Unified Description of Bulk and Interface-Enhanced Spin Pumping

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We describe a mechanism for generating nonequilibrium electron-spin accumulation in semiconductors or metals by rf magnetic field pumping. With a semiclassical model we show that a rotating applied magnetic field (or the precessing magnetization inside a weak ferromagnet) generates a dc spin accumulation. For bulk systems this spin accumulation is in general given by a small fraction of $\hbar\omega$, where ω is the rotation or precession frequency. With the addition of a neighboring, field-free region, and allowing for the diffusion of spins across the interface, the spin accumulation is dramatically enhanced towards $\hbar\omega$ near the interface. The interface-enhanced spin accumulation obtained within our bulk-oriented model is surprisingly similar to predictions based on interface-scattering theory [A. Brataas *et al.*, Phys. Rev. B **66**, 060404(R) (2002)].

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A new direction in the field of spin electronics concerns the interaction between spin currents and the magnetization dynamics of a ferromagnetic electrode into or out of which the spin currents flow. In spin-pumping devices [1–3], electron spins are pumped from a precessing ferromagnet (driven by a resonant rf magnetic field), into an adjoining nonmagnetic metal, without accompanying charge currents. The so-called spin battery [2] is based on the interface spin-mixing conductance [4], which describes the coherent reflection and spin rotation of electrons within an exchange length (typically a few nanometers in conventional ferromagnets) of the interface. It has been experimentally verified by measuring the effect of adjoining metal layers on the Gilbert damping constant of a ferromagnetic layer [5,6] due to the transfer of angular momentum away from the ferromagnetic and into adjacent metals [7]. For the spin battery, under specific conditions the universal value of $\hbar\omega$ is obtained for the spin accumulation, where ω is the precession frequency of the magnetization. On the other hand, a bulk-oriented approach based on mean-field theory [8] has been used to relate the intrinsic Gilbert damping to the spin relaxation processes resulting from the generation of nonequilibrium spin accumulation in the ferromagnet.

We present a new mechanism for bulk and interface-enhanced spin pumping. In the model, spin pumping is treated fundamentally as a bulk mechanism, and we find that dc spin accumulation can be generated inside nonmagnetic materials with an external, rf-frequency, rotating magnetic field. This provides a new technique for generating large spin accumulation values that involves neither charge currents nor ferromagnetic sources. We also examine a hybrid system, in which spins are allowed to diffuse from a "pumped" region into a neighboring region without pumping fields, and find the spin accumulation is dramatically enhanced near the interface. We compare our results for the hybrid system to the predictions for the spin battery by Brataas *et al.* [2] (who use a fundamentally different theory, based on interface scattering) by employing a simple exchange-field model of a weak ferromagnet, and find comparable spin accumulation values near the interface.

Our approach is based on classical dynamics of spin ensembles in a diffusive system. We start with Bloch-type equations written in terms of the electrochemical potentials for the three spin directions, $\vec{f} = (f_x, f_y, f_z)$ that describe the space- and time-dependent, nonequilibrium spin accumulation $\vec{f}(x, t)$ [9]. Here we will restrict the description to one spatial dimension x. The basis of our description is the following equation for the dynamics of $\vec{f}(x, t)$ in a rotating magnetic field $\vec{B}(x, t)$ [10,11]:

$$-\frac{\partial \vec{f}}{\partial t} + \vec{I}(x, t) = -D\nabla^2 \vec{f} + \frac{\vec{f}}{\tau} - \frac{g\mu_B}{\hbar}\vec{B} \times \vec{f}, \qquad (1)$$

where D is the diffusion constant and $\vec{I}(x, t)$ is a source term which will be described in the next paragraph. The length scales relevant to the problem are the spin diffusion length $\lambda = \sqrt{D\tau}$, the Larmor precession length $\lambda_B = \sqrt{2\pi D/|\vec{\omega}_B|}$, where $\hbar\vec{\omega}_B = g\mu_B\vec{B}$, and a length $\lambda_\omega = \sqrt{2\pi D/\omega}$ corresponding to the distance the spins can diffuse in one period $\frac{2\pi}{\omega}$. Equation (1), with the left hand side equal to zero, has been shown to give an accurate description of spin dynamics in metallic systems for time-independent magnetic fields [11,12].

