Aging of spin glass semiconductor Cd$_{1-x}$Mn$_x$Te without Cd defects under photo illumination

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Abstract. The aging behaviour of spin glass semiconductor Cd$_{1-x}$Mn$_x$Te subjected to bond perturbation was studied by means of the direct change in the spin–spin interaction $\Delta J$ using photo illumination, without change in temperature. Under the $\Delta J$ perturbation, the maximum of relaxation rate of magnetization $S$ at the time $t_{\text{max}}$ is suppressed and $t_{\text{max}}$ becomes longer with increasing the light intensity. The former is consistent with the droplet picture, but the latter indicates that the $\Delta J$ perturbation accelerates the magnetic relaxation so as to contradict with the chaotic nature of spin configuration under the perturbation.

1. Introduction
Spin glass behavior is sensitive to the change in the spin-spin interaction. The unique phenomenon of spin glass, i.e., aging or memory-rejuvenation effect, has been studied by magnetization response under the change in spin-spin interaction through temperature perturbation $\Delta T$, e.g., the temperature cycle of $T \rightarrow T+\Delta T \rightarrow T$ [1]. However, it is difficult to evaluate the intrinsic effects of temperature perturbation, because the temperature perturbation causes ambiguous changes in thermally activated dynamics. Recently, the aging of spin glass under direct change in spin-spin interaction $\Delta J$, called bond perturbation, using photo illumination has been studied in diluted magnetic semiconductors Cd$_{1-x}$Mn$_x$Te [2]. However, there were some problems in the previous study, i.e., the light intensity contribution for the relaxation rate of magnetization $S$ was scarcely observed and the S/N ratio in measurement data was insufficient to perform the detailed analysis. The former may be attributed to photo absorption by Cd defects in the sample without heat treatment in Cd atmosphere [3] and the latter to the insufficient stability of light intensity.

In this study, the magnetization relaxation of Cd$_{1-x}$Mn$_x$Te sample, which is annealed in Cd atmosphere to reduce Cd defects, is studied under photo illumination, where the stability of light intensity is significantly improved. We demonstrate the $\Delta J$ perturbation through photo illumination so as to intrinsically change the spin glass dynamics.

2. Experimental
The Cd$_{1-x}$Mn$_x$Te single crystals were grown by the vertical Bridgman technique. Two samples with Mn compositions of $x = 0.30$ and 0.40 have been studied. The composition of the samples was evaluated by means of electron probe microanalysis and X-ray diffraction using Vergard’s law. The samples were annealed in Cd atmosphere at 600–800°C for 3–120 hours to reduce Cd defects. The photo absorption spectra of the samples were obtained by a Fourier transform spectrometer BOMEN.
DA8 to check whether or not the photo absorption due to Cd defects is observed. Two orthogonally-
polarized semiconductor lasers with wavelengths of $\lambda = 532\text{nm} (2.33\text{eV})$ and $834\text{nm} (1.49\text{eV})$ were used as the light source, where the band gap energy $E_g$ of the samples is between the two energies. Light was guided to the sample through a quartz optical fiber. The light intensity was controlled by the PID system using a polarizer with stepping motor. The magnetization relaxation was measured by a Quantum Design MPMS5 superconducting quantum interference device (SQUID) magnetometer.

Figure 1 shows the photo illumination protocol in the present experiment, where $t_w (= 500\text{sec})$ is wait time, $t_{\text{heat}} (= 500\text{sec})$ and $t_{\text{cool}} (= 500\text{sec})$ are the durations necessary to reach the target states, and $t_{\text{cool}} (= 200\text{sec})$ is the duration of photo illumination. The temperature increase, which is induced with the photo illumination, is canceled out by the synchronous cooling in SQUID system using a temperature control apparatus. Magnetization measurement is performed in $H = 100\text{Oe}$.

![Figure 1. The magnetic measurement protocol under photo illumination. The sample was first cooled to $T_m = 9K$ and aged during $t_w = 500\text{sec}$ (initial aging stage). The perturbation was subsequently added during $t_{\text{heat}}+ t_{p} + t_{\text{cool}} = 1200\text{sec}$ (perturbation stage) using photo illumination. After the stop of perturbation, $H$ of 100Oe was applied and the magnetization was measured as a function of $t$ (healing stage).](image)

3. Results and Discussion

3.1. Heat treatment in Cd atmosphere

Figure 2 shows the heat treatment temperature dependence of Mn concentration on the sample surface of Cd$_{0.70}$Mn$_{0.30}$Te, where the heat treatment time is 3 hours. The Mn atoms separate out from the matrix by heat treatment in Cd atmosphere at temperatures above 600°C. Therefore, we performed the heat treatment of the sample at a temperature slightly lower than 600°C.

