Dynamic Exchange Coupling in Magnetic Bilayers

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A long-ranged dynamic interaction between ferromagnetic films separated by normal-metal spacers is reported, which is communicated by nonequilibrium spin currents. It is measured by ferromagnetic resonance (FMR) and explained by an adiabatic spin-pump theory. In FMR the spin-pump mechanism of spatially separated magnetic moments leads to an appreciable increase in the FMR line width when the resonance fields are well apart, and results in a dramatic line-width narrowing when the FMR fields approach each other.

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The giant magnetoresistance[1] accompanying realignment of magnetic configurations in metallic multilayers by an external magnetic field is routinely employed in magnetic read heads and is essential for high-density nonvolatile magnetic random-access memories. These typically consist of ferromagnetic/normal/ferromagnetic (F/N/F) metal hybrid structures, i.e., magnetic bilayers which are an essential building block of the so-called spin valves. The static Ruderman-Kittel-Kasuya-Yosida (RKKY) interlayer exchange between ferromagnets in magnetic multilayers[2] is suppressed in these devices by a sufficiently thick nonmagnetic spacer N or a tunnel barrier. The interest of the community shifts increasingly from the static to the dynamic properties of the magnetization[3]. This is partly motivated by curiosity, partly by the fact that the magnetization switching characteristics in memory devices is a real technological issue. A good grasp of the fundamental physics of the magnetization dynamics becomes of essential importance to sustain the exponential growth of device performance factors.

In this Letter we study the largely unexplored dynamics of magnetic bilayers in a regime when there is no discernible static interaction between the magnetization vectors. Surprisingly, the magnetizations still turn out to be coupled, which we explain by emission and absorption of nonequilibrium spin currents. Under special conditions the two magnetizations are resonantly coupled by spin currents and carry out a synchronous motion, quite analogous to two connected pendulums. This dynamic interaction is an entirely new concept and physically very different from the static RKKY coupling. E.g., the former does not oscillate as a function of thickness and its range is exponentially limited by the spin-flip relaxation length of spacer layers and algebraically by the elastic mean free path. This coupling can have profound effects on magnetic relaxation and switching behavior in hybrid structures and devices.

The unit vector \( \mathbf{m} = \mathbf{M}/M \) of the magnetization \( \mathbf{M}(t) \) of a ferromagnet changes its direction in the presence of a noncollinear magnetic field. The motion of \( \mathbf{m} \) in a single domain is described by the Landau-Lifshitz-Gilbert (LLG) equation

\[
\frac{d\mathbf{m}}{dt} = -\gamma \mathbf{m} \times \mathbf{H}_{\text{eff}} + \alpha \mathbf{m} \times \frac{d\mathbf{m}}{dt},
\]

with \( \gamma \) being the absolute value of the gyromagnetic ratio. The first term on the right-hand side represents the torque induced by the effective magnetic field \( \mathbf{H}_{\text{eff}} = -\partial F/\partial \mathbf{M} \), where the free-energy functional \( F[\mathbf{M}] \) consists of the Zeeman energy, magnetic anisotropies, and exchange interactions[4]. The second term in Eq. (1) is the Gilbert damping torque which governs the relaxation towards equilibrium. The intrinsic damping in bulk metallic ferromagnets, \( \alpha^{(0)} \), typically 0.002-0.025, appears to be governed by spin-orbit interactions[5] in the 3d transition metals. The magnetization vector can be forced into a resonant precession motion by microwave stimulation. This ferromagnetic resonance (FMR) is measured via the absorption of microwave power using a small rf field at a frequency \( \omega \) polarized perpendicular to the static magnetic moment as a function of the applied dc magnetic field, see the right inset in Fig. 1. The absorption is given by the imaginary part of the susceptibility \( \chi'' \) of the rf magnetization component along the rf driving field. This FMR signal has a Lorentzian line shape with a width \( \Delta H = (2/\sqrt{3}) \alpha \omega/\gamma \) when defined by the inflection points (i.e., the extrema of \( d\chi''/dH \)), see the left inset in Fig. 1.

