ULTRAFAST OPTICAL DIAGNOSTICS OF SURFACE/INTERFACE MAGNETIZATISM AND MAGNETIZATION REORIENTATION IN Fe FILM AND NANOPARTICLE ARRAYS

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Introduction

In the last decade the research in magnetism is shifting from bulk materials towards thin films and nanostructures. Not only have fabrication techniques improved to the extent that it has become possible to make flat magnetic layers of only a few atomic layers thick, but also nanostructures with even smaller dimensions, such as wires and dots [1]. Nanomagnets behave very differently from their bulk counterparts. This is mainly because new crystallographic structures can be stabilized at the nanoscale and, the atoms at the surfaces and edges occupy a larger fraction of the total volume. These atoms experience electric and magnetic fields of different symmetry compared to those within the interior of the material. The atoms possessing a different magnetic moment and magneto - crystalline anisotropy are expected to modify significantly the overall behavior of the nanoparticles. For example, as the size of the structural building units becomes comparable with a characteristic intrinsic magnetic length scale of the system (domain wall width, magnetic exchange length, or critical single domain size), novel effects may emerge. The exchange bias and the giant magnetoresistance in multilayer thin films [2], enhanced magnetocaloric effects in nanocomposites [3], and exchange-spring behavior in hard-soft exchange-coupled nanomagnets [4] are just a few examples.

The ongoing studies of nanomagnets are making it more apparent that the knowledge of their surface /interface properties and their dynamical responses play a decisive role in understanding fundamental magnetic properties of nanoscale metarials and in the enhanced performance of nanoscale devices. The very interest is now devoted to the characterization of the surface/interface and the dynamical magnetic properties of such small entities and it has become clear that more work needs to be done to understand and control the physical parameters involved. One primary issue is the precessional switching of the magnetization in moderate magnetic fields. This circumvents the need for complex devices for the generation of ultrafast magnetic field pulses with strong enough amplitude and could bring the switching speed into the femtosecond regime. Another issue is the surface/interface magnetism and the sources of spin-flipping in the multilayers and/or the arrays of nanoelements, which may occur either in the magnetic units or at the interfaces between the units. In the past, magnetization precession and spin flipping of nanoscale magnetic elements have been measured or inferred by techniques such as ferromagnetic resonance and spin-polarized electron spectroscopies [5]. However, investigations of magnetization dynamics have been largely limited and there are still fundamental questions regarding collective spin excitations and spin-dependent transport in these systems. In particular, experimental data on collective magnetic excitations and spin relaxations, affected by intergranular exchange interactions, in arrays of nanoelements are scarce. Since the intergranular interactions directly mediate magnetic percolation in the magnetic nanostructures, the spin and magnetization dynamics are strongly dependent on the topological distribution of magnetic elements. As collective spin excitations and relaxation dynamics appear to play an essential role for the device properties, a detailed understanding of

the surface /interface magnetism and spin coherence is needed.

In this paper, we report the magnetization reversal and demagnetization processes of the Fe interface layer in thin films and nanoparticles of Fe/Al and Fe/Pt by the magnetization-induced second-harmonic generation techniques. The results are compared with those of the bulk magnetization as obtained from magneto-optic Kerr effect (MOKE). We realize that switching and demagnetization characteristics are distinctly different between bulk and interface layers because of the interface-derived anisotropy and the dipole interactions. In particular, the surface and interface magnetism will dominate the behaviors of nanoscale structures. The all-optical pump-probe technique using the second-harmonic generation (SHG) and the Kerr effect was a major breakthrough in time-resolved studies of magnetization dynamics, which extended time resolution into the sub picosecond range [6, 7]. In contrast to frequency domain techniques, time resolved measurements allow direct observation of the basic time constants of magnetization dynamics with inherent time resolution using femtosecond laser pulses. The magnetization precession and relaxation after intensive pump-laser pulse excitation is followed by ultrafast demagnetization, which happens shortly after the pump laser reaches the sample, and by the coherent and incoherent collective relaxation modes, which happen on the ns time scale. The detection of magnetization precession and relaxation is mostly based on the magnetic contrast of the probe light in the visible wavelength regime. The polarization change of the light reflected from the ferromagnetic sample is proportional to the magnetization of the material due to the spin-orbit coupling. The *femtosecond*-laser pulses can induce magnetization switching and control the magnetization reversal in nanostructures. The recent development of compact ultrafast laser systems and successful incorporation of ultrafast all-optical magnetization reversal might spur the realization of a new generation of ultrafast magnetic recording devices.

Experimental

Materials:

The Fe and Fe/Al films were prepared on chemically etched Si (111) substrates in a magnetron sputtering system with a base pressure of 4.6×10^{-5} Pa, and the argon gas pressure was kept at 0.3 Pa during sputtering. The deposition rates were 0.08 nm/s for Fe and 0.1 nm/s for Al, respectively. Fe nanodot arrays were prepared by electron beam evaporation through porous anodic alumina templates followed by the removal of the templates [8]. The nominal total thickness was varied from 50 Å to 200 Å.

