Ultrafast spin dynamics and critical behavior in half-metallic ferromagnet: \SrFeMoO6

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Abstract

Ultrafast spin dynamics in ferromagnetic half-metallic compound \SrFeMoO6 is investigated by pump-probe measurements of magneto-optical Kerr effect. Half-metallic nature of this material gives rise to anomalous thermal insulation between spins and electrons, and allows us to pursue the spin dynamics from a few to several hundred picoseconds after the optical excitation. The optically detected magnetization dynamics clearly shows the crossover from microscopic photo-induced demagnetization to macroscopic critical behavior with universal power law divergence of relaxation time for wide dynamical critical region.

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Control and manipulation of spins by ultrafast optical excitation, which gives rise to photo-induced magnetization change and magnetic phase transitions in dilute magnetic semiconductor quantum structures \[1\]–\[3\], doped semiconductors \[4\] and ferromagnetic metals \[6\]–\[10\], have attracted considerable attention. Recent study on the magnetization dynamics in the photo-excited Ni films with nonlinear optical techniques has revealed ultrafast spin process within 50 fs \[9\]. Strongly correlated electron systems with half-metallic nature, which have perfectly spin polarized conducting electrons at the ground state \[11\], are promising candidates for the study of the photo-induced spin dynamics. These materials have been found to possess exotic physical properties such as colossal magnetoresistance, which have strong application potential \[12\]. The strong coupling between spin, charge and lattice degrees of freedom in strongly correlated systems makes it possible to manipulate the magnetic properties via cooperative effects induced by optical excitation. In particular, the evidence of photo-induced phase transition accompanied with magnetization changes have been recently reported \[13\]–\[14\]. In order to understand the nature of these phenomena, it is crucial to investigate the temporal evolution of the spin system in the picosecond time scale. Although some attempts have been made by employing pump-probe spectroscopy \[15\], to the best of our knowledge direct investigation of the ultrafast spin dynamics in half-metallic materials has not been reported so far. Such an investigation can be carried out by exploiting the time resolved magneto-optical Kerr effect (MOKE), which has been shown to be a powerful tool to study the ultrafast dynamics of magnetization \[6\]–\[16\].

In the present paper we report on the ultrafast pump-probe MOKE and reflectivity study of dynamics of spin and electron systems in the ordered double perovskite Sr$_2$FeMoO$_6$. Since Fe$^{3+}$(3$d^5$; $t_{2g}e_g^2$, \(S = 5/2\)) and Mo$^{5+}$(4$d^1$; $t_{2g}^1$, \(S = 1/2\)) couple antiferromagentically via interatomic exchange interaction and the down-spin electron of Mo$^{5+}$ is considered itinerant (upper panel of Fig. 1), a conducting ferrimagnetic ground state with half-metallic nature is expected for this material. The density functional calculation \[17\] also shows that the occupied up-spin band mainly consists of Fe 3$d$ electrons, while the Fermi level exists within the down-spin band composed of Fe $t_{2g}$ and Mo $t_{2g}$ electrons. The temperature depen-
dence of magnetization and resistivity measurements under the magnetic field have shown
the ferromagnetic phase transition with the Curie temperature $T_C \sim 410-450$ K \cite{18}. Also,
this material in the form of polycrystalline ceramics shows inter-grain tunneling type giant
magnetoresistance at room temperature because of its spin polarization \cite{17}. Another im-
portant property of Sr$_2$FeMoO$_6$ is the enhancement of MOKE due to spin-orbit coupling of
$t_{2g}$ electrons in the heavy Mo-atom \cite{19}. The strong MOKE signal enables us to investigate
the spin dynamics over a wide temporal range from sub-picosecond to nanosecond and to
obtain the critical exponent of the relaxation time at the magnetic phase transition.

