

Magnetization dynamics induced by ultra-short optical pulses

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The interplay of interface and bulk magnetization dynamics of Cr-capped Fe(001) thin films have been studied by the time-resolved Kerr effect (TR-MOKE) and time-resolved magnetization-induced second harmonic generation (TR-MSHG) using an all-optical pump-probe technique. Long-lived (≈ 1 ns) MOKE and MSHG oscillations are excited by ultra-short (≈ 0.15 ps) optical pulses and reveal a similar behavior of the interface and bulk magnetization dynamics. The in-plane amplitude of the optically excited magnetization precession is determined and gives evidence that the observed dynamics reaches beyond the usually considered linear range. These results are of importance for the application of ultra-fast optical switching schemes in opto-magnetic devices.

The magnetization dynamics is currently intensively studied both in thin magnetic films and bulk crystals with emphasis on the dynamic response induced by ultra-short optical pulses [1]. The practical interest in this type of excitation mechanism is driven by the possibility of ultra-fast optical switching of local magnetic areas, which might be employed in new types of opto-magnetic devices for information and data processing technology. From a more fundamental point of view this optical approach enables the investigation of the microscopic mechanisms governing the excitation of magnetization dynamics on a very short time scale and allows testing the applicability, the limits, and validity of the classical Landau-Lifshitz-Gilbert (LLG) formalism.

The magnetization dynamics probed by the TR-MOKE reflects the properties of the bulk magnetization, because the Kerr signal stems from the entire thickness of the film, provided it is less than the information depth of light. The dynamic behavior of the interfacial magnetization, however, can selectively be probed by means of TR-MSHG. In this method a pump beam (frequency ω) excites magnetization precession and a probe beam is used to generate a MSHG signal $I_{2\omega}$, the amplitude of which depends on the transient magnetic state at a time delay Δt after the excitation. (M)SHG in centrosymmetric structures originates from a very narrow region of one or two monolayers at the surface or interface, where the inversion symmetry is broken. Previous TR-MOKE and TR-MSHG studies [2, 3] of epitaxial Fe/AlGaAs

films surprisingly revealed different dynamic behavior of bulk and interfacial magnetization. This finding was interpreted as an indication that the interfacial and bulk magnetization contributions in Fe/AlGaAs are to some extent decoupled. However, the microscopic origin has not been addressed, and the general validity of this observation remained unclear. Here, we employed both TR-MOKE and TR-MSHG to study interfacial and bulk magnetization dynamics induced by ultra-short laser pulses in epitaxial GaAs(100)/Ag/Fe/Cr structures [4].

The Fe films were grown by molecular beam epitaxy onto GaAs(001) substrates with an Ag(150 nm)/Fe(1 nm) buffer layer and are covered by a Cr(2 nm) protective cap layer. Fe and Cr layers crystallize in the *bcc* structure, which for Fe gives rise to cubic magnetocrystalline anisotropy. The strong demagnetizing field of the thin film geometry confines the static magnetization predominantly in the film plane. Therefore, the overall magnetic anisotropy can be described by an effective in-plane, four-fold anisotropy energy. The dynamic response of the magnetization \mathbf{M} resulting in TR-MSHG and TR-MOKE signals was induced by 150 fs optical pump pulses at $\lambda = 800$ nm generated by a regenerative amplifier with 1 kHz repetition rate. For TR-MSHG a photon counting technique was used to record the SHG intensity of the probe beam at the

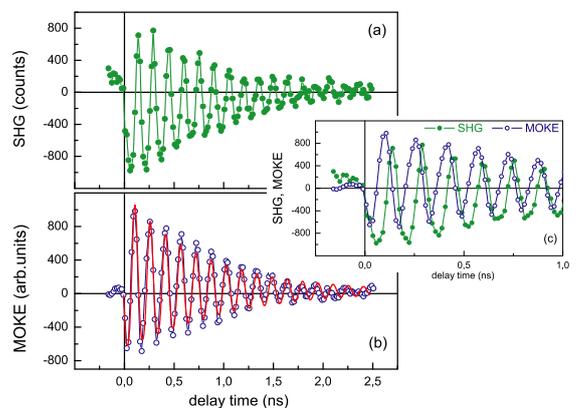


FIG. 1: (a) MSHG and (b) MOKE time profiles in *pp* polarization combination for a Fe(26 nm)/Cr(2 nm) bilayer. An offset of 2500 counts is subtracted in (a). The red line is a calculated trace (see text). Inset: TR-MOKE and TR-MSHG signals are shifted by a 90° .

