Dynamic Ferromagnetic Proximity Effect in Photoexcited Semiconductors

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The spin dynamics of photoexcited carriers in semiconductors in contact with a ferromagnet is treated theoretically and compared with time-dependent Faraday rotation experiments. The long-time response of the system is found to be governed by the first tens of picoseconds in which the excited plasma interacts strongly with the intrinsic interface between the semiconductor and the ferromagnet in spite of the existence of a Schottky barrier in equilibrium.

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Magnetoelectronics, the science and technology of using ferromagnets in electronic circuits, is divided into two subfields, metal-based [1] and semiconductor-based magnetoelectronics [2], with little common ground. Metal researchers focus mainly on topics derived from giant magnetoresistance, the large difference in the dc conductance for parallel and antiparallel magnetization configurations of magnetic multilayers, where theoretical understanding has progressed to the stage of materials-specific predictions [3]. The dynamics of the magnetization vectors in the presence of charge and spin currents has recently received a lot of attention [4–7].

Semiconductor-based magnetoelectronics is motivated by the prospect of integrating new functionalities with conventional semiconductor electronics. The emphasis has been on the basic problem of spin injection into semiconductors; theoretical understanding is less advanced, and detailed electronic structure calculations are just starting [8,9]. Unlike metals, semiconductors can be studied by optical spectroscopies such as the powerful time-resolved Faraday or Kerr rotation techniques, in which a selected component of a spin-polarized excitation cloud in the semiconductor can be monitored on ps time scales [10]. In n-doped GaAs, these experiments revealed long spin coherence of the order of μs [10]. When the semiconductor is in contact with a ferromagnet, an initially unpolarized electron distribution prepared using linearly polarized light was found to very quickly acquire a spin polarization—the dynamic ferromagnetic proximity (DFP) effect [11–13]. In turn, DFP efficiently imprints spin information from a ferromagnet onto nuclear spins by dynamic nuclear polarization, opening new options for quantum information storage.

In this Letter we show that metal and semiconductor-based magnetoelectronics can both be understood in terms of coherent spin accumulations. Specifically, the DFP can be treated by the same formalism that successfully describes the dynamics of the magnetization vector in metallic hybrids [6,14]. The experiments can be understood in terms of a time-dependent interaction between the conduction-band electrons and the ferromagnet in a ''fireball-afterglow'' scenario. Photoexcited holes are instrumental in helping the electrons to overcome the Schottky barrier between metal and semiconductor in the first $\lesssim 50$ ps (''fireball'' regime) and induce the proximity effect. The interaction weakens with vanishing hole density, thus preventing fast decay of the created spin accumulation in the ''afterglow.''

Two groups have already contributed important insights into this problem. Ciuti *et al.* [15] interpreted the DFP in terms of a spin-dependent reflection of electrons at a ferromagnetic interface through a Schottky barrier in equilibrium but did not address the time dependence of the problem. Gridnev [16] did investigate the dynamics of the photoexcited carriers but postulated a phenomenological relaxation tensor with a specific anisotropy that we find difficult to justify. We show how both approaches can be unified and extended by *ab initio* magnetoelectronic circuit theory [17–20].

We first summarize the experimental evidence [12,13]. Initially, a $\sim$100 fs pulse with frequency close to the band gap is absorbed by the semiconductor (100 nm of GaAs). The polarization state is then monitored by time-dependent Faraday rotation measurements of the coherent spin precession in an applied magnetic field. The homogeneously excited carriers (fireball) thermalize within a ps, in which the holes also lose any initial spin polarization. The interaction time scale with the ferromagnet (Fe or MnAs) can be deduced from the rise time of the polarization after excitation with a linearly polarized (LP) light pulse to be $\leq$50 ps [12]. For long delay times (afterglow), the spin relaxation is very slow ($>$2 ns), comparable to GaAs reference samples in the absence of a ferromagnet. The sample can be also excited by circularly polarized (CP) light, in which case the fireball is polarized from the outset. Dynamic nuclear polarization (DNP) by the hyperfine interaction can be detected by deviations of the precession frequency from the bare

Larmor frequency, i.e., a modified g-factor [11]. DNP should vanish when the external magnetic field is normal to the spin accumulation. It therefore remains to be explained that Epstein *et al.* [12] observe a modified g-factor for this configuration that differs for LP and CP excitation. Interesting additional information relates to the material dependence, indicating that the polarization induced by Fe is of the opposite sign to that induced by MnAs [12], and to the modulation of the afterglow Larmor frequency by an applied bias [13].