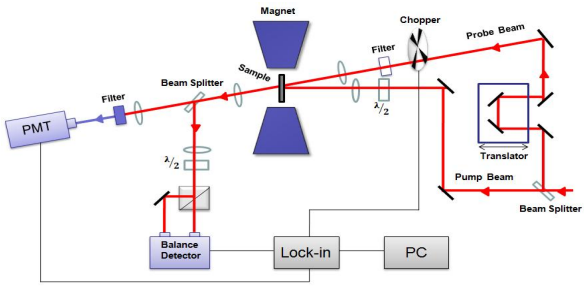

Fig. 1: Experimental setup used for the time-resolved MOFE and MSHG measurements

Setups for magnetization dynamics:

Information about spin dynamics is obtained from pump-probe magneto-optical Kerr/Faraday effect measurements. In our experiments, the laser pulses delivered by the mode-locked Ti-Sapphire system with a pulse width of ~ 70 fs were focused on the sample. The wavelength was tuned to ~ 800 nm with its peak energy of ~ 10 nJ per pulse, repeating at 82 MHz. The schematic diagram is shown in Fig. 1. The pump beam created a non-equilibrium spin population, and the subsequent magnetization precession and spin-relaxation were probed by transient Kerr-rotation (or Faraday rotation) of a linearly-polarized probe beam as a function of time delay between the two pulses. The pump beam perturbs the spin states by “thermal” pumping, in which a hot electron gas is excited, which heats the lattice and causes a change of the magnetization anisotropy. The pump beam is modulated by an

optical chopper. The probe beam is monitored either in transmission or reflection. The resulting Kerr (in reflection) or Faraday (in transmission) signals induced by the pump (chopper) frequency are picked up via a balanced photoreceiver and a lock-in amplifier. For MSHG measurements, the s- or p-polarization was used for the incoming light with an angle of incidence of about 45 degrees. Regarding the outgoing light from the sample: the fundamental light was reflected and only the SH light generated in the sample was detected by a photo multiple tube with a lock-in amplifier.

Results and Discussion

To demonstrate the capability of MSHG technique on studying the surface and interface effects on magnetic properties of thin films, we first show our results on MSHG measurements on a polycrystalline sputtered ultrathin *Fe* film sample. The application of MOKE to ultrathin films sometimes is named surface magnetooptical Kerr effect. However, the measured Kerr signal is an average over the entire layer stack within the penetration depth of the light of several 10 nm for metallic films. There is no special surface or interface sensitivity. Often the magnetic properties at surfaces and interfaces are different from those of the interior of the magnetic film. These effects are largely masked by the Kerr signal from the interior of the films in MOKE measurements. Recently, optical SHG has been developed as a sensitive method to probe surface and interface structures. For centrosymmetric materials, SHG is forbidden in the bulk within the electric dipole approximation and therefore the second harmonic (SH) light is generated near the surface or interface, where the inversion symmetry is necessarily broken.

The magnetization-induced variations of the SHG intensity and rotation of the second-harmonic wave polarization has been revealed to exceed the linear magneto-optical Kerr effect by orders of magnitude. Therefore MSHG is attracting attention as an ultra-sensitive tool for investigating the surfaces and interfaces of magnetic film and superlattice structures.

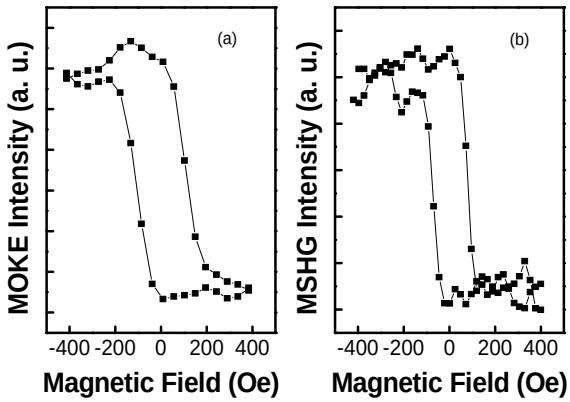

Fig. 2 MSHG and MOKE hysteresis curves: MSHG intensity for Fe/Al film measured in longitudinal geometry, with p-polarized incidence and an analyzer angle of 45 degrees relative to s-polarization. (a) shows MOKE loop: 25-nm Fe film with a cap layer of Al (5-nm). (b) shows its relative MSHG loops.

