Observation of the spin-charge thermal isolation of ferromagnetic $Ga_{0.94}Mn_{0.06}As$ by time-resolved magneto-optical measurements

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The dynamics of magnetization under femtosecond optical excitation is studied in a ferromagnetic semiconductor $Ga_{0.94}Mn_{0.06}As$ with a time-resolved magneto-optical Kerr effect measurement with two-color probe beams. The transient reflectivity change indicates the rapid rise of the carrier temperature and relaxation to a quasithermal equilibrium within a few ps, while a very slow rise of the spin temperature of the order of 500 ps is observed. This anomalous behavior originates from the thermal isolation between the charge and spin systems due to the spin polarization of carriers (holes) contributing to ferromagnetism. This constitutes experimental proof of the half-metallic nature of ferromagnetic $Ga_{0.94}Mn_{0.06}As$ arising from the large exchange energy of carriers compared to Fermi energy.

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Carrier-induced ferromagnetism is one of the most visited spontaneous symmetry-breaking phenomena in the solid-state physics. The underlying physics of this phenomenon deeply involves fundamental aspects of quantum physics including interactions between localized and delocalized carriers, where many-body quantum correlation plays an essential role.

The discovery of diluted magnetic semiconductors (DMS's) with high Curie temperature, which have become available due to recent advances in molecular beam epitaxy and transition-metal ion doping techniques, has opened new horizons in exploring of carrier-mediated ferromagnetism.[1–3] In particular, the compatibility of DMS's with conventional semiconductor technology is very promising to employ the spin degree of freedom in practical devices. Correspondingly, along with extensive efforts to clarify the microscopic mechanism of the ferromagnetism, much work has been devoted to the search for systems with higher T_c.[4–8]

In the initial stage of the research for the origin of the ferromagnetism, two mechanisms of the interaction between the local moments were considered. The first was based on the Ruderman-Kittel-Kasuya-Yoshida (RKKY) mechanism, in which the ferromagnetic interaction of localized spins is mediated by nearly free carriers.[4] This model reproduces the observed concentration dependence of Curie temperatures in II-VI DMS.[9]

However, recent photoemission experiments in Mn-doped GaAs have revealed a rather low density of state, which indicates a localization of carriers[10] and thus we need to consider a mechanism beyond the simple RKKY model. The second one was based on the double-exchange mechanism which is suggested by a band calculation of $In_{1-x}Mn_xAs$.[5] In this approach, the ferromagnetic coupling between localized spins is induced by the presence of carriers with strong *d*-band-like character. These carriers originate from transition- metal impurities and essentially contribute to the conductivity of the material. This model was also confronted with the difficulty from both the photoemission and soft x-ray absorption data,[11] which indicate that the holes have very little *d* component. Hence at present researchers are looking for a third way which compromises the above two approaches and many hypotheses have been proposed.

The pronounced spin polarization of conducting carriers in several ferromagnetic materials, which are often referred to as half-metals in which the density of state is nonzero for only one spin direction at the Fermi surface, makes these materials very attractive for spintronics applications.[12] Among high-T_c ferromagnetic semiconductors, a half-metallic nature has been suggested for Mn-doped GaAs. Such a suggestion is supported by results of polarized electro luminescence[13] and tunneling magnetoresistance measurements[14] along with *ab initio* calculation of the band structure of this material.[15] The half-metallicity is very important not only for applications but also for putting a strong constraint on the theories to explain the ferromagnetism. Nevertheless, since the exchange interaction energy is of the same order as the Fermi and potential fluctuation energy, the existing experimental data do not allow us to arrive at a decisive conclusion on the electronic structure of Mn-doped GaAs and its half-metallicity.

Simultaneous probing of the spin and carrier dynamics by time-resolved magneto-optical Kerr effect (TR-MOKE) measurements gives us a chance to reveal the half-metallic nature of the material. The TR-MOKE technique was first applied to the investigation of the picosecond photoinduced demagnetization of Ni.[16] The phenomenon is analyzed by a ''three-temperature model'' which describes the dynamical process of the thermal interaction among sub system (charge, spin, and lattice systems). We have employed the TR-MOKE technique to investigate the photoinduced spin and charge dynamics in the ordered double perovskite Sr_2FeMoO_6[17], which is known to show very large magneto resistance at room temperature. In particular, we have discovered that the response of the spin system is much slower than the charge system and shown that such a spin-charge thermal isolation originates from the half-metallic nature of Sr_2FeMoO_6. In

turn, the observation of the spin-charge thermal isolation may be suggested as an experimental criterion of the half-metallic nature of a material.

