4-30-2014

Spin relaxation time dependence on optical pumping intensity in GaAs:Mn

V. Burobina
University of Utah

Christian Binek
University of Nebraska-Lincoln, cbinek@unl.edu

Spin relaxation time dependence on optical pumping intensity in GaAs:Mn

V. Burobina and Ch. Binek

Articles you may be interested in
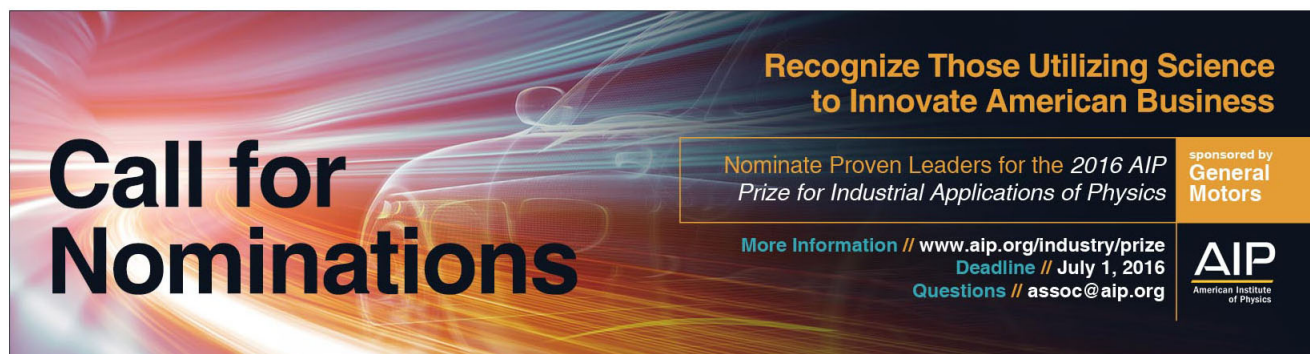
Temperature dependent electric field control of the electron spin relaxation in (111)A GaAs quantum wells
Appl. Phys. Lett. **102**, 242408 (2013); 10.1063/1.4811555

All-optical four-state magnetization reversal in (Ga,Mn)As ferromagnetic semiconductors
Appl. Phys. Lett. **99**, 091111 (2011); 10.1063/1.3634031

Electron spin dynamics in heavily Mn-doped (Ga,Mn)As
Appl. Phys. Lett. **97**, 262109 (2010); 10.1063/1.3531754

Coherent magnetization precession in GaMnAs induced by ultrafast optical excitation
Appl. Phys. Lett. **91**, 112506 (2007); 10.1063/1.2785114

Magnetization-induced optical nonlinearity in ferromagnetic GaMnAs
J. Appl. Phys. **101**, 063519 (2007); 10.1063/1.2696930

Spin relaxation time dependence on optical pumping intensity in GaAs:Mn

V. Burobina[1] and Ch. Binek[2]

[1]*Department of Physics and Astronomy, University of Utah, 115 South 1400 East, Salt Lake City, Utah 84112-0830, USA*

[2]*Department of Physics and Astronomy, Nebraska Center for Materials and Nanoscience, Theodore Jorgensen Hall, 855 North 16th Street, University of Nebraska, P.O. Box 880299, Lincoln, Nebraska 68588-0299, USA*

We analyze the dependence of electron spin relaxation time on optical pumping intensity in a partially compensated acceptor semiconductor GaAs:Mn using analytic solutions for the kinetic equations of the charge carrier concentrations. Our results are applied to previous experimental data of spin-relaxation time vs. excitation power for magnetic concentrations of approximately $10^{17}\,\mathrm{cm}^{-3}$. The agreement of our analytic solutions with the experimental data supports the mechanism of the earlier-reported atypically long electron-spin relaxation time in the magnetic semiconductor. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4874218]

