Laser-induced magnetization precession in the magnetite Fe_3O_4 in the vicinity of a spin-reorientation transition

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Abstract. Using time-resolved magneto-optical pump-probe technique we demonstrate excitation of magnetization precession in a single crystalline bulk magnetite Fe_3O_4 below and in the vicinity of the Verwey and spin-reorientation (SR) phase transitions. Pronounced temperature dependence of the precession amplitude is observed suggesting that the excitation occurs via laser-driven spin-reorientation transition. Similarity observed between the characteristic features of the laser-induced ultrafast SR and Verwey transitions suggests that they both rely on the same microscopic processes.

1. Introduction

Nowadays an active experimental and theoretical research in magnetism is focused on a search of effective ways to change a magnetic state of a matter at ultrafast timescales [1]. At a spin-reorientation (SR) transition, the equilibrium orientation of magnetization is changed because of the laser-induced changes of the magnetic anisotropy [2]. It has been previously shown that the ultrafast laser heating can trigger SR transitions at a picosecond time scale [3], thus allowing ultrafast control of the media under study [4-5]. Recently, non-thermal approach to ultrafast SR transition was also realized by means of picosecond THz pulses [6].

So far, the ultrafast laser-driven SR transition has been demonstrated only in one group of magnetic rare-earth orthoferrites. In these compounds, the ultrafast SR transition relies on a laser-induced heating and consequent change of the electronic state of rare-earth ions and this mechanism limits the speed of transition to several picoseconds [7]. Thus, it remains unclear whether the SR transitions can be triggered by laser pulses in other types of materials, where these transitions are not related to this specific mechanism.

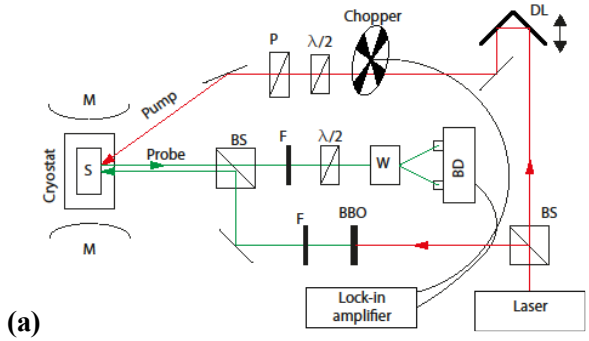
One of the model compounds possessing SR transition is the magnetite Fe_3O_4. The SR transition in single crystals of Fe_3O_4 occurs at T_{SR}=130 K and is inherently related to the changes of anisotropy from uniaxial to cubic one as the temperature is increased (for a review see [8]). The distinctive feature of magnetite is that the SR transition is observed only 7 K higher than the structural and metal-insulator transitions (Verwey transition, T_V=123 K) [9], at which the crystal structure changes from the low-temperature monoclinic to the high-temperature cubic one. Relation between equilibrium SR and Verwey transitions is still under debate [10]. Recently, the feasibility of triggering ultrafast non-thermal structural and insulator-metal transitions in Fe_3O_4 was reported [11]. It is, therefore, of high interest to examine whether the laser-driven SR transition in this material may have a non-thermal origin, and, thus, may be intrinsically fast.

Here we investigate feasibility of triggering ultrafast SR transition in magnetite Fe_3O_4. Using magneto-optical pump-probe technique we observed a laser-induced precession of magnetization at temperatures below T_{SR}, which is a typical signature of the laser-driven ultrafast SR transition.

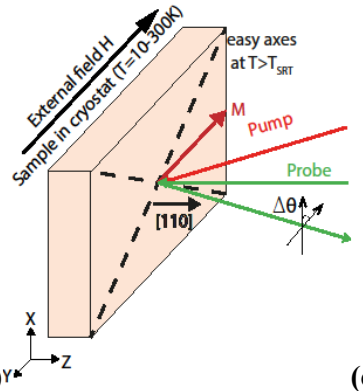
2. Experimental

We used time-resolved polar magneto-optical Kerr effect (TRMOKE) technique. Scheme of the setup and geometry of the experiment are shown in figure 1a and figure 1b, respectively. Laser pulses with a duration of 170 fs, a central wavelength of 1028nm, and a repetition rate of 100 kHz were used as pump pulses. Probe pulses central wavelength was of 514 nm. Pump pulse fluencies were in a range of 0.15 – 8.5 mJ/cm^2. Probe pulse fluencies were ~40 times lower. The measurements were carried out in the temperature range of 80 – 160 K, i.e. below, in a vicinity and above the Verwey (123 K) and SR (T_{SR}=130 K) transitions.