The MOKE measurements are carried out on single crystal Sr$_2$FeMoO$_6$, grown by
floating-zone method \cite{18}, in polar Kerr configuration under the magnetic field of 2000
Oe, where the magnetization is nearly saturated at room temperature [15,16], utilizing po-
larization modulation by a piezo-elastic modulator (CaF$_2$). Figure 1 (a) shows the spectral
profiles of ellipticity $\eta$ and rotation angle $\theta$ at room temperature. A very large MOKE sig-
nal, one order of magnitude larger than that of the doped manganites \cite{20} is observed. The
MOKE signal is proportional to $f \cdot M$, where $M$ is the magnetization and $f$ is determined by
the complex refractive index at the probe frequency. Correspondingly, the magneto-optical
spectra show resonance, known as the plasma enhancement effect \cite{21}, around 1eV, which
is close to the plasma edge \cite{18}. The temperature dependence of the $\eta$, probed at a photon
energy of 0.95 eV, clearly shows the magnetic phase transition at 450 K, which is the $T_C$
of the present sample \cite{22}. Since the reflectivity is almost temperature independent in this
temperature range, the sample magnetization can be monitored with the $\eta$.

For the pump-probe measurements, a Ti:Sapphire regenerative amplifier system (1 kHz
repetition rate) with an Optical Parametric Amplifier (OPA) is used as light source. The
second harmonic of the amplified pulses with a pulse duration of 200 fs, photon energy
of 3.1 eV and a maximum fluence of 90 $\mu$J/cm$^2$ is used as pump pulse, and its energy is
close to the charge transfer excitation from O 2$p$ to Fe/Mo $t_{2g}$ band with down-spin (see
the upper panel of Fig.1). The probe pulses from the OPA are tuned to 0.95 eV, at which
the ellipticity dominates the MOKE signal rather than the rotation effect (Fig.1 (b)). The
pump-probe MOKE measurements are also carried out in polar Kerr configuration and the polarization change of the reflected light from the sample is measured by a balanced detection scheme shown in Fig. 2 (a). By synchronizing the chopper for the pump beam with the regenerative amplifier, the balanced signal is detected and analyzed shot by shot using boxcar integrator and A/D converter. The photo-induced Kerr ellipticity change is measured as the difference between magnetization reversal signals, i.e., \( \Delta \eta_{Kerr} = \frac{1}{2} [\Delta \eta(M) - \Delta \eta(-M)] \) by changing the sign of the magnetic field in order to eliminate the contribution from the pump induced optical anisotropy. A sensitivity of \( 10^{-3} \) deg is achieved in our measurement system. The signal is observed to be proportional to the pump beam intensity in all pump-probe measurements.

The inset in Fig. 2 (a) shows the transient reflection change \( \Delta R/R \), measured at 300 K. It shows a sharp reduction in the reflectivity during the pump pulse duration, followed by a fast relaxation within \( 2 \sim 3 \) ps (region (1)) and a fairly long time plateau up to few tens of nanosecond (region (2)). The reflectivity returns to the initial state in 1 ms by heat diffusion (region (3)). Figures 2 (b) and (c) show the \( \Delta R/R \) and ellipticity change \( \Delta \eta_{Kerr} \) for different temperatures, indicating that the temperature dependence is negligible in \( \Delta R/R \), while that is significant in \( \Delta \eta_{Kerr} \). The temporal evolution of \( \Delta \eta_{Kerr} \) up to 500 ps is shown in Fig.3 (a) for different temperatures. One can observe from Fig. 3(a), that below the Curie point, it can be fitted by \( \Delta \eta_{Kerr}(t) = \Delta \eta_{step} + (\Delta \eta_{max} - \Delta \eta_{step})(1 - \exp[-t/\tau_{spin}]) \), where \( \Delta \eta_{step} \) describes the instantaneous decrease in the \( \Delta \eta_{Kerr} \), while \( \Delta \eta_{max} \) is the asymptotic value at the quasi-equilibrium state. The signal, \( -\Delta \eta_{max} \), increases drastically close to \( T_C \) as shown in Fig.3 (b). Our measurements reveal nearly linear increase of \( -\Delta \eta_{max} \) with the pump intensity.

