Invited Paper

Contributions from coherent and incoherent lattice excitations to ultrafast optical control of magnetic anisotropy of metallic films

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ABSTRACT

Spin-lattice coupling is one of the most prominent interactions mediating response of spin ensemble to ultrafast optical excitation. Here we exploit optically generated coherent and incoherent phonons to drive coherent spin dynamics, i.e. precession, in thin films of magnetostrictive metal Galfenol. We demonstrate unambiguously that coherent phonons, also seen as dynamical strain generated due to picosecond lattice temperature raise, give raise to magnetic anisotropy changes of the optically excited magnetic film; and this contribution may be comparable to or even dominate over the contribution from the temperature increase itself, considered as incoherent phonons.

Keywords: laser-induced magnetization dynamics, magnetostrictive materials, magnetic anisotropy

1. INTRODUCTION

Changing magnetic anisotropy by femtosecond laser pulses is among the most efficient and universal approaches allowing controlling magnetic state of matter on a (sub)picosecond time scale.[1] In the most of the cases the laser-induced change of the magnetic anisotropy manifests itself in magnetization precession triggered by a sudden change of the effective magnetic field, which strength and direction are dependent on the anisotropy itself and the external magnetic field. As a result, controlling magnetic anisotropy by light enables generation of uniform magnetization precession,[2] propagating spin waves,[3] and coherent magnetization switching.[4]

Since magnetocrystalline anisotropy originates from the spin-lattice coupling, optically-induced generation of collective excitations in a lattice, or phonons, paves a way towards control of magnetic anisotropy. In a thin metallic films both coherent and incoherent phonons can be effectively generated as a result of an impact of a femtosecond laser pulses. Indeed, impact of a femtosecond laser pulse on a thin metallic films results in a rapid increase of the electron temperature. This is followed by an increase of the lattice temperature, i.e. generation of non-coherent phonons, as well as by emergence of the thermal stress in the near surface region.[5] The latter induces the strain, seen also as coherent phonons, in two forms, persistent anisotropic quasiuniform strain and the picosecond strain pulses propagating away from the surface with sound velocities. The effect

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of non-coherent phonons, or lattice heating, on the magnetic anisotropy of metallic films has been extensively studied by a number of authors.[6–10] Analogously, propagating picosecond stain pulses were demonstrated to alter the magnetic anisotropy in thin magnetic films[11–13] via inverse magnetosctriction. However, the effect of the persistent quasiuniform strain, which is intrinsically present in laser-excited metallic films, on their magnetic anisotropy remains largely unexplored, to the best of our knowledge.

Here we report on experimental and theoretical study of the magnetic anisotropy change in the magnetostrictive metallic Galfenol ($Fe_{0.81}Ga_{0.19}$) film subjected to the action of femtosecond laser pulses. The choice of the film grown on a low-symmetry substrate GaAs (311) enabled us to distinguish unambiguously between the change of the magnetic anisotropy driven by laser pulse-induced persistent strain and that occurring due to lattice heating. While both processes result in excitation of magnetization precession, we show that the trajectory of the precession differs depending on the underlying mechanism of the anisotropy change. Furthermore, we demonstrate that the strain-driven change of magnetic anisotropy dominates the response of the metallic film when the external magnetic field strength is large and the heating-driven mechanism becomes ineffective.

2. EXPERIMENTAL

Polycrystalline 100 nm thick film of a Galfenol alloy $Fe_{0.81}Ga_{0.19}$ was grown on the (311)-oriented GaAs substrate (Fig. 1(a)) by the magnetron sputtering technique. The misorientation of crystallographic axes of crystallites, average size of which was of a few nanometers, was not exceeding a few degrees. Therefore, below we consider the studied film as the single crystalline one. The SQUID measurements confirmed that the easy magnetization axis of the film is oriented in the film plane along the $[0\bar{1}1]$ crystallographic axis (y-axis). In our experiments external DC magnetic field $\mathbf{B}$ was applied in the sample plane along the magnetization hard axis, which lies along $[\bar{2}33]$ crystallographic direction (x-axis). In this geometry magnetization $\mathbf{M}$ orients along the applied field if the latter strength exceeds B=150 mT. At lower field strength magnetization is along an intermediate direction between the x- and y-axes.

