only the long range contribution and all hole bands. (c) The short-dashed is the same as (b), but includes only the hh-hh scattering transitions. (d) The top solid line includes only the short-range contribution to the electron-hole exchange interaction and all relevant transitions between hole bands.

3. MICROSCOPIC ELECTRON DYNAMICS DUE TO EXCHANGE SCATTERING

We now turn towards a derivation of the microscopic spin-dependent carrier dynamics that underlies the concept of the spin relaxation time derived in the previous section. The purpose of this derivation is to make the presentation self-contained and pay attention to the screening properties of the exchange interaction. In particular, all the parameters needed to describe the exchange interaction quantitatively are included in this section and the appendices, and the steps for a numerical solution of the Boltzmann equation are presented in some detail in Appendix B.

3.1 Hamiltonian and dynamical equations

Because we have the application to p-doped GaAs in mind, we treat the case of electronic dynamics in the presence of a high density of thermalized holes. We do not include electron-phonon interactions, and do not treat the hole dynamics and hole-hole Coulomb explicitly. Rather, we assume that the hole system is always in equilibrium with the lattice, i.e., we have equilibrium hole distributions at the lattice temperature. Furthermore, electron-electron scattering is neglected because electron-hole scattering is the dominant scattering mechanism due to the low electronic density. Under these assumptions, the starting point is the basic Hamiltonian

$$H = H_{\text{kin}} + H_{\text{Coul}} + H_{\text{exc}},$$

which includes the electron kinetic energy and the electron-hole Coulomb interaction as well as the electron-hole exchange interaction. The kinetic and the direct electron-hole Coulomb term have the familiar form

$$H_{\text{kin}} = \sum_{sk} \epsilon_{sk} c^\dagger_{sk} c_{sk}$$

and

$$H_{\text{Coul}} = \frac{1}{2} \sum_{kk'qjjs'j'} 2 \langle j', s' | V_{\text{Coul}}(q) | j, s \rangle c^\dagger_{j',kk'} c_{j,kk} c^\dagger_{j',s'k'k} c_{j,sk}.$$
In Eqs. (13) and (14), $c_s$, $c_j^\dagger$ and $c_j$, $c_j^\dagger$ are conduction and valence-band destruction and creation operators, respectively. Conduction-electron states are labeled by the electron spin projection quantum number $s = \pm 1/2$, whereas valence-band electrons are labeled by the projection quantum numbers $j = \pm 1/2, \pm 3/2$ corresponding to the total angular momentum $J = 3/2$. In this paper, we will keep the conduction-valence (cv) picture for carrier states and the index structure of direct and exchange terms in the Hamiltonian and throughout the calculations. Only in the final expressions for the scattering kernels a change to electron and hole distributions is made by replacing $n_{j,k} \longrightarrow 1 - n_{j,k}$ and $\varepsilon_{j,k} \longrightarrow -\varepsilon_{j,k}$. However, the linear and angular momentum labels, $j$ and $k$, as well as the interaction matrix elements from the conduction-valence band picture are retained. For a complete transformation to the electron-hole picture, one would have to change the angular and linear momenta to $-j$ and $-k$, define hole operators and calculate the interaction matrix elements using the time-reversed valence-band wave functions, which leads to a switch of the $j$ and $j'$ labels in the matrix elements. The matrix element for the direct Coulomb interaction is

$$\langle j', s'|V_{\text{Coul}}(\vec{q})|j, s \rangle \equiv \delta_{j',j}\delta_{s',s}v_q$$

(15)

with the Coulomb potential energy

$$v_q = \frac{1}{L^3} \frac{e^2}{\varepsilon_{\text{br}}}$$

(16)

in SI units, where $\varepsilon_{\text{br}}$ is the dimensionless background dielectric constant. The electron-hole exchange interaction is described by

$$H_{\text{exc}} = \sum_{k,k',q,j,j',s,s'} \langle j', s'|V_{\text{exc}}(\vec{q})|s,j \rangle c_j^\dagger c_{j',k+\vec{q}}^\dagger c_{s',k'} c_s c_{k,s'}$$

(17)

Note in particular that the Hamiltonian of the exchange interaction differs from its direct interaction counterpart in the last two creation operators and the order of the band indices in the matrix element. The matrix element of the exchange interaction consists of a “long range” and a “short range” part

$$\langle j', s'|V_{\text{exc}}(\vec{q})|s,j \rangle = \langle j', s'|V_{\text{exc}}^{\text{sr}}(\vec{q})|s,j \rangle + \langle j', s'|V_{\text{exc}}^{\text{lr}}(\vec{q})|s,j \rangle$$

