Ultrafast optically induced spin dynamics in single-crystal Fe ultra thin films and patterned dot arrays

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Introduction

The information data storage and communication technologies have been growing explosively and have pushed timescales to the GHz/THz regime. Femtosecond spin dynamics experiments provide important information on spin-scattering processes at ultrafast timescales, which is essential for technological applications. The straightforward method for changing the magnetisation of a magnetic sample is through the manipulation of fast magnetic field pulses. This manipulation has a limited bandwidth set by the rise time of the magnetic field pulses which is typically tens of picoseconds [1]. On a shorter timescale (THz), the spin dynamic process can be induced by excitation using ultrafast laser pulses. The idea is that a rapid elevation of the spin temperature produces a decrease of the magnetisation as the Curie temperature is approached. Beaurepaire and his co-workers reported their pioneering work on ultrafast magnetisation dynamics induced by femtosecond laser pulses in ferromagnetic transition metals in 1996 [2]. They observed that the time-resolved magneto-optical contrast dropped on a timescale of 1-2ps by ~50% following the absorption of a 60 fs laser pulse. The rapid dropping was interpreted as nonequilibrium heating of the spins, electrons, and the lattice resulting in an ultrafast demagnetisation of nickel. This result surprised the scientific community because until then these processes were believed to be governed by spin-lattice relaxation which occurs at least two orders of magnitude more slowly and has been considered to be the speed limit of magneto-optical technology.

In this report, we have performed time-resolved pump-probe MOKE (magneto-optical Kerr effect) measurements on single crystal Fe ultra thin films and patterned dot arrays with feature sizes down to 50nm using a stroboscopic set-up. We address the factors controlling the recovery of the magnetization after the femtosecond laser excitation, the role of the anisotropy and the patterning effect in the ultrafast spin dynamics process.

Experiments

A time resolved MOKE (TRMOKE) setup capable of separating the longitudinal and polar MOKE responses is used for pump-probe measurements. An ultrafast Ti:Sapphire laser is used to deliver s-polarized optical pulses of 150fs duration, central wavelength of 800nm and repetition rate of 1kHz. The wavelength of the pulses in the probe path is set to 400nm whilst the wavelength of the pulses in the pump path is maintained at 800nm. The laser power of the pump beam can be varied by a neutral density wheel in the pump beam path. The pump and probe laser pulses are focused onto the same spot on the sample using a single lens and the spot diameter is around 100μm. The spot may be positioned with micrometer precision on the sample using a x-y-z translation stage and CCD camera setup. The dynamic responses were measured at room temperature using a lock-in amplifier and optical chopper placed in the pump path. The sample used for the measurements presented here is a high quality epitaxial bcc Fe film with thickness of 10ML, grown on GaAs(100) substrate by molecular beam epitaxy. The film has been grown at room temperature to have bulk-like magnetic moments down to monolayer thickness [3]. The film is capped with a 15ML thick Cr film. Using focused ion beam (FIB) etching parts of the film are patterned into 100μm square arrays of 50nm square dots with 50nm separation.

Results and discussions

MOKE measurements have determined that the Fe film has uniaxial anisotropy with the edges of the 50nm square dots parallel to the hard/easy magnetic axis of the sample as shown in Fig 1 a,b. On the other hand a modified magnetic anisotropy is observed for the Fe dot array. Along both the easy and hard axes of the film the MOKE loops from the Fe dot array now show the characteristics of intermediate axes as shown in Fig 1 c,d. The modified magnetic anisotropy of the Fe dot array is expected to be a combination of shape anisotropy and possible change in intrinsic anisotropy due to the patterning.