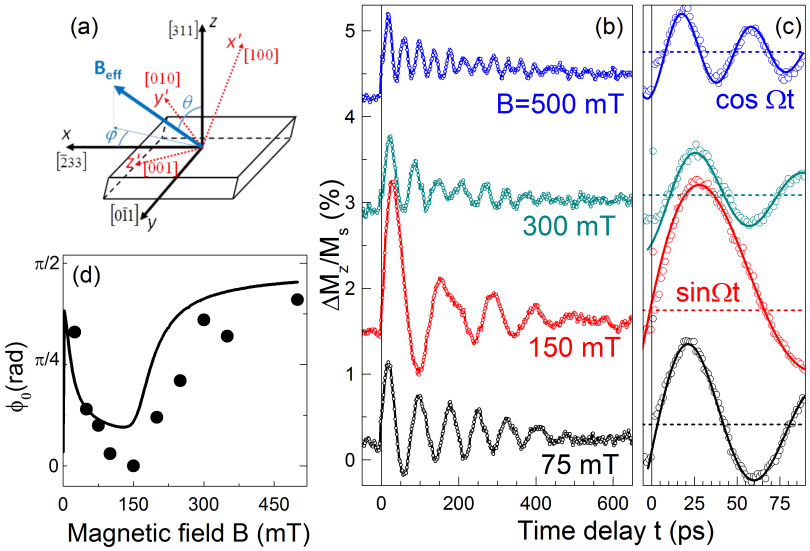


Figure 1. (a) Schematic presentation of the Galfenol film grown on the (311) GaAs substrate. x'-, y'- and z'-axes are directed along the crystallographic [100], [010] and [001] axes, respectively. (b) Laser-induced changes of the normalized out-of-plane component of magnetization M_z/M_s as a function of the pump-probe time delay t measured at various strengthes of the applied magnetic field B. (c) The same dependences measured with higher resolution (symbols) and their fit (solid lines) using Eq. 1. (d) Field dependence of the initial phase of the oscillation ϕ_0 of the out-of-plane component of magnetization as obtained from the fit using Eq. 1(symbols) and from calculation (solid line).

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Studies of the laser-induced magnetization dynamics in the $Fe_{0.81}Ga_{0.19}$ film were performed using the conventional femtosecond pump-probe technique described in details elsewhere.[14] Linearly polarized pump pulses with a duration of 200 fs, central wavelength of 1030 nm, and fluence of 2-20 mJ/cm^2 were used to excite the Galfenol film. Linearly polarized probe pulses split from the same beam and having the flunece of 10 μJ/cm^2 were used to monitor the temporal evolution of the out-of-plane component M_z of the film magnetization via changes in magneto-optical Kerr rotation (MOKE). All measurements were performed at room temperature.

3. RESULTS AND DISCUSSION

Fig. 1(b) shows the temporal evolution of the MOKE signal following excitation of the sample by femtosecond laser pulses. There are clear oscillatory component in the observed signal, which can be approximated by the function

$$\theta(t) = Ae^{-t/\tau}\sin(2\pi\Omega t + \phi_0). \quad (1)$$

As can be seen the frequency Ω and the amplitude A of the oscillations are dependent on the applied field strength thus confirming that the latter originates from the laser-induced magnetization precession. The most striking result is that the initial phase ϕ_0 of the oscillations possesses nontrivial field dependence (Fig. 1(c)). This dependence shown in detail in Fig. 1(d) captures how the trace of the magnetization precession changes when the field is increased. Keeping in mind that at $t = 0$ both the magnetization easy axis and the applied field $\mathbf{B}$ are oriented in the film plane, one can conclude that pure *sine*-like ($\phi_0 = 0$) temporal evolution of the MOKE signal at the applied field of B=150,T corresponds to the magnetization precessing around the transient effective field $\mathbf{B}_{\text{eff}}$, which lies in the sample plane. By contrast, pure *cosine*-like ($\phi_0 = \pi/2$) behavior of the laser-induced MOKE corresponds to the precession of the magnetization around $\mathbf{B}_{\text{eff}}$, having finite out-of-plane component.

The laser-induced magnetization precession in the Galfenol film grown on a (311)-GaAs substrate can be described by Landau-Lifshitz equation[15]

$$\frac{d\mathbf{m}}{dt} = -\gamma\mathbf{m}\times\mathbf{B}_{\text{eff}}(\mathbf{m},t), \quad (2)$$

where $\mathbf{m} = \mathbf{M}/M_s$ is the normalized magnetization, γ is the gyromagnetic ratio, and $\mathbf{B}_{\text{eff}}(\mathbf{m},t) = -\nabla_M F_M(\mathbf{m},t)$ is the time-dependent effective magnetic field. The magnetic part of the free energy of the Galfenol film grown on the (311)-GaAs substrate can be expressed as

$$\begin{aligned} F_M(\mathbf{m},t) &= -\mathbf{m}\cdot\mathbf{B} + B_d m_z^2 + K_1\left(m_{x'}^2 m_{y'}^2 + m_{z'}^2 m_{y'}^2 + m_{x'}^2 m_{z'}^2\right) - K_u m_y^2 + \\ &\quad b_1(\varepsilon_{x'x'}m_{x'}^2 + \varepsilon_{y'y'}m_{y'}^2 + \varepsilon_{z'z'}m_{z'}^2) + b_2(\varepsilon_{x'y'}m_{x'}m_{y'} + \varepsilon_{x'z'}m_{x'}m_{z'} + \varepsilon_{y'z'}m_{y'}m_{z'}). \end{aligned} \quad (3)$$

Here for a sake of convenience Zeeman, shape and uniaxial anisotropy terms are written in the coordinate frame associated with the film, i.e the z-axis is directed along the sample normal. Cubic anisotropy term and the magneto-elastic terms are written in the frame given by the crystallographic axes (Fig. 1(a)). The equilibrium values of the saturation magnetization M_s=1.59 T, the magnetic anisotropy constants K_1=30 mT, K_u=45 mT, the magneto-elastic coefficients b_1=-6 T, b_2=2 T were found using literature data[16–18] as well as from the fit of the field dependence of the precession frequency.[14] Strain components ε_{ij} are considered to be zero at equilibrium.

