Detection of coherent and incoherent spin dynamics during the magnetic switching process using vector-resolved nonlinear magneto-optics

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It is usually assumed that magnetic switching proceeds via coherent rotation under conditions of high symmetry. There is no *a priori* reason to expect an inhomogeneous response when a uniform magnetic torque is applied to a homogeneous ferromagnet. We test this assumption using vector- and time-resolved nonlinear magneto-optic measurements on a continuous Ni–Fe film. While coherent dynamics are observed when the magnetization **M** is initially oriented along the easy axis (the preferred axis of **M** in the absence of external fields), we find evidence for inhomogeneous spin dynamics when **M** is initially oriented perpendicular to the easy axis, which suggests the generation of incoherent spin waves during the magnetic reorientation process. The inhomogeneity is sufficient to reduce the spatially averaged magnetic moment within the measured area by almost 50%.
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We describe a measurement technique that relies upon a nonlinear form of the magneto-optic Kerr effect[1,2] to detect the time-resolved spin dynamics at the surface of a thin ferromagnetic film. Using this technique, we have successfully imaged the dynamics of the surface magnetization vector when subject to a fast magnetic field pulse generated with a microwave waveguide. A particular strength of the vector-resolved measurement is the ability to verify the homogeneity of the spin dynamics during the switching process. Surprisingly, we find that the degree of homogeneity is a strong function of the initial magnetic orientation in a thin metallic film, which suggests that a nonlinear coupling between the magnetization and spin-wave modes could be a source of difficulty in high-speed switching and the operation of future magnetoelectronic devices.

Among the numerous methods that exist for the measurement of magnetic switching speeds, one of the most useful has been stroboscopic magneto-optical imaging, facilitated by the increased availability of ultrafast lasers.[3,4] The second-harmonic magneto-optic Kerr effect (SHMOKE) has been used to measure precessional switching in thin films of the alloy Permalloy ($Ni_{81}Fe_{19}$).[1] SHMOKE is sensitive to the rotation of the magnetization vector **M** in Permalloy in response to a slowly swept magnetic field.[2] We combine these two capacities of SHMOKE to determine the motion of **M** in response to a magnetic field pulse. A full description of the vector calibration procedure is provided in Ref. 4.

Permalloy is an ideal material for these measurements because of its uniaxial anisotropy and low switching fields. The uniaxial anisotropy keeps **M** aligned along an in-plane easy axis in the absence of an applied field. Application of a dc magnetic field in a direction orthogonal to the easy axis, or ‘‘hard’’ axis, rotates the magnetization in a continuous fashion into the field direction. By calibrating the SHMOKE signal in response to a slowly swept magnetic field and fitting the data to conventional models of uniaxial anisotropy,[5] we determine the appropriate fitting parameters to provide a one-to-one correspondence between the ellipsometric state of the second-harmonic light and the magnetization components parallel (M_x) and perpendicular (M_y) to the plane-of-incidence.

The polarization angle is measured with a photoelastic modulator and lock-in amplifier. A photon counter is used to measure the second harmonic intensity, with typical yields of 10^3 photons per second. Integration times of 3–4 h are required to achieve signal-to-noise ratios for m_x and m_y of >100 and ~ 25, respectively, when acquiring a time trace with 100 ps time steps over a 5 ns time interval.

To determine the dynamic response, the sample (a 50-nm-thick film of $Ni_{81}Fe_{19}$ sputter deposited on a sapphire substrate 100 μm thick) is subjected to fast magnetic field pulses, generated by current pulses propagating in an underlying 450 μm wide coplanar waveguide that is separated from the magnetic film by the sapphire substrate. The 5 μm laser spot is centered relative to the waveguide width, insuring a high degree of field uniformity at the measured region. Using the Karlqvist equation[6] for the field produced by a finite-width current sheet, we calculate that the field pulse magnitude at the sample should vary by less than 1 A/m (10^{-2} Oe) over a 25 μm span near the middle of the waveguide. Data are obtained with optical sampling methods. The time resolution is limited to 50 ps by the electronic jitter of the pulse generation system, with a commensurate 3 dB bandwidth of 7 GHz.

