Dynamic Exchange Coupling in Magnetic Bilayers

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A long-range 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 and explained by an adiabatic spin-pump theory. In such a resonance the spin-pump mechanism of spatially separated magnetic moments leads to an appreciable increase in the resonant linewidth when the resonance fields are well apart, and results in a dramatic linewidth narrowing when the resonant 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. For example, 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

$$\frac{d\mathbf{m}}{dt} = -\gamma \mathbf{m} \times \mathbf{H}_{\rm eff} + \alpha \mathbf{m} \times \frac{d\mathbf{m}}{dt}, \qquad (1)$$

with γ 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 $\mathbf{H}_{\rm eff} = -\partial F/\partial \mathbf{M}$, 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 ω polarized perpendicular to the static magnetic moment as a function of the applied dc magnetic field; see the right inset of Fig. 1. The absorption is given by the imaginary part of the susceptibility χ'' 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 of 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

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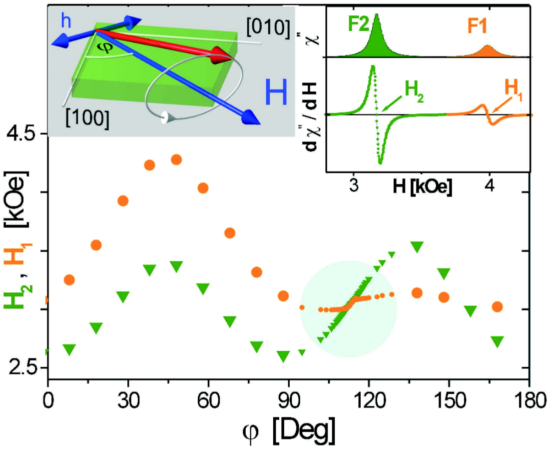

FIG. 1 (color online). Dependence of the FMR resonance fields H_1 (circles) and H_2 (triangles) for the thin Fe film $F1$ and the thick Fe film $F2$, respectively, on the angle φ of the external dc magnetic field with respect to the Fe [100] crystallographic axis. The sketch of the in-plane measurement in the left inset shows how the rf magnetic field (double-pointed arrow) drives the magnetization (on a scale grossly exaggerated for easy viewing). In the right inset we plot the measured absorption peaks for layers $F1$ and $F2$ at $\varphi = 60°$.

moving magnetization. This causes additional magnetic damping, provided that the spin-flip relaxation rate of normal metal is high [8]. This Letter focuses on the discovery of novel dynamic effects in $F1/N/F2$ structures in the limit when the spin-flip scattering in N is weak. Let us first sketch the basic physics. A precessing magnetization $\mathbf{m}_i$ "pumps" a spin current $\mathbf{I}_{si}^{\rm pump} \perp \mathbf{m}_i$ into the normal metal [8]. We focus on weakly excited magnetic bilayers close to the parallel alignment, so that $\mathbf{I}_{si}^{\rm pump} \perp \mathbf{m}_j$ 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, $\lambda_{\rm sc} = \pi/|k_{\rm F}^{\uparrow} - k_{\rm F}^{\downarrow}|$, which is determined by the spin-dependent Fermi wave vectors $k_{\rm F}^{\uparrow,\downarrow}$ and is smaller than a nanometer for $3d$ metals [9]. A transverse spin current ejected by one ferromagnet can therefore be absorbed at the interface to the neighboring ferromagnet, thereby exerting a torque $\boldsymbol{\tau}$. 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 $\lambda_{\rm sc}$ 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 $\mathbf{m}_i$ pumps spin angular momentum at the rate [8]

$$\mathbf{I}_{si}^{\rm pump} = \frac{\hbar}{4\pi} g^{\uparrow\downarrow} \mathbf{m}_i \times \frac{d\mathbf{m}_i}{dt}, \qquad (2)$$

