Spin glasses of the system $Zn_x Cd_{1-x} Cr_2Se_4$ with cubic magnetocrystalline anisotropy

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Spin glasses of the system $Zn_x Cd_{1-x} Cr_2Se_4$ with different values of the cubic magnetocrystalline anisotropy were investigated. It is shown that this anisotropy influences the quantitative characteristics of spin glasses.

I. INTRODUCTION

Single crystals of the system $Zn_x Cd_{1-x} Cr_2Se_4$ are ferromagnetic at $x < 0.3$, are antiferromagnetic at $x > 0.5$, and are spin glasses at $x \approx 0.4$ (Refs. 1 and 2). The magnetic subsystem of these spin glasses is made up of magnetic ions of one type, Cr^{3+}, arranged strictly periodically in octahedral positions of a spinel crystal structure, and only the random placement of the diamagnetic ions Cd^{2+} and Zn^{2+} leads to spatial fluctuations of the signs and magnitudes of the superexchange interaction between the magnetic chromium ions. Such a magnetic system corresponds exactly to the "random bond" model, in contrast to the "classical" spin glasses, whose magnetic ions are randomly located.

In addition, spin glasses of the $Zn_x Cd_{1-x} Cr_2Se_4$ system have a cubic magnetocrystalline anisotropy whose value can be varied smoothly from 10^3 to 10^5 erg/cm^3 by doping with a small amount of silver (~ 5 mol.%).[3] By investigating samples with different concentrations of the Ag impurity it is therefore possible to track in experiment the influence of the magnetocrystalline anisotropy on the spin-glass properties. This investigation is of particular interest, since it has been shown in recent theoretical papers[4,5] that in the model of Heisenberg magnets with short-range action it is important to take into account the anisotropic interactions in the description of spin-glass properties.

We have investigated the temperature dependences of the dynamic magnetic susceptibility and the remanent-magnetization relaxation processes. To determine the influence of the magnetocrystalline anisotropy, we investigated spin glasses of two types.

Type I comprised samples of the $Zn_x Cd_{1-x} Cr_2Se_4$ system with $x = 0.46$, doped with $\sim 5\%$ Ag. They have a cubic magnetocrystalline anisotropy with a first constant $K_1 \sim 10^5$ erg/cm^3.

Type II are undoped samples of the $Zn_x Cd_{1-x} Cr_2Se_4$ system with $x = 0.4$ and $K_1 \sim 10^3$ erg/cm^3.

The constants of the cubic magnetocrystalline anisotropy of the samples investigated in the present study were determined by one of us in Ref. 3. The dynamic magnetic susceptibility was measured with the setup described in Ref. 6, and the relaxation processes of the remanent magnetization were investigated with a rotating-sample magnetometer described in Ref. 7.

II. EXPERIMENTAL RESULTS

1. Magnetic susceptibility

It was established in Ref. 7 that the paramagnetic Curie points of samples of type I and II are close in value, 85 K and 79 K respectively. The paramagnetic Curie point is a quantitative characteristic of the average energy of the exchange interactions. The average exchange interaction energies in the samples of both types are therefore approximately equal. The "freezing" temperatures T_f of these samples, however, differ by almost a factor of 2, with $T_f \approx 37$ K for strongly anisotropic samples as against $T_f \approx 21$ K for weakly anisotropic ones. Such a difference between the "freezing" temperatures can be explained by assuming that the magnetocrystalline anisotropy influences the value of T_f, which increases with increase of the magnetocrystalline anisotropy. This result agrees qualitatively with the deductions of the theoretical papers.[4,5]

It is known that the "freezing" temperature of a spin glass depends on the applied constant magnetic field H. Most theoretical papers cite power-law dependences, with the shift of the freezing temperature proportional to the field raised to the power δ:

$$T_f(0) - T_f(H) \propto H^\delta. \tag{1}$$

For the Ising model in the Alameida-Thouless theory we have $\delta = 2/3$ (Ref. 8), and for the Heisenberg model of a spin glass the Gabay-Toulouse theory predicts a similar relation but with $\delta = 2$ (Ref. 8). To determine the law that governs the shift of the "freezing" temperature, investigations were made of the temperature dependences of the dynamic magnetic susceptibilities of samples of both types in different constant magnetic fields. The plots obtained for the samples of both types are shown in Fig. 1. The coordinates are $\ln H$ and $\ln[T_f(0) - T_f(H)]$. It was found that the changes of T_f for samples of both types are described by a power law. The exponent for the strongly anisotropic samples is close to unity, whereas for weakly anisotropic samples it is approximately equal to 2. This result agrees with the theoretical premises, since the Ising model corresponds to an infinitely large anisotropy while the Heisenberg model must be used to describe a system with weak anisotropy.

2. Investigation of remanent magnetization

We report in this section the results of an investigation of the remanent magnetization of strongly anisotropic (type I) and weakly anisotropic (type II) spin glasses of the system $Zn_x Cd_{1-x} Cr_2Se_4$, using a rotating-sample magnetometer.