Although TR-MOKE is a unique method to probe the ultrafast dynamics of magnetization, the extraction of the pure magnetization component from the MOKE signal is an important experimental problem because a photo induced change of the refractive index also contributes to the pump-induced TR-MOKE signal. In the experiment with ferromagnetic metal films, the magnetization component of the signal has been extracted by comparing the results of linear and nonlinear magneto-optical measurements in both reflection and transmission geometries.[18]

In this paper, we propose and develop a two-color probe method of TR-MOKE applicable to opaque DMS's, which allows us to extract the magnetization component in reflection geometry. With this method, we investigate the magnetization dynamics in $Ga_{0.94}Mn_{0.06}As$ which has the highest Curie temperature, $T_c = 110$ K among ferromagnetic semiconductors.[8] The extracted magnetization dynamics reveals that spins thermalize much more slowly in comparison with holes, indicating the half-metallicity of this material, supporting some band calculations[15] based on a large exchange energy. A sample of $Ga_{0.94}Mn_{0.06}As$ with 1.05 μm thickness was grown by molecular beam epitaxy on a GaAs[001]substrate and annealed at 280° C.

The measurements of the linear spectra and TR-MOKE were performed in the polar Kerr configuration under a magnetic field of 0.2 T by alternating the field direction in a liquid N_2 flow cryostat. A detailed description of the experiment was given in the previous paper.[17] In the TR-MOKE measurements, a Ti-sapphire regenerative amplifier system with a photon energy of 1.55 eV, a pulse duration of 150 fs, and a repetition rate of 1 kHz was used as a light source. The pulses at the fundamental and doubled frequencies were used as a probe and a pump, respectively. The intensity of the pump pulse was 45 μJ/cm^2, which corresponds to a carrier density of 6.2×10^{19}cm^{-3}. We also performed a TR-MOKE measurement at different probe photon energy under the same pump condition. Part of the white light continuum obtained with sapphire crystal was spectrally filtered and used for the probe pulse with 1.77 eV photon energy. The stability of the light source limits the sensitivity of our measurements, typically 1 mdeg at 1.77 eV, and 0.2 mdeg at 1.55 eV. The temporal resolution of 0.3 ps was limited by the pump pulse duration.

Figure 1 shows the Kerr rotation and ellipticity spectrum at 77 K. The arrows in Fig. 1 indicate the photon energies of the probe pulses (1.55 and 1.77 eV) used in the TR-MOKE measurement. Figure 2(a) shows the dependence of the differential reflectivity at 1.55 eV under a 0.2-T magnetic field at 95 K on the time delay between the pump and probe pulses. One can observe a resolution-limited rapid reflectivity change, while a very slow relaxation is observed after several 100 ps. The temporal evolution of the Kerr rotation $\Delta\theta$ and ellipticity $\Delta\eta$ at 1.55 eV and 1.77 eV [Figs. 2(b) and 2(c), respectively] are more complicated in comparison with that of Sr_2FeMoO_6.[17] In particular, $\Delta\theta$ at 1.55 eV is initially negative, however, it changes its sign at a later time. Furthermore, the strong dependence of the temporal profiles on the probe frequency clearly shows that TR-MOKE signals do not directly give information of magnetization. In order to analyze these signals, let us present the Kerr rotation and ellipticity in the following form:

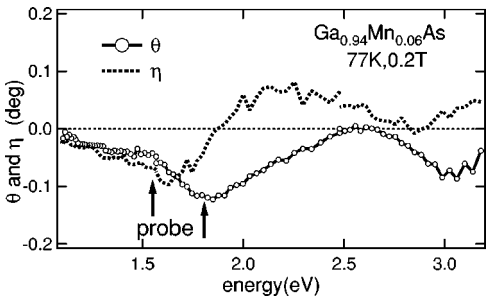

FIG. 1. Magneto-optical Kerr spectrum on $Ga_{0.94}Mn_{0.06}As$ under a magnetic field of 0.2 T at 77 K. The open circles show Kerr rotation. The dotted line shows the Kerr ellipticity. Two arrows show the photon energy of the probe pulse used in the TR-MOKE measurement.