The key new ingredient of our approach is the source term:

$$\vec{I}(x, t) = -g\mu_B \frac{\partial}{\partial t}\vec{B}(x, t). \qquad (2)$$

It describes the rate at which the oscillating magnetic field, via the Zeeman energy, pushes spins aligned with the magnetic field below the Fermi level, and antialigned spins above the Fermi level. This mechanism can be seen as a source of locally injected, time-dependent spin currents [13]. If one considers a magnetic field rotating in a plane with angular frequency ω, then the in-plane spin accumulation reflects the balance between the injection rate [via Eq. (2)] and the relaxation rate. If there were no diffusion or precession, then the spin accumulation would follow the field, but out of phase by an angle $\phi = \tan^{-1}(\omega\tau)^{-1}$. Precession of this spin accumulation around the field will generate a component of spin accumulation perpendicular to the plane of rotation.

Figure 1(a) shows the basic field configuration we will consider: a rotating magnetic field B_{xy} and a perpendicular, static field B_z. The field vector is $\vec{B} = (B_{xy}\cos\omega t, B_{xy}\sin\omega t, B_z)$ in Cartesian coordinates. We first consider a system that is formed by region I *alone* [Fig. 1(b)]; i.e., a uniform (spatially invariant) system without boundaries. Dropping the diffusion term and rewriting Eq. (1) in a more concise form:

$$\tau\dot{\vec{f}} = -\vec{f} + \vec{\omega}_B\tau \times \vec{f} - \hbar\dot{\vec{\omega}}_B\tau, \tag{3}$$

where $\vec{\omega}_B = (\omega_{xy}\cos\omega t, \omega_{xy}\sin\omega t, \omega_z)$. Since the induced spin accumulations f_x and f_y will, in the steady state, oscillate with the same frequency as B_x and B_y, it will be advantageous to transform to a coordinate system in which the x and y axes rotate about the z axis with angular frequency $\vec{\omega} = \omega\hat{z}$, so that $\vec{B} = (B_{xy}, 0, B_z)$. The time derivative of a vector $\vec{r}$ in the stationary frame transforms as $(\dot{\vec{r}})_{\text{lab}} \Rightarrow (\dot{\vec{r}})_{\text{rot}} + \vec{\omega} \times (\vec{r})_{\text{rot}}$. Using this relation, Eq. (3) now becomes

$$\tau\dot{\vec{f}} = -\vec{f} + (\vec{\omega}_B - \vec{\omega})\tau \times \vec{f} - \hbar(\dot{\vec{\omega}}_B + \vec{\omega} \times \vec{\omega}_B)\tau, \tag{4}$$

In the last term, we have $\dot{\vec{\omega}}_B = 0$ in this frame and $\vec{\omega} \times \vec{\omega}_B = (0, \omega\omega_{xy}, 0)$. For the stationary analysis we set $\dot{\vec{f}} = 0$, and obtain the result [14]:

$$f_z = -\frac{(\omega_{xy}\tau)^2}{1 + (\omega_{xy}\tau)^2 + ((\omega_z - \omega)\tau)^2}\hbar\omega. \tag{5}$$

The other components are related to f_z: $f_x = -\frac{(\omega_z - \omega)}{\omega_{xy}} f_z$ and $f_y = (\omega_{xy}\tau)^{-1} f_z$.

With Eq. (5), we predict that a rotating magnetic field will produce dc spin accumulation perpendicular to the plane of rotation, which will be, in general, a small fraction of the universal value $\hbar\omega$. The value is supressed by the Larmor precession around the rotating field (ω_{xy}) as well as around the effective z field ($\omega_z - \omega$). The suppression from the effective z field can be removed when in resonance, $\omega_z = \omega$, in which case the full universal value is achieved in the limit of long relaxation time, such that $(\omega_{xy}\tau)^2 \gg 1$. Experimentally, this means that in a material with long spin relaxation times even small rotating magnetic fields on the order of milli-Tesla can produce $\hbar\omega$ spin accumulation, on the order of micro-electron volts at gigahertz frequencies.