Let us consider a semiconductor (SC) film in which a nonequilibrium electron chemical potential $\langle\mu| = \langle\mu_c, \vec{\mu}_s|$ is excited with charge and spin components μ_c and $\vec{\mu}_s \equiv \langle\mu_x, \mu_y, \mu_z|$ respectively (in energy units). The bilayer parallel to the yz plane (see Fig. 1) consists of a semiconductor in contact with a metallic ferromagnetic film (F) with fixed single-domain magnetization in the direction of the unit vector $\vec{m}$. Because of its relatively huge density of states, a metallic ferromagnet may be treated as a reservoir in equilibrium. A current $\langle I| = \langle I_c, \vec{I}_s|$ (in units of reciprocal time), with charge and spin components I_c and $\vec{I}_s$, respectively, flows through the ferromagnet/semiconductor (F|SC) interface, which is governed by the spin-dependent (dimensionless) conductances $g_{\uparrow\uparrow}$ and $g_{\downarrow\downarrow}$ as well as the complex spin-mixing conductance $g_{\uparrow\downarrow}$ [17]. Physically, the real part of the mixing conductance expresses the angular momentum transfer to and from the ferromagnet, such as the strength of the spin-current induced magnetization torque [4,18] or nonlocal Gilbert damping [14], whereas the imaginary part is an effective magnetic field [21–23]. The microscopic expression for the conductances is Landauer like

$$g_{\sigma\sigma'} = \sum_{nm}[\delta_{nm} - r^{\sigma}_{nm}(r^{\sigma'}_{nm})^*], \tag{1}$$

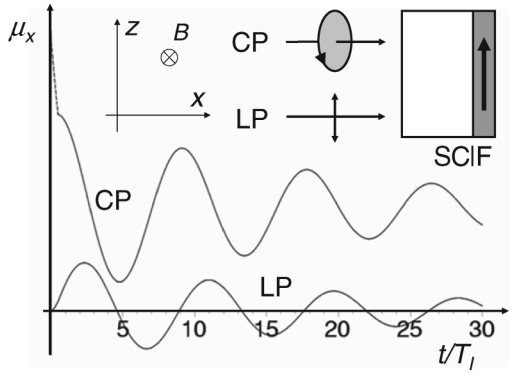

FIG. 1. Spin dynamics in the fireball-afterglow scenario of an excited semiconductor in proximity with a ferromagnet polarized in the z direction. A (DNP enhanced) magnetic field 0.24 T is applied in the y direction. Plotted is $\mu_x(t)$ in arbitrary units for CP (shifted upwards by 0.6) and LP excitation, with the wave vector in the x direction. Time is measured in units of T_I. The transition from fireball to afterglow with $\breve{T}_I = 10T_I$ is taken to be abrupt at $t = T_I/2$.

where the reflection coefficient r^{σ}_{nm} of an electron in the SC with spin σ at the SC|F contact between nth and mth transverse modes is accessible to *ab initio* calculations [8,9,19,20]. The time dependence of the system is governed by charge and spin conservation [17]:

$$-2h\mathcal{D}\left(\frac{d\mu_c}{dt}\right)_{\text{bias}} = (g_{\uparrow\uparrow} + g_{\downarrow\downarrow})(\mu_c - e\varphi) + (g_{\uparrow\uparrow} - g_{\downarrow\downarrow})(\vec{m}\cdot\vec{\mu}_s), \tag{2}$$