A complete vector description of the magnetization requires measurement of the component perpendicular to the film plane, or M_z. However, the precessional motion is highly elliptical for the films under study, with an ellipticity ratio $M_z/M_y \sim 0.02$. Therefore, we assume $M_z \sim 0$ for the vectorial analysis of the data.

An important feature of such vectorial measurements is the ability to determine the coherence of the magnetic response. If all the spins that contribute to the magnetic moment within the measured spot do not precess in phase with each other, there must be a net reduction in the average

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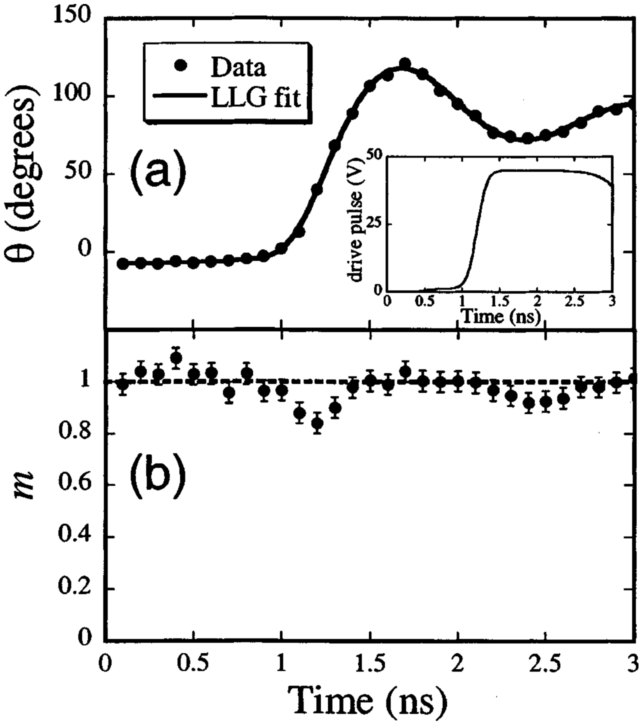

FIG. 1. Time-resolved SHMOKE data for Permalloy with a dc bias field $H_b = 80$ A/m (1 Oe) applied along the anisotropy axis and an orthogonal step pulse of 1.2 kA/m (15 Oe). The voltage wave form of the microwave pulse used to excite the sample is shown as an inset in (a). Both the time trace of (a) the in-plane angle θ and (b) the magnitude $m = M/M_s$ are shown. $\theta = 0$ is the direction of the applied bias field. The field geometry is shown as an inset in (b). The data for θ are fitted to the classical LLG equation of magnetic motion with reasonable results.

