Spin relaxation of photogenerated degenerate electron distributions in GaAs

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Spin-relaxation time of photogenerated degenerate electron distributions in GaAs is directly measured with the use of a mode-locked ruby laser and streak camera system. It is shown both theoretically and experimentally that the dominant relaxation mechanism is due to the spin precession mechanism of D'Yakonov-Perel'. The calculated spin-relaxation-rate dependence of $N_e^{2/3}$ agrees well with the measured dependency of $N_e^{2/3}$.

Recently, there has been considerable interest in studying the electron-spin-relaxation kinetics in semiconductors. Most of the work has used the circularly polarized luminescence as a measure of the electron-spin polarization. In this Communication, we will present theoretical and experimental evidence that the mechanism for spin relaxation in semiconductors with degenerate electron distributions is through the D'Yakonov-Perel' mechanism. This occurs because the strong narrowing due to the very rapid momentum relaxation is overcome by the large splitting of the conduction band ($\alpha k^2$) at the Fermi surface. The observed spin-relaxation rate and that calculated assuming the D'Yakonov-Perel' mechanism is less than that calculated assuming the spin exchange between electrons and holes using the parameters assumed by Fishman and Lampel. This discrepancy may be due to the change in this rate due to the degenerate hole distribution which has been pointed out recently by Kleinman and Miller (KM).

Previous workers have performed spin experiments and calculations for nondegenerate electron distributions since these are the only electron distributions attainable using the usual techniques for measuring the electron-spin relaxation with cw excitation. We have performed experiments with a mode-locked ruby laser and a streak camera system which allows us to obtain varying electron densities which are all degenerate in the temperature range (70–100 K) of our experiments. These experiments have allowed us to test our calculation of the various spin-relaxation mechanisms under conditions of electron degeneracy. Only the D'Yakonov-Perel' mechanism which accounts for the relaxation through the precession of electron spins in the $\alpha$-dependent pseudomagnetic field of the crystal has the proper dependence on the electron-hole concentration.

Fishman and Lampel have provided a good summary of the various proposed spin-relaxation mechanisms and their electron energy dependence. They then calculated the temperature dependence of these mechanisms by integrating them over a Maxwellian distribution. The approximations made by Fishman and Lampel in doing this are not valid for degenerate distributions. We have instead extended the theory and integrated these mechanisms over Fermi electron distributions using Sommerfield's approximation for the Fermi-Dirac integral because we are looking at a broad band of energies:

$$F_j \left( \frac{n}{kT} \right) = \int \frac{(e/kT)d\epsilon/kT}{1 + \exp[(\epsilon - e)/kT]} = \left( \frac{n/kT}{1 + 1} \right)^{j+1}$$

where $e$ is the electron energy above the bottom of the conduction band, $k$ is Boltzmann's constant, $T$ is the temperature in kelvin, and $n$ is the Fermi (or quasi-Fermi, where appropriate) level. This approximation is strictly valid for $n/kT > 20$. Our experiments span the range from $n/kT = 10$ to $n/kT > 20$. At the lower end of the range, there is about a 20% error which is within the experimental error.

The problem remains of how to determine the momentum relaxation terms in the D'Yakonov-Perel' mechanism and the Elliot-Yafet mechanism. We can ignore the Elliot-Yafet mechanism since the energy depence is too weak to explain our experimental results and becomes even stronger with any increase in the scattering rate due to the high electron energies and densities. We have approximated the dependence of the D'Yakonov-Perel' mechanism by considering only electron-electron collisions which dominate at these electron densities. We can then consider the collision rate by taking into account the influence of the decreasing mean free path (at high densities) and increasing electron velocity, i.e., $1/\tau_e = N_e \sqrt{e}/\tau_0$ (Refs. 3 and 10). Starting from the expression given by Fishman and Lampel [Eq. (16), Ref. 3], using the dependence of electron collision time $\tau_e$ and correcting for the isotropic scattering factor rather than the anisotropic factor valid for impurity dominated scattering, we obtain upon integration over the electron distribution

$$\frac{1}{T_e} = \frac{2}{3} \frac{\tau_0}{(E_0)^3} \frac{1}{N_e} (4.7 \times 10^{12})(kT)^{3/2} F_j(n/kT) \int F_j(n/kT)$$


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where \( \tau_0 \) is a collision rate scaling factor whose value we shall obtain from the experiment, \( E_g \) is the band gap, and \( F_j(n/kT) \) is the Fermi-Dirac integral of order \( j \). Using Sommerfeld’s approximation this reduces to

\[
\frac{1}{T_s} = 4 \left( \frac{E_g}{N_e} \right)^3 \frac{1}{N_e} (4.7 \times 10^{27}) \frac{h^{5/2}}{N_e} .
\]