We investigated the thermoremanent magnetization obtained after cooling the samples from the paramagnetic temperature region to 4.2 K in a constant magnetic field H). We designate this type of remanent magnetization by TRM
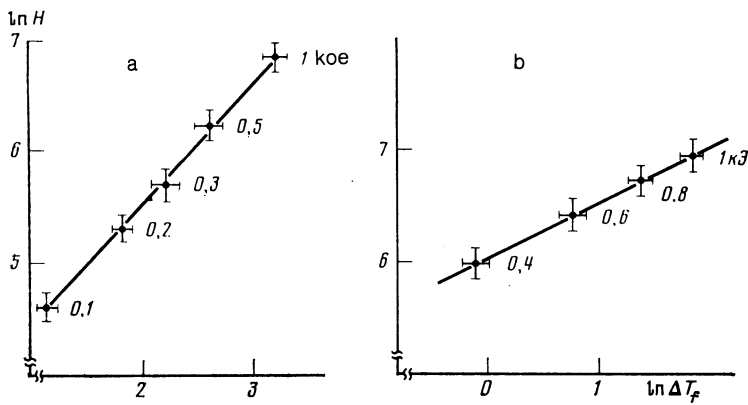
FIG. 1. Shift of "freezing" temperature vs the magnetic field for samples of type I (a) and type II (b). The coordinates are $\ln H$ and $\ln(\Delta T_f) = \ln[T_{f(0)} - T_{f(H)}]$. The parameter δ is equal to 0.96 ± 0.1 for samples of type I and to 2.14 ± 0.2 for samples of type II.

(4.2 K, H). We investigated also the isothermal remanent magnetization obtained by isothermal magnetization of the samples in a field for a time t_H at a temperature T. This type of remanent magnetization will be designated IRM (T,H,t_H). Prior to the isothermal magnetization, the sample was either cooled from a temperature higher than the "freezing" temperature to a temperature T in a zero field, or demagnetized at a temperature T by a rotating magnetic field whose strength decreased smoothly from 7 kOe to zero within in a time on the order of a minute. The influence of the rotating field was investigated in Ref. 7. In addition, we use M_R to designate the residual magnetization at the instant of time t after turning off the magnetizing field, and M_{RS} to designate the saturated remanent magnetization, i.e, the one produced by a sufficiently strong field such that $TRM \approx IRM$, while the remanent magnetization itself is independent of the magnetizing field.

2.1. Dependences of the remanent magnetization on the magnetizing field

It was found that the dependences of TRM and IRM on H at 4.2 K take for the samples of both types a form typical of spin glasses. The dependence of TRM (4.2 K) on H has a maximum, whereas the IRM (4.2 K) dependence takes the form of a typical saturation curve. The saturated remanent magnetization M_{RS} at $T = 4.2$ K is ~ 40 G for samples of type I and ~ 3.5 G for samples of type II. Note that the saturation magnetization of both strongly and weakly anisotropic samples ~ 360 G.

2.2 Relaxation of remanent magnetization of spin glasses of the system $Zn_x Cd_{1-x} Cr_2 Se_4$

Investigations of the relaxation of the remanent magnetization at 4.2 K have shown [9] that the remanent-magnetization relaxation of samples of both types, produced by different methods (TRM or IRM in different magnetizing field and at different magnetization durations (for IRM) is described by a power law, with the exponent α dependent on the parameters of the magnetization process, i.e., the relaxation of the residual magnetization is described by the power law

$$M_R = M(t/\tau)^{-\alpha}, \tag{2}$$

where M and τ are parameters that have respectively the dimensions of magnetization and time, while t is the time elapsed after the magnetizing field is turned off.

It turned out that $\alpha_{TRM} \approx \alpha_{IRM}$ in sufficiently strong

fields H, but with decreasing field the values of α_{TRM} decrease monotonically, while those of α_{IRM} increase (Fig. 2). Note that under identical conditions the exponent α is much smaller for samples of type I, i.e., the remanent magnetization of strongly anisotropic samples relaxes more slowly. The exponent α_{RS} corresponding to saturated remanent magnetization at 4.2 K is equal to 0.05 and 0.15 for samples I and II, respectively.

For magnetizing fields weaker than the remanent-magnetization saturation field. The value of IRM and the corresponding exponent α_{IRM} depend on the magnetization duration, viz., IRM increases with increase of t_H while α_{IRM} decreases Fig. 3). It should be noted that the relaxation of the remanent magnetization with time cannot be used to determine simultaneously all the parameters M, τ, and α of the relaxation law. It follows from (2) that $\ln M_R = \ln(M\tau^\alpha) - \alpha \ln t$, i.e., the experimental log-log plot of $M_R(t)$ yields the exponent α and the value of $\ln M\tau^\alpha$, but it is impossible to determine from the latter separately the parameters M and τ. Additional data are therefore necessary to determine M and τ.

