

## Spin-exchange scattering in semiconductors

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We calculate the spin-coherence lifetime of an electron in a semiconductor due to exchange scattering from neutral donors. We consider the process where a band electron scatters from a donor, and the electron bound to the neutral donor has the opposite spin of the band electron. They exchange places in the scattering, which is a process that effectively flips the electron spin of the two electrons. The lifetime is calculated in two and in three dimensions using the Born approximation.

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### I. INTRODUCTION

We calculate the rate of spin-exchange scattering of conduction-band electrons. We consider the process of exchange scattering of the band electron from electrons bound to donors. If the bound electron initially has the opposite spin of the band electron, exchange scattering is a process that effectively flips the spin of the conduction electron. A similar process is the spin-exchange scattering from an electron that is part of an exciton. We consider the process in both two and three dimensions and evaluate the scattering rate using the Born approximation. The identical process can happen for holes when they scatter from other holes bound to acceptors or excitons.

There is much interest in spintronics: using spin currents for a variety of electronic processes.<sup>1-5</sup> In all these devices, the spin-relaxation time is very important. Spin coherence is also important for many optical experiments.<sup>6-9</sup> A theory of spin-flip scattering was presented by D'yakonov and Perel',<sup>10</sup> based on an earlier theory by Dresselhaus.<sup>11</sup> It uses the spin-orbit interaction and is a small relativistic effect. The spin-flip process discussed by us is not based on a relativistic process and is a much larger effect; the lifetimes we find are much shorter.

The calculation is done in both two and in three dimensions. Three-dimensional results are of interest for bulk devices. Many other applications are done in quantum wells or superlattices. There the electron motion is largely two dimensions. So we calculate the rate process also in two dimensions. Both cases depend upon the density  $n_B$  of scattering centers, which is the density of donors.

In metals, spin-flip scattering of a band electron by a localized electron is a well-known process.<sup>12,13</sup> It causes the Kondo effect<sup>12</sup> among other phenomena. There the process is characterized by an effective interaction  $V_{sd} = J\vec{S} \cdot \vec{s}$ , where  $J$  is the interaction and the two factors of  $\vec{s}, \vec{S}$  are the local spin and band electron spin. Here we use the Born approximation<sup>14</sup> to calculate  $J$  using hydrogen wave functions. We find that the cross section for scattering has a very large dependence on the energy of the band electron.

Electron scattering from neutral donors was investigated years ago in relation to the mobility of electrons and holes in semiconductors.<sup>15-18</sup> They used a variety of methods, including variational techniques,<sup>16</sup> resonance scattering,<sup>17</sup> and phase shifts.<sup>18</sup> They assumed the system had only two elec-

trons and computed the cross section for the spin singlet and spin triplet arrangements. If one constructs a many-electron eigenfunction, for many band electrons and many bound electrons, then each pair does not have the singlet/triplet symmetry. For that reason, our calculation differs from those in these earlier papers.

### II. RATE EQUATIONS

The spin-exchange scattering will cause a mutual spin flip between a bound spin and a conduction-band spin, provided they have opposite initial spin states. Later, they can flip back. So we need to consider the rate equations for the time dependence of the spin polarization. Let  $n_\sigma(t)$  be the time dependence of the density of conduction-band spins with  $\sigma = \uparrow, \downarrow$ , where the axis defining up and down is arbitrary. The total conduction-band density  $n = n_\uparrow + n_\downarrow$  is independent of time. Similarly, let  $n_{B\sigma}(t)$  be the time dependence of the density of electron spins bound to donors. This total  $n_B = n_{B\uparrow} + n_{B\downarrow}$  is also constant. In Secs. III and IV we employ the golden rule to calculate the rate  $\gamma$  of spin flip by the spin-exchange process:  $\gamma$  has the dimensions of  $(\text{length})^d/s$ , where  $d$  is the dimension. Then the rate equations for the evolution of spin polarization are

$$\frac{d}{dt}n_\uparrow(t) = -\gamma[n_\uparrow(t)n_{B\downarrow}(t) - n_\downarrow(t)n_{B\uparrow}(t)], \quad (1)$$

$$= -\frac{d}{dt}n_\downarrow(t) = \frac{d}{dt}n_{B\downarrow}(t) = -\frac{d}{dt}n_{B\uparrow}(t). \quad (2)$$