(18)

where the long range part is given by

$$\langle j', s'|V_{\text{exc}}^{\text{lr}}(\vec{q})|s,j \rangle = v_q \frac{\hbar^2}{2m_0e^2} (\vec{q} \cdot \vec{p}_{s,j})(\vec{q} \cdot \vec{p}_{s',j'}) = v_q (\vec{q} \cdot \vec{x}_{s,j})(\vec{q} \cdot \vec{x}_{s',j'}$$

(19)

In Eq. (19), $\vec{p}_{s,j}$ is the momentum and $\vec{x}_{s,j}$ the dipole operator matrix element between the conduction electron state $s$, $k = 0$ and the valence-band state $j$, $k = 0$. The energies $\epsilon_0$ are the electronic energies at the $\Gamma$ point, $k = 0$, where the conduction and valence electron bands are respectively degenerate. Using the standard basis for the coordinate representation of conduction ($S = 1/2$) and valence ($J = 3/2$) electrons, the short range part can be brought into the form

$$\langle j', s'|V_{\text{exc}}^{\text{sr}}(q)|s,j \rangle = \frac{3}{2} \Delta_{\text{exc}}^{\text{sr}} \langle j', s' \rangle \left[ \frac{3}{4} + \langle \vec{J} \cdot \vec{S} \rangle \right] |j, s \rangle$$

(20)

In Eq. (20), $\Delta_{\text{exc}}^{\text{sr}}$ denotes the excitonic exchange splitting, and $\vec{J}$ and $\vec{S}$ are the hole and electron angular momentum operators, respectively. Explicit matrix expression for Eqs. (19) and (20) are compiled in Appendix A.

In order to describe the scattering dynamics on a microscopic footing, exchange and direct Coulomb interaction need to be included in the equations of motion,

$$\frac{\partial}{\partial t} n_s(k) = \frac{\partial}{\partial t} n_s(k)\bigg|_{\text{Coul}} + \frac{\partial}{\partial t} n_s(k)\bigg|_{\text{exc}}$$

(21)

We therefore present a derivation of the exchange scattering in a fashion analogous to the derivation of the Boltzmann equation for direct electron-hole Coulomb scattering in the random-phase approximation (RPA).
Employing a Green’s function approach\cite{21,22} and using a quasiparticle ansatz one obtains in the electron-hole picture

\[
\frac{\partial}{\partial t} n_s(k) \bigg|_{\text{exc}} = \sum_{k'} \left[ -W_{k,k'}^{\text{exc}} (1 - n_{-s,k'}) n_{s,k} + W_{k,k'}^{\text{exc}} (1 - n_{s,k}) \right]
\]

\[
\frac{\partial}{\partial t} n_s(k) \bigg|_{\text{dir}} = \sum_{k'} \left[ -W_{k,k'}^{\text{dir}} (1 - n_{s,k'}) n_{s,k} + W_{k,k'}^{\text{dir}} (1 - n_{s,k}) \right]
\]

where

\[
W_{k,k'}^{\text{exc}} = \frac{2\pi}{\hbar} \sum_q \sum_{j,j'} \left| \langle j,-s|V_{\text{exc}}(q,E_G + \epsilon_{-s,k'} + \epsilon_{j',k'+q})|s j' \rangle \right|^2 n_{j,k+q} (1 - n_{j',k'+q})
\]

\[
W_{k,k'}^{\text{dir}} = \frac{2\pi}{\hbar} \sum_q \sum_{j,j'} \left| \langle j,-s|V_{\text{exc}}(q,E_s - \epsilon_{-s,k})|s j' \rangle \right|^2 n_{j,k+q} (1 - n_{j',k'+q}) \delta(\epsilon_{j,k+q} + \epsilon_{s,k} - \epsilon_{s,k'} - \epsilon_{j',k'+q})
\]

Note that, instead of writing separate expressions for the in and out-scattering kernels, we have displayed the symmetry of the in and out-scattering processes by introducing only one kernel \( W_{k,k'} \). This symmetric form with one kernel is especially convenient for a numerical solution because it avoids the separate calculation of two kernels, which may introduce numerical problems if one violates the balance between in and out scattering, which is responsible for the conservation of the carrier density.