I. INTRODUCTION

During the last three decades, there has been increasing interest in experimental and theoretical studies in the field of spintronics.[1–3] A major challenge in spintronics is the optimization of electron-spin lifetime. A lower bound for the required electron-spin lifetime is given by spin-polarized carrier transport across characteristic length scales such as the conducting channel in transistor-type architectures. The success of engineering spintronics depends on a deeper understanding of fundamental spin interactions in solid-state materials.[4] Dilute magnetic semiconductors (DMS) have been intensively studied since 1980s.[5,6] The current renaissance of interest in magnetic semiconductors is due to their various functionalities relevant to spintronic devices.[7] DMS demonstrate a number of spin-dependent optical and transport properties caused by large, spontaneous spin splitting of electronic bands. These relatively strong effects of exchange interaction are not readily available in magnetically undoped semiconducting materials.

Long spin relaxation times for electrons on the order of 100 ns are typically observed in *n*-type GaAs.[8–10] In *p*-type GaAs, electron spin relaxation time is usually less than 1 ns. Astakhov *et al.*[11] reported that *p*-type GaAs with enhanced spin relaxation time by two orders of magnitude can be realized by doping with manganese acceptor concentration on the order of $10^{17}\mathrm{cm}^{-3}$. The ferromagnetism of GaAs:Mn is very sensitive to the growth conditions as well as post-growth processing of the sample.[12] The concentration of shallow donor and shallow acceptor impurities is an important parameter influencing the spin relaxation time. Specifically, the concentration of shallow donor impurities needs to be larger than the concentration of shallow acceptor impurities to ensure ionized (negatively charged) Mn impurity states. The electron-spin flip rate is inversely proportional to the square of the concentration of photoexcited holes on manganese because the spins of the photoexcited holes couple antiferromagnetically to the spins of Mn ions. This antiferromagnetic coupling reduces the effective magnetic field, which is detrimental to spin relaxation time through scattering via precession of the electron spins in the conduction band. Reduction of the precession frequency through reduction of the effective magnetic field increases the electron spin relaxation time.[11]

II. DEPENDENCE OF SPIN RELAXATION TIME ON CONCENTRATION OF PHOTOEXCITED HOLES

In this article, we calculate the dependence of the concentration of photoexcited holes, q, on manganese as a function of optical pumping power using a rate-equation approach. The results have been adapted to allow for a comparison with experimental data of spin-relaxation time, τ_s, vs. excitation power, P. The spin-relaxation time has been measured in the partially compensated acceptor semiconductor of GaAs:Mn with the magnetic concentration of approximately $10^{17}\mathrm{cm}^{-3}$. The theoretical q vs P dependence can be mapped onto a τ_s vs. P dependence motivated by the fact that the concentration of photoexcited holes determines the effective magnetic field, B_{eff}, produced by the spins of the manganese ions. According to Dyakonov and Perel,[13,14] the inverse spin-relaxation time is given by $\tau_s^{-1} = \frac{2}{3}\langle\omega_f^2\rangle\tau_c$, where ω_f is the frequency of the electron-spin precession in the effective fluctuating magnetic field, B_{eff}, and τ_c is the characteristic time of this local field fluctuation where $B_{eff} \propto \omega_f$, and τ_c is virtually independent of the pumping power. The main detrimental impact on spin lifetime results from the presence of B_{eff}. The mechanism which reduces B_{eff}, and therefore increases τ_s, is the formation of antiferromagnetically coupled spins of ionized Mn atoms with the spin of the photoexcited holes. It is thus reasonable to assume that B_{eff} decreases with increasing number, q, of photoexcited holes. We employ the simplest empirical ansatz, $B_{eff}(q) = \frac{B_{eff}(0)}{b\,q+1}$, which is consistent with a decrease of B_{eff} for increasing q and an asymptotic approach $B_{eff} \rightarrow 0$ in the limit of large q. This ansatz allows us to map q vs P onto τ_s vs. P, via an analytic fitting function which we apply to fit the experimental τ_s vs. P data.
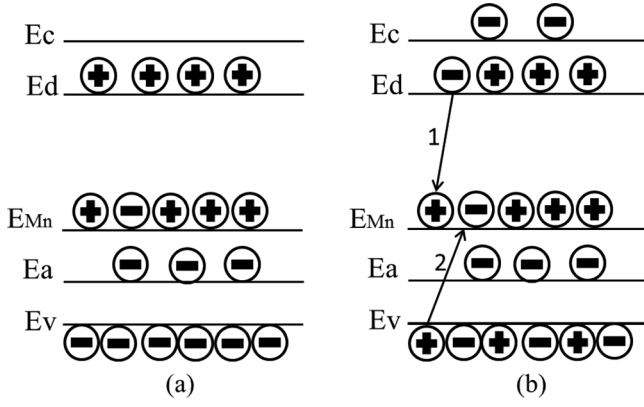
FIG. 1. (a) Energy diagram of GaAs:Mn with a manganese concentration of $7.8 \times 10^{17} \mathrm{cm}^{-3}$ at $T = 8 \mathrm{K}$. Circles with minus (plus) sign represent electrons (holes). (b) Energy diagram of electrons and holes in GaAs:Mn under the influence continuous optical excitation. Arrow 1 shows the transition of an electron from the donor level to the Mn level. Arrow 2 shows the transition of a hole from the valence band to the Mn level.