$$-2h\mathcal{D}\left(\frac{d\vec{\mu}_s}{dt}\right)_{\text{bias}} = 2\text{Re}g_{\uparrow\downarrow}\vec{\mu}_s - 2\text{Im}g_{\uparrow\downarrow}(\vec{m}\times\vec{\mu}_s) + [(g_{\uparrow\uparrow} - g_{\downarrow\downarrow})(\mu_c - e\varphi) + (g_{\uparrow\uparrow} + g_{\downarrow\downarrow} - 2\text{Re}g_{\uparrow\downarrow})(\vec{m}\cdot\vec{\mu}_s)]\vec{m}, \tag{3}$$

where $\mathcal{D}$ is the SC single-spin energy density of states. The electrostatic potential φ due to an applied bias and/or a charge imbalance between electrons and holes will be disregarded in the following (see below). In the presence of a magnetic field $\vec{B}$, the sum of externally applied and hyperfine (Overhauser) fields with ordered nuclear spins, we have to add

$$\left(\frac{d\vec{\mu}_s}{dt}\right)_{\text{field}} = \frac{g_e\mu_B}{\hbar}\vec{B}\times\vec{\mu}_s, \tag{4}$$

where g_e is the electron g-factor (≈ -0.4 in GaAs) and μ_B is the Bohr magneton. These equations can be summarized in terms of a 4×4 matrix equation:

$$-T_I\frac{d|\mu\rangle}{dt} = \mathbf{\Gamma}|\mu\rangle, \tag{5}$$

where $T_I = 2h\mathcal{D}/g$ is an interface-mediated relaxation time in terms of the total conductance $g = g_{\uparrow\uparrow} + g_{\downarrow\downarrow}$. Choosing $\vec{m}$ parallel to the z axis,

$$\mathbf{\Gamma} = \begin{pmatrix} 1 & 0 & 0 & p \\ 0 & \eta_r & \eta_i + \Omega_z & -\Omega_y \\ 0 & -\eta_i - \Omega_z & \eta_r & \Omega_x \\ p & \Omega_y & -\Omega_x & 1 \end{pmatrix}. \tag{6}$$

$|\Omega_\alpha|/T_I = |g_e|\mu_B B_\alpha/\hbar$ is the Larmor frequency, the mixing conductance has been normalized as $\eta = 2g_{\uparrow\downarrow}/g$ with subscripts i and r denoting its real and imaginary part, and the polarization is defined as $p = (g_{\uparrow\uparrow} - g_{\downarrow\downarrow})/g$. Equation (5) can be solved easily [16] for the boundary conditions corresponding to LP excitation $\langle\mu^{\text{LP}}(0)| = \langle 1, 0, 0, 0|$ or CP excitation with wave vector in the x direction $\langle\mu^{\text{CP}}(0)| = \langle 1, 1, 0, 0|$.

When the conductance is expressed in terms of an interface transparency parameter κ times the intrinsic SC single-spin Sharvin conductance of a ballistic constriction of area A, $g = \kappa(2\pi m^*\varepsilon A/h^2)$, we can write

$$T_I \approx \frac{3.5}{\kappa}\frac{L_{\mathrm{SC}}}{100\ \mathrm{nm}}\sqrt{\frac{m^*}{0.067m_0}\frac{10\ \mathrm{meV}}{\varepsilon}}\ \mathrm{ps}. \quad (7)$$

For an Ohmic interface with $\kappa \simeq 0.1$, an SC layer thickness of $L_{\mathrm{SC}} = 100$ nm and at a characteristic electron kinetic energy (depending on doping and excitation density) of $\varepsilon = 10$ meV, the time constant for GaAs (effective mass $m^* = 0.067m_0$) is $T_I \sim 35$ ps, of the order of the experimental rise time of the proximity effect [12]. At long-time scales, experiments find $\check{T}_I > 2$ ns, which corresponds to a strongly reduced transparency of $\check{\kappa} \lesssim 0.002$, introducing the embellishment $\check{}$ to indicate parameters in the long-time regime.

With a few exceptions (notably InAs), Schottky barriers are formed at metal-semiconductor interfaces when interface states in the gap of the semiconductor become filled giving rise to space charges. Photoexcited holes are strongly attracted by the barrier, thereby dragging the electrons with them [24] and/or screen the barrier. The observed large κ in the fireball regime reflects the facilitation of electron transport to the SC|F interface by the holes. In the limit of predominant ambipolar electron-hole transport (and in the absence of an applied bias), which should apply for the low doping densities in the experiments, we may neglect the electrostatic potential φ, but the electron conductance g might be affected by the scattering of the holes.

