Spin dynamics of semiconductor electrons in the hybrid ferromagnet/semiconductor structure

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Submitted 11 December 2002
Resubmitted 20 January 2003

We analyze the recent experimental study by R. J. Epstein et al. [Phys. Rev. **B65**, 121202 (2002)] on spin dynamics of semiconductor electrons in the hybrid ferromagnet/semiconductor structure by using a simple model based on the Bloch equations. A comparison between the model calculations and the experimental observations shows that the spin relaxation rate is strongly anisotropic. We interpret this anisotropy as a manifestation of the exchange interaction between metallic and semiconductor electrons at the ferromagnet/semiconductor interface.

PACS: 42.50.Md, 72.25.–b, 75.70.–i

In recent years, much interest has been aimed at the physics of electron spin dynamics in bulk semiconductors and heterostructures [1]. An understanding of spin relaxation mechanisms is important because of potential use of spin degrees of freedom in magnetoelectronics and quantum computation [2].

A hybrid ferromagnet/semiconductor (F/S) structure is often considered as a key element of magnetoelectronics. Usually, when considering the transport of spin-polarized electrons from a ferromagnetic metal into a semiconductor, it is tacitly assumed that the spin relaxation rate in the semiconductor is independent of the proximity of the ferromagnet. In most cases it is a good approximation, but it is questionable for samples in which the exchange coupling between metal and semiconductor electrons reaches a noticeable magnitude. It is well known that an exchange interaction is able to provide an efficient channel for the spin relaxation. The BAP mechanism of the spin relaxation in semiconductors is the well known example [3].

In the recent experiment [4] it was observed by the method of time-resolved Faraday rotation that in the hybrid F/S structures, namely, Fe/n–GaAs and MnAs/n–GaAs, the proximity of the ferromagnetic metal affects the dynamical behavior of the spin polarization of semiconductor electrons. It was found that in an external in-plane magnetic field a short circularly polarized (CP) or linearly polarized (LP) pump laser pulse, with energy tuned near the GaAs band gap, induces the component of the spin polarization of semiconductor electrons $S_x\ (t)$:

$$S_x(t) = A_0 e^{-t/T_2} \cos(\omega t + \phi), \qquad (1)$$

where A_0 is the amplitude, T_2 is the effective transverse spin lifetime, $\omega = g\mu_B B/\hbar$ is the Larmor precession frequency, ϕ is the phase of the spin precession and the x axis is directed along the sample normal. In the case of CP pump pulse, which creates spin polarized (optically oriented) along a pump path electrons, the well known [5] result with $\phi = 0$ was reproduced. Although a LP pump creates spin unpolarized carries, the spin oscillations given by Eq. (1) with $\phi \approx -90^\circ$ were also observed. This phenomenon was qualitatively explained in Ref. [4], as manifestation of the exchange interaction between the ferromagnet and semiconductor electrons . Despite the absence of the spin polarization in the semiconductor just after the arrival of a pump pulse, it arises due to the interlayer exchange interaction and oscillates with the Larmor frequency ω for several nanoseconds (to be exact, we note that the term "spin interaction" rather than "exchange interaction" was used in Ref. [4]).

Although the physical origin of the observed effect was justified in Ref. [4] in sufficient detail, many features of the effect remain unexplained. First, we should understand why the oscillation frequency ω is determined solely by the external magnetic field $\mathbf{B}$ and unaffected by the exchange interaction, whereas the amplitude A_0 of the spin oscillations varies (in the case of LP pump) with the angle α between $\mathbf{B}$ and ferromagnet's magnetization $\mathbf{M}$ as $\sin\alpha$. Second, it should be explained why the spin relaxation time for a LP pump ($\sim$ 4 ns) differs from that for a CP pump ($\sim$ 2 ns). Finally, the obtained spin relaxation times are comparable by the order of magnitude with those obtained for n-GaAs without ferromagnetic layer [6]. This looks so that despite the pronounced ex-

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change interaction between the metal and semiconductor electrons, this interaction has little effect on the spin relaxation rate. Of coarse, we keep in mind that the spin relaxation rate is strongly influenced by impurities and defects and, as a consequence, is sample dependent, so that the comparison between different samples should be made with care. Nonetheless, the experiment [4] raises a question about the influence of the exchange interaction at the F/S interface on the spin relaxation rate of semiconductor electrons.