FIG. 2. Experimental dependence of electron spin relaxation time, τ_s, vs. excitation power, P, for the p-type GaAs:Mn (circles), and best fit of Eq. (12) (line).

III. RATE EQUATIONS: PHOTOEXCITED HOLE CONCENTRATION VERSUS EXCITATION POWER

The dependence of electron spin relaxation time, τ_s, on excitation power, P, at a temperature of 8 K presented in Fig. 3 of Ref. 11 demonstrates that τ_s increases with an increase of P, and tends to saturate for $P > 10$ mW. The electronic spin relaxation in GaAs:Mn was studied using the Hanle effect under optical pumping conditions.[13] In the p-type GaAs:Mn with a manganese concentration of $7.83 \times 10^{17} \mathrm{cm}^{-3}$, the donor concentration N_d is larger than the shallow acceptor concentration N_a, $N_d - N_a = 14.90 \times 10^{15} \mathrm{cm}^{-3}$, which causes a total ionization of shallow acceptor atoms and a partial ionization of manganese at a temperature of 8 K near the ground state (Fig. 1(a)). Under the influence of circularly polarized light provided by a titanium-sapphire laser operating at 800 nm, the photoexcited electrons in the conductivity band are captured by the trap of the donor impurities, and then recombine with manganese holes generating charge centers of manganese[11] (arrow 1 Fig. 1(b)). Thus, in addition to the applied external magnetic field, the electrons and holes experience an effective magnetic field from the manganese ions. This produces a Zeeman splitting. If the holes from the valence band fill the ionized acceptors of manganese (arrow 2 Fig. 2(b)), the hole spins will couple antiferromagnetically with the manganese spins. The effective magnetic fields generated by the manganese ions, therefore, will be compensated and will not significantly impact the electron spin relaxation. The electron-spin relaxation time will be prolonged with increasing concentration of photoexcited holes on manganese according to $\tau_s \propto 1/\omega_f^2 \propto (1 + bq)^2 / B_{eff}(0)^2$. To verify this mechanism, we calculated the dependence of the concentration of photoexcited holes on manganese vs. excitation power, map it onto $\tau_s(P)$, and compare the analytic result with the experimental data of electron spin relaxation time vs. excitation power.

