Spin relaxation of hot electrons in the $\Gamma$-valley of zinc-blende semiconductors

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We present a technique to calculate the spin relaxation of hot electrons during their energy thermalization in the $\Gamma$-valley of zinc-blende semiconductors. The results of this model match recent experimental data and they can be used to check the applicability of spin related boundary conditions across a semiconductor/ferromagnet junction in reverse bias conditions (spin injection).

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Spin relaxation has an important role in semiconductor spintronics devices [1]. Recent measurements by Crooker et al. have shown that the spin polarization across a reverse bias GaAs/Fe junction drops appreciably already in moderate bias-voltage levels [2]. Saikin et al. have studied the spin relaxation of injected hot electrons for relatively large bias conditions and across a wide GaAs depletion region [3]. The authors have included zone edge phonon interactions in a Monte Carlo modeling and simulated the spatial spin distribution of injected hot electrons in the $\Gamma$, $X$ and $L$ valleys of GaAs. These effects were then verified by the electroluminescence of Fe-based spin light emitting diodes [4]. Ivchenko et al. have studied the polarization relaxation of photo-excited hot excitons during a cascade of longitudinal optical (LO) phonon emissions [5]. They have used a method of invariants to determine the general form of the density matrix. None of these models nor the conventional spin relaxation description of quasi-thermal electrons [6, 7] is suitable for studying the spin relaxation in moderate bias conditions [2]. In this paper, we fill this gap and describe a transparent cascade spin relaxation process during the ultrafast energy thermalization in the $\Gamma$-valley of zinc-blende semiconductors. The energy thermalization is governed by emission of long wavelength LO-phonons [9]. The results of this model match the experimental data and they can be used to evaluate the applicability of the boundary conditions in modeling spintronics devices.

The Dyakonov-Perel mechanism [6] is by large the dominant spin relaxation process during thermalization of hot electrons. However, the assumptions that are used in calculating the spin relaxation time of quasi-thermal electrons [7] are not valid for hot electrons. The broken assumptions are that (I) the density matrix of hot electrons is far from equilibrium and often is anisotropic, (II) the scattering is not elastic, and (III) the spin precession of hot electrons due to the intrinsic magnetic field is non negligible even within the momentum relaxation time (along certain high symmetry directions).

The spin behavior of hot electrons should be studied after each momentum scattering rather than in the asymptotic limit (after many scattering events). The reason is that hot electrons reach the bottom of the conduction band after a few LO-phonon emissions and altogether in about a picosecond. We begin by writing the intrinsic spin dynamics in the $\Gamma$-valley of a zinc-blende semiconductor [6],

$$\frac{dS}{dt} = \Omega \times S,$$  \hspace{1cm} (1) $$\Omega_j(k) = \alpha \hbar^2 (2m_e^3 E_g)^{-1/2} k_j (k_i^2 - k_m^2).$$ \hspace{1cm} (2)

The intrinsic Larmor frequency vector, $\Omega$, has a cubic dependence in the wavevector components $\{x, y, z\}$. The $\{j, \ell, m\}$ subscripts denote any cyclic permutation of $\{x, y, z\}$. $E_g$ is the energy-gap, $m_e$ is the electron’s effective mass, and $\alpha$ is a dimensionless parameter that relates to the strength of the spin-orbit coupling in the semiconductor.

We assume that the initial energy of a hot electron is well above the $\Gamma$ point but still below the energy of the other valleys in the conduction band. In this case the energy thermalization is governed by emission of long wavelength LO-phonons via the Fröhlich interaction [9]. After each of the ultra-fast momentum scattering events the direction and magnitude of $\Omega$ are randomized. The probability distribution that an electron with a wavevector $k$ will be scattered into wavevector $k'$ is,

$$I(k \rightarrow k') = \frac{B(k) \delta(\varepsilon(k') + \varepsilon_{LO} - \varepsilon(k))}{k^2 + k'^2 - 2k'k \cos(\theta_{k', k})}.$$ \hspace{1cm} (3)

The denominator describes the phonon wavevector dependence of the Fröhlich interaction, $|\langle k'|H_F|k\rangle|^2 \propto |k' - k|^2$. We have neglected the weak dispersion relation between the energy and wavevector in the LO-phonon branch, $\varepsilon_{LO}(k') \approx \varepsilon_{LO}(k)$.

The momentum scattering events are represented by a homogeneous Poisson process where the spin distribution of a hot electron immediately before its $n^{th}$ scattering event is denoted by,

$$S_{n-1}(k) = \frac{1}{\tau_{LO}(k)} \int_0^\infty dt e^{-t/\tau_{LO}(k)} S_{n-1}(k, t),$$ \hspace{1cm} (5)
where \( S_{n-1}(k, t) \) is the solution of Eq. (11) with an initial condition \( S_{n-1}(k, 0) \) which denotes the spin distribution of an injected electron immediately after its \((n-1)^{th}\) scattering event. Using the probability distribution in Eq. (3), the spin distribution immediately after the \(n^{th}\) scattering event is denoted by,

\[
S_n(k', 0) = \frac{1}{(2\pi)^3} \int d^3k \tilde{S}_{n-1}(k) f(k \rightarrow k'). \tag{6}
\]

This cascade process is complete by defining the initial spin distribution of the hot electrons. Due to the importance of spin relaxation in spintronics devices, we consider spin injection from a ferromagnetic contact into a semiconductor. Thus, the initial spin distribution is governed by the difference of the spin dependent transmission coefficients,

\[
S_0(k, t = 0) \propto |t_\uparrow(k, V)|^2 - |t_\downarrow(k, V)|^2, \tag{7}
\]

where \( V \) denotes the reverse voltage bias across the semiconductor/ferromagnet junction. We use a simple effective mass model (\( \varepsilon(k) = \hbar^2 k^2/2m_sc \)) to calculate the spin dependent transmission coefficients due to tunneling across a thin triangular-like Schottky barrier [10]. The results of the following discussion are not qualitatively affected by the choice of initial distribution.