The nonlinear Eq. (1) has a simple solution. The following equations define the fractional magnetizations  $m(t)$  for conduction electrons and  $m_B(t)$  for bound electrons:

$$n_\uparrow(t) = \frac{n}{2}[1 + m(t)], \quad n_{B\uparrow}(t) = \frac{n_B}{2}[1 + m_B(t)], \quad (3)$$

$$n_\downarrow(t) = \frac{n}{2}[1 - m(t)], \quad n_{B\downarrow}(t) = \frac{n_B}{2}[1 - m_B(t)]. \quad (4)$$

Then Eqs. (1) and (2) become linear,

$$\frac{d}{dt}m(t) = -\gamma n_B(m - m_B), \quad (5)$$

$$\frac{d}{dt}m_B(t) = \gamma n(m - m_B). \quad (6)$$

The solutions to these rate equations are

$$m(t) = \frac{1}{n + n_B}[B_0 + n_B A_0 e^{-\lambda t}], \quad (7)$$

$$m_B(t) = \frac{1}{n + n_B}[B_0 - n A_0 e^{-\lambda t}], \quad (8)$$

$$\lambda = \gamma(n + n_B), \quad (9)$$

where the constants [ $A_0 = m(0) - m_B(0)$ ,  $B_0 = nm + n_B m_B$ ] are set by the initial conditions.

As an example, assume that the bound electrons are initially unpolarized [ $m_B(t=0)=0$ ], while the conduction-band electrons have an initial polarization of  $m_0$ . These initial conditions give  $A_0 = m_0$ ,  $B_0 = nm_0$ ,

$$m(t) = \frac{m_0}{n + n_B}[n + n_B e^{-\lambda t}], \quad (10)$$

$$m_B(t) = \frac{nm_0}{n + n_B}[1 - e^{-\lambda t}]. \quad (11)$$

Note that regardless of initial conditions, the two systems have the same fractional polarization for times  $\lambda t \gg 1$ ,

$$m(t = \infty) = \frac{B_0}{n + n_B} = m_B(t = \infty). \quad (12)$$

Also note that if  $n \gg n_B$  then the final fraction  $m(\infty) \approx m_0 = m(0)$ , and the spin polarization of the conduction electrons does not change much. After the characteristic time  $t_c = 1/\lambda$ , the spin system will reach a steady state where  $m \approx m_B$  and no more spin depolarization occurs by this mechanism. Then further spin relaxation occurs by the spin-orbit channel described in Ref. 10.

As a simple example, consider a system with  $n_B = 100$  bound electrons, which initially have  $m_B(0) = 0$ ,  $n_{B\uparrow} = n_{B\downarrow} = 50$ . Inject a single conduction spin-up electron. In time it will exchange scatter and flip to the down position. Then there will be 51 bound electrons with spin up. The conduction electron, now with spin down, will flip from one of these 51 down spins and go back to having spin up. The conduction electron spends slightly longer with the up-spin arrangement compared to the down-spin arrangement, since there are 50 down spins to scatter from when it is up but 51 up spins to scatter from when it is down. The average polarization is then about 1%. Sections III and IV calculate the constant  $\gamma$  in two and three dimensions.

### III. THREE DIMENSIONS

The rate of spin-exchange scattering is given by<sup>14,19</sup>

$$\gamma = \frac{2\pi}{\hbar} \int \frac{d^3 k_f}{(2\pi)^3} M^2 \delta(\varepsilon_i - \varepsilon_f). \quad (13)$$

The matrix element has the form<sup>14,19</sup>

$$M(\mathbf{k}_i, \mathbf{k}_f) = \frac{e^2}{\varepsilon_0} \int d^3 r_1 d^3 r_2 \phi_{1s}(r_1) \phi_{1s}(r_2) e^{i(\mathbf{k}_i \cdot \mathbf{r}_1 - \mathbf{k}_f \cdot \mathbf{r}_2)} \times \left[ \frac{1}{r_1} - \frac{1}{r_{12}} \right], \quad (14)$$

$$\phi_{1s}(r) = \frac{1}{\sqrt{\pi a_0^3}} \exp(-r/a_0), \quad (15)$$

where  $a_0$  is the effective Bohr radius of the donor in the effective-mass approximation. In GaAs it is around  $a_0 = 10$  nm.<sup>20</sup> We assume that the scattering is elastic, so  $k_f = k_i$  and they differ only in their angle.