$$\theta = f_\theta \cdot M, \quad \eta = f_\eta \cdot M, \tag{1}$$

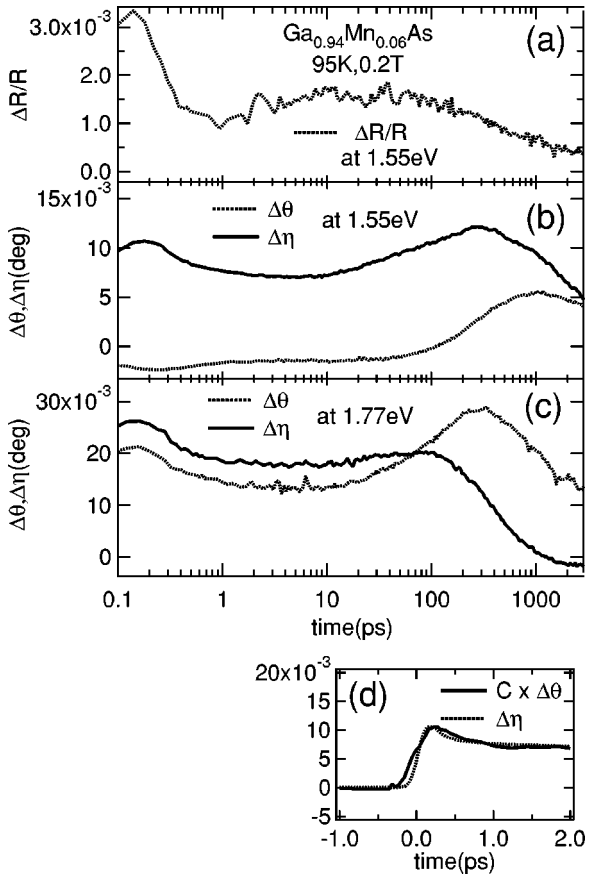

FIG. 2. Temporal evolution of reflectivity change $\Delta R/R$ (a) and Kerr rotation $\Delta\theta$ and ellipticity $\Delta\eta$ at 1.55 eV (b) and 1.77 eV(c) probe photon energy, respectively, under a magnetic field of 0.2 T at 95 K. The pump photon energy is 3.1 eV. The pump fluence is 45 μJ/cm^2. A small inset (d) shows $\Delta\theta$ multiplied by some constant and $\Delta\eta$ in the initial process at 1.55 eV

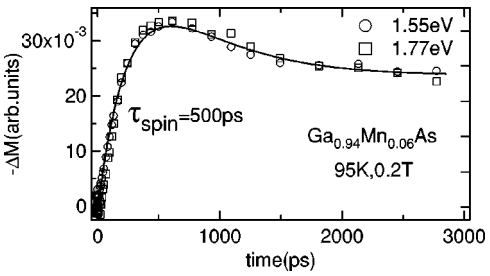

FIG. 3. Temporal evolution of the extracted magnetization component (the open circles are from $\Delta\theta$ and $\Delta\eta$ at 1.55 eV, and the open squares are from $\Delta\theta$ and $\Delta\eta$ at 1.77 eV). The open circles and squares are normalized at the maximum point. A positive direction shows demagnetization. A solid line shows a result obtained using the three-temperature model considering thermal diffusion.

where M is the magnetization, while f_θ and f_η depend on the electronic properties of the material and can be presented in terms of the refractive index and absorption coefficient. Correspondingly, the photo induced change in θ and η consists of two components

$$\Delta\theta(t) \approx f_\theta \ \Delta M(t) + \Delta f_\theta(t) M,$$

$$\Delta\eta(t) \approx f_\eta \ \Delta M(t) + \Delta f_\eta(t) M. \qquad (2)$$

At the probe frequencies of the present experiment, $\theta<0$ and $\eta<0$ (see Fig. 1), i.e., both coefficients $f_\theta<0$ and $f_\eta<0$. Thus, the reduction of magnetization leads the positive signal for both $\Delta\theta$ and $\Delta\eta$ from the first terms of Eq. (2). The second terms Δf_θ and Δf_η are the photo induced change of Kerr signals associated with the complex refractive index change ($\Delta\tilde{n}$) caused by photoexcitation. At a probe energy of 1.55 eV [Fig. 2(b)], we observe $\Delta\theta<0$ and $\Delta\eta>0$ right after the excitation. This indicates that the rapid changes in θ and η are governed by the second term of Eq. (2).

Figure. 2(d) presents temporal profiles of $\Delta\theta$ and $\Delta\eta$ at probe 1.55 eV [Fig. 2(b)] within the 2-ps time interval. By multiplying $\Delta\theta$ by -4.6, both curves fit well. This indicates that in this time interval, both $\Delta\theta$ and $\Delta\eta$ are governed by a single dynamical parameter. Therefore, we can take $\Delta f_\eta(t) = C\Delta f_\theta(t)$ where C is a constant in the whole temporal region. By substituting this equation into Eqs. (2) we readily arrive at the conclusion that the value of $C\Delta\theta - \Delta\eta$ is proportional to the magnetization change $\Delta M(t)$:

$$(Cf_\theta - f_\eta)\Delta M(t) = C\Delta\theta(t) - \Delta\eta(t). \qquad (3)$$

Equation (3) shows that we can extract ΔM from the measured values of the MOKE and TR-MOKE signals and parameter C, which depend on the probe frequency.[19] To examine the validity of this procedure, we analyzed the data for the probe at 1.77 eV to obtain the parameter C and extracted ΔM. Figure 3 shows that the normalized $C\Delta\theta - \Delta\eta$ at 1.55 eV (open circles) and 1.77 eV (open squares) show the same temporal behavior. The chosen values of C are -4.6 and 1.2 for 1.55 eV and 1.77 eV, respectively. Our results clearly show that we can extract the magnetization component from the TR-MOKE signals.

