

All-Optical Probe of Coherent Spin Waves

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A novel, all-optical method to excite and detect spin waves in magnetic materials is presented. By exploiting the temperature dependence of the magnetic anisotropy, an ultrashort laser pulse is efficiently converted in a picosecond “anisotropy field” pulse that triggers a coherent precession of the magnetization. Recording the temporal evolution of the precessing spins by a time-delayed probe-pulse provides a quantitative method to study locally the magnetic anisotropy, as well as switching and damping phenomena in micromagnetic structures. Applications to nickel and permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) films are discussed, particularly showing the possibility to explore standing spin waves in thin films.

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The recent development of multilayered structures with strongly magnetic field dependent electrical properties has opened the way for a whole new class of magnetic devices. In these devices, both the charge information and the spin information of electrons are used, so-called spintronics [1,2]. For example, an enormous research effort is presently devoted to the development of a magnetic random access memory, a new type of nonvolatile computer memory in which data are stored in large arrays of submicron magnetic tunnel junctions or spin valves [1–3]. To characterize the magnetic layers in these structures, which are of micrometer or submicrometer dimensions, a highly sensitive probe is needed that can locally measure their magnetic properties. Since the fast magnetic switching behavior is one of the most prominent current issues, measuring the dynamic magnetic properties is of great importance. Among these, damping of the magnetization precession is one of the crucial—though poorly understood—phenomena that greatly affects the switching speed.

In this Letter, we present an all-optical technique for the dynamic characterization of micromagnetic devices. By exploiting the temperature dependence of the magnetic anisotropy, a coherent spin wave is launched using an intense laser pulse. The precessional motion of the excited spins is then optically measured in the time domain with a subpicosecond ($<10^{-12}$ s) resolution, allowing for the quantitative determination of damping and anisotropy parameters. It is found that even for materials with a very low crystalline anisotropy (e.g., permalloy) a precession can be initiated, using only the shape anisotropy of the film. Moreover, since the optical excitation is nonuniform in depth, standing spin waves can be excited in thin films. This way, additional information on spin-spin interactions can be obtained. The spatial resolution of the technique is limited only by the wave character of light, allowing for the study of sub- μm structures.

Our all-optical approach has a number of interesting advantages over existing techniques. For example, ferromagnetic resonance (FMR) [4] is a widely used method

for the dynamic characterization of magnetic materials. Although normally used as a nonlocal technique, local versions of FMR have been developed using scanning probe techniques [5] and by modulating the anisotropy with a laser [6]. However, since measurements are taken in the frequency domain, information on damping can only indirectly be obtained from the broadening of the absorption lines. Also Brillouin light scattering, a microscopic technique for the detection of spin modes [7], works in the frequency domain. Recently, techniques have become available to measure the switching and precession of the magnetization locally and in the time domain [8,9]. A magnetic field pulse with a short (several picoseconds) rise time is used to excite the spin system, followed by a time-delayed laser pulse to optically measure the spin response as a function of time. The application of short field pulses requires lithographically defined structures on the sample, and this limits the applicability of the technique for standard characterization. In contrast, our all-optical method offers a flexible and generally applicable technique, with the additional advantage of its sensitivity to standing spin-wave phenomena.

In our setup, shown in Fig. 1a, a sample is placed between two magnet poles and in the focus of a lens. Using an intense laser pump-pulse at $\lambda = 780$ nm and of 0.1 ps duration, the material at the focal point (~ 10 μm) is almost instantaneously heated. The effect on the magnetization is measured by a much weaker, time-delayed probe-pulse using the magneto-optical Kerr effect; i.e., upon reflection the probe-pulse will undergo a change in polarization proportional to the magnetization. In the particular (polar) optical geometry used, mainly the out-of-plane component of \vec{M} , M_z , is detected. Vectorial schemes are available, however, to extend the technique to a full orientational mapping of the magnetization dynamics [9]. By varying the time delay between pump and probe, the magnetization can be measured as a function of time after excitation. Since for ferromagnets the magnitude of the magnetization decreases with temperature, this setup