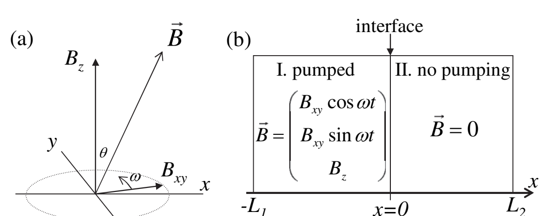

FIG. 1. (a) Magnetic field vector diagram. An applied rf field B_{xy} rotates with angular frequency ω in the x-y plane. A static magnetic field B_z is applied perpendicular to B_{xy} generating a field vector $\vec{B}$ that rotates around B_z with cone angle θ. (b) The hybrid system with an interface at $x = 0$. The fields just described are applied in the pumped region I, $x < 0$. $\vec{B} = 0$ in region II, $x > 0$.

We now turn to discussing the hybrid system of Fig. 1(b): the rotating field is only applied in the space $x < 0$ (the pumped region I), whereas for $x > 0$ there are no magnetic fields (region II). We will consider two physical realizations for the pumped region I: spin pumping in a nonmagnetic material with an applied, rotating magnetic field of magnitude $B_{xy} = 10$ mT, and $B_z = 0$ (both region I and II are nonmagnetic conductors); and, in order to make a connection with conventional spin-pumping theories, we utilize a simple model of a weak ferromagnet with $B_{xy} = 1$ T and $B_z = 100$ T representing the exchange interaction corresponding to small angle magnetization precession around an easy axis (region II is a nonmagnetic conductor). This exchange interaction approach, in which the magnetization of the localized d moments couples to the s electrons and polarizes them, has been used in early spin-pumping theory [15] (which was applied to transmission electron-spin resonance), and more recently in the mean-field-theory description of Gilbert damping [8,16].

The system is solved as a boundary-value problem governed by Eq. (4) with the diffusion term reinstated:

$$\lambda^2\nabla^2\vec{f} = \vec{f} - (\vec{\omega}_B - \vec{\omega})\tau \times \vec{f} + \hbar\vec{\omega} \times \vec{\omega}_B\tau. \tag{6}$$

This equation can either be solved numerically directly, for instance, using a finite element method solver, or by using the boundary conditions to determine the six constants of integration (for each region) of the general solution [17]. For simplicity in our model system we have assumed that the spin diffusion lengths, times, and diffusion constants are the same for both regions. With these assumptions the boundary conditions at the interface reduce to continuity of $\vec{f}$ and $\nabla\vec{f}$ at $x = 0$. When the regions are unbounded, the outer boundary conditions are that in region I the bulk result [Eq. (5) and below] is recovered; in region II, $\vec{f}(x \gg 0) = 0$. For bounded regions, $\nabla\vec{f} = 0$ at the boundaries so that there is no leakage of spin current. In the calculations the diffusion constant has been fixed at $D = 20$ cm^2/s (such as for Al) so the spin diffusion length λ scales as $\sqrt{\tau}$, and $\omega = 10$ GHz.

We first consider the case where both regions are unbounded. Figure 2 shows the results of the calculation for the two different field pumping regimes, plotting the spa-

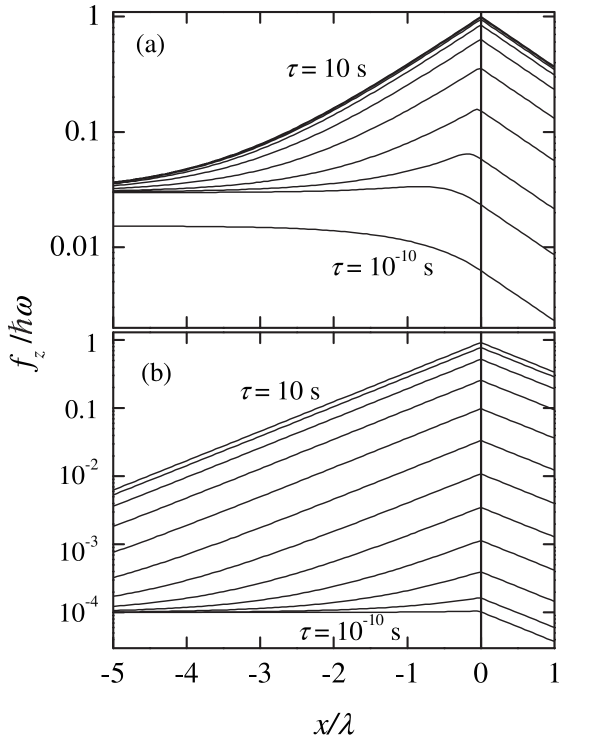

FIG. 2. The spin accumulation $f_z/\hbar\omega$ as a function of x/λ away from the interface, for τ ranging in decades from 100 ps to 10 s. Both regions are unbounded. For (a), $B_{xy} = 10$ mT and $B_z = 0$, for (b) $B_{xy} = 1$ T and $B_z = 100$ T.