Now we discuss the temporal responses of $\Delta R/R$ shown in Fig. 2(a) and Δf_θ and Δf_η of Eq. (2). In semiconductors, these changes are driven by the photoinduced change in the complex refractive index caused by the band filling effects and successive carrier heating. In $Ga_{1-x}Mn_xAs$ as well as other low-temperature-grown GaAs, the excess arsenic atoms form high-density As antisites ($\sim 10^{19}$ cm^{-3}), which act as deep donor levels. The photogenerated electrons are immediately trapped in such levels and nonradiatively recombined within 1 ps.[20] It is known that the additional acceptor doping lowers the Fermi level and promotes the ionization of deep donor levels, which enhances electron trapping. In the present experiment, the high-density Mn doping brings acceptor levels with density as high as 10^{19} cm^{-3}. Thus the trapping and nonradiative recombination of photoexcited carriers should be very efficient and the carrier lifetime is less than 1 ps.[21,22] The resolution-limited fast decay of $\Delta R/R$ within 1 ps corresponds to such a rapid non radiative recombination process. The nonradiative process heats up the lattice and preexisting holes. Comparing with the temperature dependence of the linear reflectivity spectrum, we estimate the temperature increase of the holes after the fast decay as 3–5 K. Considering the small heat capacity of holes, which is four orders smaller than that of the lattice, the estimated small temperature increase of holes indicates that the holes and lattice should have reached a thermal equilibrium during the nonradiative recombination processes. The slow decay observed in $\Delta R/R$ over several hundred ps is due to thermal diffusion.

After the nonradiative recombination, $\Delta\tilde{n}$ induced by the temperature increase of holes governs the temporal responses of $\Delta R/R$ and Δf_θ and Δf_η. As we have noted, all the signals are described with a single dynamical parameter shown in Fig. 2(d).

Now we discuss the origin of the slow thermalization of the extracted magnetization in Fig. 3. The observed slow thermalization of spin systems is rather unusual for GaAs-based semiconductors because the valence band is strongly affected by the spin-orbit interaction and the heavy-hole and light-hole bands are degenerated at the Γ point. The spin relaxation time for the hole in pure GaAs is known to be about 110 fs (Ref. 23). Moreover, with doping the magnetic impurity,[24] the spin relaxation time decreases due to the spin-flip scattering by magnetic impurities.

However, our experimental findings indicate that the spin temperature rises slowly towards the hole and lattice temperature. According to the three-temperature model,[16] the rising speed of the spin temperature is determined by the heat capacities and heat flow rates. The heat capacity of the spin system is 18 J/K m^3, which is much smaller than those of lattice and hole systems, 1.0×10^6 J/K m^3 and 1.8×10^2 J/K m^3, respectively.[25] Therefore, we need to consider anomalous thermal isolation of spin systems. This resembles what we have observed in the half-metallic ferromagnet Sr_2FeMoO_6.[17]

After the rapid nonradiative relaxation, the subsequent carrier scattering mainly occurs near the Fermi surface. Therefore, the demagnetization rate of carriers strongly de-

pends on their spin polarization near the Fermi level. In particular, if the density of states for one spin direction is much smaller than that of the other direction, the spin flipping is inefficient. This situation occurs for a system where the exchange splitting of the carrier bands is larger than the Fermi energy, leading to the half-metallic electronic structure. Therefore, the observed slow demagnetization invokes this condition which holds for $Ga_{0.94}Mn_{0.06}As$ as well as for Sr_2FeMoO_6.

The observed temporal responses can be numerically simulated by using the three-temperature model.[16] The solid curve of Fig. 3 shows that this model can reproduce the temporal evolution of spin systems within a wide time interval. The rising time of the spin temperature is found to be 500 ps. It is necessary to emphasize that the obtained coupling constant between the charge and spin systems is much smaller than that in Ni. The observed spin relaxation at longer time delay might be due to spin-lattice coupling, which stems from the magnetic anisotropy.[26]

In conclusion, we observe the magnetization dynamics in $Ga_{0.94}Mn_{0.06}As$ over a wide temporal range from ps to ns using two-color TR-MOKE measurements. We discover that the spin system thermalizes much slower than the hole and lattice systems, indicating spin-charge thermal isolation. This experimental finding confirms that $Ga_{0.94}Mn_{0.06}As$ has a half-metallic electronic structure, making it a prospective material for spintronic applications. The half-metallicity indicates a large exchange energy of the carriers compared to the Fermi energy, which plays an essential role for the magnetism and transport properties of this material.

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