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Relaxation of magnetization in $Cd_{1-x}Mn_xTe$ diluted magnetic semiconductors under illumination

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Relaxation of thermoremanent magnetization (TRM) of $Cd_{1-x}Mn_xTe$ diluted magnetic semiconductors (DMS) in the spin-glass state have been studied under light illumination. The relaxation of TRM can be described well by a power law decay, $M(t)=M(t_0)t^{-\alpha}$ ($t>t_0$, $t_0\sim 2$ s). The variations of the decay parameter α with the illumination light intensity has been measured and a relation which indicates that α is proportional to the photogenerated carrier concentration n has been observed.

The diluted magnetic semiconductors (DMS) are a group of materials which have attracted a great deal of attention for many years because of their unique properties for understanding many fundamental physics as well as their promising practical applications.[1,2] The magnetic behavior of different kinds of DMS show many common characteristics, which can be understood on the basis of a random array of localized magnetic moments coupled by isotropic antiferromagnetic interaction.[3] Many aspects of this interaction between magnetic ions are still under investigation. One of the interesting and important subjects in the DMS is the spin-glass (SG) state at low temperatures. It is well established in DMS that there exists a paramagnetic to a SG transition at low temperatures because of randomness of magnetic ion distribution and spin–spin interactions. The transition temperature or freezing temperature T_f depends on magnetic ion composition,[4,5] magnetic field,[6] and history of the system. Although some work has been devoted to study SG dynamics in the DMS previously,[7–10] the understandings of the dynamic process of SG formation and transformation, as well as the relaxation in the SG state are still far from complete.

In this paper, we report the studies of the relaxation of thermoremanent magnetization (TRM) of $Cd_{1-x}Mn_xTe$ in the SG state under light illumination. A power law time dependence of TRM relaxation has been observed and the decay parameter at different excitation light intensities has been measured. The main results reported in this paper are the investigations of the spin–spin interaction of the magnetic ions in the SG state as affected by photogenerated electrons and holes. We have measured TRM under light illumination at different conditions and the results have been compared to those obtained in the dark.

The sample used for this study was a $Cd_{0.74}Mn_{0.26}Te$ single crystal provided by Cleveland Crystal, Inc. It was grown by temperature gradient technique and nominally undoped. However, photoluminescence measurements of a bound exciton transition indicated a low concentration of impurities in the samples. The size of the sample was a few cubic millimeters. The magnetization was measured by a superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS5). In order to study the effects of photoexcitation to the relaxation of TRM, a quartz fiber optic probe was used with the SQUID. The fiber optic probe was made of an optic fiber bundle with a diameter of about 2 mm. One end of the fiber optic probe was in the sample chamber within a few mm from the sample and the other end was outside the SQUID. A mercury lamp with a set of neutral density filters was used as an excitation source.

We have measured the susceptibility of a $Cd_{0.74}Mn_{0.26}Te$ sample under the field-cooled (FC) and zero-field-cooled (ZFC) conditions to determine the freezing temperatures T_f of the SG state. The data show that T_f is about 4.2 ± 0.2 K, which is consistent with previous measurements.[5–7,11] In this paper, we only report relaxation of TRM in the SG state, or at temperatures below T_f. We did not observe any remanent magnetization above T_f.

Figure 1 is the plot of relaxation of magnetization at $T=1.7$ K and measured at $B=100$ Oe. The experiment was performed by cooling down the system with the magnetic field on ($B=1$ T), i.e., under FC condition, then by reducing the field to $B=100$ Oe after the temperature has reached 1.7

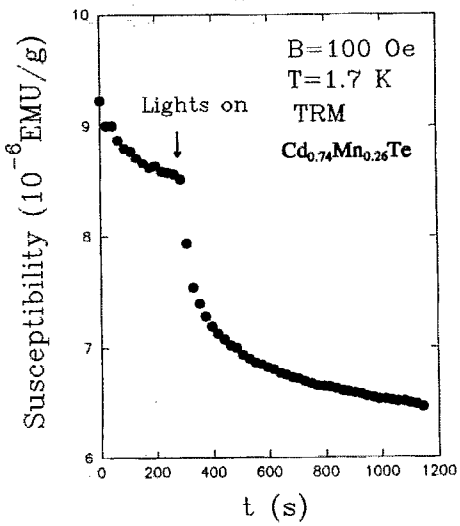

FIG. 1. Decay of magnetic susceptibility of TRM of a $Cd_{0.74}Mn_{0.26}Te$ sample in the dark and under illumination in the spin-glass state at $T=1.7$ K measured at a magnetic field of $B=100$ Oe. The arrow indicates the moment the light illumination was turned on. The applied magnetic field during the cooling down was 1 T.