The two terms in Eq. (14) are evaluated separately,

$$M(\mathbf{k}_i, \mathbf{k}_f) = M_N(k) - M_x(\mathbf{k}_i, \mathbf{k}_f), \quad (16)$$

$$M_N(k) = \frac{32\pi e^2 a_0^2}{\varepsilon_0 (1 + p^2)^3}, \quad p = ka_0. \quad (17)$$

In order to evaluate  $M_x$ , convert all variables to a dimensionless form:  $\mathbf{p}_j = \mathbf{k}_j a_0$ ,  $\tilde{\rho}_j = \mathbf{r}_j / a_0$ ,

$$M_x = \frac{32\pi e^2 a_0^2}{\varepsilon_0} L(\mathbf{p}_i, \mathbf{p}_f), \quad (18)$$

$$L(\mathbf{p}_i, \mathbf{p}_f) = \frac{1}{32\pi^2} \int d^3 \rho_1 d^3 \rho_2 e^{-\rho_1 - \rho_2} e^{i(\mathbf{p}_i \cdot \tilde{\rho}_1 - \mathbf{p}_f \cdot \tilde{\rho}_2)} \frac{1}{\rho_{12}}. \quad (19)$$

Evaluate this at zero wave vector and get

$$L(0,0) = \frac{5}{8}. \quad (20)$$

Note that the two terms in the matrix element do not cancel,

$$M(0,0) = \frac{32\pi e^2 a_0^2}{\varepsilon_0} \left[ 1 - \frac{5}{8} \right] = \frac{12\pi e^2 a_0^2}{\varepsilon_0}. \quad (21)$$

The formulas for the scattering rate are now

$$\gamma(p) = 288\pi \frac{E_{\text{Ry}}}{\hbar} (a_0^3) I(p), \quad (22)$$

$$I(p) = \frac{64p}{9} \int \frac{d\Omega}{4\pi} \left[ \frac{1}{(1+p^2)^3} - L(\mathbf{p}_i, \mathbf{p}_f) \right]^2, \quad (23)$$

where  $d\Omega$  is the integral over  $4\pi$  solid angle. The large prefactor of  $288\pi$  comes from the factor of  $12\pi$  in the matrix element. In GaAs (Ref. 20)  $E_{\text{Ry}} = 5.9$  MeV. Then the numerical value of the prefactor is

$$\gamma_0 = 288\pi \frac{E_{\text{Ry}}}{\hbar} = 8.1 \times 10^{15}/\text{s}. \quad (24)$$

In order to estimate values of  $p = ka_0$ , consider the thermal energy using numbers for GaAs,

$$\frac{\hbar^2 k^2}{2m^*} = \frac{3}{2} k_B T, \quad p^2 = 0.022T. \quad (25)$$

This gives  $p^2 = 0.09$  at  $T = 4$  K and  $p^2 = 1.7$  at  $T = 77$  K. At low temperature,  $p^2$  is small and  $L \approx 5/8$ . At higher tempera-

tures, the values of  $p^2$  increase and  $L$  decreases.

One way to evaluate the integral is by Fourier transforming the Coulomb interaction, which gives

$$L(p, \theta) = \frac{2}{\pi} \int_{-\infty}^{\infty} dq \int \frac{d\Omega}{4\pi} \frac{1}{[1 + (\mathbf{p}_i + \mathbf{q})^2]^2 [1 + (\mathbf{p}_f + \mathbf{q})^2]^2}. \quad (26)$$

Write the denominator as

$$L(p, \theta) = \frac{2}{\pi} \int_{-\infty}^{\infty} dq \int \frac{d\Omega}{4\pi} \frac{1}{[(q + q_i)^2 + a_i^2]^2 [(q + q_f)^2 + a_f^2]^2}, \quad (27)$$

$$q_j = p v_j, \quad a_j^2 = 1 + p^2(1 - v_j^2), \quad (28)$$

$$v_i = \hat{p}_i \cdot \hat{q}, \quad v_f = \hat{p}_f \cdot \hat{q}, \quad \cos(\theta) = \hat{p}_i \cdot \hat{p}_f. \quad (29)$$

The integral over  $dq$  can be evaluated by contour integration,

$$L(p, \theta) = \int \frac{d\Omega}{4\pi} \frac{1}{a_i^2 a_f^2 D^2} \left[ \frac{a_i^3 + a_f^3}{a_i a_f} + \frac{4(a_i + a_f)^3}{D} \right], \quad (30)$$

$$D = (a_i + a_f)^2 + (q_i - q_f)^2. \quad (31)$$

For forward scattering one gets that

$$L(p, 0) = \frac{1}{8} \left[ \frac{1}{1 + p^2} + \frac{4/3}{(1 + p^2)^2} + \frac{8/3}{(1 + p^2)^3} \right]. \quad (32)$$

For back scattering,  $v_f = -v_i$ ,  $D = 4(1 + p^2)$ , and

$$L(p, \pi) = \frac{1}{8(1 + p^2)^3} \left[ 1 + \frac{4}{p} \sin^{-1} \left( \frac{p}{\sqrt{1 + p^2}} \right) \right]. \quad (33)$$

By comparing Eqs. (32) and (33), one sees that  $L(p, \theta)$  is largest at  $\theta=0$  and decreases with increasing angle.