tial dependence of $f_z/\hbar\omega$ near the interface for values of τ ranging over many decades. The most striking feature in Fig. 2 is that the spin accumulation at the interface is strongly enhanced over the bulk, uniform system value (at $x \ll 0$), and in the limit of very large τ achieves the universal value $\hbar\omega$, similar to the results of Ref. [2]. The calculations seem to describe a spin current injected at the interface which diffuses into both regions, even though the pumping occurs uniformly throughout region I.

In the unbounded system this effect is universal only for unphysically large τ, so we now try bounding the system in order to optimize the effect. In what follows we will focus only on the weak ferromagnet model; however, most of the results apply generally to both systems. Figure 3 shows the spin accumulation at the interface obtained for symmetric bounding of the regions at $x = \pm L$ as a function of L/λ_ω, over decades of τ from 10^{-10} to 10 s. The spin accumulation is optimized when the sample dimension $L \simeq 0.6\lambda_\omega$, independent of τ.

The spin accumulation obtained for different specific bounding lengths as a function of τ is summarized in Fig. 4. The most optimal spin accumulation was found for bounding region I at $L_1 \simeq 0.6\lambda_B$, and region II at $L_2 \simeq 0.6\lambda_\omega$ (curve E in Fig. 4). The interface spin accumulation in the optimized configuration may be compared to the interface-scattering matrix model result [2]: $f_z = \hbar\omega \frac{\sin^2\theta}{\sin^2\theta + \eta}$, where θ is the magnetization precession cone angle and $\eta = (\tau_i/\tau)\tanh(L/\lambda)/(L/\lambda)$ is a reduction factor which depends on the normal metal properties and the spin-injection rate τ_i (which is inversely proportional to the mixing conductance $g_{\uparrow\downarrow}$). We take $\theta \simeq \frac{B_{xy}}{B_z} = \frac{1}{100}$ and evaluate η for $L = \lambda_\omega$, and find that η depends on τ as $\eta \propto \frac{1}{\sqrt{\tau}} \tanh\sqrt{2\pi/\omega\tau}$. This functional dependence on τ agrees well with curves D and E, as shown for the fit to curve E (with 10^{-5} as the fitting parameter for η), even though our model is derived for an entirely different regime.

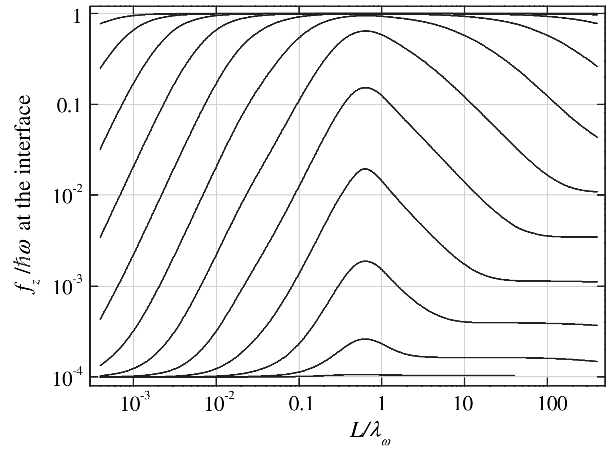

FIG. 3. Spin accumulation at the interface for the symmetrically bounded system plotted as a function of the normalized bounding length L/λ_ω, where $\lambda_\omega = \sqrt{2\pi D/\omega} \simeq 1.1$ μm, for τ ranging in decades from 10^{-10} (bottom curve) to 10 s (top curve).