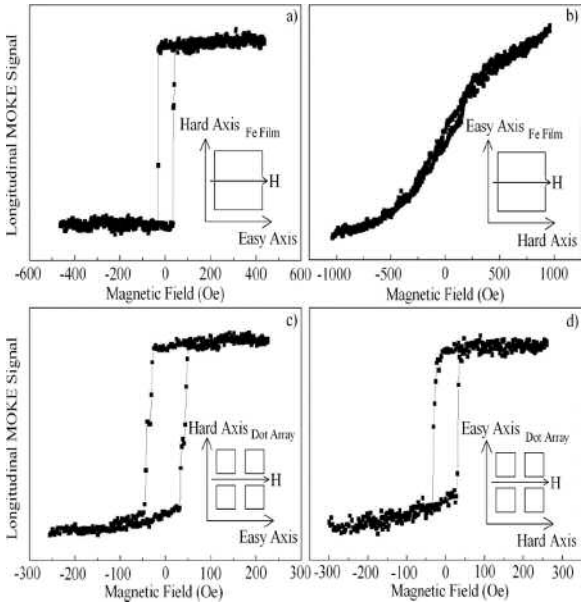

Figure 1. Longitudinal MOKE loops of the Fe film, measured along a) the easy axis and b) the hard axis, and Fe dot array measured with magnetic field applied along c) easy axis and d) hard axis of the Fe film.

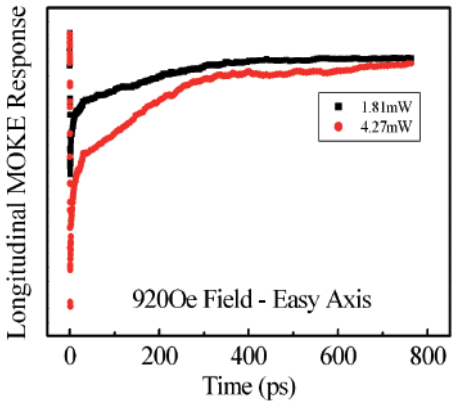

Figure 2. Longitudinal TRMOKE responses of the Fe film with magnetic field applied along the easy magnetic axis under two different pump beam power.

Spin dynamics responses of the Fe film with the magnetic field applied in the sample plane and along the easy magnetic axis are shown in Fig. 2. Here the magnetic field strength is kept fixed at 920 Oe and two different pump powers, 1.81mW and 4.27mW, are used for the measurements. The TRMOKE data from the Fe film shows that after femtosecond optical excitation an ultrafast demagnetization process is observed in the first 400fs, and then the magnetization recovers monotonically with no precession. The higher the optical excitation power is used, the higher the demagnetization rate is achieved, and the slower the magnetization recovers. This experimental result fits well with the theoretical prediction using an atomistic model. The TRMOKE measurements along the hard axis of the Fe film show similar characteristics as those obtained along the easy axis.

Spin dynamics responses of the Fe dot array with the magnetic field applied in the sample plane and along the hard magnetic axis of the Fe film are shown in Fig. 3. Here again the magnetic field strength is kept fixed at 920 Oe and the pump power is varied from 1.39 mW to 4.12 mW for the measurements. The TRMOKE data from the dot arrays magnetized along the hard axis of the Fe film again show an ultrafast demagnetization in the first 400fs, then the monotonical recovery of the magnetization without precession. The demagnetization rate of the array keeps increasing with the increase of the pump power till

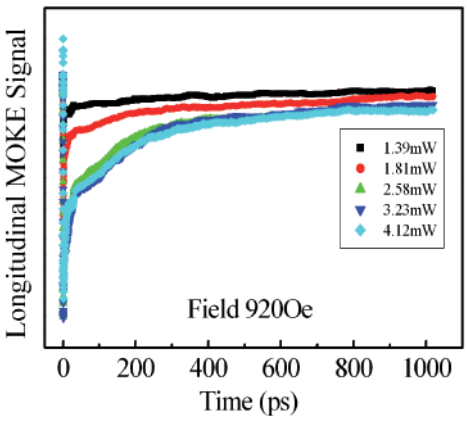

Figure 3. Longitudinal TRMOKE data of Fe dot array of feature size of 50nm magnetised along the hard axis of the Fe film under various pump powers.

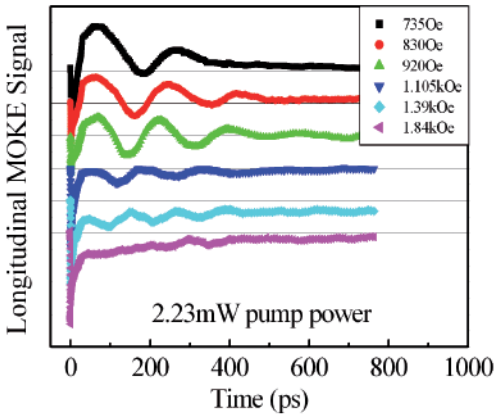

Figure 4. Longitudinal TRMOKE data of Fe dot array of feature size of 50nm at a fix pump power of 2.23mW with magnetic field of various strength applied along the easy axis of the Fe film.

the pump power approaching 2.58 mW, which indicates a possible total demagnetization of the array above this power value. The further the magnetization is reduced, the slower the magnetization recovers, similar to the results of the Fe film.