For a general scattering angle  $\theta$  one can numerically evaluate Eq. (30). Another way to evaluate the integral is to write it as

$$L(p, \theta) = \sum_{n=0} b_n(p) P_n(\theta), \quad (34)$$

$$b_0 = \frac{5 + p^2}{8(1 + p^2)^3}. \quad (35)$$

The other  $b_n$  are much smaller, so an adequate approximation to the cross section in three dimensions is to set  $L \approx b_0$  and get isotropic scattering,

$$I(p) = \frac{p(3 - p^2)^2}{3^2(1 + p^2)^6}, \quad (36)$$

where  $p = ka_0$ . Since  $I(0) = 0$ , the maximum value of this function is at  $p = 0.283$  where  $I \approx 1/6$ . That is the value of  $p^2 = 0.08$  when at thermal energies at  $T = 4$  K.

#### IV. TWO DIMENSIONS

The electron-scattering rate in two dimensions is given by

$$\gamma = \frac{2\pi}{\hbar} \int \frac{d^2 k_f}{(2\pi)^2} M^2 \delta(\varepsilon_i - \varepsilon_f). \quad (37)$$

The matrix element for exchange scattering has the form

$$M(\mathbf{k}_i, \mathbf{k}_f) = \frac{e^2}{\varepsilon_0} \int d^2 r_1 d^2 r_2 \phi_{1s}(r_1) \phi_{1s}(r_2) e^{i(\mathbf{k}_i \cdot \mathbf{r}_1 - \mathbf{k}_f \cdot \mathbf{r}_2)} \times \left[ \frac{1}{r_1} - \frac{1}{r_{12}} \right], \quad (38)$$

$$\phi_{1s}(r) = \frac{\sqrt{8}}{\sqrt{\pi} a_0} \exp(-2r/a_0), \quad (39)$$

where  $a_0$  is the effective Bohr radius of the donor in three dimensions. We assume the scattering is elastic, so  $k_f = k_i$  and they differ only in their angle. We define zero angle as the direction of  $\mathbf{k}_i$ , so that the direction of  $\mathbf{k}_f$  is the scattering angle  $\theta$ . The matrix element has two terms, and the first one is an easy integral. Defining  $p = ka_0/2$ ,

$$M_N(p) = \frac{4\pi e^2 a_0}{\varepsilon_0 (1 + p^2)^2}. \quad (40)$$

In order to evaluate  $M_x$ , convert all variables to a dimensionless form:  $\mathbf{p}_j = \mathbf{k}_j a_0/2$ ,  $\vec{\rho}_j = 2\mathbf{r}_j/a_0$ ,

$$M_x = \frac{4\pi e^2 a_0}{\varepsilon_0} L(p, \theta), \quad (41)$$

$$L(p, \theta) = \frac{1}{4\pi^2} \int d^2 \rho_1 d^2 \rho_2 e^{-\rho_1 - \rho_2} e^{i(\mathbf{p}_i \cdot \vec{\rho}_1 - \mathbf{p}_f \cdot \vec{\rho}_2)} \frac{1}{\rho_{12}}. \quad (42)$$

The integral  $L$  is evaluated in Fourier transform (FT) space. Taking the two-dimensional FT of the Coulomb potential, we get the integral in dimensionless form ( $x = qa_0/2$ ,  $y = ka_0/2$ ),

$$L(y, \theta) = \int_0^\infty dx \int_0^{2\pi} \frac{d\phi}{2\pi} G(x, y, \phi) G(x, y, \theta - \phi), \quad (43)$$

$$G(x, y, \phi) = \frac{1}{[1 + x^2 + y^2 + 2xy \cos(\phi)]^{3/2}}. \quad (44)$$

This integral is hard to evaluate. It can be done for a few special cases. The first is setting  $k=0$  ( $y=0$ ),

$$L(0, 0) = \int_0^\infty \frac{dx}{[1 + x^2]^3} = \frac{3\pi}{16}. \quad (45)$$

Note that this is not equal to  $M_N(0)$  and the two terms do not cancel at forward scattering. The exchange-scattering constants are

$$\gamma(y) = 32\pi^2 \frac{E_{RY}}{\hbar} (a_0^2) I(y), \quad (46)$$

$$I(y) = \int_0^{2\pi} \frac{d\theta}{2\pi} \left[ \frac{1}{(1+y^2)^2} - L(y, \theta) \right]^2, \quad (47)$$

where  $E_{\text{Ry}}$  is the binding energy of the three-dimensional donor. The prefactor of the above expression is

$$32\pi^2 \frac{E_{\text{Ry}}}{\hbar} = 2.8 \times 10^{15}/\text{s}. \quad (48)$$