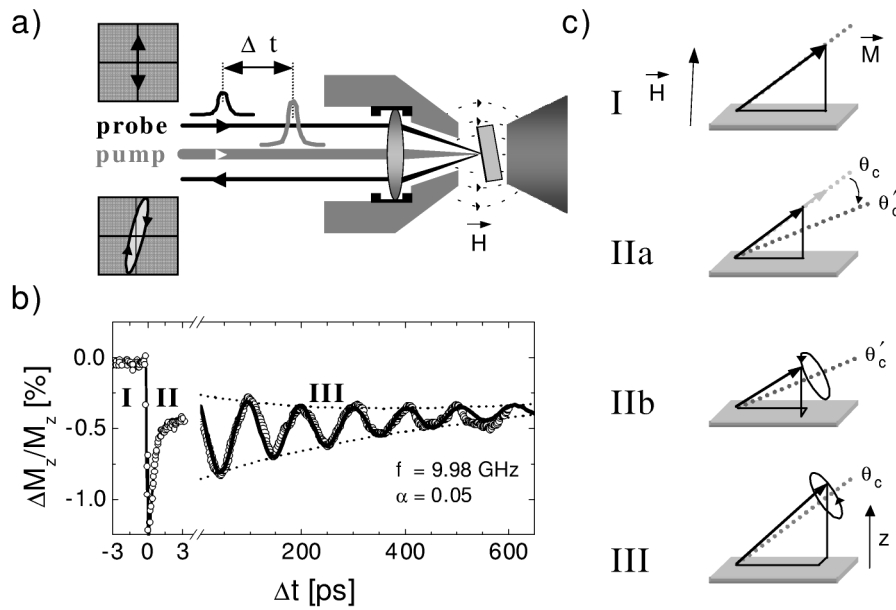


FIG. 1. (a) Schematic pump-probe setup. The magnetization is measured by the polarization state of the reflected probe pulse. (b) Typical measurement on a 7 nm Ni film (open circles: data; thick line: fit) displaying the perpendicular component of the magnetization, M_z , as a function of delay time Δt . The different stages are indicated by numbers. (c) The stages of the excitation process: (I) $\Delta t < 0$, the magnetization \vec{M} points in equilibrium direction (dotted line), (IIa) $\Delta t = 0$, the magnitude of \vec{M} and the anisotropy change due to heating, thereby altering the equilibrium orientation, (IIb) $0 < \Delta t < 10$ ps, \vec{M} starts to precess around its new equilibrium, (III) $\Delta t > 10$ ps, heat has diffused away, the magnitude of \vec{M} and anisotropy are restored, but the precession continues because of the initial displacement of \vec{M} .

has been used for studying ultrafast demagnetization phenomena on a subpicosecond time scale [10,11].

In previous work it has been demonstrated that for certain special systems the heat generated by a laser pulse can alter not only the magnitude of the magnetization but also its orientation. Examples of the latter are our previous work on epitaxial Cu/Ni/Cu films with a special canted configuration [12], and experiments by Ju *et al.* on exchange-biased NiFe/NiO layers [13]. Here we show that using an all-optical technique coherent spin waves can be excited in practically any ferromagnetic film by slightly canting the magnetization with an external field.

A typical measurement on a 7 nm thick polycrystalline nickel layer on silicon is shown in Fig. 1b. When the pump-pulse heats the material at $\Delta t = 0$, a sharp decrease in M_z is observed. This effect is caused by a change in magnitude of the (temperature dependent) magnetization. The subsequent recovery of M_z on a time scale of a few ps is due to rapid heat diffusion into the substrate. Strikingly, long after returning to thermal equilibrium a secondary response appears as a persistent oscillation that lasts for hundreds of picoseconds. A series of detailed experiments, some of them elucidated in this Letter, show the oscillatory signal to be an optically triggered precession of the magnetization, i.e., an orientational effect. In this specific case of an in-plane magnetization, the observed behavior can be well described by the excitation mechanism depicted in Fig. 1c. By applying an appropriate external field H , the magnetization is canted out-of-plane. The canting angle θ_c is determined by the subtle balance between the external field and the anisotropy field of the film, including the shape anisotropy. Upon sudden heating by the pump-pulse, not only the magnetization decreases but also the anisotropy of the film changes. This results in a change of the equilibrium orientation from θ_c to θ'_c (IIa), triggering an initial precession of the magnetization around its new equilibrium orientation (IIb). The initial precession

