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Ultrafast reflectivity and electron dynamic properties of $Tb_{0.27}Dy_{0.73}Fe_2$ thin films

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ABSTRACT

The ultrafast optical properties and electron dynamics of $Tb_{0.27}Dy_{0.73}Fe_2$ thin film were studied using the femtosecond pump–probe technology. Experimental results revealed that the extremum of $\Delta R/R$ curves corresponding to the electron thermalization occurred around 100 fs, and with the decrease of thin film thickness, the average rate of reflectivity equilibration after extremum became faster. Amplitude of $\Delta R/R$ curves reach its maximum at 160 mW pump fluence and it turns zero at 0 mW. Echo waves were found in reflectivity waveform and the smaller the thickness of the thin film, the earlier the echo occurs. Time variation of the complex dielectric constant indicates that the absorption of thin films became greater with the increase of reflection at the surface after 0 ps delay time, but absorption of the thin film recovered to its initial state after 1 ps.

1. Introduction

Since $Tb_{0.27}Dy_{0.73}Fe_2$ alloy was found to exhibit much larger magnetostriction coefficient than other magnetostrictive materials by Clark [1] in 1973, much effort has been put into enhancing its practicability [2–5]. The major requirement for application in microsystem technology is that these materials should be prepared in the form of film and should have sufficient magnetostriction at low magnetic fields. The continuing development of magnetostrictive devices into nanoscale regime and terahertz frequency has emphasized the importance of understanding intrinsic electron dynamics on increasingly shorter time scales (picosecond and femtosecond (fs) time scale). Although the optical and electron dynamic properties of TbDyFe thin films may provide key information to their application for high-frequency MEMS sensors and actuators devices, they have not been systematically studied yet for the reason of the transition process of the reflection properties that occurred in several hundreds fs time scales and it is impossible to measure this process through the general measuring method. However, with the rapid progress of the fs laser technology in the 1990s, fs laser pump–probe technology has introduced into the domain of ultrafast measurement of the reflection transition process of thin film and bulk material successfully [6]. In this paper, the fs laser pump–probe experimental setup was designed to investigate ultrafast reflection and electron dynamics characteristics of TbDyFe thin films after the fs laser illuminated and the experiment results indicate that it was strongly influenced by different film thicknesses and different pump fluences. Also the time variation of complex dielectric constant of the thin film was calculated in order to observe the absorption characteristic of thin films.

2. Sample preparation and experimental setup

$Tb_{0.27}Dy_{0.73}Fe_2$ thin films were prepared by DC magnetron sputtering at a deposition rate of 0.3 nm/s. The substrates were $2\times2\,cm^2$ polished silicon slice (1 0 0) with a thickness of 0.2 mm without heating during sputtering. The sputtering power was 60 W and the vacuum chamber pressure was held at 10^{-5} Pa before and at 3 Pa during the argon gas flow. Thicknesses of four series of films were measured by an Ambios XP-2 profiler and the results are 200, 400, 600 and 800 nm. AFM images of the as-deposited thin films are shown in Fig. 1 in order to illustrate the surface morphology and roughness.

In our pump–probe experimental setup of studying the reflectivity and the electron dynamics of TbDyFe thin films, the transient change in the electron system as excited by an intense pump pulse is probed by measuring the intensity after reflection of the sample with a weaker (probe) laser pulse that follows the pump at a variable delay Δt. The experiments reported here were performed with a commercial Ti:sapphire laser system, operating at 800 nm and at 82 MHz repetition rate with a pulse width of

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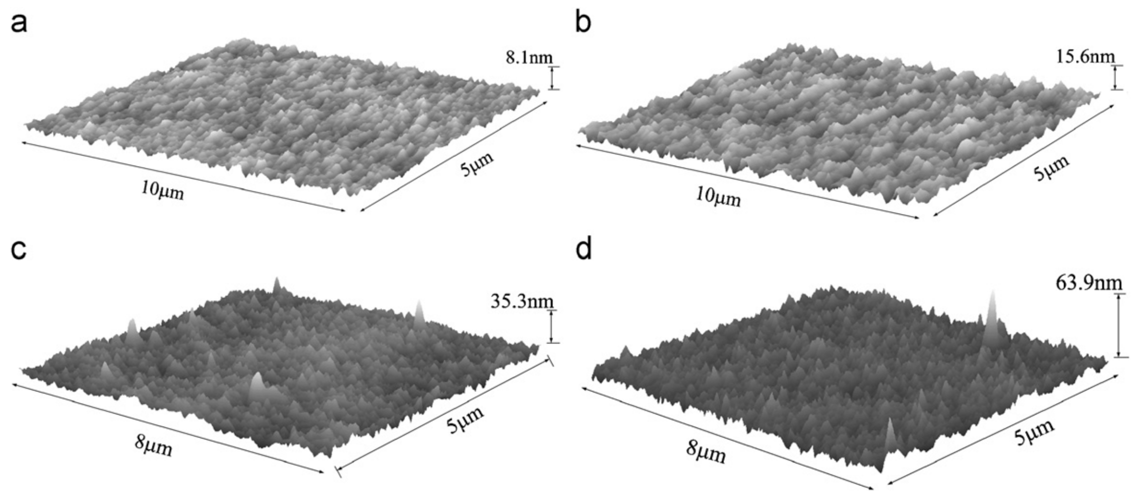

Fig. 1. AFM images of the four series thin films with different thicknesses. (a) 200 nm, (b) 400 nm, (c) 600 nm and (d) 800 nm.