where $g^{\uparrow\downarrow}$ is the dimensionless "mixing" conductance [10] of the F/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 linewidths of F/N and $F/N/F$ magnetic structures [8,15,16]. Note that $g^{\uparrow\downarrow}$ must be renormalized for the intermetallic interfaces considered here [14]. We assume identical Fi/N interfaces with real-valued $g^{\uparrow\downarrow}$, 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 $g^{\uparrow\downarrow}$, 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 2: $\boldsymbol{\tau}_1 = (\mathbf{I}_{s2}^{\rm pump} - \mathbf{I}_{s1}^{\rm pump})/2 = -\boldsymbol{\tau}_2$. When one ferromagnet is stationary (see the left drawing in Fig. 2) the dynamics of the other film, Fi, is governed by the LLG equation with a damping parameter $\alpha_i = \alpha_i^{(0)} + \alpha_i'$ enhanced with respect to the intrinsic value $\alpha_i^{(0)}$ by $\alpha_i' = \gamma\hbar g^{\uparrow\downarrow}/(8\pi\mu_i)$, where μ_i is the total magnetic moment of Fi. Since μ_i

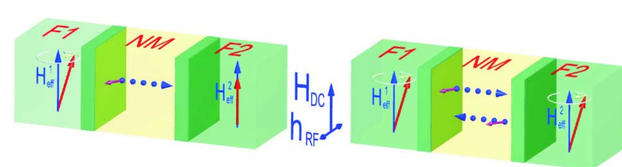

FIG. 2 (color online). A cartoon of the dynamic coupling phenomenon. In the left drawing, layer $F1$ is at a resonance and its precessing magnetic moment pumps spin current into the spacer, while $F2$ is detuned from its FMR. In the right drawing, both films resonate at the same external field, inducing spin currents in opposite directions. The short arrows in N indicate the instantaneous direction of the spin angular momentum $\propto \mathbf{m}_i \times d\mathbf{m}_i/dt$ carried away by the spin currents. Darker areas in Fi around the interfaces represent the narrow regions in which the transverse spin momentum is absorbed.

scales linearly with the volume of the ferromagnet and $g^{\uparrow\downarrow}$ scales with its interface area, α_i' is inversely proportional to the film thickness.

When both magnetizations are allowed to precess (see the right drawing in Fig. 2) the LLG equation expanded to include the spin torque reads

$$\frac{d\mathbf{m}_i}{dt} = -\gamma \mathbf{m}_i \times \mathbf{H}_{\text{eff}}^i + \alpha_i^{(0)} \mathbf{m}_i \times \frac{d\mathbf{m}_i}{dt} + \alpha_i' \left[\mathbf{m}_i \times \frac{d\mathbf{m}_i}{dt} - \mathbf{m}_j \times \frac{d\mathbf{m}_j}{dt} \right], \quad (3)$$

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 matched resonance conditions. In addition, let us assume the resonance precession is circular. If we linearize Eq. (3) in terms of small deviations $\mathbf{u}_i(t) = \mathbf{m}_i(t) - \mathbf{m}_i^{(0)}$ of the magnetization direction $\mathbf{m}_i$ from its equilibrium value $\mathbf{m}_i^{(0)}$, we find that the average magnetization deviation $\mathbf{u} = (\mathbf{u}_1\mu_1 + \mathbf{u}_2\mu_2)/(\mu_1 + \mu_2)$ is damped with the intrinsic Gilbert parameter $\alpha^{(0)}$, whereas the difference $\Delta\mathbf{u} = \mathbf{u}_1 - \mathbf{u}_2$ relaxes with enhanced damping constant $\alpha = \alpha^{(0)} + \alpha_1' + \alpha_2'$.