The most striking feature is the very slow spin thermalization observed in \( \Delta \eta_{Kerr} \) signal in comparison with the electron thermalization observed in transient \( \Delta R/R \) data. In \( \text{Sr}_2\text{FeMoO}_6 \), the behavior of electrons is similar to that of ferromagnetic nickel [8]. Specifically, the electron temperature rises rapidly by the optical excitation and it relaxes within 2 to 3 picoseconds to reach quasi-equilibrium temperature, which is 8-10 K, obtained from
$\Delta R/R$, higher than the initial temperature. The fast decay of transient reflectivity indicates that the local heat transfer from electron to the lattice system is completed within a few picoseconds, accompanied by the lattice heat-up to reach quasi-equilibrium temperature. On the other hand, the behavior of the spin system in Sr$_2$FeMoO$_6$ looks very different from the behavior of the electronic system. Specifically, the very slow spin thermalization (see Fig. 2(c)), which is pronounced at higher temperature, indicates the anomalously small heat exchange between electrons and spins in Sr$_2$FeMoO$_6$. Such an electron-spin thermal insulation can be attributed to the nature of the half-metallic electronic structure where conducting electrons are perfectly spin polarized in the down-spin band and isolated from the insulating up-spin band as shown schematically in the upper panel of Fig. 1. Thermal motion of electrons around the Fermi level in the spin polarized conduction band does not increase the spin temperature.

From the observed results, we have the following scenario for the temporal evolution of electron, lattice and spin system in Sr$_2$FeMoO$_6$. Initially, during the photo-excitation ($\leq 1$ ps), the electron system is heated-up and rapidly thermalized due to electron-electron interaction. In this first stage the ellipticity shows a sharp decrease ($\Delta \eta_{\text{step}}$). In the next stage, the electron system relaxes by its energy transfer to the lattice system. The electron and lattice systems reach quasi-equilibrium state ($\sim 5$ps) by the electron-phonon interaction, leaving the spin system at its initial temperature. After that, the spin slowly relaxes toward this quasi-equilibrium state through weak heat exchange with the reservoir at quasi-equilibrium temperature. Finally, the system returns to the initial state by heat diffusion.

The sharp decrease in the ellipticity is the major feature of the initial stage of the optical relaxation in Sr$_2$FeMoO$_6$ (see inset of Fig. 2). The ratio $\Delta \eta_{\text{step}}/\eta$ shows a weak temperature dependence (see Fig. 3(c)). Since MOKE signal is proportional to $f \cdot M$, both photo-induced change in the refractive index ($\Delta f/f \sim \Delta R/R$) as well as the photo-induced magnetization change ($\Delta M/M$) contribute to $\Delta \eta_{\text{step}}/\eta$. Though the relative instantaneous changes in the ellipticity and reflectivity are of the same order, $\Delta \eta_{\text{step}}/\eta \sim \Delta R/R \sim 0.01$, the subsequent temporal evolution of $\Delta \eta_{Kerr}$ in the picosecond time scale is very different from that of $\Delta R$. 

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This indicates the direct demagnetization by resonant optical excitation. However, as it has been discussed in recent papers on Ni [6–9], it is premature to directly connect the MOKE signal with demagnetization in such ultrafast time scale.

We now discuss the temporal evolution of the MOKE signal in the second stage, when the electron and lattice system have reached the quasi-equilibrium (plateau region (2) in the inset of Fig. 2). The dramatic increase in $\Delta \eta_{\text{max}}$ (see Fig. 3(b)) and the relaxation time $\tau_{\text{spin}}$ as the temperature approaches $T_C$ indicate that the time resolved signal directly reflects the critical behavior of magnetization at the ferromagnetic phase transition. The spin temperature at the quasi-equilibrium, which can be estimated from Fig. 3(b) and the temperature dependence of the $\eta$ (Fig. 1(a)), is in good agreement with the electron temperature estimated from $\Delta R/R$.

It is necessary to emphasize that the time-resolved MOKE measurements give us an unique opportunity to study the critical dynamics of spin system independently from other degrees of freedom and obtain the critical characteristics of the ferromagnetic phase transition. The dynamics of the second order phase transition can be described by the dynamical scaling theory [23–25], which allows us to relate the critical behavior of the kinetic parameters (e.g. the relaxation time) to the critical exponents of the static parameters (e.g. correlation length) on the both sides of the critical point. The theory predicts that in the vicinity of $T_C$, the relaxation time of the order parameter can be described as $\tau \propto |T - T_C|^{-z\nu}$, where $\nu$ and $z$ denote the critical exponent of the correlation length and the dynamical critical exponent respectively. Figure 4 shows the temperature dependence of $\tau_{\text{spin}}$ as a function of $|T' - T_C|$, where $T'$ denotes the quasi-equilibrium temperature at the plateau region (temperature region (2) in the inset of Fig.2(a)). One can clearly observe the power law divergence with $z\nu = 1.22 \pm 0.06$ for the spin relaxation time in the vicinity of $T_C$ [26]. It should be emphasized that the power law behavior is established in the time scale of few tens of picosecond, while the width of the dynamical critical region is much higher than for conventional metals [27]. The theoretical calculation for the three dimensional Ising and Heisenberg models give $z\nu \approx 1.30$ [28] and 1.37 [29], respectively, while the two-dimensional Ising model gives
$z\nu \approx 2.165$ $^{30}$. Therefore, our measurements clearly indicate three dimensionality of the spin system in Sr$_2$FeMoO$_6$.