The occurrence of an interband energy in the dynamical exchange interaction is analogous to the dynamically screened of the direct Coulomb interaction \( V_{\text{exc}} \), and can be derived in exactly the same fashion\cite{21,22}. The result for the dynamical exchange interaction is

\[
\langle j s'|V_{\text{exc}}(q,\omega)|s j' \rangle = \langle j s'|V_{\text{exc}}(q)|s j' \rangle + \sum_{s1j1} \langle j s1|V_{\text{exc}}(q)|s j1 \rangle \Pi_{s1j1}(q,\omega) \langle j1 s'|V_{\text{exc}}(q,\omega)|s1 j' \rangle
\]

with the retarded RPA electron-hole polarization function

\[
\Pi_{s j}(q,\omega) = -i\hbar \sum_{k} G_{s,k+q}(\omega) G_{j,k}(\omega) = \sum_{k} \frac{n_{s,k+q} + n_{j,k}}{\hbar \omega - (\epsilon_{s,k+q} + \epsilon_{j,k} + E_G) + i\hbar\gamma}
\]

Here, a zero-density contribution that occurs on transforming into the electron-hole picture has been subtracted. In Eq. (27) an interband energy enters the denominator. On evaluating Eq. (27) for the case of GaAs, the product of the exchange interaction and the polarization function in Eq. (26) turns out to be so small that the effect of the screening term on the exchange interaction is negligible for the numerical results in this paper. Therefore only the unscreened exchange interaction will be used in the following, i.e., \( V_{\text{exc}}(q,\omega) \rightarrow V_{\text{exc}}(q) \). Even though the effect of screening on the exchange interaction is comparatively weak for the case of GaAs considered here, it might become more important in narrow band-gap systems. In the following numerical results we will also use a statically screened Coulomb interaction \( V_{\text{scr}}(q,\omega) \rightarrow \varepsilon^{-1}(q)v_q \). A discussion of the static approximation is given by Collet.\cite{20} Although dynamical screening effects can become important for quantitative results on direct Coulomb scattering, for the purposes of this paper it is mainly important that the direct electron-hole Coulomb scattering occurs much faster than the electron-hole exchange scattering.

The connection to Boltzmann equation\cite{1} used previously is made in the following way. In the general Boltzmann equation\cite{22}, which contains distribution functions and energies that may be anisotropic, one assumes spherical symmetry and employs an effective mass description for electrons and holes. Then the carrier distributions depend on the energy, i.e.,

\[
n_{\nu,k} \rightarrow n_{\nu}(E) \text{ where } E = \frac{\hbar^2}{2m_\ell} k^2
\]
for \( \nu = \) electrons (e), heavy holes (HH), and light holes (LH). As mentioned above we assume further a high density of equilibrated, unpolarized holes. Then the distribution functions can be expressed as Fermi functions
\[
n_{j,k} = f(\epsilon_{k}^{\text{HH}} - \mu) \quad \text{for} \quad j = \pm 3/2 \quad \text{and} \quad n_{j,k} = f(\epsilon_{k}^{\text{LH}} - \mu) \quad \text{for} \quad j = \pm 1/2
\]
and the \( \vec{q} \) summation can be converted into an integral that can be evaluated mostly analytically as shown in Appendix B. The chemical potential \( \mu \) is fixed by the temperature and density of the equilibrated holes. Due to the assumption of unpolarized holes and the symmetry properties of the interaction matrix element, which are discussed in the Appendix A, the kernel is a function \( W(E, E') \) independent of the electron spin. Writing the wave-vector summations in Eq. (22) as energy integrals including the density of states, one finally obtains Eq. (1), on which the analysis of the BAP spin-relaxation time is based.