A very different optically-induced spin dynamic process occurs when the 50nm dot array is magnetized along the easy axis of the Fe film [5] as shown in Fig. 4. Here the pump power is kept fixed at 2.23mW and the magnetic field strength is varied from 735Oe up to 1.84kOe for the different measurements. After femtosecond optical excitation, following an ultrafast demagnetization in the first 400fs, the magnetization begins to recover monotonically and after about 50ps a damped precessional response is observed before the magnetization fully recovers to the initial state. The frequency of the precession increases with the increase of bias field strength whilst the damping of the precession decreases with the increase of bias field strength.

The precession frequency as a function of bias field strength is shown in the upper panel of Fig. 5. Frequency is nearly linear with the applied field. Using the measured precession frequency, f, and the exponential decay time τ, the damping constant, α, is calculated using the relation $\alpha=1/(2\pi f\tau)$ [4]. Thus the damping constant is obtained as a function of bias field as shown in the lower panel of Fig 5, with higher field values resulting in lower damping factor.

One of the features of the results shown in Fig. 4 is that the longitudinal MOKE response for the magnetic fields of 920Oe and below rises above the horizontal line, which represents the initial longitudinal state before the excitation of the pump pulse. Since the longitudinal responses are proportional to the magnetization component along the applied field direction, this suggests that the initial magnetization is not aligned along the field direction. The shape anisotropy together with the intrinsic magnetocrystalline anisotropy of the Fe dot array and applied field result in an effective magnetic field and the initial magnetization must lie along this effective field direction. The effective field direction is tilted away from the applied field direction, giving rise to the magnetization precession. Upon the sudden excitation with the pump

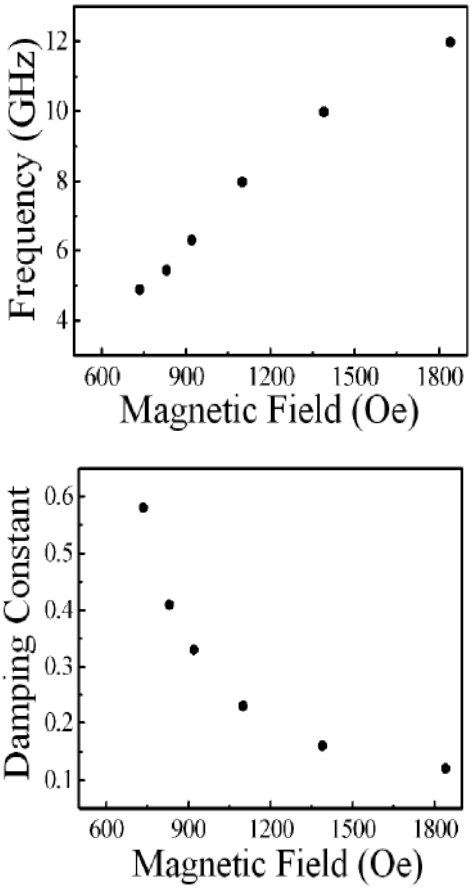

Figure 5. Measured precession frequency (upper panel) and damping constant as a function of bias field (lower panel) of the data shown in Fig 4.

pulse, rapid rise of the temperature causes not only the reduction of the ferromagnetic order but also a change in the anisotropy of the sample. This results in a change of the equilibrium orientation during the recovery process of the ultrafast demagnetization, triggering the magnetization precession around the final equilibrium orientation as shown in Fig. 4. The difference in the magnetization recovery process of the Fe dot array compared to the Fe film is expected to be related to the modified magnetic anisotropies as evidenced in Fig. 1. This shows that the modified magnetic anisotropies of the Fe dot array play a key role in triggering the magnetization precession in nano-patterned structures. However no precessions are observed in the dot array while magnetized along the hard axis of the Fe film as shown in Fig 3. From the above argument, this would suggest that when the sample is magnetised along this direction, the sudden change in the anisotropy induced by the laser excitation alter the orientation of the effective field to a much less degree so that the precessions, if there are any, are not profound. More experiments are needed to clarify this issue.