(3)

Note that there is no temperature dependence to \( 1/T_s \) in this approximation since the electron distributions are nearly independent of the temperature in the heavily degenerate regime at these temperatures. We can obtain the dependence of the electron density on the Fermi level from \( F_{1/2} \) (Ref. 11):

\[
N_e = 4\pi \left[ \frac{2m_e kT}{\hbar^2} \right]^{3/2} F_{1/2} \left( \frac{n}{kT} \right) \approx \frac{8\pi}{3} \left[ \frac{2m_e}{\hbar^2} \right]^{3/2} n^{3/2},
\]

(4)

where \( N_e \) is the electron density, \( m_e \) is the electron effective mass, and \( h \) is Planck’s constant. When the Fermi level dependence [Eq. (4)] is substituted into Eq. (3) we obtain the spin-relaxation rate

\[
\frac{1}{T_s} = \frac{1}{4} \left( \frac{E_g}{N_e} \right)^3 \left[ \frac{3}{8\pi} \left( \frac{2m_e}{\hbar^2} \right)^{3/2} (4.7 \times 10^{27}) \frac{h^{3/2}}{N_e} \right].
\]

(5)

Note that the units of \( E_g \) are now ergs.

The experimental arrangement and procedure have been described previously\(^\text{12}\) with the exception of two modifications. A photodiode and beam splitter have been inserted to enable us to measure the excitation energy of each laser pulse. In addition, the orthogonal polarizers at the streak camera slit have been replaced with a Wollaston prism which provides greater sensitivity, allowing measurements at lower electron-hole concentrations. Figure 1 shows the experimental data for three different samples of Zn-doped GaAs (\( N_d \approx 6 \times 10^{16}, 7 \times 10^{17}, \) and \( 1.1 \times 10^{18} \) cm\(^{-3}\)). The solid line is a least-squares fit to the data with a slope of \(-0.63\) [note \( T_s \) is plotted and, not \( 1/T_s \) as in Eq. (5)]. The uncertainty in the excitation energy within a sample group is approximately 10% but increases to approximately 25% between samples due to uncertainties in the focal-spot diameter on the sample. The uncertainty in the measured spin-relaxation time is shown on the graph by the error bars. The densities being referred to in the abscissa of Fig. 1 is the average of the carrier density over the entire absorption length corresponding to 1.8-ev excitation photon energy. This contributes to less than 25% uncertainty in the spin-relaxation time which is within the experimental error.

The fitted dependence of the data on electron concentration \( (aN_d^{0.63}) \) agrees well with the calculated \( 3/2 \) power dependence. The measured collision rate scaling factor \( \tau_0 \) is \( 3.64 \times 10^{-2} \) sec cm\(^{-2}\) which corresponds to a momentum relaxation time of \( 1.2 \times 10^{-13} \) sec at an electron concentration of \( 10^{18} \) cm\(^{-3}\). This value is consistent with calculated values\(^\text{13}\) and values obtained indirectly from hot electron-relaxation measurements in GaAs\(^\text{14}\).

The essential difference between our work and the work done by Kleinman and Miller\(^\text{4}\) is in regard to the photon fluence of excitation used, which changes the essential physical condition. We have created degenerate electron distributions over the essentially nondegenerate background hole distribution at our temperature range while the KM\(^\text{4}\) experiment employed nondegenerate electron distributions and degenerate hole distributions. The virtual photon mechanism (KM) does not account for the observed spin-relaxation rate of \( N_d^{2/3} \); instead it predicts a linear dependence on \( N_d \) and a strong temperature dependence. Furthermore, Kleinman and Miller measurements\(^\text{4}\) do not agree with the measurements of Clark\(^\text{15}\) et al. under similar experimental conditions. The temperature range used in our experiment is limited by stimulated emission and higher-order recombination processes at low temperatures and transitions from the split-off band at high temperatures.
Several of the other spin-relaxation mechanisms yield values that are close to our measured values for spin-relaxation rate. However, they yield power dependences on the electron-hole concentration for degenerate distribution that are quite different [Elliott and Yafet (EY) is $\frac{2}{3}$ and Bir, Aronov, and Pikus (BAP) is $\frac{4}{7}$] than the measured 0.63 dependence and calculated $\frac{2}{3}$ dependence for the D'Yakonov-Perel' mechanism.

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††Spin relaxation due to scattering of electrons from screened ionized impurities as proposed by Elliott and Yafet (EY) and exchange interaction between holes and electrons as proposed by Bir, Aronov, and Pikus (BAP).