### 3.2 Numerical Results for Spin-Polarization Dynamics

In this Section, the dynamics of spin polarized electrons under the influence of exchange (spin-flip) and direct (non spin-flip) electron-hole scattering processes are numerically evaluated using Eqs. (22), (24) and the static (non-screened) exchange interaction matrix element (18). Here, we analyze the energy and spin-resolved electron dynamics for excitation conditions that would be used in experiments designed to measure the energy-resolved polarization dynamics. One of our goals here is to show, which energy-resolved quantities can be extracted from typical experiments. For experiments with sub-picosecond excitation pulses, the detailed microscopic description of the coherent carrier-creation process is again neglected because it occurs on a shorter time scale than for the the spin-dependent dynamics. As a model for such an experiment that creates a nonequilibrium electron distribution by a short laser pulse, we consider as an initial condition a Lorentzian electron distribution peaked around an electronic energy \( \epsilon_{k_{L}} \), or modulus of the electron wave vector \( k_{L} \). In an experiment, the wave vector \( k_{L} \) and the energy would be determined by the laser wavelength and the band structure. We restrict ourselves to excited electron densities that are small enough so that the neglect of the electron-electron interactions is justified. The initial low-density (nondegenerate) electron distribution is chosen with initial spin polarization of 0.5 and with the same temperature as the holes.

![Figure 4](image_url)

Figure 4 shows the dynamical results for the spin polarization \( P(E, t) \) defined in Eq. (8). The curves were calculated numerically as described in Appendix B with both the short-range and the long-range contribution to the electron-hole exchange interaction matrix element taken into account. The parameters are again those given in Table 1. We assume unpolarized holes with density \( N_{A} = 5 \times 10^{18} \text{cm}^{-3} \) and temperature \( T_{h} = 75 \text{K} \). The spin polarization is monitored at the energy \( E \), at which the carriers are initially created in the form of a nonequilibrium electron distribution peaked at \( E \). At a carrier energy \( E = 5 \text{meV} \), i.e., close to the bottom of the electron band, the decay of the spin polarization after the excitation of the electrons is almost exponential. For higher peak energies of the excited electrons, already starting below an excess energy of 70 meV, the dynamics is not described by an exponential drop at all. Instead, the \( P(E, t) \) rises after an initial steep drop. Only for
longer times, the spin polarization dynamics resembles an exponential decay with time constants of about 305 ps, which is identical to the time constant of the decay of the total spin polarization and almost identical to the BAP spin relaxation-time as shown in Figs. 1 and 2. The reason for this behavior is the interplay between the spin-flip exchange scattering and the direct scattering. If the polarization is determined at an energy where electrons undergo in and out-scattering processes, a quasi-equilibrium is reached due to direct electron-hole scattering events on a timescale faster than the spin-flip scattering, so that the spin-flip scattering occurs for effectively equilibrated electrons. For these, the concept of a spin relaxation time exists, as shown in Section 2. For electrons with higher energies out-scattering processes, dominate for several ten picoseconds up to more than 100 ps. On this timescale, the initial peaked electronic distribution has been already been washed out, but no quasi-equilibrium has been established for electrons at these energies. The polarization dynamics is therefore due to a combination of spin-conserving and spin-flip scattering processes, and the dynamics depend on the form of the actual nonequilibrium electron distribution. This non-exponential dynamics occurs until a quasi-equilibrium is established due to direct scattering processes. Then the spin polarization decay becomes exponential. Thus experiments exciting and probing the electronic spin polarization at higher energies will not yield energy-resolved information about the the energy resolved spin-relaxation time due to the exchange interaction as defined in Eq. (11).

Figure 5 shows the same scenario for a hole temperature of $T = 300$ K. In this case, again, the spin polarization decays exponentially only for electrons close to the bottom of the band, with a spin-relaxation time of about 70 ps, which is again almost identical to the spin-relaxation time of the total polarization and the BAP spin relaxation time. At higher energies, the initial polarization dynamics is non-exponential. However, the onset of the nonexponential dynamics occurs only for electrons with much higher excess energy, here for energies well above 200 meV because at higher energies the equilibrated “tail” of the distributions stretches out to higher energies. This indicates that the nonexponential dynamics is not as important and will not be as easily observed as it is the case for $T = 75$ K. The dynamics generally occur on a shorter time scale than in the $T = 75$ K case (note the different y axes in the two figures), but the “deviation” from the exponential decay is qualitatively different. Now the nonequilibrium dynamics leads to a slower initial decay of the polarization, before the dynamics becomes exponential when a quasi-equilibrium is established by the direct scattering processes. Since the initial dynamics is so much different for the different temperatures there is no simple rule of thumb which qualitative behavior of the spin dynamics occurs for nonequilibrium conditions. Certainly, the spin relaxation-time cannot serve even as guideline in this case, and one has to model the excitation conditions and energy-dependent measurement accurately to obtain reliable results that may be compared to a given experiment.
4. CONCLUSION