The system of kinetic equations for the hole concentration, p, in the valence band and electron concentration, n, in the conduction band in the p-type GaAs:Mn can be written as
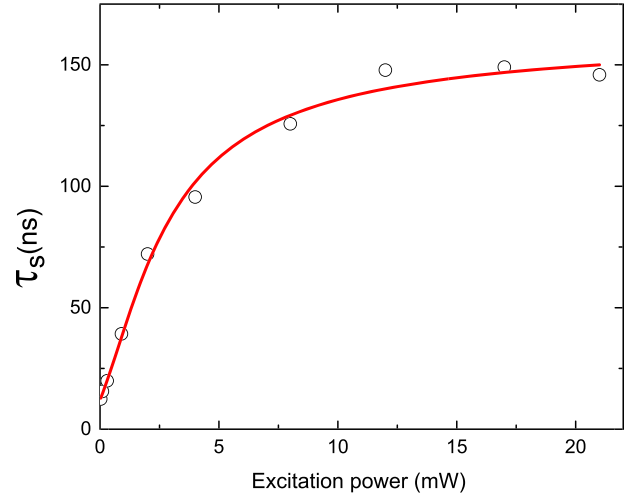
$$\dot{p} = G - W p n_{Mn} - R_x \, pn \quad \text{and}$$
$$\dot{n} = G - R_M p_{Mn} n - R_x pn, \tag{1}$$

where G is the rate at which the charge carriers are generated under circularly polarized optical pumping, W is the rate at which hole recombination occurs from the valence band to manganese levels, R_x is the band-to-band recombination rate, R_M is the rate at which electron recombination occurs from the conduction band to manganese levels, and n_{Mn} and p_{Mn} are the concentrations of electrons and holes on manganese levels, respectively. In the system (1), the concentrations of electrons and holes on the levels of shallow impurities were neglected since $N_{Mn} \gg N_d - N_a$.

The concentration of electrons in the conduction band can be expressed via the sum of the hole concentration in the valence band and the photoexcited holes, q, generated on manganese levels. It reads $n = p + q$. The number of electrons on manganese, n_{Mn}, can be written as $n_{Mn} = \delta - q$, where δ is the concentration of the ionized manganese atoms. The hole concentration on manganese, then, is $p_{Mn} = N - n_{Mn} = N - \delta + q$, where N is the total concentration of manganese atoms. The system of Eqs. (1) can be written as

$$\dot{p} = G - W p(\delta - q) - R_x p(p + q) \quad \text{and}$$
$$\dot{n} = G - R_M(N - \delta + q)(p + q) - R_x p(p + q). \tag{2}$$

Taking into account that $N \gg \delta - q$, we solved the system (2) in a steady state

$$R_M N(p + q) - W p(\delta - q) = 0.$$

Therefore,

$$p = \frac{R_{Mn} N q}{W(\delta - q) - R_M N} \tag{3}$$

or

$$q = \frac{p(W\delta - R_M N)}{R_M N + Wp}. \tag{4}$$

The value q can be placed in the expression for $\dot{n}$ of (2). In the steady-state conditions, this expression will be

$$R_x W p^2(\delta + p) + R_M N W p(\delta + p) - G(R_M N + Wp) = 0. \tag{5}$$

The resulting expression can be written as

$$R_x W \delta^3 \left(\frac{p}{\delta}\right)^2 \left(1 + \frac{p}{\delta}\right) + R_M N \delta^2 W \left(\frac{p}{\delta}\right)\left(1 + \frac{p}{\delta}\right)$$
$$- G\left(R_M N + W\delta\left(\frac{p}{\delta}\right)\right) = 0.$$

If $\frac{p}{\delta} = y$, then

$$R_x \delta W \delta y^2(1 + y) + R_M N \delta W y(1 + y) - \frac{G}{\delta}(R_M N + W \delta y) = 0. \tag{6}$$

Expression (6) can be modified as follows:

$$R_x \delta y^2(1 + y) + R_M N y(1 + y) - \frac{G}{\delta}\left(\frac{R_M N}{W\delta} + y\right) = 0,$$

which yields

$$\frac{G}{\delta} = \frac{R_x \delta y^2(1 + y) + R_M N y(1 + y)}{\frac{R_M N}{W\delta} + y}. \tag{7}$$