In this paper we consider the spin dynamics of semiconductor electrons in the vicinity of the F/S interface and analyze the experimental observations of Ref. [4] by using the semi-phenomenological Bloch equations. In accordance with the conditions of the experiment, we assume the external magnetic field $\mathbf{B}$ and the ferromagnet's magnetization $\mathbf{M}$ to be parallel to the F/S interface. We choose the x and y axes to be directed along the interface normal and the magnetization $\mathbf{M}$, respectively (see Ref. [4] for details). The thickness of the n-GaAs layer (100 nm) in the heterostructure used in the experiment was much less than the laser wavelength ($\approx$ 820 nm). For this reason, we neglect the variation of the spin polarization $\mathbf{S}(x,t)$ of semiconductor electrons across the layer and replace it some effective value $\mathbf{S}(t)$. Next, we take into account the exchange interaction between the metal and semiconductor electrons by means of an effective exchange field $\mathbf{B}_{ex}$, which is parallel (or antiparallel) to $\mathbf{M}$ and independent of x. As a consequence of these simplifications, we obtain a model, in which homogeneous spin polarization $\mathbf{S}(t)$ varies under the action of the external magnetic field $\mathbf{B}$ and the exchange field $\mathbf{B}_{ex}$ from an initial value $\mathbf{S}_0$ to the equilibrium one $\mathbf{S}_e = \chi(\mathbf{B}_{ex} + \mathbf{B})/g\mu_B$, where χ is the magnetic susceptibility, g is the electron g factor and μ_B is the Bohr magneton. This model is rather crude and can not be applied to the analysis of those properties of the spin dynamics which depend on the distribution of the spin polarization across the film (see the discussion below). However, we believe that some experimentally observed features of the spin dynamics are determined mostly by the geometrical characteristics of the system, such as the relative orientation of the external and exchange magnetic fields.

The equation of motion for the spin polarization $\mathbf{S}(t)$ is

$$d\mathbf{S}/dt = \mathbf{\Omega} \times \mathbf{S} - \hat{R} \cdot (\mathbf{S} - \mathbf{S}_e), \tag{2}$$

where $\mathbf{\Omega} = g\mu_B(\mathbf{B}_{ex} + \mathbf{B})/\hbar$ is the vector of the electron spin precession frequency and $\hat{R}$ is the relaxation tensor. In Ref. [4] it was shown that the external magnetic field $\mathbf{B}$ is modified by a nuclear hyperfine field $\mathbf{B}_n$. Because $\mathbf{B}_n$ is parallel to $\mathbf{B}$, we assume that the nuclear field is included in $\mathbf{B}$. The form of the relaxation tensor is crucial for the following consideration. We choose the relaxation tensor $\hat{R}$ to be diagonal for a given coordinate system: $R_{ij} = \delta_{ij}\Gamma_i$. This assumption physically means that the exchange field $\mathbf{B}_{ex}$ has much larger influence on the spin relaxation than the external magnetic field $\mathbf{B}$. We shall justify this by comparison of our model calculations with the experimental observations. It might seem that the anisotropy of the spin relaxation is redundant here, since the solution of Eq. (2) for $S_x(t)$ with the isotropic relaxation tensor $R_{ij} = \delta_{ij}/T_2$ has the required form, Eq. (1). However, for such solution $\omega = \Omega \propto |\mathbf{B} + \mathbf{B}_{ex}|$ that contradicts to the experiment. Moreover, as we shall show, for a LP pump ($\mathbf{S}_0 = 0$) and isotropic tensor $\hat{R}$, Eq. (2) gives $S_x(t) \equiv 0$. Thus, the anisotropy of the spin relaxation is essential for the explanation of the experimental observations.

We are interested in the experimentally measured component of the spin polarization $S_x(t)$. The anisotropy of the spin relaxation somewhat complicates the solution of Eq. (2) for $S_x(t)$, which differs now from Eq. (1) and has the form

$$S_x(t) = C_1 e^{\lambda_1 t} + C_2 e^{\lambda_2 t} + C_3 e^{\lambda_3 t}, \tag{3}$$

where λ_i are the eigenvalues of the matrix $H_{ij} = e_{ikj}\Omega_k - \Gamma_i\delta_{ij}$, e_{ikj} is the unit antisymmetric tensor of rank three. The coefficients C_i should be determined from the initial conditions for the spin polarization $\mathbf{S}(t)$. Expression (3) for $S_x(t)$ describes both LP and CP pumps, with different initial conditions in either case and, as a consequence, with different coefficients C_i. Assuming $S_x(0) = S_{0,x}$ and $S_y(0) = S_z(0) = 0$ we obtain

$$C_1 = \frac{S_{0,x} D_1 + \Omega_z S_{e,y}(\Gamma_y - \Gamma_z)}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}, \tag{4}$$

where

$$D_1 = \Gamma_x^2 - \Omega^2 + \Gamma_x(\lambda_2 + \lambda_3) + \lambda_2\lambda_3. \tag{5}$$