We have investigated how spin relaxation-times can be used to describe the spin polarization dynamics of electrons due to the electron-hole exchange interaction, i.e., the Bir-Aronov-Pikus mechanism. It is found that there is only one \textit{ensemble averaged} spin relaxation time that can be accessed by optical experiments, such as Faraday rotation, differential transmission, or 2-photon photoemission, even if these experiments have an energy resolution that is smaller than the spread of excited electronic energies. In order to derive a meaningful spin-relaxation time, one has to assume that a nondegenerate electronic distribution is kept a quasi-equilibrium by the direct scattering processes while the exchange scattering effects the spin polarization. Thus the energy-dependence of the spin relaxation dynamics is washed out by the thermal average. If one tries to get around measuring the ensemble average and intends to probe the electronic spin polarization at energies, at which there is initially no quasi-equilibrium established, one still does not get a unique energy dependent spin relaxation-time, but rather a complicated spin polarization dynamics that is determined by the interplay of direct and spin-flip scattering processes. It is therefore not possible to extract an energy-dependent spin relaxation-time because the of the direct scattering mechanisms, although an energy-dependent energy relaxation time can be defined, as is usually done for electronic lifetimes.

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APPENDIX A. MATRIX ELEMENTS

The exchange interaction between electrons and holes consists of a short-range and long-range contribution. In the following, electron-hole states are labeled by conduction-valence band angular momentum labels \((j\sigma) = (-\frac{3}{2} \uparrow), (-\frac{3}{2} \downarrow), (-\frac{1}{2} \uparrow), (-\frac{1}{2} \downarrow), (+\frac{1}{2} \uparrow), (+\frac{1}{2} \downarrow), (+\frac{3}{2} \uparrow), (+\frac{3}{2} \downarrow)\)

The \textit{long-range part} to the exchange interaction matrix results from the evaluation of Eq. (19) in the standard angular-momentum basis and reads:

\[
V_{\text{exc}}^{lr}(\vec{q}) = v_q |r_{ev}|^2 \begin{pmatrix}
0 & 0 & 0 & \frac{1}{\sqrt{\eta}} q^2 y_1 & 0 & \frac{1}{\sqrt{\eta}} q + q_z & 0 & -\frac{1}{\sqrt{\eta}} q^2 z^1 \\
0 & \frac{1}{2} q^2 y_1 & 0 & \frac{1}{\sqrt{\eta}} q z + q^2 y_1 & 0 & -\frac{1}{2} q^2 y_1 & 0 & 0 \\
0 & 0 & \frac{1}{\sqrt{\eta}} q z + q^2 y_1 & \frac{1}{\sqrt{\eta}} q z & 0 & -\frac{1}{\sqrt{\eta}} q z & 0 & 0 \\
\frac{1}{\sqrt{\eta}} q^2 y_1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
-\frac{1}{\sqrt{\eta}} q^2 z^1 & 0 & \frac{1}{\sqrt{\eta}} q^2 z^1 & 0 & 0 & 0 & 0 & 0 \\
\frac{1}{\sqrt{\eta}} q^2 z & -\frac{1}{2} q^2 y_1 & -\frac{1}{\sqrt{\eta}} q^2 y_1 & 0 & 0 & 0 & 0 & 0 \\
\frac{1}{\sqrt{\eta}} q z & 0 & -\frac{1}{\sqrt{\eta}} q z & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

(29)

Here, \(q^2 = q_x^2 + q_y^2\) is modulus of the transverse component of \(\vec{q}\), and \(q_z\) is the \(z\) component. Then

\[
q_{\parallel} = q_x \pm iq_y = q_{\perp} e^{\pm i\varphi}
\]

(30)

where \(\varphi\) is the azimuthal angle of \(\vec{q}\).

Pikus and Bi\textsuperscript{23} also show how the \textit{short-range contribution} arises directly from the Coulomb interaction. Here, one evaluates the following exchange Coulomb matrix elements

\[
\langle j' k'_1, s' k'_2 | v | s k_3, j k_4 \rangle = \sum_{\sigma, \sigma'} \int \varphi_{j' k'_1} (\vec{x} \sigma)^* \varphi_{s' k'_2} (\vec{x}' \sigma')^* v(\vec{x} - \vec{x}') \varphi_{s k_3} (\vec{x} \sigma) \varphi_{j k_4} (\vec{x}' \sigma') d^3 x d^3 x'
\]

(31)

The carrier states are of the usual form

\[
\varphi_{\alpha k} (\vec{x}, \sigma) = \frac{1}{\sqrt{L^3}} \exp(-ik \cdot \vec{x}) u_{\alpha} (\vec{x} \sigma),
\]

(32)
where \( u_\alpha(\vec{x}\sigma) \) are the Bloch functions at \( k = 0 \). For electrons, \( \alpha = s \), and for holes, \( \alpha = j \), so that the angular momentum dependence of the \( u \) is determined by their angular momentum quantum number.