A qualitative explanation for the interface enhancement in our model is as follows. A pumped spin accumulation in region I diffuses into region II (spin current), where it results in spin accumulation near the interface. Subsequently, this induces a flow of spin current back into region I. For spins that diffuse back within the relaxation time, the z component will be hardly affected. However,

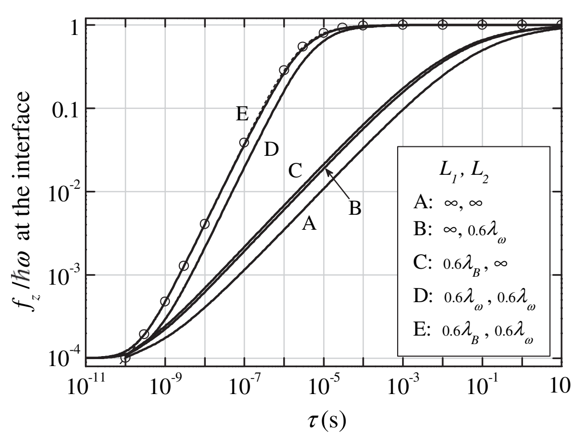

FIG. 4. The spin accumulation at the interface vs τ for specific bounding lengths indicated as L_1, L_2 (curves A through E). The open circles are based on Ref. [2] with $\eta = 10^{-5} \frac{1}{\sqrt{\tau}} \tanh\sqrt{\frac{2\pi}{\omega\tau}}$ (see text for details).

the x and y components of spins that flow back have a distribution of spin angles with respect to the rotating field in region I, due to the distribution of dwell times of spins in region II. This averages out the influence of the x and y components near the interface. The overall effect is similar to considering a uniform system [Eq. (5)] with anisotropic relaxation times τ_{xy} and τ_z:

$$f_z = -\frac{\omega_{xy}^2 \tau_{xy} \tau_z}{1 + \omega_{xy}^2 \tau_{xy} \tau_z + ((\omega_z - \omega)\tau_{xy})^2} \hbar\omega. \tag{7}$$

The relevant limit is now $\tau_{xy} \ll \tau_z$, so the influence of the effective z field $(\omega_z - \omega)$ term in the denominator is reduced, and the universal value $\hbar\omega$ can be achieved off-resonance in the limit $\omega_{xy}^2 \tau_{xy} \tau_z \gg 1$. In the hybrid system, this averaging process is optimized when the dimension of region II is roughly the same as λ_ω (Fig. 3). This length scale also quantifies the "sharpness" requirement for the interface: the pumping field should be rolled off over a distance less than λ_ω in order for the interface enhancement to occur. In our calculations with $D = 20\ \mathrm{cm}^2/\mathrm{s}$ and $\omega = 10$ GHz, then $\lambda_\omega \approx 1.1\ \mu$m. However, in a GaAs quantum well where the diffusion constant D can be extremely large ($10^5\ \mathrm{cm}^2/\mathrm{s}$), λ_ω can be of the order 100 μm.

We conclude with a short discussion of the experimental feasibility of applying the rf fields and detecting the predicted spin accumulations. A high-intensity, rf magnetic field can be applied with on-chip, coplanar waveguide (CPW) or stripline techniques, such as for ferromagnetic resonance experiments on small ferromagnetic elements [18,19]. Near the shorted end of the CPW we have been able to deliver a rf field of order 10 mT. Rotating fields can be applied locally with two parallel strip lines or a cross-point architecture with rf signals out of phase. For the bulk effect, linear rf fields can produce spin accumulation for finite B_z near resonance [17].

Our model of spin pumping in a nonmagnetic region by a rotating magnetic field can be applied quite generally to diffusive systems, such as metals and semiconductors. For the bulk spin accumulation effect, a numerical calculation for Al metal with realistic experimental parameters of linear rf field 10 mT at $\omega = 2\pi \times 16$ GHz and $B_z = 0.57$ T, gives a spin accumulation 0.5 μeV. This magnitude of spin accumulation can be detected with a ferromagnetic electrode contacted to the Al [12]. If we consider a semiconductor with τ as large as 100 ns and experimentally achievable $\omega = 2\pi \times 100$ GHz, then a spin accumulation close to the universal result of $\hbar\omega$ corresponding to 0.7 meV can be achieved even in the nonresonant case. For semiconductor systems, in particular, optical Kerr microscopy has been used to detect very small spin accumulations [20].

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