30 fs and single pulse energy of 6 nJ. In pump–probe experiments, the main laser beam was split into two parts with a ratio of 10:1. Both of them were focused on the sample by means of a convex lens. A CCD camera, equipped with a microscope objective, was used to optimize the overlap of the spots. The incidence angle was 45° for the p-polarized probe pulse and 90° for the s-polarized pump pulse. When measuring the transient reflectivity, the pump beam is modulated with a chopper and the induced intensity changes in the reflected probe are detected using a lock-in amplifier and then the experimental data were picked up by a computer.

3. Result and discussion

As shown in Fig. 2, the relative change in reflectivity, $\Delta R/R$, is plotted versus pump–probe delay Δt. Before 0 ps, the probe pulse arrives at the surface prior to the pump pulse and the reflectivity change remains constant at a low level. When the pump and the probe pulses arrive at the surface simultaneously at 0 ps, a strong reflectivity change with sharp peak can be identified. For trailing edge of the sharp transition, the average rate of equilibration reflectivity after extremum, from 2 ps of 800 nm to 0.3 ps of 200 nm, with the decrease in thickness is faster.

In the experiment, there is no thermal insulated barrier layer between the thin film and the silicon substrate. Hot electron transport and heat diffusion led to energy loss from the laser-irradiated volume. From the three-temperature model of ferromagnetic metal [6], after laser excitation, the combined effect of mass electron–electron scattering events led to the fast thermalization of electron gas and electron spin effect. Former research of ferromagnetic metal [7,8] demonstrated that the reflectivity relative change, $\Delta R/R$, was approximately proportional to the electron temperature change; therefore, electron thermalization was expected to present a sharp peak around zero probe delay (with a rise time given by the temporal time resolution). When the energy of highly excited electrons got close to the Fermi level (photon energy is 1.6 eV in our experiment), the electron–phonon scattering processes start to dominate. Temperature difference between the electron (Te), lattice (Tl) and spin (Ts) system acted as the driving force behind energy transfer, Te was decreased and eventually equalized the temperature among the three systems. The electron temperature equilibration can be regarded as exponential decays process, the reflectivity change also decay exponentially as shown in Fig. 2 and became steady after 0.3–2 ps. Although the electron–phonon relaxation situated in the picoseconds range play a principal role, following the specific features of the spin-polarized iron containing ions' principal role in the electron–phonon interactions play inharmonic interactions [9] and lead to the irregular fluctuation of reflectivity curves in Fig. 2.

Contribution of the electron thermalization generated an abrupt change of reflectivity. Considering different reflectivity measurements and the temporal resolution, $\Delta R/R$ extremum occurred around 100 fs. It indicated that this value was decreased obviously when compared with the former research result of ferromagnetic thin film of 220 fs [8] (using 85 fs pulses), 260 fs [6] (60 fs pulses) and 280 fs [10] (150 fs pulses). Here, we may ascribe this phenomenon to the local 4f electrons in rare-earth ions that doped in thin film and the improvement of temporal resolution of the pump–probe setup. With the increase of film thickness, hot electron transport will induce energy diffusion from the laser-irradiated surface layer to the deeper level and more electron, lattice and spin systems will be involved in the process of temperature recovery. It could cause the equilibration process of reflectivity curves more tardily as clearly shown in Fig. 2.

The wavelike phenomenon was observed in the equilibration process of the reflectivity change, particularly in 200 and 400 nm thin films. The undulation curves were related to strain waves in the sample that are caused by rapid heating of the thin film. Rapid laser heating generates thermal stress in thin films and then the elastic stress pulse was induced by lattice offset through electron–phonon scattering. The reflected part of a compressive stress pulse, which was generated at the film substrate interface, propagates out of the film surface, which is called the echo wave [11]. When we compared it with the four graphs in Fig. 2, one can see that the thickness of the thin film determines the echo time, but echo wave in 800 nm was not obvious because the longer the decay distance in the thin film, the weaker the amplitude of the echo wave. Moreover, the echo waves have smaller intensity, close to zero, and became irregular when compared with Refs. [8,12]; the reason is partly echo waves in Fig. 2 were generated from the reflection and scattering of the defects near the film surface.

The reflectivity curves are proportional to the electronic temperature, as it can be verified by plotting the linear behavior of the reflectivity signal versus the different pump fluence. Fig. 3