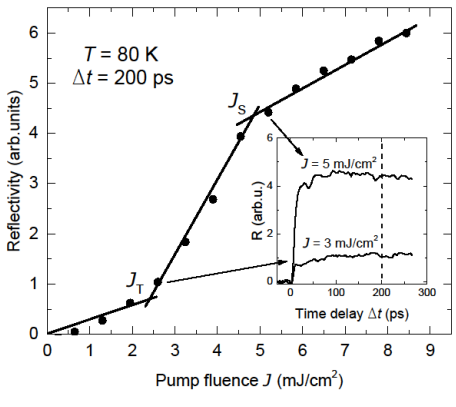
In experiments pump pulses excite the dynamics of magnetization which is revealed by measuring polarization rotation $\Delta\theta$ of the linearly polarized probe pulses reflected from the samples as a function of the pump-probe time delay Δt. In the experimental geometry shown in figure 1a, probe polarization changes are proportional to the magnetization component M_Z normal to the sample surface. Sample under study was a bulk single crystalline Fe_3O_4 grown by the floating zone method. The thick plane-parallel plate was cut perpendicularly to the [110] crystallographic axis (figure 1b). The external magnetic field H=0.3 T was applied along the [001] axis, which is the easy axis of magnetization at $T<T_{SR}$. Orientation of the easy axes at $T>T_{SR}$ are shown in figure 1b by the dashed lines. The sample surface was polished to the optical quality. The sample was fixed in a cold-finger cryostat cooled with the liquid nitrogen.



(a)



(b)



(c)

Figure 1. **(a)** Experimental setup. S – sample in the cryostat; M – electromagnet; Laser– the Yb:KGW femtosecond amplifier; DL – delay line; BBO – nonlinear crystal β-BaB_2O_4 for conversion of the probe pulses wavelength; BS – beam splitter; F – short-pass filter; λ/2 – halfwave plate; P – polarizer; W – Wollaston prism; BD – balance detector. **(b)** Geometry of the experiment. **(c)** Dependence of the pump-induced reflectivity changes on the pump fluence measured at Δt=200 ps at T=80 K. Inset shows pump-induced change of reflectivity at the two values of pump fluencies.

3. Results and discussion

First, we verified that pump pulses used in our experiment could induce the ultrafast insulator-metal transition, which is confirmed via pump-induced changes of the sample reflectivity. Figure 1c shows the pump-induced reflectivity change measured at the 200 ps delay after the excitation at T=80 K, i.e. below the Verwey transition. Inset in figure 1c shows typical temporal pump-induced evolutions of the reflectivity observed at the two pump fluencies. Nonmonotonous fluence dependence of the reflectivity change (figure 1c) is a typical manifestation of the laser-induced insulator-metal transition [11], where J_T is the threshold fluence and J_S is the saturation fluence at which the excited material is completely turned into the metallic state.