The coefficients C_2 and C_3 can be obtained from Eqs. (4) and (5) by the cyclic permutations of the indices 1, 2 and 3. The eigenvalues λ_i can be determined from the third order algebraic equation:

$$(\lambda + \Gamma_x)(\lambda + \Gamma_y)(\lambda + \Gamma_z) + \Omega_y^2(\lambda + \Gamma_y) + \Omega_z^2(\lambda + \Gamma_z) = 0. \tag{6}$$

Analytical expressions for the eigenvalues λ_i are rather cumbersome and therefore not given here. In the following, we shall calculate λ_i approximately for particular relations between the spin relaxation rates Γ_i which are relevant to the experiment.

As is seen from Eq. (4), there are two physically different sources for the nonzero spin polarization $S_x(t)$. The first one is well known and originates from the initial spin polarization (optical orientation) $S_{0,x}$ created by a pump pulse. This is the case of CP pump. The second source is not related with the optical orientation and relevant to a LP pump. It comes into play when the external magnetic field is not parallel to the magnetization ($\Omega_z \neq 0$) and, besides, the spin relaxation in the interface plane is anisotropic, that is, $\Gamma_y \neq \Gamma_z$. Physically, this means that under these conditions, the emerging spin polarization $\mathbf{S}(t)$ is not parallel to $\mathbf{\Omega}$, thus creating $S_x(t)$. Our primary interest here lies in the case of LP pump, since it reveals some important properties of the exchange coupling between the ferromagnet and the semiconductor.

A distinctive feature of the spin oscillations induced by LP pump is the dependence of their amplitude A_0 on the angle α between the external magnetic field $\mathbf{B}$ and the exchange field $\mathbf{B_{ex}}$. It was found experimentally that $A_0 \propto \sin\alpha$. Our model calculations also predict some dependence of A_0 on α. As is seen from Eq. (4), the form of this dependence is determined by the product $\Omega_z S_{e,y}$, which, in turn, depends on the relation between the external magnetic field and the exchange field. Since $\Omega_z \propto \sin\alpha$ and the same angular dependence was observed for $S_x(t)$ in the experiment, we conclude that $S_{e,y} = \chi(B_{ex} + B\cos\alpha)$ must be independent of α (within the experimental accuracy). This occurs only if $B_{ex} \gg B$. Thus, the observed dependence of $S_x(t)$ on α indicates that the exchange field is much larger than the external magnetic field ~ 1 T, which was used in the experiment.

Now we turn to the temporal behavior of $S_x(t)$. As we have already note, in general case Eq. (3) can not be reduced to the decaying oscillations, Eq. (1), observed in the experiment. However, we shall show that under special relations between the parameters of the considered system, Eq. (3) leads to almost the same temporal behavior of $S_x(t)$ as Eq. (1) and, at the same time, gives true dependence of the Larmor frequency and the amplitude of the spin oscillations on the external magnetic field. Let us consider this point in more detail. As we consider the oscillatory behavior of the spin polarization $S_x(t)$, two (say, λ_1 and λ_2) of the three eigenvalues in Eq. (3) must be complex and inevitably complex conjugate, $\lambda_1 = \lambda_2^*$. Taking into account that $C_1 = C_2^*$, we conclude that the first two terms in Eq. (3) give the decaying oscillations similar to Eq. (1) with $1/T_2 = -\mathrm{Re}\ \lambda_1 = -\mathrm{Re}\ \lambda_2$, $\omega = \mathrm{Im}\ \lambda_1 = -\mathrm{Im}\ \lambda_2$ and $\phi = \arg C_1$. In order for Eq. (3) can be reduced to Eq. (1), we must have a reason to neglect the third term in Eq. (3). This is possible in two cases. The first one is trivial and takes place if $C_3 = 0$. One can easily see from Eqs. (4)–(6) that this occurs if the in-plane relaxation is isotropic ($\Gamma_y = \Gamma_z$). As we have already noted, this case is not relevant to the experiment. Another situation, when the third term in Eq. (3) can be neglected, occurs if $|\lambda_3| \gg 1/T_2$. Then, this term rapidly approaches to zero with time. In order to find the conditions under which λ_3 is large, we should analyze the dependence of the eigenvalues λ_i on the parameters Γ_i and Ω_i in case of strong anisotropy of the spin relaxation. To this end we compare the behavior of the terms in equation Eq. (6) when Γ_z increases. We notice that the second term in Eq. (6) can be neglected when $\Gamma_z \to \infty$ and then $\lambda_i^{(0)} = \lambda_i(\Gamma_z \to \infty)$ are given by