We divide (7) by $R_M N$

$$\frac{G}{\delta R_M N} = \left(\frac{R_x \delta W \delta y^2(1 + y)}{R_M N R_M N} + \frac{W \delta y(1 + y)}{R_M N}\right)\frac{1}{1 + \frac{W\delta y}{R_M N}}.$$

Let $\frac{G}{\delta R_M N} = g$, $\frac{R_x}{R_M} = A$, $\frac{W}{R_M} = C$, and $\frac{\delta}{N} = \frac{N_d - N_a}{N_{Mn}}$. Then

$$g = \frac{ACF^2 y^2(1 + y) + CFy(1 + y)}{1 + CFy}, \tag{8}$$

where $F = \frac{N_d - N_a}{N_{Mn}} = 1.9 \times 10^{-2}$ for the p-type GaAs:Mn.

The value g characterizes the excitation power of the p-type GaAs:Mn under circularly polarized excitation provided by a titanium-sapphire laser.

Next, we divided expression (4) by δ

$$\frac{q}{\delta} = \frac{p}{\delta} \times \frac{\left(1 - \frac{R_M N}{W\delta}\right)}{\left(\frac{R_M N}{W\delta} + \frac{p}{\delta}\right)}.$$

Let $\frac{q}{\delta} = z$, then $z = y\frac{(CF-1)}{1+CFy}$, where z is the function of filling of manganese ions with the valence band holes. The concentration of the photoexcited holes on manganese is

$$q = \delta y \frac{(CF - 1)}{1 + CFy}. \tag{9}$$

IV. COMPARISON OF EXPERIMENTAL WITH THEORETICAL RESULTS

The rate equations above provide Eqs. (8) and (9) which relate $q(y)$ and $g(y)$ through the parameter y. Eliminating the parameter y provides the explicit $q = q(g)$ dependence. Substitution of $q = q(g)$ into $\tau_s(q)$ yields the function $\tau_s = \tau_s(g)$, which allows for comparison with the experimental data. Next, we complete those steps in detail.

In order to obtain a simple analytic function, $\tau_s(g)$, we approximate Eq. (8) into

$$g = \frac{y(1 + y)}{g_1 + y}, \tag{10}$$

where $g_1 := \frac{1}{CF}$ is a parameter characterizing the relation between the frequency of electron recombination from the conduction band to manganese levels and the frequency of hole recombination from the valence band to manganese levels. $Fy \ll 1$ has been used utilizing the fact that F is of the order of 1/100 for the p-type GaAs:Mn. Solving Eq. (9) with respect to y yields $y = q\frac{g_1}{\delta(1-g_1)-q}$. Substituting $y(q)$ into Eq. (10) and solving for q yields

$$q = \frac{\delta}{2}\left(1 + g - \sqrt{1 + g(2g_1 + g - 2)}\right). \tag{11}$$

With Eq. (11), we selected the root of the quadratic equation, which guarantees that the concentration of photoexcited holes in the absence of excitation power is zero $q(g = 0) = 0$. Note that in the limit of very large excitation power, this solution also guarantees that the concentration of photoexcited holes levels off at the equipartition value $q(g \rightarrow \infty) \rightarrow \frac{\delta}{2}$.

We substitute $q(g)$ from Eq. (11) into $\tau_s = \tau_s(0)$ $(1 + bq)^2$ and obtain

$$\tau_s = \tau_s(0)\left(1 + \left(\sqrt{\frac{\tau_s(\infty)}{\tau_s(0)}} - 1\right)\right.$$
$$\left. \times \left(1 + g - \sqrt{1 + g(2g_1 + g - 2)}\right)\right)^2. \tag{12}$$