When two or more ferromagnets are in electrical contact via nonmagnetic metal layers, interesting new effects occur. Transport of spins accompanying an applied electric current driven through a magnetic multilayer causes a torque on the magnetizations[6], which at sufficiently high current densities leads to spontaneous magnetization-precession and switching phenomena[7]. Even in the absence of an applied charge current, spins...
are injected into the normal metal by a ferromagnet with moving magnetization. This causes additional magnetic damping, provided that the spin-flip relaxation rate of normal metal is high [8]. The present Letter focuses on the discovery of novel dynamic effects in $F1/N$ structures in the limit when the spin-flip scattering in $N$ is weak. Let us first sketch the basic physics. A precessing magnetization $m_i$ "pumps" a spin current $I_{\text{pump}}^{i} \perp m_i$ into the normal metal [8]. We focus on weakly excited magnetic bilayers close to the parallel alignment, so that $I_{\text{pump}}^{i} \perp m_i$ for arbitrary $i,j = 1,2$. The spin momentum perpendicular to the magnetization direction cannot penetrate a ferromagnetic film beyond the (transverse) spin-coherence length, $\lambda_{\text{sc}} = \pi/|k_F^\uparrow - k_F^\downarrow|$, which is determined by the spin-dependent Fermi wave vectors $k_F^{\uparrow\downarrow}$ and is smaller than a nanometer for 3$d$ metals [8]. A transverse spin current ejected by one ferromagnet can therefore be absorbed at the interface to the neighboring ferromagnet, thereby exerting a torque $\tau$. Each magnet thus acts as a spin sink which can dissipate the transverse spin current ejected by the other layer.

The theoretical basis of this picture is the adiabatic spin-pumping mechanism [8] and magnetoelectronic circuit theory [10]. $N$ is assumed thick enough to suppress any RKKY [2] pin-hole [11], and magnetostatic (Néel-type) [12] interactions. We consider ultrathin films with a constant magnetization vector across the film thickness [4], which are nonetheless thicker than $\lambda_{\text{sc}}$ and, therefore, completely absorb transverse spin currents. In the experiments described below, $N$ is thinner than the electron mean free path, so that the electron motion inside the spacer is ballistic. Precessing $m_i$ pumps spin angular momentum at the rate

$$I_{\text{pump}}^{i} = \frac{h}{4\pi g_{\uparrow\downarrow}^i} m_i \times \frac{dm_i}{dt},$$

where $g_{\uparrow\downarrow}^i$ is the dimensionless "mixing" conductance [10] of the $F/i/N$ interfaces, which can be obtained via $ab$ initio calculations of the scattering matrix [13] or measured via the angular magnetoresistance of spin valves [14] as well as FMR line widths of $F/N$ and $F/N/F$ magnetic structures [8,15,16]. Note that $g_{\uparrow\downarrow}^i$ must be renormalized for the intermetallic interfaces considered here [14]. We assume identical $Fi/N$ interfaces with real-valued $g_{\uparrow\downarrow}^i$, as suggested by calculations for various $F/N$ combinations [13]. When the spacer is not ballistic, its diffuse resistance can simply be absorbed into the value of $g_{\uparrow\downarrow}^i$, which should then be interpreted as the mixing conductance of an $F/N$ interface in series with the half of the spacer. When, furthermore, the spacer is thicker than the spin-diffusion length, the spin-pumping exchange between the magnetic layers becomes exponentially suppressed with the spacer thickness [8].

Alloy disorder at the interfaces scrambles the distribution function. Disregarding spin-flip scattering in the normal metal, an incoming spin current on one side leaves the normal-metal node by equal outgoing spin currents to the right and left [14]. (As the interfacial scrambling is only partial and the spacer is ballistic, the last statement should not be taken literally, but as an effective theory which is valid after renormalizing the interfacial conductance parameters.) On typical FMR time scales, this process occurs practically instantaneously. The net spin torque at one interface is therefore just the difference of the pumped spin currents divided by two: $\tau_i = (I_{\text{pump}}^{i} - I_{\text{pump}}^{j})/2 = -\tau_2$. When one ferromagnet is stationary, see the left drawing in Fig. 2, the LLG equation expanded to include the spin torque reads

$$\frac{d\mathbf{m}_i}{dt} = -\gamma \mathbf{m}_i \times \mathbf{H}_{\text{eff}} + \alpha_i^{(0)} \mathbf{m}_i \times \frac{d\mathbf{m}_i}{dt} + \alpha_i^{(\gamma)} \mathbf{m}_i \times \frac{d\mathbf{m}_i}{dt},$$

where $j = 1(2)$ if $i = 2(1)$. As a simple example, consider a system in the parallel configuration, $\mathbf{m}_1^{(0)} = \mathbf{m}_2^{(0)}$, with
molecular beam epitaxy (MBE) [16, 17]. Fe(001) films were grown on 4x6 reconstructed GaAs(001) substrates by MBE. The interface magnetic anisotropies allowed us to separate the FMR fields of the two Fe layers with resonance-field differences that can exceed 5 times the FMR line widths, see Fig. 1. Hence, the FMR measurements for F1 in double layers can be carried out with a nearly static F2.