Fig. 5 is the precessional responses from the 50nm dot array magnetized along the easy axis of the Fe film under two different pump powers. The TRMOKE data shows that the higher the excitation power used, the bigger the precessional amplitude is observed. There are no obvious changes in the precessional frequency and the damping constant.

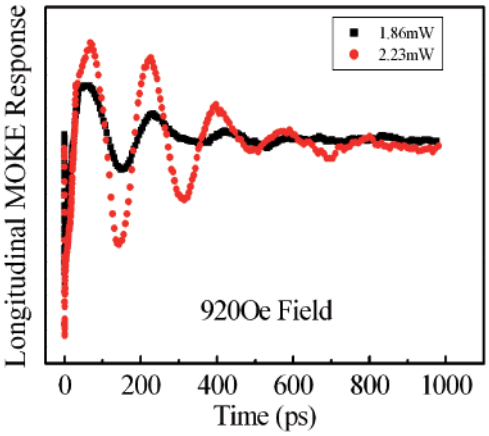

Figure 6. Longitudinal TRMOKE data of Fe dot array of feature size of 50nm magnetised along the easy axis of the Fe film under various pump powers.

Computational modeling

Computational modeling of the laser heating process has been carried out using an atomistic model [6] based on the Heisenberg form of exchange with spin dynamics described by the Landau-Lifshitz-Gilbert equation augmented by a random field term introducing thermal effects. A simple model is used to calculate the temperature of the conduction electrons (T_e) and lattice (T_{ph}). The latter heats up very slowly due to the large heat capacity, whereas T_e increases to values in excess of 1000K (depending on the laser input power). Thus it is reasonable to conclude that the laser energy is transferred to the spin system via the conduction electrons.

Figure 7 shows calculated results of the recovery time for the magnetization as a function of the minimum magnetization achieved during the heating process. Consistent with the experiments τ increases with decreasing M_{min}. This is an important finding, and the effect results from the increasingly disordered magnetic state after the demagnetization process. As M_{min} decreases an increasingly large number of nuclei of small magnetized regions are formed oriented in random directions. The slow recovery of the magnetization results from frustration effects arising from competition between regions of

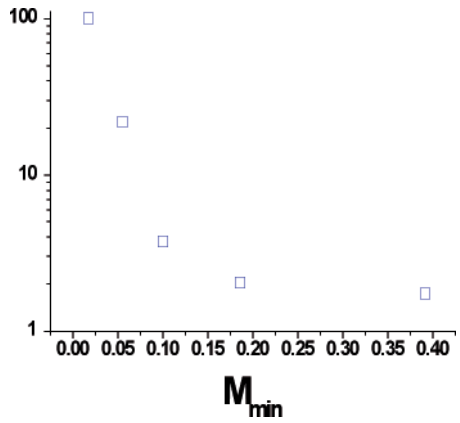

Figure 7. Recovery time for the magnetization as a function of the minimum magnetization achieved during the heating process.

magnetization recovering in different directions. A detailed comparison of theory and experiment is in progress, but this may require the extension of the atomistic calculations to larger scale using a combination of micromagnetics and the Landau-Lifshitz-Bloch (LLB) equation. The LLB equation has the property that it does not conserve the norm of the magnetization and is an excellent candidate for multi-scale calculations of pulsed laser processes[7].

Conclusion

The optically induced ultrafast response of magnetization of epitaxial bcc Fe ultrathin film (10ML) and patterned dot array with 50nm squares has been investigated by TRMOKE measurements. MOKE measurements of the Fe film have revealed a clear uniaxial anisotropy which is much reduced in patterned nanodot arrays as a result of patterning. A precessional response is observed from the Fe dot array only. In contrast to the responses of the Fe dot array, no precession is observed for the Fe thin film. Thus we have demonstrated in this work that the modified magnetic anisotropy of nano-patterned structures plays an important role in determining the magnetization relaxation dynamics following femtosecond optical excitation, which should be significant for understanding and controlling the fast switching in patterned magnetic recording media and spintronics devices.

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