Equation (12) becomes the final fitting function when introducing the scale-factor λ according to $g = \lambda P$ to fit the experimental relaxation times vs. excitation power P. $\tau_s(\infty)$ and $\tau_s(0)$ are the relaxation times in the limit of very large and small excitation power. Extrapolating them from the experimental data shown in Fig. 2 (circles) yields $\tau_s(\infty) \approx 149.1$ ns, $\tau_s(0) \approx 12.4$ ns and allows us to fit the data with just two free-fitting parameters g_1 and λ. Fig. 2 (line) shows the result of the best fit, which determines the fit parameters to $g_1 = 0.93 \pm 0.015$ and $\lambda = 0.38 \pm 0.03$ 1/mW. Our simple approach is capable of capturing the essentials of the τ_s vs. P dependence. Any remaining minor derivations of the fit from the data are reasonable when considering the approximations involved, most notably the potential excitation power dependence of τ_c, which has been neglected for simplicity.

The agreement between the experimental $\tau_s(P)$ and the fit of Eq. (12) demonstrates that the increase of the

electron-spin relaxation time is indeed related to the concentration of photoexcited holes on manganese under the increase of the excitation power. This confirms that the reduction of effective field fluctuations increases the spin lifetime. The electron spin relaxation time in the p-type GaAs:Mn is prolonged under the filling of the manganese ions with the valence band holes.

The maximum value of experimental electron spin relaxation time reaches approximately 150 ns instead of 160 ns, as was stated by the authors in Ref. 11. We extracted this data from their experimental logarithmic plot of electron spin relaxation time vs. excitation power. The exact value of maximum electron spin relaxation time is 149.06 ns.

V. CONCLUSION

We used a rate-equation approach to study the electron spin relaxation time on optical pumping intensity in a partially compensated acceptor semiconductor GaAs:Mn. Assuming a relationship between the concentration of photoexcited holes and the spin relaxation time, we discovered significant agreement between the theoretical dependence of the concentration of photoexcited holes on the excitation power and the experimental dependence of the electron spin relaxation time on the excitation power. Our investigation confirms that antiferromagnetic coupling between the spins of the photoexcited holes and the spins of Mn ions gives rise to an atypically long electron spin relaxation time in the p-type GaAs:Mn with a manganese concentration of $7.83 \times 10^{17} \mathrm{cm}^{-3}$, reaching approximately 150 ns.

[1]J. Fabian, A. Matos-Abiague, C. Ertler, P. Stano, and I. Žutić, Acta Phys. Slovaca **57**, 565 (2007).
[2]D. D. Awschalom and M. E. Flatté, Nat. Phys. **3**, 153 (2007).
[3]R. Jansen, Nature Mater. **11**, 400 (2012).
[4]M. Farle and H. Zabel, *Magnetic Nanostructures: Spin Dynamics and Spin Transport*, Springer Tracts in Modern Physics Vol. 246 (Springer, New York, 2013).
[5]T. Dietl, Nature Mater. **9**, 965 (2010).
[6]T. Jungwirth, J. Sinova, J. Mašek, J. Kučera, and A. H. MacDonald, Rev. Mod. Phys. **78**(3), 809 (2006).
[7]B. Endres *et al.*, Nat. Commun. **4**, 2068 (2013).
[8]R. I. Dzhioev *et al.*, Phys. Solid State **39**, 1765 (1997).
[9]J. M. Kikkawa and D. D. Awshalom, Phys. Rev. Lett. **80**, 4313 (1998).
[10]R. I. Dzhioev *et al.*, JETP Lett. **74**, 182 (2001).
[11]G. V. Astakhov *et al.*, Phys. Rev. Lett. **101**, 076602 (2008).
[12]H. Munekata, H. Ohno, S. von Molnár, A. Segmüller, L. L. Chang, and L. Esaki, Phys. Rev. Lett. **63**(17), 1849 (1989).
[13]M. I. Dyakonov and V. I. Perel, Sov. Phys. JETP **38**, 177 (1974).
[14]*Optical Orientation*, edited by F. Meyer and B. P. Zakharchenya (North-Holland, Amsterdam, 1984).