$$\lambda_{1,2}^{(0)} \approx -\frac{\Gamma_x + \Gamma_y}{2} \pm i\sqrt{\Omega_z^2 - \frac{(\Gamma_x - \Gamma_y)^2}{4}}, \qquad \lambda_3^{(0)} \approx -\Gamma_z. \tag{7}$$

Thus, for a large Γ_z we obtain λ_i with the required properties: λ_3 is large and the Larmor frequency $\omega \approx \Omega_z \propto B_z$. It is this dependence of ω on the external magnetic field was observed in the experiment. Note that the effective spin relaxation rate $1/T_2 = (\Gamma_x + \Gamma_y)/2$ is independent of Γ_z. It follows from Eq. (4) that the coefficients C_1 and C_2 becomes purely imaginary when λ_3 increases. Consequently, Eq. (3) reduces to Eq. (1) with $\phi = \pm 90°$, where the sign, plus or minus, depends on the sign of the exchange field, i.e., on whether the exchange coupling at the interface is ferromagnetic or antiferromagnetic.

In order to obtain restrictions, imposed on the parameters Γ_i and Ω_i, under which $\lambda_i \simeq \lambda_i^{(0)}$ we estimate corrections $\delta\lambda_i$ to $\lambda_i^{(0)}$ for large but finite Γ_z and require that $|\delta\lambda_i| \ll |\lambda_i^{(0)}|$. Substituting $\lambda_i = \lambda_i^{(0)} + \delta\lambda_i$ into Eq. (6) and assuming $\Omega_z \gg \Gamma_x \simeq \Gamma_y$ we obtain $\delta\lambda_3 = -\delta\lambda_{1,2} \simeq \Omega_y^2/\Gamma_z$. The condition $|\delta\lambda_3| \ll |\lambda_3^{(0)}|$ gives $\Gamma_z \gg \Omega_y$ but from the second condition $|\delta\lambda_{1,2}| \ll |\lambda_{1,2}^{(0)}|$ we obtain the more strong inequality $\Gamma_z \gg \Omega_y^2/\Gamma_x$. The last relation is a strict mathematical definition of the term "the strong anisotropy of the spin relaxation" for a given problem.

What temporal behavior of $S_x(t)$ do we obtain if Γ_y rather than Γ_z increases? Interchanging the subscripts y and z in Eqs. (7) we again obtain Eq. (1), but with the Larmor frequency $\omega = \Omega_y \propto B_{ex} + B_y$. This is inconsistent with the experiment, where $\omega \propto B_z$.

Thus, the observation of the oscillations of the spin polarization for a LP pump, given by Eq. (1), indicates that the relaxation rate Γ_z significantly exceeds Γ_x and Γ_y. This fact is not evident from the experimental data, since the rate of decay of the Faraday rotation $1/T_2 = (\Gamma_x + \Gamma_y)/2$ is independent of Γ_z. Hence, the increase of Γ_z leaves T_2 unchanged.

To get a more quantitative characterization of the spin relaxation anisotropy we consider a set of inequalities which are satisfied by Γ_i and Ω_i: (i) $\Gamma_z \gg \Omega_y^2/\Gamma_x$, (ii) $\Omega_y \gg \Omega_z$, and (iii) $\omega \gg 1/T_2$. The first of these inequalities allows us to reduce Eq. (3) to Eq. (1), the second is obtained from the fit of the angular dependence of $S_x(t)$ and the third follows from the experiment, where $\omega \simeq 10$ ns^{-1} at $B = 0.25$ T and $1/T_2 \simeq 0.25$ ns^{-1}. Since the restrictions, imposed on the parameters Γ_i and Ω_i, are determined by the three inequalities (i)-(iii), the lower bound for the relaxation rate Γ_z is allowed to vary over the whole range, which we estimate as $\sim (10^3 \div 10^5)/T_2$ and the corresponding range for the relaxation time $T_z \sim 0.01 \div 1$ ps.