The FMR line width of F1 increases in the presence of F2. The difference $\Delta H'$ in the FMR line widths between the magnetic bilayer and single-layer structures is nearly inversely proportional to the thin-film thickness $d_F$ [16], proving that $\Delta H'$ originates at the F1/N interface. Secondly, $\Delta H'$ is linearly dependent on microwave frequency for both the in-plane (the saturation magnetization parallel to the film surface) and perpendicular (the saturation magnetization perpendicular to the film surface) configurations, strongly implying that the additional contribution to the FMR line width can be described strictly as an interface Gilbert damping [16]. At the FMR, the film precessions are driven by an applied rf field. When the resonance fields are different, one layer (say F1) is at resonance with maximum precessional amplitude while the other layer (F2) is off resonance with small precessional amplitude, see Fig. 2. The spin-pump current for F1 reaches its maximum while F2 does not emit a significant spin current at all. F2 acts as a spin sink causing the nonlocal damping for F1. The N/F2 interface provides a “spin-momentum brake” for the F1 magnetization. The corresponding additional Gilbert parameter $\alpha'$ for a 16 ML Fe is significant, being similar in magnitude to the intrinsic Gilbert damping in isolated Fe films, $\alpha^{(0)} = 0.0044$.

These assertions can be tested by employing the in-plane uniaxial anisotropy in F1 to intentionally tune the resonance fields for F1 and F2 into a crossover which is shown in the shaded area of Fig. 1. When the resonance fields are identical, $H_1 = H_2$, the rf magnetization components of F1 and F2 are parallel to each other, see the right drawing in Fig. 2. The total spin currents across the F1/N and N/F2 interfaces therefore vanish resulting in zero excess damping for F1 and F2, see Eq. 3, which is experimentally verified, as shown in Fig. 3. For a theoretical analysis, we solved Eq. 3 and determined the total FMR signal as a function of the difference between the resonance fields $H_2 - H_1$. The theoretical predictions are compared with measurements in Fig. 3. The remarkable good agreement between the experimental results and theoretical predictions provides strong evidence that the dynamic exchange coupling not only contributes to the damping but leads to a new collective behavior of magnetic hybrid structures.

We have additionally carried out our measurements on samples with Au spacer thickness between 14 and 100.
FIG. 3: Comparison of theory (solid lines) with RT measurements (symbols) close to and at the crossover of the FMR fields, marked by the shaded area in Fig. 1. The left and right frames show FMR signals for the field difference, $H_2 - H_1$, of -78 Oe and +161 Oe, respectively. The theoretical results are parameterized by the full set of magnetic parameters which were measured independently [16]. The magnitude of the spin-pump current was determined by the line width at large separation of the FMR peaks. The middle frame displays the effective FMR line width of magnetic layers for the signals fitted by two Lorentzians as a function of the external field. At $H_1 = H_2$, the FMR line widths reached their minimum values at the level of intrinsic Gilbert damping of isolated films. The calculations in the middle frame did not take small variations of the intrinsic damping with angle $\varphi$ into account, which resulted in deviations between theory and experiment for larger $|H_1 - H_2|$. Note that $\Delta H_1$ first increases before attaining its minimum, which is due to excitation of the antisymmetric collective mode.

monolayers. The weak dependence of the FMR response on the spacer thickness fully supports our picture of the long-ranged dynamic interaction.

In conclusion, we found decisive experimental and theoretical evidence for a new type of exchange interaction between ferromagnetic films coupled via normal metals. In contrast to the well-known oscillatory exchange interaction in the ground state, this coupling is dynamic in nature and long ranged. Precessing magnetizations feel each other through the spacer by exchanging nonequilibrium spin currents. When the resonance frequencies of the ferromagnetic banks differ, their motion remains asynchronous and net spin currents persist. However, when the ferromagnets have identical resonance frequencies, the coupling quickly synchronizes their motion and equalizes the spin currents. Since these currents flow in opposite directions, the net flow across both $F1/N$ and $N/F2$ interfaces vanishes in this case. The lifetime of the arising collective motion is limited only by the intrinsic local damping. These effects can be well demonstrated in FMR measurements.

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