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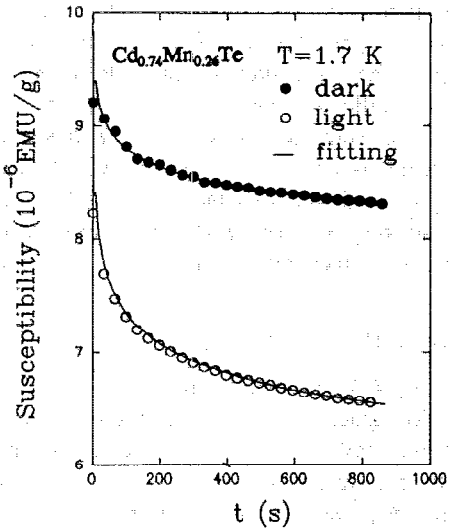

FIG. 2. Decay of magnetic susceptibility of TRM of a $Cd_{0.74}Mn_{0.26}Te$ sample in the dark (●●●) and under illumination (○○○) at $T=1.7$ K measured at a magnetic field of $B=100$ Oe. The solid lines are the least-squares fit by the power law decay of Eq. (1).

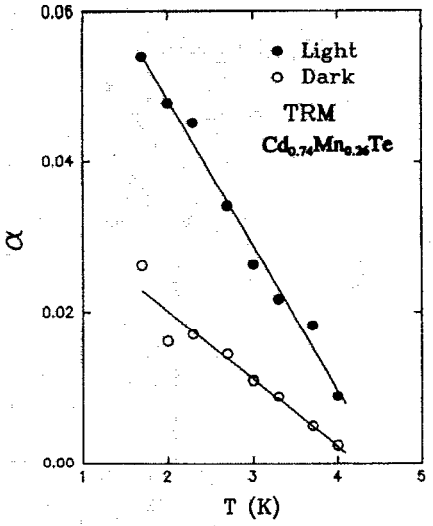

FIG. 3. The decay parameter α as a function of temperature in the dark (○○○) and under the illumination (●●●) for a $Cd_{0.74}Mn_{0.26}Te$ sample. The solid lines are the least-squares fit using $\alpha=a-bT$, where a is 3.8×10^{-2} and 8.7×10^{-2} and b is 9.0×10^{-3} and 1.9×10^{-2}/K in the dark and under illumination, respectively. Extrapolate these two lines to $\alpha=0$ gives $T_f=4.2\pm0.2$ and 4.5 ± 0.2 K for the dark and under illumination, respectively.

K. Shown in Fig. 1 is an initial decay measured in the dark for about 300 s and a subsequent decay measured after the moment the light excitation was turned on as indicated by an arrow in the figure. As we can see from Fig. 1, the decay of the magnetization under illumination proceeds faster compared to that in the dark.

Figure 2 shows the results of relaxation of magnetization measured at 100 Oe in the dark (●●●) and under illumination (○○○) at $T=1.7$ K for a $Cd_{0.74}Mn_{0.26}Te$ sample together with a least-squares fit using a power law decay (solid lines). We have to point out that the initial condition for the relaxation depends on the waiting period before the measurement. In order to minimize the difference in the initial condition, we begin to collect the relaxation data under illumination and in the dark at the same time so that direct comparison between two cases can be made here. Previously, a power law decay of magnetization in DMS has been observed.[12] Here, we also find that the experimental results shown in Fig. 2 can be described quite well by a power law decay,

$$M(t)=M_0t^{-\alpha}(t>t_0,t_0\sim2\ \text{s}), \tag{1}$$

both in the dark and under illumination, where α is the decay parameter. The fitted values in Fig. 2 are $\alpha=2.6\times10^{-2}$ and 5.4×10^{-2} and $M_0=9.9\times10^{-6}$ and 9.4×10^{-6} emu/g for the conditions in the dark and under illumination, respectively. The decay parameter α is directly correlated with the decay rate. Therefore, the experimental results indicates that the decay rate is about a factor of 2 larger under illumination than that in the dark.

Figure 3 are the plots of decay parameter α as a function of temperature in the dark (○○○) and under illumination (●●●) for $Cd_{0.74}Mn_{0.26}Te$. The decay parameter α decreases linearly with increasing of temperature for both cases. At all temperatures, the magnetization relaxes faster under illumination compared to that in the dark. The difference between α under illumination and in the dark becomes smaller as temperature approaches to T_f. Eventually, α approaches to zero at the freezing temperature T_f. Dependence of α on temperature T can be described by a linear relation, $\alpha=a-bT$ $(T<T_f)$. Extrapolating from $\alpha=0$, we obtain T_f to be 4.2 ± 0.2 and 4.5 ± 0.2 K in the dark and under illumination, respectively. We have also measured the magnetization of ZFC vs temperature under illumination and in the dark, we cannot determine whether or not T_f has been changed under illumination because the difference between the values of T_f in the dark and under illumination is within the experimental uncertainty. However, the values of T_f extrapolated from $\alpha=0$ in Fig. 3 are consistent with the freezing temperature $T_f(=4.2\pm0.2$ K) obtained from the FC susceptibility measurement within the experimental uncertainty. The results in Fig. 3 tells us that we can also determine the freezing temperature T_f of the SG state by measuring the temperature dependence of the relaxation parameters of magnetization.