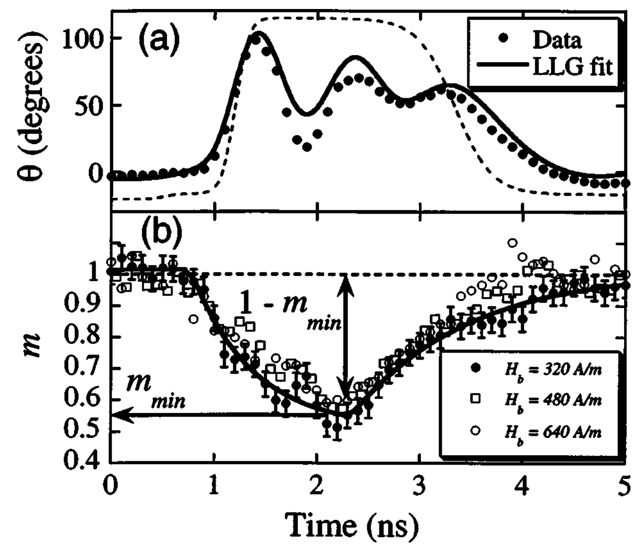

FIG. 2. Time-resolved SHMOKE data for Permalloy with a dc bias field $H_b = 320$ A/m (4 Oe) applied orthogonal to the anisotropy axis (transverse bias) and a step pulse of 1.2 kA/m (15 Oe) along the anisotropy axis. The step pulse duration is 2 ns. A trace of the step wave form is superimposed on the data as the dotted curve in (a). The sample is identical to that used to obtain the data in Fig. 2. Both the time trace of (a) the in-plane angle θ and (b) the magnitude $m = M/M_s$ are plotted with solid dots. $\theta = 0$ is defined as the initial orientation of **M** in the direction of the applied bias field. Additional magnitude data are shown for dc bias fields of 480 A/m (open squares) and 640 A/m (open circles). The data for m show a pronounced dip that recovers on a time scale of a few nanoseconds. The depth of the dip is extracted by exponential fitting to the data. The data for θ are fitted to the classical LLG equation of magnetic motion with poor results.

moment.[7] It is a fundamental assumption in the derivation of the Landau–Lifshitz–Gilbert (LLG) torque equation that **M** is uniform over the volume to be modeled.[8] Coherence at the experimental length scale is crucial to the validity of using the LLG equations to fit dynamics in ferromagnetic systems. Presumably, LLG could be used to simulate the dynamics at a finer length scale than our measured spot through the use of micromagnetic modeling methods.[9] However, such an indirect extraction of dynamical parameters such as the damping constant is no longer trivial and is subject to the details of the numerical simulation.[10] It is therefore imperative that one ascertain whether **M** responds uniformly in a given experimental geometry before one can directly extract the LLG damping parameter from the data using a single-domain model.

We first measured the magnetic response of our sample to a slowly swept field applied along the hard axis. The relative magnitude of the magnetization $m = M/M_s$ remained constant at its saturation value (within error bars of $\pm 7\%$) during the field sweep, as expected for coherent rotation of the magnetization under adiabatic conditions. The dynamic response for this sample, in this particular field geometry, is similarly uniform, as shown in Fig. 1. Here, the sample is subjected to a field pulse of 1200 A/m (15 Oe) along the hard axis. The field pulse has an onset time of 150 ps and duration of 2 ns. A dc field of 80 A/m (1 Oe) is applied along the easy axis to provide a preferred direction for **M**. By doing so, **M** should return to the same initial direction following termination of the field pulse. The magnetic response is spatially uniform over the volume of the illuminated spot (except for a slight dip at $t = 1.2$ ns), even though the magnetization is undergoing large-angle precessional motion. In this case, fitting of the data with LLG is reasonably justified and permits an unambiguous extraction of the phenomenological LLG damping parameter $\alpha = 0.016$, in agreement with previously measured values.[3]

An alternative measurement geometry can be achieved by rotating the sample by 90°, so that the magnetic field from the waveguide is oriented along the film's easy axis. The dc bias field is then applied along the hard axis to rotate the magnetization toward the hard axis. The magnetization is aligned parallel to the hard axis if the bias field is equal to (or greater than) the anisotropy field of $H_k = 320$ A/m (4 Oe). We applied a hard-axis bias field H_b of at least H_k. This prevents any possibility that the switching process may proceed via a process of nucleation and growth of domains, a notoriously irreproducible mechanism for magnetization reversal.[11] We then apply a field pulse (of the same magnitude used to obtain the data in Fig. 1) along the easy axis with the resulting response shown in Fig. 2. The magnetization response is highly incoherent: the magnitude of the magnetization is no longer constant during the switching process. Instead, m dips to almost 50% of saturation within 1 ns after the onset of the field pulse. Similar behavior was observed when the measurement was repeated at other locations over the waveguide center conductor. We therefore conclude that a single-domain LLG model is not appropriate to characterize the average dynamics of the measured volume in this configuration.