The contribution due of the exponential factors can be treated as in the spin-diagonal Coulomb matrix element by dividing the integral over the crystal into integrals over unit cells and pulling the exponential factors out of the unit cell integrals. The remaining integrals over the unit cell are evaluated by inserting the explicit expressions for the \( u \) whose angular momentum dependence is given by their total angular momentum projection quantum number \( j \), i.e., by linear combinations of spherical harmonics. Expanding the interaction \( v(\vec{x} - \vec{x}') = \sum_l V_l(r, r') \sum_{m=-l}^l Y_m^l(\vec{x}_l V_m^l(\vec{x}_l) \ldots \), the angular momentum integrals can be calculated and the remaining integrals over \( r \) and \( r' \) can be absorbed into a quantity that is accessible experimentally. One finds in the same matrix representation as Eq. (29)

\[
V_{ex}^{sr} = \frac{1}{L^3} \begin{pmatrix}
0 & 0 & 0 & \frac{1}{2} \sqrt{3} & 0 & 0 & 0 \\
0 & \frac{1}{2} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 1 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 1 & 0 & 0 & \frac{\sqrt{3}}{2} \\
0 & 0 & 1 & 0 & 0 & \frac{1}{2} & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{3}{2} & 0 \\
0 & 0 & 0 & 0 & \frac{1}{2} \sqrt{3} & 0 & 0
\end{pmatrix} \frac{3}{2} V_{ac} \Delta_{SR} \quad (33)
\]

where

\[
\Delta_{SR} = \sum_{\sigma, \sigma'} \int_{\text{unit cell}} u_{\alpha}(\vec{x}\sigma)^* u_{\beta}(\vec{x}'\sigma')^* [v(\vec{x} - \vec{x}')]_{\text{periodic}} u_{\alpha}(\vec{x}\sigma) u_{\beta}(\vec{x}'\sigma') d^3 x d^3 x' \quad (34)
\]

is the exchange splitting energy, \( V_{ac} \) is the volume of the unit cell and \( L^3 \) the crystal volume. The Coulomb potential \( v \) appearing in Eq. (34) is lattice periodic because it results from a summation over reciprocal lattice vectors. The energy \( \Delta_{SR} \) is related to the 1s-exciton splitting (into one triplet and one quintuplet) \( \Delta_{SR}^{\text{exciton}} \) due to the exchange interaction by

\[
V_{ac} \Delta_{SR} = \frac{1}{2} \pi a_B^3 \Delta_{SR}^{\text{exciton}},
\]

where \( a_B \) is the excitonic Bohr radius.

Using the matrix expressions (29) and (33) in (22) one calculates the squared matrix elements for electronic spin-flip scattering due to transitions between the different hole bands. For instance, for heavy-hole to heavy-hole scattering one has

\[
|\langle 3/2, \downarrow | V_{ac}(\vec{q}) | \uparrow, 3/2 \rangle|^2 = |v_q |r_{cv}|^2 \left(1 - \frac{1}{2} q_z^2 \right)^2 = \frac{(e^2}{\varepsilon_0 q^2}) |r_{cv}|^2 q_z^2 \left(1 - \frac{1}{2} q_z^2 \right)^2 = \alpha^2 (1 - z^2)^2 \quad (36)
\]

where \( \alpha = \frac{\pi^2}{\varepsilon_0} |r_{cv}|^2 \), and we have defined \( z = q_z/q \) so that \( q_z^2 = q^2 - q_x^2 \equiv q^2(1 - z^2) \). In a similar fashion one obtains for heavy-hole to light-hole scattering (two contributions in the scattering kernel)

\[
\frac{1}{3} \left( \frac{\alpha}{2} (1 - z^2) + \frac{3}{2} \Delta_{SR} \right) ^2 \quad (37)
\]

and for light-hole to light-hole scattering

\[
\left( \frac{2}{3} z^2 \alpha + \Delta_{SR} \right)^2 + (1 - z^2) \frac{\alpha^2}{36} \quad (38)
\]