Measuring the spin torques requires independent control of the precessional motion of the two F layers, with FMR absorption linewidths of isolated films dominated by the intrinsic Gilbert damping. Both conditions were met by high-quality crystalline Fe(001) films grown on 4×6 reconstructed GaAs(001) substrates by molecular beam epitaxy [16,17]. Fe(001) films were deposited at room temperature (RT) from a thermal source at a base pressure of less than 2×10^{-10} Torr and the deposition rate was $\sim$1 ML (monolayer)/min. For the experiments discussed below, single Fe ultrathin films with thicknesses $d_F = 11, 16, 21, 31$ ML were grown directly on GaAs(001) and covered by a 20 ML protective Au(001) cap layer. The magnetic anisotropies as measured by FMR are described by a constant bulk term and an interface contribution inversely proportional to d_F. The Fe ultrathin films grown on GaAs(001) and covered by gold have magnetic properties nearly identical to those in bulk Fe, modified only by sharply defined interface anisotropies. The in-plane uniaxial anisotropy arises from electron hybridization between the As dangling bonds and the iron interface atoms. These Fe films were then regrown as one element of a magnetic bilayer structure and in the following referred to as $F1$ layers. They were separated from a thick Fe layer, $F2$, of 40 ML thickness by a 40 ML Au spacer. The magnetic bilayers were covered by 20 ML of protective Au(001). The complete structures are therefore GaAs/Fe(8, 11, 16, 21, 31)/40Au/40Fe/20Au(001), where the integers represent the number of MLs. The electron mean free path in thick films of gold is 38 nm [17] and, consequently, the spin transport even in the 40 ML (8 nm) Au spacer is purely ballistic. 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 linewidths; see Fig. 1. Hence, the FMR measurements for $F1$ in double layers can be carried out with a nearly static $F2$.

The FMR linewidth of $F1$ increases in the presence of $F2$. The difference $\Delta H'$ in the FMR linewidths 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. Second, $\Delta H'$ 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 linewidth 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 α' 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 MLs. 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

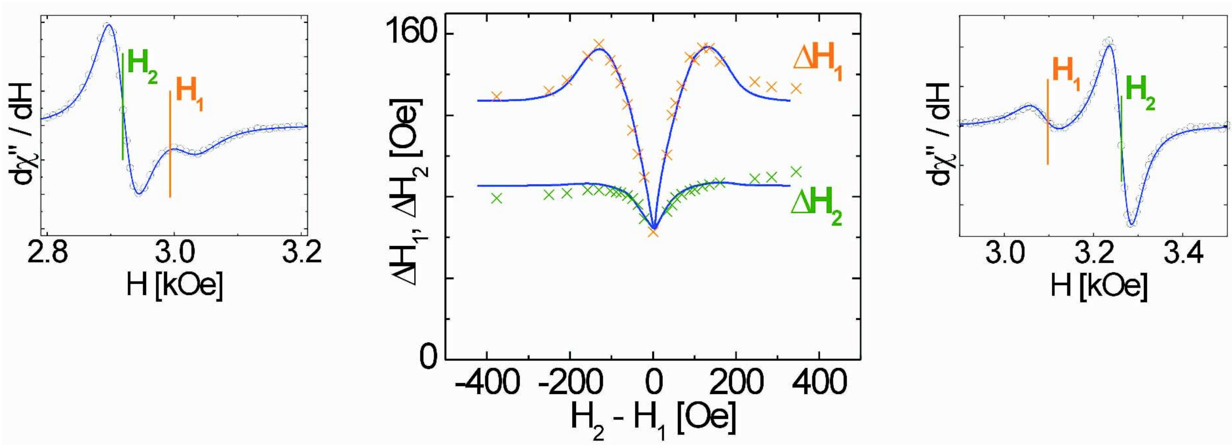

FIG. 3 (color online). Comparison of theory (solid lines) with RT measurements (symbols) close to and at the crossover of the FMR fields, marked by the shaded area in Fig. 1. The left and right frames show FMR signals for the field difference, $H_2 - H_1$, of -78 Oe and $+161$ Oe, respectively. The theoretical results are parametrized by the full set of magnetic parameters which were measured independently [16]. The magnitude of the spin-pump current was determined by the linewidth at large separation of the FMR peaks. The middle frame displays the effective FMR linewidth of magnetic layers for the signals fitted by two Lorentzians as a function of the external field. At $H_1 = H_2$, the FMR linewidths 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 φ into account, which resulted in deviations between theory and experiment for larger $|H_1 - H_2|$. Note that ΔH_1 first increases before attaining its minimum, which is due to excitation of the antisymmetric collective mode.

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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