![Figure 2. Heat treatment temperature dependence of Mn concentration on the sample surface. The nominal Mn concentration in the sample is 15.0%](image)

Figure 3 shows the photo absorption spectrum at 72K of Cd$_{0.60}$Mn$_{0.40}$Te for various heat treatment times, where the heat treatment temperature is 593°C. The band gap energy $E_g \approx 2.1\text{eV}$ is evaluated. The photo absorption by Cd defects is observed at 1.4eV in the sample without heat treatment. The photo absorption by Cd defects intrinsically disappears under the heat treatments for 30 and 120 hours, although the heat treatment for 3 hours is insufficient to reduce the Cd defects. However the 120 hours heat-treated sample shows two peaks, corresponding to spin glass freezing, in the magnetic data (Figure 4. a), i.e., the heat treatment for 120 hours resulted in the inhomogeneity of Mn composition. The heat treatment for 30 hours was most suitable because the single peak corresponding to spin glass freezing appeared in the magnetic data (Figure 4. b). Thus, the sample, which is heat-treated at 593°C for 30 hours, is used in the present study.
3.2. Laser intensity control by PID system

Figure 5 shows the fluctuation of light intensity of orthogonally-polarized semiconductor laser with $\lambda = 532\text{nm}$ used in the present study and Figure 6 shows the magnetization relaxation of Cd$_{0.60}$Mn$_{0.40}$Te in $H = 100\text{Oe}$ at 9K after cooling in $H = 100\text{Oe}$ under photo illumination. The fluctuation of $\pm 2.5\%$ in the laser intensity without any control significantly affects magnetization relaxation as shown in inset in Figure 6. This is attributed to the accidental temperature perturbation to the sample during photo illumination. The fluctuation of less than $\pm 0.1\%$ in the light intensity controlled by PID system brings about the magnetization relaxation data which are indistinguishable with the data without photo illumination. Thus we claim that the present stability of light intensity is sufficient to realize the change in spin-spin interaction $\Delta J$ through photo illumination without any other perturbation.

Figure 5. Time dependence of the light intensity. The light intensity fluctuation is decreased from $\pm 2.5\%$ to $\pm 0.1\%$ by the PID control.

Figure 6. Magnetization relaxation in $H = 100\text{Oe}$ at 9K after cooling in $H = 100\text{Oe}$ under photo illumination.
3.3. Aging under photo illumination

Figure 7 shows the time dependent relaxation rate $S$ under photo illumination obtained according to the experimental protocol in Figure 1, where the light intensities are 1.4, 0.3 and 0.0 mW. The data obtained without photo illumination shows a maximum at $t_{\text{max}}$, which is corresponding to $t_w = 1700\text{sec}$. Under photo illumination with $\lambda = 532\text{nm}$, the systematic change in $S$ dependent on the light intensity is observed, i.e., the value of $S$ at $t_{\text{max}}$ decreases and $t_{\text{max}}$ becomes longer with increasing light intensity. In addition, this behaviour is not observed under photo illumination with $\lambda = 834\text{nm}$ at which the photo excitation should not be occurred in the present sample. Thus, this is first demonstration of the $\Delta J$ perturbation through photo illumination so as to affect the spin glass dynamics without contribution from the other perturbation.

The suppression of $S$ at $t_{\text{max}}$ indicates that magnetic domain, in which the spin configuration is equilibrated under the initial condition, breaks by $\Delta J$ perturbation as predicted in the droplet picture [4]. It is consistent with the previous experimental study of $\Delta T$ perturbation and the simulations of $\Delta J$ perturbation [5]. The increment of $t_{\text{max}}$ with increasing intensity, however, indicates that the magnetic relaxation accelerates by $\Delta J$ perturbation, i.e., the magnetic domain continuously grows with $\Delta J$ perturbation, which may be inconsistent with the droplet picture. This interesting feature in spin glass dynamics should be intrinsic to verify the feasibility of scenario based on the droplet picture, although the mechanism is not clear in the present stage.

![Figure 7. Aging measurements under photo illumination which light intensities are 1.4, 0.3 and 0.0 mW. The wavelengths are 532nm (2.33eV) and 834nm (1.49eV).](image)

4. Conclusion

The reduction of Cd defects in spin glass semiconductor Cd$_{1-x}$Mn$_x$Te by heat treatment in Cd atmosphere at 593°C for 30 hours and the improved stability of light intensity achieve the spin glass aging which is perturbed only by $\Delta J$ effect through photo illumination without contribution from the other perturbation.

By $\Delta J$ perturbation, the $S$ at $t_{\text{max}}$ is suppressed and $t_{\text{max}}$ becomes longer with increasing light intensity. The former is consistent with the droplet picture, but the latter indicates that the $\Delta J$ perturbation accelerates the magnetic relaxation so as to contradict with the chaotic nature of spin configuration under the perturbation. This feature should be studied in detail to clarify the intrinsic nature of spin glass dynamics.

References

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