In general, under the action of the laser pulse the rapid increase of the electron temperature leads to ultrafast demagnetization,[19] along with a rapid increase of the lattice temperature and thermal stress generation. Since in our experiment both easy magnetization axis and external magnetic field lie in the film plane the change of the demagnetizing field due to ultrafast demagnetization is not expected to trigger the precession.[2] Therefore, we ascribe the observed precession excitation to the rapid change of magnetic anisotropy. The non-monotonous dependence of the phase ϕ_0 of the M_z oscillations suggests that there is a competition between different mechanism responsible for the change of magnetic anisotropy.

We modelled the impact of the a 200 fs laser pulse of a fluence of 10 mJ/cm^2 on the (311) Galfenol film as an instantaneous step-like increase of the lattice temperature by 120 K, which is justified by the fact that the characteristic time of such change is of a few picosecond, while relaxation to the equilibrium value requires microseconds (see Ref. 14 for details). The corresponding thermal stress leads to the persistent compressive

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$\varepsilon_{zz} = 1.2 \cdot 10^{-3}$ and shear $\varepsilon_{xz} = -4 \cdot 10^{-4}$ strain.[20] With these values we were able to model using Eqs. 2-3 the precession of magnetization excited due to strain-induced magnetic anisotropy change. Additionally, fitting the experimental data to these equations yielded the changes of the cubic and uniaxial anisotropy constants ΔK_1=-4.75 mT and ΔK_u=-2.2 mT. This lead a good agreement with the experimental data, as illustrated in Fig. 1(d) for the case of the initial phase of the oscillations ϕ_0.

Therefore, employing analytical calculations and minimal fitting procedure we were able to describe the response of the (311) Galfenol film to the action of the laser pulse and distinguish two mechanisms responsible for the magnetization precession excitation. In particular, from the analysis it follows that both laser-induced changes of magnetocrystalline anisotropy constants ΔK_1, ΔK_u and persistent strain ε_{zz}, ε_{xz} lead to the in-plane and out-of-plane tilt of the effective magnetic field $\mathbf{B}_{\text{eff}}(\mathbf{m}, t)$. However, the ratio between these tilts depends strongly on the strength of the applied magnetic field. At B=150 mT the in-plane tilt related to ΔK_1, ΔK_u dominates. In experiment it is seen as the pure *sine*-like oscillations of the out-of-plane component of magnetization (Fig. 1(c)). By contrast, at the applied field of B=500 mT the out-of-plane tilt of $\mathbf{B}_{\text{eff}}$ due to the laser-induced strain governs the magnetization response. This is seen as the pure *cosine*-like oscillations of the out-of-plane component of magnetization (Fig. 1(c)).

Importantly, our analysis shows that the laser-induced change of ΔK_1, ΔK_u effectively trigger the precession in the range of relatively low strength of magnetic fields applied along the hard magnetization axis, while at high fields the effect of the laser-induced strain dominates. Indeed, magnetic anisotropy favors orientation of the magnetization along the easy direction, while applied magnetic field pulls it towards the hard one. The laser-induced lattice heating changes K_1 and K_u, thus mostly reducing the anisotropy strength without changing the easy axis orientation. When the applied magnetic field is low, there is a delicate balance between the anisotropy and applied field, which is easily altered by reduction of K_1 and K_u due to lattice heating. By contrast, at high magnetic field the orientation of the magnetization is dictated mostly by applied field and, thus, is less sensitive to laser-induced changes of K_1 and K_u. The effect of the laser-induced strain on the magnetic anisotropy in the low-symmetry film is somewhat different. The compressive ε_{zz} and shear ε_{xz} strains alter the direction of the easy magnetization axis and, therefore, affect the magnetization direction even at relatively high applied magnetic fields. This conclusion is confirmed further by studying experimentally and analytically laser-induced magnetization precession in a Galfenol film grown on a high-symmetry GaAs substrate, where the laser-generated strain does not alter the magnetization axis orientation.[14] As a result, the laser-induced changes of ΔK_1, ΔK_u define the response of such sample.

4. CONCLUSIONS

In conclusion, we have demonstrated experimentally laser-induced excitation of the magnetization precession in magnetostrictive Galfenol film grown on the (311)-oriented GaAs substrate. By analysing the effect of laser-induced increase of the lattice temperature and laser-induced strain on the magnetic anisotropy we show that both mechanisms can effectively trigger the magnetization precession. The increase of the lattice temperature, i.e. incoherent phonons generation, alters the magnetocrystalline anisotropy constants. The laser-induced compressive and shear persistent strains, also seen as coherent phonons, change the magnetic anisotropy via inverse magnetostriction. We determine conditions at which either of mechanisms dominates, and show that it is the low symmetry of the studied film which enabled control of magnetic anisotropy via laser-induced persistent strain.

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