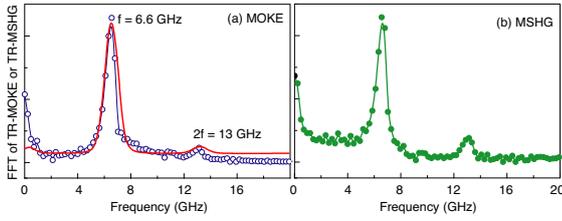


FIG. 2: Fourier transforms (FFT) of (a) TR-MOKE and (b) TR-MSHG signals for a Fe(26 nm)/Cr(2 nm) bilayer measured under the same conditions as in Fig. 1.

double frequency ($\lambda = 400$ nm). The fundamental light at $\lambda=800$ nm was rejected by a blue filter in the reflected beam. In the case of TR-MOKE, we employed lock-in technique and a differential photodetector. In both approaches the probe beam incidence angle was $\theta \approx 10^\circ$. The diameter of the illuminated area was about 1 and 0.3 mm with an average power of 10 and 3 mW for the pump and probe beam, respectively. The measurements of TR-MSHG and TR-MOKE were carried out at a magnetic field of 0.5 kOe oriented close to an in-plane hard-axis direction.

Figure 1 shows MOKE and MSHG time profiles of an Fe(26 nm)/Cr(2 nm) film for the *pp* polarization combination. The main oscillations in both TR-MOKE and TR-MSHG occur at the same frequency, but their phase is shifted by $\approx 90^\circ$. The Fourier transforms of the time profiles in Fig. 2 yield a main frequency $f = 6.6$ GHz, which corresponds to the frequency of the uniform precession mode. In addition, we observe weak intensities at the double frequency $2f$. For the analysis of the experimental results we use expressions linking the MSHG intensity to components of the interfacial magnetization \mathbf{M} , which are based on effective nonlinear susceptibilities [5]. The MSHG intensity for the *pp* polarization combination is given by the interfacial magnetization component M_y and contains both linear and quadratic contributions

$$I_{2\omega} = a + bM_y + cM_y^2. \quad (1)$$

The coefficients a , b , and c are related to the effective nonlinear susceptibilities and can be determined from the field dependencies of the static MSHG in *pp* configuration (Fig. 3). For this procedure the M_x and M_y components as a function of the applied are described within a Stoner-Wolfarth model taking into account the Zeeman and four-fold in-plane magnetic anisotropy energies. The calculated MSHG field dependence is superimposed in Fig. 3 as a red line. Since the static field dependence and optically induced time-dependent variations of the MSHG response were measured at the same experimental conditions the coefficients a , b , and c from Fig. 3 may also be used to calculate the time traces and Fourier spectra of the TR-MSHG and TR-MOKE oscillations. For these calculations we assume that the magnetization follows an exponentially damped precession. The results of the calculations are shown as red lines in Figs. 1 and 2. The best agreement between the experimental and calculated data is obtained at a max-

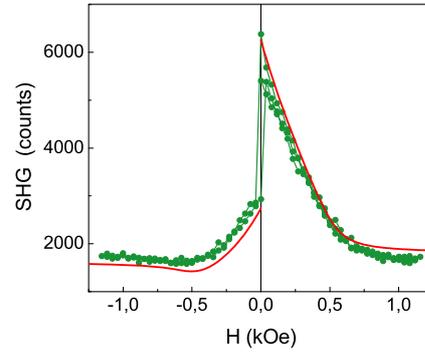


FIG. 3: Magnetic field dependence of the MSHG intensity for a Fe(26 nm)/Cr(2 nm) bilayer in the longitudinal geometry. The red line shows a fit from which the coefficients a , b , and c in Eq. (1) are determined.

imum oscillation amplitude of 13° . This large amplitude value means that at the given excitation power the system may be already at the threshold of the applicability of the LLG equations. As it is known, these equations are linearized and only valid for small deflections of the magnetization vector from the equilibrium value. The Fourier spectrum of the calculated MSHG oscillations shown in Fig. 2(a) displays a strong first harmonic and a much weaker $2f$ second harmonic contribution due to the nonlinearity of the $I_{2\omega}(M_y)$ function in Eq. (1). The phase shift observed between MOKE and MSHG oscillations (Fig. 1) is a clear evidence that the linear response relates to the polar Kerr effect and is caused by time variations of the magnetization component M_z normal to the film plane. On the other hand, the nonlinear MSHG response is related to the M_y component [Eq. (1)].

Our investigations show that in GaAs(001)/Ag/Fe/Cr—in contrast to AlGaAs/Fe films [2, 3]—the oscillations of the interfacial and bulk magnetizations appear at the same frequency corresponding to the uniform mode frequency and, thus, reveal a similar magnetization dynamics. A possible source for this discrepancy may be related to the different nature of the samples, i.e. the properties of the interfaces between a metal and semiconductor (Fe/AlGaAs) and between two metals (Ag/Fe and Fe/Cr). The semiconductor/metal interface has a stronger tendency for intermixing, which can have a significant influence on the magnetic properties in the entire interface-near region. The large amplitude of the oscillations of up to 13° gives evidence that the dynamical behavior of the system is at the limit of the linearized LLG equation's applicability and that next order effects should be considered when developing novel opto-magnetic switching schemes.

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