At long time scales, the holes disappear into the ferromagnet or recombine with electrons in the semiconductor, but the electron spin accumulation persists [15]. In this afterglow, remaining space charges vanish when the sample is earthed and net charge transport is suppressed. Equation (2) thus vanishes and

$$-\check{T}_I \frac{d\vec{\mu}_s}{dt} = \check{\mathbf{\Gamma}}_s \vec{\mu}_s \quad (8)$$

with

$$\check{\mathbf{\Gamma}}_s = \begin{pmatrix} \check{\eta}_r & \check{\eta}_i + \check{\Omega}_z & -\check{\Omega}_y \\ -\check{\eta}_i - \check{\Omega}_z & \check{\eta}_r & \check{\Omega}_x \\ \check{\Omega}_y & -\check{\Omega}_x & 1-\check{p}^2 \end{pmatrix}, \quad (9)$$

where the parameters are now governed by the full Schottky barrier.

The kinetic equations are valid when the system is diffuse or chaotic (as a result of interface roughness or bulk disorder). The ferromagnetic elements should have an exchange splitting Δ which is large enough that the magnetic coherence length $\lambda_c = \hbar/\sqrt{2m\Delta} < \min(\ell_F, L_F)$, where L_F is the thickness of the ferromagnetic layer (typically 50 nm) and ℓ_F is the mean free path. These conditions are usually fulfilled in hybrid systems except for very thin layers with nearly perfect interfaces. The spin relaxation time in GaAs is taken to be very long. We also require that $L_{\mathrm{SC}} < \sqrt{2\hbar v_{\mathrm{F}}\ell_{\mathrm{SC}}/3g_e\mu_B B_{\mathrm{ext}}}$ for diffuse systems [21] or $L_{\mathrm{SC}} < \hbar v_{\mathrm{SC},f}/g_e\mu_B B_{\mathrm{ext}}$ for ballistic systems, where ℓ_{SC} is the mean free path, $v_{\mathrm{SC},f}$ is the Fermi velocity, and B_{ext} is the externally applied field. For samples with $L_{\mathrm{SC}} = 100$ nm the applied magnetic fields should therefore not much exceed 5 T.

Thus a microscopic justification can be given for Gridnev's phenomenological relaxation tensor [16]. But whereas we can explain how the longitudinal relaxation [$\sim g$ or $\sim\check{g}(1-\check{p}^2)$] can differ from the transverse components [$\sim \mathrm{Re}g_{\uparrow\downarrow}$ or $\sim\mathrm{Re}\check{g}_{\uparrow\downarrow}$], Gridnev postulated a large difference between the two components *normal* to $\vec{m}$. Such large magnetic anisotropies can be excluded for Fe and the scenario sketched by Gridnev cannot be a generic explanation for all experiments. To explain the proximity polarization, Gridnev's 3×3 Bloch equation *must* be extended to the 4×4 kinetic Eq. (5) that includes a charge current component.

First-principles calculations of the bare interface conductance (with the Schottky barrier assumed quenched) provide a first indication of the transport properties in the first tens of ps. We choose here the Fe|InAs system, which apart from the Schottky barrier, is very similar to Fe|GaAs, and as such, of great interest in itself. Table I summarizes results obtained from first-principles scattering matrix calculations [9]. We find the reversal of polarization sign with disorder as noted before [9], which may explain the negative polarization found for Fe|GaAs [12]. The real part of the mixing conductance η_r is close to unity similar to metallic interfaces, but in contrast to these, the imaginary part η_i is strongly enhanced. The latter can be explained by the focus on a small number of states with wave vectors close to the origin, which prevents the averaging to zero found in metals [20].