The physical picture of the motion of the spin polarization is very simple. When Γ_z is large, S_z rapidly approaches to its equilibrium value $S_{e,z}$ due to the large damping. This means that $\mathbf{S}(t)$ precesses around the z axis. The frequency of such precession is determined by the z-component of the magnetic field and independent of the exchange field.

So far we considered the case of LP pump. The case of CP pump can be considered quite analogously. Now the coefficients C_i in Eq. (3) should be calculated taking into account that the initial spin polarization $S_{0,x}$ is nonzero, since a CP pump creates spin polarized electrons. Assuming again the validity of Eqs. (7), we obtain that $S_x(t)$ is given by Eq. (1) with the phase ϕ depending on the magnitude of the initial spin polarization $S_{0,x}$. When $S_{0,x}$ increases, then $\phi \to 0$, and Eq. (1) reproduces the well known decaying spin oscillations in a transverse magnetic field [5]. It is these oscillations were observed in Ref. [4] for a CP pump. Though in the case of CP pump Eq. (1) follows from the Bloch equation for the isotropic in-plane relaxation, the frequency ω remains to be dependent on the anisotropy. It was found in the experiment that the oscillation frequencies are the same for both pump polarizations. This means that the anisotropy of the spin relaxation is equally essential for the explanation of the experimental observations for both pump polarizations.

If we compare the relaxation times for LP and CP pumps, we see a distinction between our model calculations and the experimental data. Our calculations give equal relaxation times $T_2 = 2(\Gamma_x + \Gamma_y)^{-1}$ for LP and CP pumps, while the relaxation times measured experimentally differ: $T_2 = 4$ ns for a LP pump and $T_2 = 2$ ns for a CP pump. A possible reason for this discrepancy is the inhomogeneity of the spin polarization combined with the different origin of the spin polarization for LP and CP pumps. As opposed to the case of LP pump, the spin polarization for a CP pump exists and without the exchange coupling due to the optical orientation. This leads to the different profiles of $S_x(x,t)$ across the film for LP and CP pumps. Since the exchange coupling decreases with distance from the interface, the variation of $S_x(x,t)$ with x affects the relaxation rates and, in turn, leads to the different T_2 for LP and CP pumps. Our model fails to describe this effect, since it does not take into account the spatial variations of the exchange coupling and the spin polarization.

In conclusion, we have analyzed experimental results of Ref. [4] on the spin dynamics in the hybrid F/S structure by using semi-phenomenological Bloch equations. It has been shown, that the qualitative agreement between the model calculations and the experimental observations can be achieved only under certain conditions imposed on the parameters of the system. First, we have shown that the exchange field, representing the effect of exchange coupling between the ferromagnet and the semiconductor on semiconductor electrons, significantly exceeds the external magnetic field. Second, the spin relaxation rate of semiconductor electrons near the F/S interface is strongly anisotropic. The spin relaxation rate depends on the orientation of the spin polarization in the interface plane and reaches its maximum when the spin polarization is perpendicular to the ferromagnet's magnetization. In a sense, this anisotropy is hidden, since it weakly affects the rate of decay of the Faraday rotation beating and manifests itself only through the form of the beating. For this reason we have obtained only a rough estimate of the anisotropy. The maximum value of the spin relaxation rate exceeds the bulk one approximately by a factor of $10^3 \div 10^5$. The corresponding relaxation times are of the order of $0.01 \div 1$ ps. Such fast spin relaxation is typical for magnetic semiconductors. This indicates that the exchange coupling between the ferromagnet and the semiconductor provides an additional channel for the spin relaxation. We may speculate that the anisotropy of the spin relaxation is related with the anisotropy of the surface spin excitation spectrum of the ferromagnet. This point requires an additional theoretical and experimental investigations.

The author thanks K. A. Chao for reading the manuscript and criticism. This work was supported by the RFBR and by the programs of Russian Ministry of Science.

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