is not visible in Fig. 1b, region II, since its displacement $\Delta\vec{M}$ is approximately parallel to the surface, and the small effect on M_z is completely masked by the strong change in magnitude of \vec{M} . Heat diffusion into the film quickly removes the excess heat, and, after about 10 ps for metallic films, the original equilibrium angle is restored. However, at this point the magnetization is still not in equilibrium due to its initial displacement, and will continue to precess for hundreds of picoseconds (III).

The solid line in Fig. 1b is a fit to the data, giving a precession frequency $f = 9.98$ GHz and a phenomenological (Gilbert) damping parameter $\alpha = 0.05$. By repeating the experiment at different pump intensities it was checked that only the amplitude of the precession is dependent on the absorbed power. This confirms that the induced change of anisotropy affects only the first picoseconds and that the measured precession occurs in the original anisotropy field, thus revealing the equilibrium magnetic properties. Additionally, it follows that the exact excitation mechanism is not of importance for the result; i.e., the picosecond excitation determines the amplitude and phase of the final precession, but not its precession frequency and damping.

To unambiguously demonstrate the precessional nature of the oscillation a comparison has been made with traditional FMR measurements. Since FMR is based on the resonant absorption of microwave radiation by a ferromagnet, it is essentially nonlocal, and the experiments have to be performed on uniform layers of Ni. In the FMR experiment, the microwave radiation frequency was fixed at 9.45 GHz and the (in-plane) magnetic field at which resonance occurs was measured; see Fig. 2 (inset). In the all-optical FMR experiment, the frequency can be directly measured as a function of field, as shown in Fig. 2 for a field tilted about 10° out-of-plane. The FMR measurement, represented by the single solid square, predicts a precession frequency of 9.45 GHz at 0.18 T, in very good agreement with the all-optical experiment. The theoretical

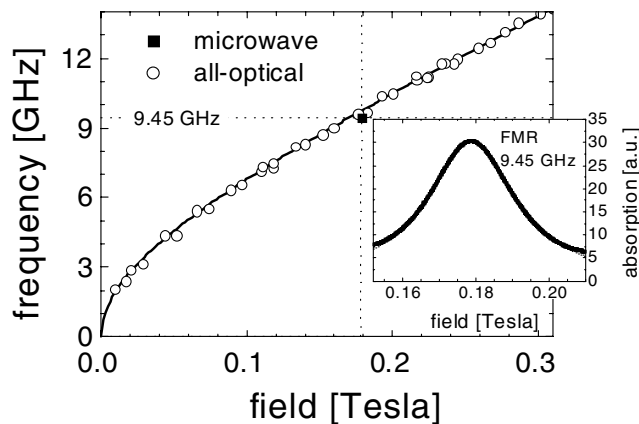


FIG. 2. Optically determined precession frequency versus in-plane field for a Ni layer, the solid line is a fit to the data. Inset: Microwave FMR measurement on the same layer, yielding a resonance field of 0.18 T at 9.45 GHz, depicted by the solid square in the main diagram.

field dependence of the precession frequency for this film with an in-plane anisotropy is given by