The turbulent magnetic state that reduces the measured

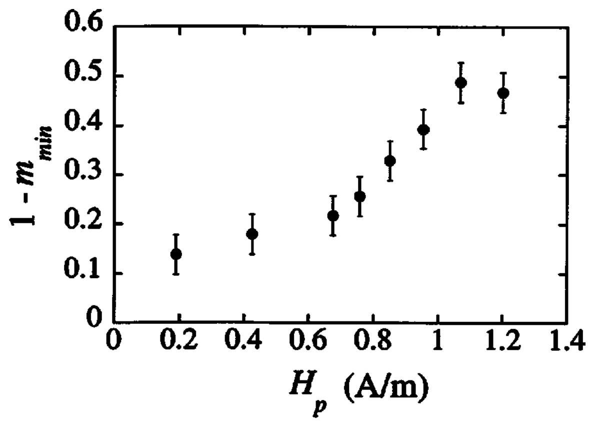

FIG. 3. Maximum reduction in $m=M/M_s$ as a function of pulse amplitude. For the vertical axis, $m_{\min}$ is the minimum value of m during the dynamic response to the field pulse.

moment is the result of spin–spin relaxation processes, which give rise to spin-wave modes. The spin waves in question must have wave numbers in excess of $10^4\ \mathrm{cm}^{-1}$ to reduce the spatially averaged value of m within the measured spot. Because the applied field pulse is highly uniform over the volume of the measured spot and the sample is prepared in a coherent initial state, the magnetic response must exhibit uniform precession immediately after the application of the pulse. With increasing time, however, the uniform mode breaks down into nonuniform excitations, characterized by wavelengths shorter than the illuminated spot. This results in a reduced measurement of m. In turn, these nonuniform modes decay via a variety of possible mechanisms. The possible mechanisms for spin-wave decay include magnon-phonon[12] and magnon-electron[13] scattering. The recovery of m to unity in Fig. 3 occurs with a longitudinal relaxation time of $T_1=1.1$ ns. This is the time scale required for the eventual decay of the induced spin waves.

Marginal stability is one possible explanation for the observed reduction in m. When biased along the hard axis with $H_b>H_k$, the net effective field that stiffens the individual spins is H_b-H_k.[14] When $H_b\sim H_k$, the magnetization is highly susceptible to any sample imperfections that perturb the nominally uniform state. However, it is possible to stiffen the spin system to an arbitrary degree when $H_b>H_k$. To test this hypothesis, we applied bias fields as large as 880 A/m (11 Oe) with little quantitative change in the temporal behavior of m. The data for $H_b=480$ and 640 A/m (6 and 8 Oe) are shown in Fig. 2. The insensitivity of our results to the exact value of H_b (even for a bias field of twice the anisotropy strength) eliminates marginal stability as a plausible explanation for the observed incoherent response. It is the orientation of **M** that is important for the instigation of incoherent spin dynamics.

Nonlinear mechanisms strongly affect ferromagnetic spin–spin relaxation.[15,16] We varied the amplitude of the field pulse to determine whether nonlinear effects play a role in our measurements. The maximum reduction in m is plotted as a function of the pulse amplitude in Fig. 3 for $H_b=320$ A/m (4 Oe). Indeed, the dependence on field amplitude is highly nonlinear, implying that nonlinear spinwave generation is the reason for the observed reduction of m. Micromagnetic simulations for the case of negligible intrinsic damping have predicted such nonlinear effects when large-angle magnetic reorientation is induced with an applied field pulse.[17]

Nonuniformities in the pulse field near the edges of the waveguide could spawn spin waves that propagate toward the region where the measurements are made. For magnetostatic surface wave modes[18] in a 50-nm-thick film and $k=10^4\ \mathrm{cm}^{-1}$, the calculated group velocity is $\nu_g=1.4\times10^4$ m/s. The requisite time for such a spin wave to propagate from the edge of the waveguide to the measured spot is therefore 16 ns, making it unlikely that waves generated at the waveguide edge would contribute to the observed reduction of m.

In conclusion, we find that the large-angle dynamic reorientation of magnetization at precessional time scales can occur coherently or incoherently, depending on the initial orientation of the magnetization relative to the anisotropy axis. While the fitting of time-resolved magnetodynamic data with LLG is a common means of analysis for the extraction of relevant material parameters,[1,3] our vector-resolved results clearly show that the actual magnetization dynamics can differ significantly from the coherent response presumed by LLG. When the dynamics are coherent, fitting of the data with LLG is valid. Even when the dynamics are not coherent, we find that the magnitude of **M** is still amenable to analysis. This permits the determination of the longitudinal relaxation time T_1 that describes the decay of incoherent spin wave modes within the sample.

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