An evaluation of  $L(y, \theta)$  is to do a Fourier expansion in the angle and retain only a few terms,

$$L(y, \theta) = \sum_{n=0} a_n(y) \cos(n\theta). \quad (49)$$

It is useful to define

$$J_n(x, y) = \int_0^{2\pi} \frac{d\phi}{2\pi} \frac{\cos(n\phi)}{[1+x^2+y^2+2xy\cos(\phi)]^{3/2}}, \quad (50)$$

$$a_n(y) = \varepsilon_n \int_0^\infty dx J_n(x, y)^2, \quad (51)$$

$$\varepsilon_0 = 1, \quad \varepsilon_n = 2, \quad n \geq 1. \quad (52)$$

The  $J_n$  can be expressed in terms of elliptic integrals. Let  $\phi = 2\alpha$  and

$$J_n(x, y) = \frac{2/\pi}{[1+(x+y)^2]^{3/2}} \int_0^{\pi/2} \frac{d\alpha \cos(2n\alpha)}{[1-m\cos^2(\alpha)]^{3/2}}, \quad (53)$$

$$m = \frac{4xy}{1+(x+y)^2} \leq 1, \quad (54)$$

$$a_n(0) = \frac{3\pi}{16} \delta_{n=0}. \quad (55)$$

At small values of  $y$  then  $a_n(y) \sim y^n$ . The lowest few are

$$J_0 = \frac{2/\pi}{[1+(x+y)^2]^{3/2}} \left[ K(m) + 2m \frac{dK}{dm} \right], \quad (56)$$

$$J_1 = \frac{2/\pi}{[1+(x+y)^2]^{3/2}} \left[ 2(2-m) \frac{dK}{dm} - K \right], \quad (57)$$

$$J_2 = \frac{2/\pi}{m^2[1+(x+y)^2]^{3/2}} \left[ 8(E-K) + m^2 K + 2m \frac{dK}{dm} (8-8m+m^2) \right],$$

$$K(m) = \int_0^{\pi/2} \frac{d\alpha}{\sqrt{1-m\cos^2(\alpha)}}, \quad E = \int_0^{\pi/2} d\alpha \sqrt{1-m\cos^2(\alpha)}. \quad (58)$$

We have evaluated these expressions and find, as in three dimensions, that an accurate approximation for the wave

vector dependence is to retain only the first term in the series,

$$I(y) \approx \left[ \frac{1}{(1+y^2)^2} - a_0(y) \right]^2. \quad (59)$$

In two dimensions, the function  $\gamma(y)$  has its largest value at  $y=0$  and decreases for increasing values of  $y=ka_0/2$ .

## V. DISCUSSION

We have shown that spin-exchange scattering defines a rate constant  $\gamma$ . Using this rate constant, we derived and solved a set of rate equations which determines the final degree of spin polarization of the conduction-band system. This seems to be a fast process, so that the final degree of spin polarization is reached rapidly.

The spin-exchange scattering depends upon the density  $n_B$  of scattering centers. This number will vary from sample to sample and be a very small number in nominally pure samples. The spin-polarization lifetime,

$$\frac{1}{\tau} = \gamma(n + n_B), \quad (60)$$

depends upon this density. All semiconductors have some impurities. Some may be shallow donors, while others have a larger binding energy. Since these defects can trap electrons, they serve as scattering sites that can exchange scatter with the band electron.

In three dimensions, at high doping, if one is above the Mott limit ( $n_D^{1/3} a_0 > 0.24$ ), the average binding energy of the donors goes to zero. Then  $n_B$  will be very small. Due to the random locations of the donors, some will still be clustered and lead to bound electrons. These are the donors that give rise to the well-known band tails. In many pump-probe optical experiments, a temporary density of excitons is created, which could also serve to exchange scatter a band electron.

In two dimensions, the binding energy does not vanish at any electron density, so there are always electrons bound to donors. Many experiments are done in quantum wells or superlattices. These states are midway between two and three dimensions; they are three dimensions but the confinement to the quantum well makes them quasi-two-dimensional. In this case the location of the donor is important for the calculation of the cross section: whether it is in the quantum well or outside in the barrier layer.

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