We now model the experiments of Epstein *et al.* on GaAs|MnAs (concentrating on Fig. 1 in [12]) with an applied magnetic field of 0.12 T in the y direction (in our coordinate system). Analytic solutions of Eq. (5) can be obtained for $\eta_r = 1 (= \check{\eta}_r)$ (see Table I and [20]) such that all modes are exponentially damped by a single interface relaxation time T_I. Time is measured in units

TABLE I. *Ab initio* interface transport parameters for a clean InAs|Fe (001) interface with In (or As) termination. G is the electric conductance e^2g/h, p is the polarization, κ is the ratio between G and the intrinsic SC Sharvin conductance $[5.2\times10^{-5}/(\mathrm{f}\Omega\,\mathrm{m}^2)]$, η is the relative mixing conductance, all at a kinetic energy of 20 meV in the InAs conduction band. The dirty interfaces are modeled as a monolayer of random alloy with 1/4 of the interface In (or As) replaced by Fe in a 7×7 lateral supercell.

		$G[1/(\mathrm{f}\Omega\,\mathrm{m}^2)]$	p	κ	$\mathrm{Re}\eta$	$\mathrm{Im}\eta$
Clean	Fe|InAs	1.5×10^{-5}	0.98	0.14	1.3	−1.3
	Fe|AsIn	3.6×10^{-5}	0.88	0.35	1.6	−1.05
Dirty	Fe|InAs	5.7×10^{-5}	−0.29	0.56	1.1	−0.18
	Fe|AsIn	7.4×10^{-5}	−0.22	0.71	1.3	−0.30

of T_I and the polarization is chosen to be $p = 1(= \check{p})$. In the fireball regime the quality factor $\Omega_y \ll 1$, and we adopt $\eta_i = -1(= \check{\eta}_i)$. For LP excitation the charge component relaxes in favor of the z component polarized along the magnetization direction $2\mu_z(t) = e^{-(1-p)t/T_I} - e^{-(1+p)t/T_I}$ for $\Omega_y = 0$, which is the essence of the DFP effect. The time scale on which the Schottky barrier recovers determines (together with p) the modulus of the spin accumulation in the afterglow. It is of the same order, but smaller, than T_I because of competing electron-hole recombination in the semiconductor. Our Fig. 1 is similar to Fig. 1 in [12], but additional experimental data on a short time scale are required to guide the development of a more refined model.

As mentioned above, a modified Larmor frequency has been observed [12] even when the photon wave vector is normal to the field. This is at odds with the notion that a CP excited spin accumulation should rotate around the field without net angular momentum transfer to the nuclei. This *could* be evidence for a DFP effect for the CP configuration. In the fireball, a significant η_i acts like a magnetic field in the z direction, causing the initial spin ensemble to precess into the direction of the external magnetic field, which is then able to polarize the nuclear spins. This effect is weaker for LP excitation since, in the brief fireball interval, any spin accumulation has to be generated before it can precess.

For LP excitation the Larmor frequency depends [13] on an applied bias, proving that the electrostatic potential φ, in general, cannot be neglected in Eqs. (2) and (3). This does not invalidate our qualitative arguments since, compared to the bare interface exchange the modifications needed to explain the shifts in the effective Larmor frequencies are small, beyond the accuracy of our model. The decreasing spin lifetime in the afterglow with increasing bias has been explained by inhomogeneous nuclear polarization [13], but lowering the Schottky barrier by a forward bias also reduces $\check{T}_I$.

In conclusion, we propose a physical picture for the spin dynamics of photoexcited carriers in semiconductor/ferromagnet bilayers. The experiments can be understood in terms of at least two time scales. In the first 50 ps or so, the photoexcited carriers screen the Schottky barrier efficiently and the interaction of the electrons with the ferromagnet is described by nearly intrinsic interface conductances that can be calculated from first principles. After delay times of > 100 ps, the Schottky barrier protects the semiconductor carriers from fast decay and any residual exchange interaction is very weak. More insight into the interaction of carriers in semiconductors with ferromagnets could be gained by a faster (ps) time resolution and higher applied magnetic fields. A quantitative explanation of the experiments requires self-consistent modeling of the combined electron and hole carrier dynamics as well as *ab initio* calculations of the interface scattering matrices for electrons and holes.

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