$$f = \gamma \frac{g}{4\pi} \mu_0 \sqrt{H(H + H_{az})} \quad (1)$$

with γ the gyromagnetic ratio, g the Landau splitting factor, and H_{az} the anisotropy field. With $g = 2.12$ and $\mu_0 H_{az} = 0.42$ T the measurement is well described, clearly showing the square rootlike behavior at lower fields. Using Eq. (1), the width of the FMR resonance (ΔH) can be related to the damping parameter, realizing that $\alpha = \Delta f / f_{\text{res}} = (df/dH)_{\text{res}} \Delta H / f_{\text{res}}$. This procedure yields $\alpha = 0.054$, which compares well with the value of 0.052 ± 0.003 obtained from the all-optical experiment by averaging the exponential decay of a number of time scans. The close agreement supports our claim that the all-optical method is capable of locally reproducing equilibrium magnetization dynamics; i.e., for the long-term dynamics the impact of the laser pulse equals that of a picosecond magnetic field pulse. We stress that this good agreement is not self-evident. First of all, since we optically create a highly excited material, a deviating frequency and/or damping might be anticipated. The resemblance with the microwave results means that at the laser fluencies applied, the system relaxes to near equilibrium soon enough not to affect dynamics appreciably on a 100 ps time scale. Second, spin waves laterally moving out of the irradiated area might enhance the damping. However, this was not experimentally observed with the current focus diameter of 10 μm and a typical damping time in the order of 0.1 ns.

The local character of the technique, as well as its general applicability, has been demonstrated by individually addressing elements out of lithographically defined arrays. A typical example of a measurement on a 10 nm triangular element is shown in Fig. 3. Although for permalloy the crystalline anisotropy is negligible, still a clear precession is observed. We conjecture that the precession is excited by the different magnetization dependence of the

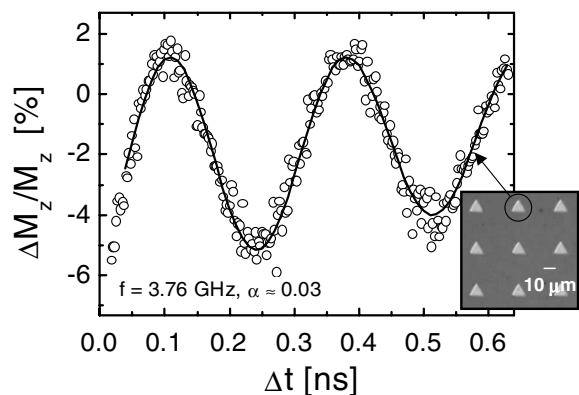


FIG. 3. Measurement on an individually addressed 10 μm permalloy element. Inset: optical microscope image of the array.

shape anisotropy ($E \sim M^2$) and the Zeeman energy in the applied field ($H \cdot \vec{M}$). In general, also strain, induced by the rapid heating of mainly the top 10–15 nm of the layer, may contribute to the excitation. However, for permalloy this contribution is expected to be small due to its low magnetostriction.

Our analysis so far has been based on a homogenous precession of the magnetization. It can be understood, however, that the approach is oversimplified if the thickness of the film is no longer negligible compared to the penetration depth of light, typically 10–15 nm for ferromagnetic transition metals. In that case, spins within the optical skin depth of the film are more intensely excited and also more efficiently detected by the delayed probe-pulse. This property will be shown to provide an intriguing additional possibility of the all-optical technique: the investigation of standing spin waves in magnetic films.

Figure 4 shows a measurement on a 40 nm thick polycrystalline Ni layer (solid dots). Instead of a single oscillation, we find a much richer response that is well described by the sum of two damped oscillations at frequencies ω_0 and ω_1 , indicated by the solid curves in the figure. Measurements on a wedge structure (see Fig. 5) show that the precession at ω_0 is thickness independent. In contrast, the oscillation at ω_1 shows a strong thickness dependence, approximately falling off as $1/L^2$ to ω_0 , with L the thickness of the layer.

This behavior is perfectly explained in terms of a fundamental and first-order excited mode of a standing spin wave within the thickness of the layer. The fundamental mode, shown graphically in Fig. 4, bottom surface, is the normal precession of the magnetization that is uniform within the thickness of the layer. Assuming open boundary conditions, the first-order standing wave has a single node in the middle of the layer and is free at the surfaces, as depicted in Fig. 4, middle. The complete precession is given by the sum of these two, Fig. 4, top, with the measured data drawn at the surface. The thickness dependence of ω_1 can be understood by looking at the magnon dispersion relation, Fig. 5 (inset). For small k vectors this relation is