The temperature dependence of this cascade process is manifested only by a slight modification of the intrinsic Larmor frequency vector (via a change in band-gap energy). However, one should recall that LO-phonon absorptions may also occur during the thermalization. The probability of an absorption process is smaller than the probability of an emission process by \( \exp\{-\varepsilon_{LO}/k_BT\}. \) Thus, in typical room temperature III-V semiconductors the absorption processes may somewhat enhance the spin relaxation of hot-electrons. In this paper, we ignore absorption processes by limiting our discussion to low temperatures. One can construct, however, a more general probabilistic model that incorporates both emission and absorption processes in the iterative procedure.

![FIG. 1: (a) Momentum scattering time in GaAs versus energy (in \( \varepsilon_{LO}=36 \text{ meV units} \)). (b) A typical normalized initial spin distribution of injected electrons from the Fermi level of Fe into GaAs versus the polar angle (with respect to the interface normal). The injected spin direction is aligned with the spin direction of majority electrons in Fe.](image1)

![FIG. 2: (color online) (a) Spin distribution over electrons’ momentum polar and azimuth angles after the 1st, 3rd and 5th successive LO-phonon emissions. The energy of the injected electrons is 0.2 eV above the conduction band \( \Gamma \) point and their spin distribution is normalized and points in the +z direction (see Fig. 1b). \( S_z \) keeps its dominant role throughout the energy thermalization process whereas \( S_y \) (not shown) and \( S_x \) are of much lower amplitude. After thermalization the electrons have lose \( \sim 50\% \) of the net injected \( S_z \) value (see text).](image2)
semiconductor/ferromagnet interface plane. The injected spin direction is aligned with the spin direction of majority electrons in Fe and we set it as the $S_z$ component \[1\]. Fig. 2 shows the evolution of the spin distribution after the first, third and fifth successive LO-phonon emissions of hot electrons whose initial energy is 0.2 eV above the semiconductor conduction band minima. Assuming that these electrons are injected from the iron’s Fermi level, then at 4K this scenario describes a GaAs/Fe junction at $-0.2$ V reverse bias-voltage (or $-0.3$ V at 300K). It takes five successive LO-phonon emissions for electrons to reach the vicinity of the $\Gamma$ point. Fig. 2 shows that after the first LO-phonon emission, the main contribution to $S_z$ is still from electrons whose motion is along the interface normal. This can be understood due to the forward scattering nature of the Fröhlich interaction (see the denominator of Eq. \(3\)). Thus, the initial distribution is not totally randomized in wavevector space after the first LO-phonon emission. This is not the case after five LO-phonon emissions where we see that the contribution to the net spin is nearly isotropic in the electron momentum. At these injection levels the spin information is largely kept when the electrons reach the bottom of the conduction band, and $S_z$ is much larger than the $S_x$ (not shown) and $S_y$ (shown) components.

The important figure of merit is to determine the net spin of the injected electrons after their ultra-fast energy relaxation. Using the effective mass approximation, the net spin after $n$ LO-phonon emissions is given by
$$\frac{k_n^2 \frac{d}{dk_n} \int d\phi \int d\theta \sin \theta \sin S_{N,z}(k_N,0)}{N \varepsilon_{LO}}$$ where $i$ enumerates the spin components. This writing assumes that initially the electrons’ energy is concentrated in a thin energy shell between $\varepsilon_0$ and $\varepsilon_0 + \Delta \varepsilon_0$. Due to the energy conservation of the Fröhlich interaction (Eq. \(3\)), the ratio between the $k_n^2 \frac{d}{dk_n}$ and $k_0^2 \frac{d}{dk_0}$ prefactors simply yields $k_n/k_0$. Thus, the fraction of the net injected spin that is left after the thermalization process is denoted by
$$R(\varepsilon_0) = \left( \frac{\varepsilon_0}{\varepsilon_0 - N \varepsilon_{LO}} \right) \frac{\int d\phi \int d\theta \sin \theta S_{N,z}(k_N,0)}{\int d\phi \int d\theta \sin \theta S_{0,z}(k_0,0)}$$

where $\varepsilon_0$ is the electrons’ injected energy above the semiconductor conduction band $\Gamma$-point and $N$ is the number of phonon emissions that are needed to reach the bottom of the band. We have considered only the $z$-direction since the injected net spin is aligned along this direction.

Fig. 3 shows the ratio between the final and initial injected net spin. The spin information is largely kept when the energy of injected electrons is less than 0.2 eV. These results are also in accordance with recent measured data by Crooker et al. [2]. The slower spin decay of the experimental results is attributed to the fact that the theoretical curve considers only the most energetic injected electrons (whose spin relaxation is fastest). In the experiment, however, electrons with lower energy also contribute to the current (the allowed injection energy is anywhere between the conduction band edge in the semiconductor and the Fe Fermi level). The application of this iterative procedure is limited if the injected electrons experience strong inter-valley scattering processes. In GaAs this limit is reached when the injection energy is $\varepsilon_0$=0.3 eV [14]. Our model shows that less than 1/5 of the net injected spin is left during the possible $n=8$ LO-phonon emissions in this limit.

In summary, we have presented a transparent technique to trace the spin evolution during the relaxation of hot electrons in the conduction band $\Gamma$-valley of zinc-blende semiconductors. We have shown that if the voltage drop across a reversed biased GaAs/Fe junction is moderate then the spin information is largely kept. This technique can be used to evaluate the applicability of the boundary conditions in modeling spintronics devices.

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