The full squared interaction matrix element in Eq. (22) is a sum of Eqs. (36)–(38), but the heavy-hole to heavy-hole contribution (36) is the dominant one for the conditions investigated in this paper. Note that there is no dependence of the in-plane angle \( \varphi \) (between \( q_x \) and \( q_y \)) for matrix elements that also have a short-range contribution. Therefore there is no explicit \( \varphi \) dependence in the terms that have combined short-range and long-range contributions.
APPENDIX B. NUMERICAL EVALUATION OF BAP SPIN RELAXATION-TIME

To obtain the BAP spin relaxation-time one evaluates the expression, with momentum indices chosen for numerical convenience and as a function of $k$ instead of $E$, but cf. Eq. (28),

$$\gamma_{xc}(k) = \frac{2\pi}{\hbar} \sum_{\tilde{q}, \tilde{p}} \sum_{j,j'} |\langle j', -s | V_{xc}(\tilde{q}) | s, j \rangle|^2 \left( 1 - n_{j,\tilde{p}+\tilde{q}} \right) n_{j',\tilde{k}+\tilde{q}} \delta (\epsilon^e_k + \epsilon^j_{\tilde{k}+\tilde{q}} - \epsilon^e_{\tilde{p}} - \epsilon^{j'}_{\tilde{p}+\tilde{q}}) \right).$$

First, the momentum sums over $\tilde{q}$ and $\tilde{p}$ are replaced by two three-dimensional integrals using spherical coordinates with the $p$ integration as the inner integral. One measures $p$ against $q$, and $q$ against $k$, denoting $z' = \cos \angle(\tilde{p}, \tilde{q})$ and $z = \cos \angle(\tilde{q}, \tilde{k})$ together with their respective azimuthal angles $\varphi'$ and $\varphi$. The $\varphi'$ and $\varphi$ integrations yield only $2\pi$ each because there is no explicit dependence on these angles in the momentum labels $\tilde{k} + \tilde{q}$ or $\tilde{k}' - \tilde{q}$, and the exchange matrix element does not depend on $\varphi'$, only on $q$ and $z'$. Then one analytically evaluates the integral over $z'$ making use of the properties of $\delta$ distribution. For these manipulations, it is convenient to introduce a unit of length, denoted by $\ell$, and make the integrations over $q$ and $k'$ dimensionless by defining $q = q\ell$ and $k' = k\ell$. It is also convenient to introduce the energy

$$E_{kin} = \frac{\hbar^2}{2m_0\ell^2}$$

(40)

to make the kinetic energies dimensionless, $\epsilon^j_{p,e}/E_{kin} = (p^2/j_{p,e})$. The masses are expressed in terms of the vacuum electron mass $m_0$. The result is (expressed in our dimensionless units and leaving the tildes off)

$$\gamma_{xc}(k) = \frac{1}{32\pi^3} \frac{1}{h\beta E_{kin}^2} \int_0^\infty dq \int_1^1 dz \sum_{j,j'} f \left( \epsilon^j_{\sqrt{k^2 + q^2 + 2kqz}} - \mu \right) |\langle j', \downarrow | V_{xc}(z) | \uparrow, j \rangle|^2 m_e m_{j'} \log \left( \frac{1 + \epsilon_{\max}^{j'}}{1 - \epsilon_{\min}^{j'}} \right)$$

(41)

In this equation, the squared interaction matrix elements Eqs. (36)–(38) are used, and we have used the following definitions. First, the Fermi-Dirac function $f(\xi) = [1 + \exp(\beta\xi)]$ and the hole chemical potential $\mu$, further

$$\zeta_{\min,\max} = \beta \left( \frac{1}{m_{j'}} p_{\min,\max}^{j'} - \mu_{j'} \right)$$

(42)

where $\mu_{j'}$ is the reduced mass of an electron and a hole in band $j'$. Further,

$$p_{\min,\max} = \sqrt{\mu \left[ \frac{1}{m_{j'}} \left( k^2 + q^2 + 2kqz \right) + \frac{1}{m_e} k^2 - \frac{M_{j'}}{m_e} q^2 \right] - \frac{\mu_{j'}}{m_e} q}$$

(43)

with $M_{j'} = m_e + m_{j'}$.

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