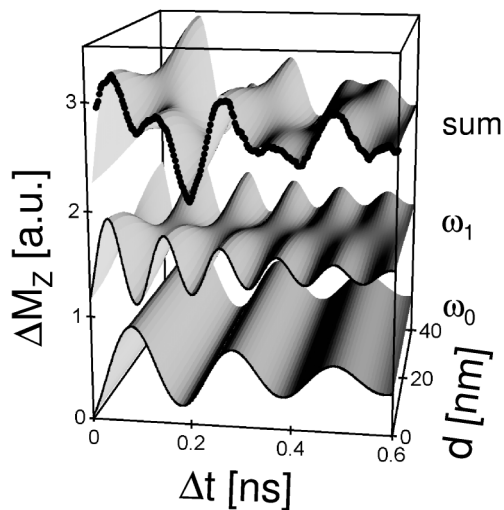


FIG. 4. Observation of standing spin waves. Because of the nonuniform heating by the pump-pulse both the fundamental precession (ω_0), which is uniform in the depth d of the layer, and the first-order standing spin wave (ω_1) are excited. From the resulting precession (sum) only the magnetization near the surface is optically measured. The measurement is thus shown at $d = 0$ nm by solid dots.

quadratic, and due to the finite thickness of the film the allowed k values are discrete. At $k = 0$ we find the uniform precession at ω_0 , whereas $k = \pi/L$ gives the first-order standing spin wave at a frequency $\omega_1 = \omega_0 + D(\pi/L)^2$, with D the spin-wave exchange constant. For the canted configuration studied here a slightly more complicated description is required. An improved model gives a value of $D = 430 \text{ meV \AA}^2$ that can be compared with values of $350\text{--}500 \text{ meV \AA}^2$ obtained from FMR and neutron diffraction [14]. So far no modes with $k \geq 2\pi/L$ have been observed. This can be attributed to the small, but finite, penetration depth of light: due to their shorter wavelength, higher order modes are less efficiently excited and measured since one averages over a skin depth of ~ 13 nm.

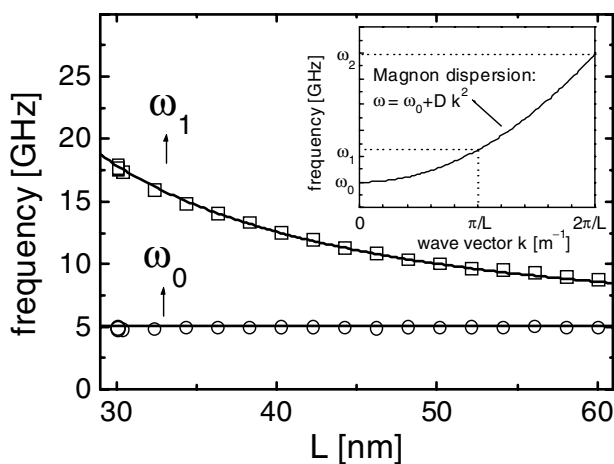


FIG. 5. Fundamental (ω_0) and first-order (ω_1) spin-wave frequency ($\omega/2\pi$) in Ni versus layer thickness L . The solid lines are fits to the data. Inset: schematic magnon dispersion relation.

For a 30 nm thick layer calculations predict a contribution of only 3% for the second-order mode, compared to a first-order contribution of 35%. An interesting aspect of the all-optical technique is that the antisymmetric mode observed in the optical experiment cannot be measured by microwave FMR, since in uniform thin films FMR selection rules allow only excitations with a net magnetic moment.

In conclusion, we have demonstrated a novel all-optical technique to measure the dynamic magnetic properties of microstructures. The key element of the approach is the temperature dependence of the anisotropy, which allows us to use the heat from an absorbed laser pulse to generate an anisotropy field pulse. Time-domain measurements on the consequently excited precession give quantitative information on the anisotropy, switching, and damping phenomena of small magnetic structures, with potential applications in the characterization of spintronic devices. We have further demonstrated that due to the nonuniform character of the excitation standing spin waves can be excited, allowing for the investigation of spin-wave dispersion. One of the future challenges is the study of lateral spin waves. Since the excitation is also laterally confined, this technique is well suited for the spatiotemporal imaging of laterally propagating spin waves, provided that sufficient spatial resolution is achieved.

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