We have presented ultrafast spin dynamics in the ordered double perovskite Sr$_2$FeMoO$_6$ by using the time-resolved MOKE technique. We have observed, for the first time, extremely slow relaxation of spins, thermally insulated from electron and lattice systems due to the half-metal nature of this material. The thermal insulation of spin system provides us a unique opportunity to examine the non-equilibrium spin dynamics near the critical point in a time scale from picosecond to nanosecond range. Crossover from ultrafast microscopic spin relaxation to macroscopic critical behavior has been clearly demonstrated. In the vicinity of the critical point, the spin relaxation time increases as $|T-T_C|^{-(1.22\pm0.06)}$, which is consistent with the theoretical prediction for the 3D ferromagnetic system. We also observe very fast decrease in the MOKE signal $\Delta\eta_{Kerr}$ caused by charge transfer optical excitation. Although the underlying physical mechanism of such an ultrafast phenomenon is not established yet, the distinct difference in the temporal profiles of the photo-induced reflectivity and ellipticity suggests the contribution from ultrafast spin dynamics to nonlinear MOKE signal.

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and the possible thermal drift effects would be pronounced. Correspondingly, in order
to minimize the possible uncertainty in the estimation of the critical exponent, we
intentionally avoid the experimental point closest to $T_C$ in Fig. 4 for fitting.

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Figure captions.

Fig. 1. Magneto-optical Kerr measurements on Sr$_2$FeMoO$_6$ under the magnetic field of 2000 Oe. (a) Kerr rotation (solid line) and ellipticity (dashed line) spectra at 300K. (b) Temperature profile of linear Kerr ellipticity probed at 0.95eV. The upper panel shows the spin configuration of Fe and Mo ions and a schematic of the electronic band structure of Sr$_2$FeMoO$_6$ based on the density-functional calculations by Sawada and Terakura (Ref. [17]).

Fig. 2. (a) Schematic of the experimental setup for the pump-probe magneto-optical Kerr measurements. Inset shows the temporal evolution of transient reflection change $\Delta R/R$ from subpicoseconds up to millisecond. Temporal evolution of the transient reflection $\Delta R/R$ (b) and Kerr ellipticity change $\Delta \eta_{Kerr}$ (c) up to 50ps, measured at 200K, 300K and 400K.

Fig. 3. (a) Temporal evolution of photo-induced Kerr ellipticity $\Delta \eta_{Kerr}$ up to 500 ps, measured at various temperatures. Solid lines are the exponential fit. Temperature profiles of $\Delta \eta_{max}$ (b) and rapid component $\Delta \eta_{step}$ normalized to linear Kerr ellipticity $\eta$ (c).

Fig. 4. Temperature dependence of spin relaxation time $\tau_{spin}$ as a function of $|T' - T_C|$. The solid line is a power law fit $|T'/T_C - 1|^{-z\nu}$ for the points near $T_C$. The fit returns $z\nu = 1.22 \pm 0.06$. 

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Sr$_2$FeMoO$_6$
H $\sim$ 2000 Oe

Fe$^{3+}$(3d)$^5$
S=5/2

Mo$^{5+}$(4d)$^1$
S=1/2

antiferromagnetic coupling

$\theta$ and $\eta$ (deg.)

probe
$\theta$ (rotation)
$\eta$ (ellipticity)

probe photon energy : 0.95eV

(a)
(b)

Fig. 1 T. Kise et al.
\[
\Delta \eta_{\text{Kerr}} = \frac{(\Delta \eta (+M) - \Delta \eta (-M))}{2}
\]
Fig. 3 T. Kise et al.
Fig. 4  T. Kise et al.