We have also measured the decay parameter α of TRM at a fixed temperature ($T=2$ K) under photoexcitation with different light intensities. Figure 4 shows a plot of α vs the square root of excitation light intensity, $I^{1/2}$. The magnetic field for the measurements was 100 Oe and the field applied during the cooling down was 1 T. As we expected, the exponent α increases as light intensity increases. Relaxation of magnetization obtained under illumination with different light intensities can be described quite well by the power law decay. We see that α increases linearly with the square root of light intensity, $\alpha\propto I^{1/2}$. The insert of Fig. 4 shows α vs light intensity I.

One obvious mechanism which may be responsible for

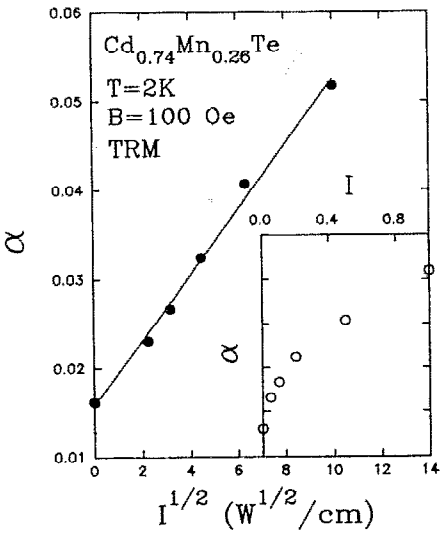

FIG. 4. Dependence of the decay parameter α of TRM on the square root of excitation light intensity $I^{1/2}$ for a $Cd_{0.74}Mn_{0.26}Te$ sample at $T=2$ K. Here $I=0$ corresponds to the dark. The solid line in the figure is the linear fit of α with $I^{1/2}$ according to Eq. (4) with fitted values $B_0(=1.57\times10^{-2})$ and $B_1(=3.68\times10^{-3})$. The insert is the plot of α vs light intensity I. The unit of the light intensity I in the inset is in 0.25 W/cm^2 and of $I^{1/2}$ in the main figure is in 0.05 W$^{1/2}$/cm. The scales of α are the same for both the main figure and the inset.

the observed light intensity dependence of α is the heating effect under light excitation. However, the systematic dependence of α on the excitation intensity cannot be explained by considering the heating effects. Furthermore, from Fig. 3, we see that the decay parameter α decreases as temperature increases in the temperature region investigated here. If the changes of α under light excitation is due to heating, we should expect that α decreases as light intensity increases, which has never been observed. These observations preclude the possibility of heating effects. Therefore, we can conclude that the observed decay rate increasing under light excitation is due to photoexcited carriers. It is clear from our experimental results that the mechanism responsible for manganese–ion coupling in DMS is carrier concentration sensitive.

Under photoexcitation, free or localized carriers (electrons and holes) are generated and subsequently recombined radiatively or nonradiatively. Photoluminescence due to carrier recombination in DMS has been observed in many experiments.[13,14] The electron concentration under illumination can be written as

$$\frac{dn}{dt}=G-Cnp/\tau, \tag{2}$$

where τ is the carrier recombination lifetime, n and p the electron and hole concentrations, and C a proportionality constant in units of cm^3. Here G is the carrier generation rate writing as $G=\alpha_a\eta I/\hbar\omega$, with $\hbar\omega$ being the excitation photon energy, I the light intensity in units of W/cm^2, α_a the absorption coefficient, and η the quantum efficiency—number of electrons generated per each absorbed photon. Under continuous and constant intensity light illumination, the photogenerated electrons and holes will be under equilibrium and so we have $dn/dt=0$. We should also have the condition $n=p$ since our samples are undoped. Then we have

$$n=(G\tau/C)^{1/2}=AI^{1/2}, \tag{3}$$

with $A=(\tau\alpha_a/C\hbar\omega)^{1/2}$. From the fact that the relaxation of magnetization proceeds faster under illumination, we can assume that the relaxation rate of magnetization is proportional to the carrier concentration through the carrier–manganese ion interactions,[14] $\alpha\propto n$. From Eq. (3), we have

$$\alpha=B_0+B_1I^{1/2}, \tag{4}$$

where B_0 is the decay parameter α in the dark ($I=0$), and B_1 (in units of cm/W$^{1/2}$) is a proportionality constant. In Fig. 4, we have plotted the least-squares fit of data to Eq. (4) as a solid line.

In conclusion, effects of carriers, which are generated by photoexcitation, to the relaxation of TRM of SG in $Cd_{0.74}Mn_{0.26}Te$ DMS have been studied. A power law decay both in the dark and under illumination has been observed. The decay parameter α as a function of light intensity has been measured at different temperatures. We found that the decay parameter under illumination increases linearly with the photogenerated carrier concentration. More detailed experimental results and discussions, including the results for different Mn concentrations, will be reported elsewhere.

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