Figure 2 shows the hysteresis curves of MSHG and MOKE intensity as a function of magnetic field in a 25-nm *Fe* film with a *Al* cap layer of ~ 5 nm. The incident fundamental light was p-polarized, and the SH light was detected behind an analyzer, which was set at 45 degrees related to s-polarization. We observe that the MOKE hysteresis loop for the 25-nm *Fe* film has a greatly different shape, compared with the MSHG curve: The coercivity of magnetic film decreases almost 40 % and a much sharper magnetization reversal is observed in the MSHG loop. The results are consistent with the fact that *MOKE gives a signal from the whole thin film, while the MSHG signal comes only from the Fe surface and Fe-Al interface.*

Moreover, we investigated the magnetization reorientation induced by subpicosecond optical pulses in the two dimensional arrays of Fe nanoparticles. Figure 3 shows the pump-probe Faraday rotation data of 50 nm *Fe* nanoparticle arrays under Voigt geometry. The probe light propagates at an angle of incidence of ~ 65^{o} with respect to the external magnetic field. The transient magnetic hysteresis loops were acquired with the probe beam

following excitation by the pump. The change of hysteresis loops was recorded for different pump-probe delays at a fixed pump fluence of ~ 25 μJ/cm^2. Before the arrival of pump pulses, we observe a simple square shaped loop as expected for the easy axis (an external magnetic field is applied parallel to the in-plane easy axis direction of the Fe arrays). After the pump pulse excitation, the Faraday rotation at high field ends loses its intensity in addition to the change of the coercivity field. The hysteresis loop tends to be a near diamond shape at 100 fs. The loop then relaxes and it recovers the original square shape at 600 fs. The development of hysteresis loops could be explained by the ultrafast magnetization reorientation. Based on the analysis of laser-induced magnetization hysteresis loops, we show a feasibility of ultrafast optical control of magnetization switching. Judging from the ultrafast dynamics much shorter than ns, we suggest that the mechanisms of optomagnetic interactions do not rely on laser-induced heating but have a nonthermal origin.

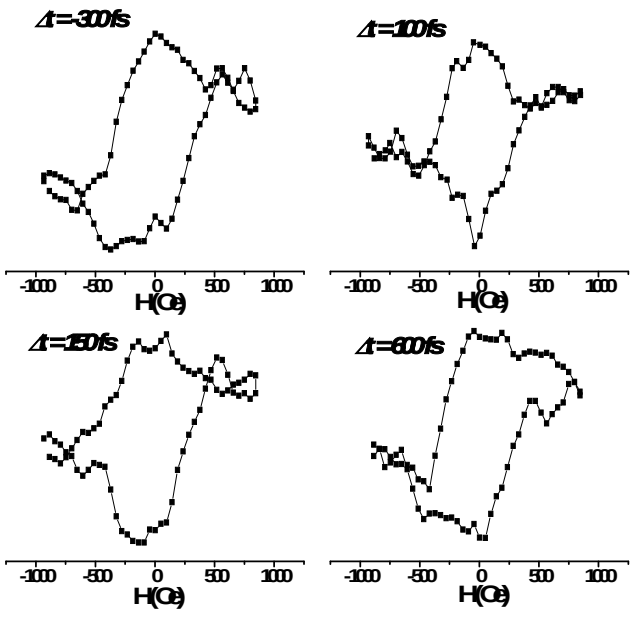

Fig. 3 Time-resolved Faraday rotation on 50-nm Fe magnetic nanoparticle arrays with a cap layer of Pt (5-nm).

Conclusion

We studied the surface/interface magnetism, magnetization reorientation and demagnetization processes in a Fe/Al thin film and 2D Fe nanoparticle arrays by the MOFE and MSHG techniques. We realize that switching and demagnetization characteristics are distinctly different between bulk and interface layers because of the interface-derived anisotropy and the dipole interactions. The all-optical pump-probe technique using the Faraday effect enables us to understand the magnetization dynamics in the sub picosecond range.

References

1. S. Sun, C. B. Murray, D. Weller, L. Folks, and A. Moser, Monodisperse FePt nanoparticles and ferromagnetic FePt nanocrystal superlattices, *Science* **287**, (2000) 1989-1992.
2. Aharoni, *Introduction to the Theory of Ferromagnetism* (Oxford University Press, Oxford, 1996).
3. R. D. McMichael, J. J. Ritter, and R. D. Shull, Enhanced magnetocaloric effect in $Gd_3Ga_{5-x}Fe_xO_{12}$, *J. Appl. Phys*, **73**, (1993) 6946 - 6948.
4. Zeng, H., Li, J., Liu, J. P., et al., Exchange-coupled nanocomposite magnets by nanoparticle self-assembly, Nature **420** (6914), (2002) 395-398.
5. Ren, Y. H., Wu, C., Yu, G., Pettiford, C., and Sun, N. X., Magnetic anisotropy and spin wave relaxation in CoFe/PtMn/CoFe trilayer films, *J. Appl. Phys*, **105**, (2009) 073910.
6. M. van Kampen, C. Jozsa, J. T. Kohlhepp, P. LeClair, L. Lagae, W. J. M. de Jonge, and B. Koopmans, All-optical probe of coherent spin waves, *Phys. Rev. Lett.* **88**, (2002) 227201.
7. Ru-Pin Pan, H. D. Wei, and Y. R. Shen, Optical second-harmonic generation from magnetized surfaces, *Phys. Rev. B* **39**, (1989) 1229 -1234.
8. Kim, C., Loedding, T, Jang, S, Zeng, H., Li, Z., Sui, Y. C., and Sellmyer, D. J., FePt nanodot arrays with perpendicular easy axis, large coercivity, and extremely high density, *Appl. Phys. Lett.* **91**, (2007) 172508.