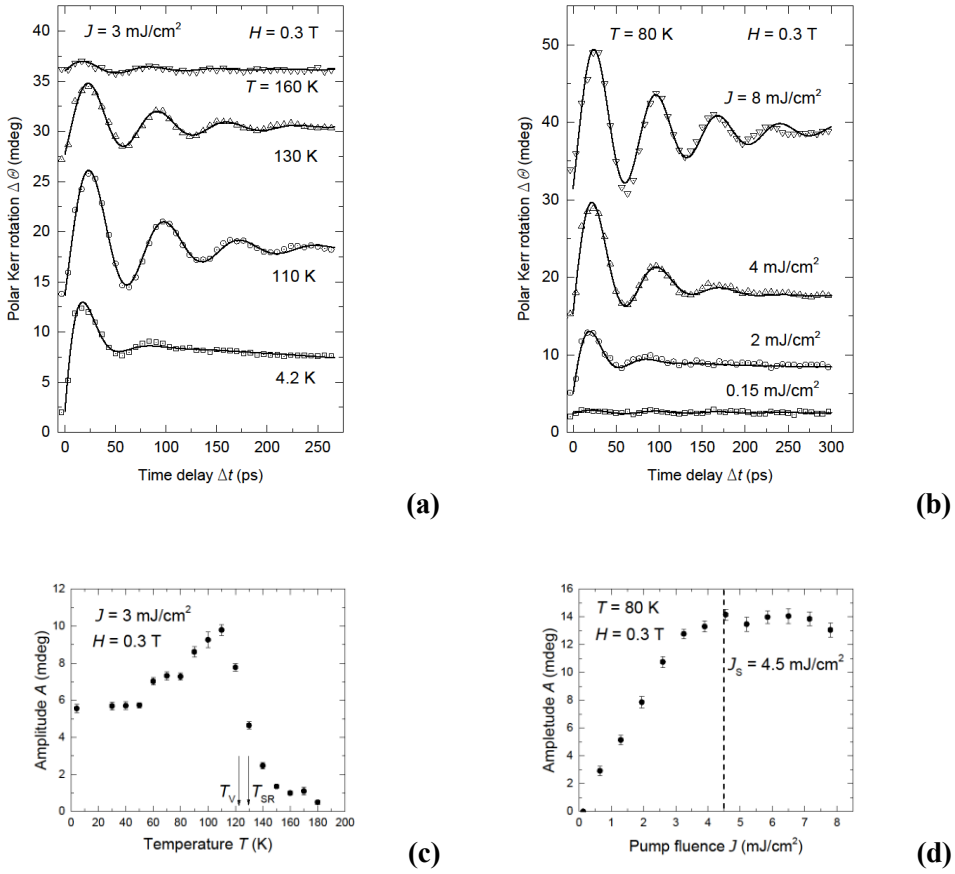


Figure 2. (a,b) TRMOKE traces measured **(a)** at different samples temperatures at the pump fluence of J=3 mJ/cm^2, and **(b)** at different pump fluencies at the sample temperature T=80 K (below T_{SR}). **(c,d)** Amplitudes of the magnetization precession **(c)** vs. the sample initial temperature at the pump fluence J=3 mJ/cm^2, and **(d)** vs. pump fluence at T=80 K.

Having confirmed the presence of ultrafast laser-induced insulator-metal transition, we proceed to laser-induced magnetization dynamics. Figure 2a and 2b shows the pump-probe time delay dependences of the probe polarization rotation measured at different temperatures and different pump fluencies (at T=80 K), respectively. Clear decaying oscillations are present in the measured signals which can be fitted by a function:

$$\Delta\theta(\Delta t) = A\sin(2\pi f + \varphi)\, e^{-\tau/t} + p(\Delta t), \qquad (1)$$

where A is the amplitude, f is the frequency, φ is the initial phase, and τ is the decay time of the oscillations. $p(\Delta t)$ accounts for the slowly varying background which origin is beyond the scope of this

paper. Frequency of oscillations obtained from the fit is in a good agreement with the ferromagnetic resonance frequency. Therefore, we can conclude that the observed oscillations originate from the laser-induced precession of magnetization.

In order to reveal the mechanism of the precession excitation, we analysed the temperature dependence of the precession amplitude (figure 2c). As one can see, with increasing the sample temperature, the amplitude of the precession increases and reaches the maximum value near the SR transitions region (T_{SR}). As the temperature is further increased, the precession amplitude rapidly decreases and finally vanishes at temperatures above 160 K. Such a behaviour allows us to conclude with confidence that the precession is excited via the laser-induced SR transition [3] triggered by the change of magnetic anisotropy occurring in response to laser excitation at a fast timescale. The simplest mechanism of laser-driven SR transition is the laser-induced increase of the lattice temperature. It is known that such changes in dielectrics and metals occur on a picoseconds time scale and, therefore, can lead to the observed precession. Estimates of the lattice temperature increase ΔT due to excitation with the J=3 mJ/cm^2 pump pulses yield ΔT=20 K at the initial sample temperature of 110 K, which corresponds to the heating up to the SR transition temperature. As can be seen in figure 2c, the maximum amplitude of the laser-induced precession is observed at this temperature, confirming the suggested scenario of the laser-driven SR transition.

It is important to emphasize that the excitation of the precession is realized even at the pump fluencies and sample initial temperatures that are not sufficient to heat up the sample over T_{SR}, e.g. at T<110 K and J=3 mJ/cm^2. Therefore, one cannot exclude that the laser-driven SR transition observed under such conditions may be related to processes which also underlay the ultrafast insulator-metal transition. The latter relies not on laser-induced heating alone, but on electronic excitations yielding destruction of the charge ordering [11]. To investigate this issue further, we have studied the pump fluence dependence of the excited precession at T=80 K (figure 2b and 2d). As one can see in figure 2d, the laser-induced precession can be reliably observed within the whole studied range of fluencies. Its amplitude linearly growth with increasing the pump fluence and reaches saturation at fluencies J~4.5 mJ/cm^2, which corresponds to the fluence required to heat the lattice up to T_{SR}. Interestingly, this fluence appears to be close to the saturation fluence for the ultrafast insulator-metal transition J_S (figure 1c). At lower fluencies the laser-driven SR may have a non-thermal nature and be related to the ultrafast charge ordering similarly to the ultrafast insulator-metal transition.

4. Conclusions

In conclusion, we have demonstrated the laser-pulse excitation of the magnetization precession in a single crystal of Fe_3O_4. The excitation is found to become more efficient once the sample initial temperature approaches the temperature region of the SR transitions, which allows us to conclude that the precession is a signature of the ultrafast laser-driven SR transition. Interestingly, we found that the pump pulse fluence dependence of the precession amplitude partly resembles that of the dynamical reflectivity changes. Since the latter occur due to ultrafast laser-induced insulator-metal transition, this observation suggests a link between the two transitions at the ultrafast time scale.

Acknowledgements

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