To determine the parameters of the relaxation law we have used in the present study a relation that follows from (2):

$$\ln M_R(t) = \ln M - \alpha \ln(t/\tau). \tag{3}$$

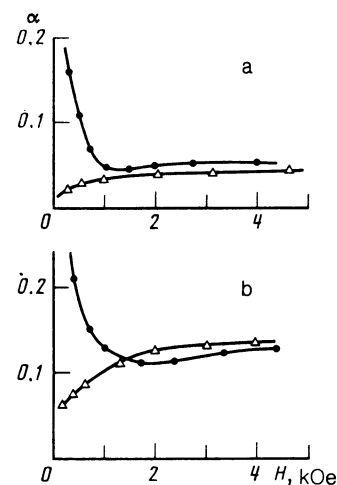Assume that only the value of α is altered by some changes of

FIG. 2. Dependence of the exponent α on the magnetizing field H for TRM ($\triangle$) and IRM ($\bullet$) at 4.2 K, $t_H = 30$ s; a—for samples of type I, b—type II.
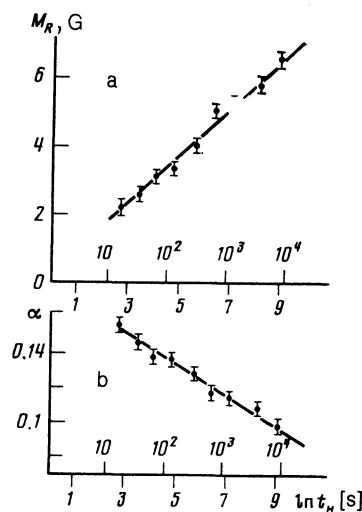
FIG. 3. Dependences on the magnetization duration t_H for samples of type I: a—of the quantity IRM (4.2 K, $H = 400$ Oe) at $t = 50$ s, b—of the value of α which corresponds to IRM (4.2 K, $H = 400$ Oe).

the magnetization conditions. If the value of M_R (t_0) at fixed t_0 and the corresponding values of α are determined after each magnetization process, and $\ln M_R$ $(t_0$ is plotted next as a function of α, it follows from (3) that the experimental points should lie on a straight line whose (3) that the experimental points should lie on a straight line whose slope yields $\ln(t_0/\tau)$ and hence the value of τ, and its intercept with the coordinate axis yields the value of $\ln M$.

2.3 Experimental determination of the parameters M and τ for type-I samples.

It turned out that the plot of $\ln M_{Rs}$ (50 s) vs α_{RS} has a pronounced linear section in the temperature range from 4.2 K to 10K (Fig. 4). This has made it possible to determine the parameters $M_0 = 120$ G $\pm 20\%$ and $\tau_0 \sim 10^{-7}$ s for the type-I samples in the indicated temperature range (the zero substrict indicates that these parameters correspond to saturated remanent magnetization); the exponent α_{RS} in this temperature range is directly proportional to the temperature, i.e., $\alpha_{RS} \approx T/T_0$, where $T_0 \approx 80$ K (Fig. 5).

Thus, in the temperature range from 4.2 K to 10 K the relaxation law for the remanent magnetization of type-I samples is
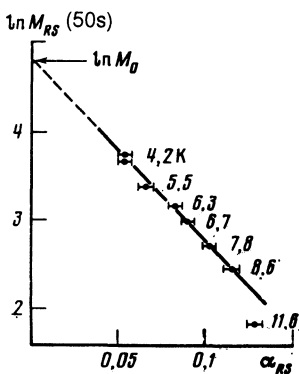

FIG. 5. Dependence of α_{RS} on the temperature for samples of type I.

$$M_{RS}[\mathrm{G}] = 120 \exp\left(-\frac{T[\mathrm{K}]}{80}\ln\frac{t[\mathrm{s}]}{10^{-7}}\right).\qquad(4)$$

It has turned out that for unsaturated isothermal remanent magnetization it is also possible to determine the relaxation-law parameters. Figure 6 shows experimental plots of $\ln M_R$ (50 s) vs α, corresponding to IRM (4.2 K; $t_H = 30$ s) at different magnetization fields and to IRM (4.2 K; $H = 400$ Oe) at different magnetization durations. This plot has a linear segment containing the points corresponding to IRM (4.2 K, $t_H = 30$ s) $H < 900$ Oe, and also the points corresponding to IRM (4.2 K, $H = 400$ Oe) at all magnetization durations. The value of τ determined from this linear section for unsaturated isothermal remanent magnetization coincides with τ_0 and amounts to $\sim 10^{-7}$ s, whereas the value of the parameter M for unsaturated isothermal remanent magnetization is approximately 50 G, or smaller than M by 2.4 times. Note from the plot of Fig. 6 it is possible to determine also the value $M_0 = 120$ G, which agrees with the one determined above.

2.4. Experimental determination of the parameters M and τ for type-II samples

We did not succeed in observing for type-II samples a linear section on the plot of $\ln M_{RS}$ (50 s) vs α_{RS} with change of temperature. Nor was a linear section observed in the temperature dependence of α_{RS}. It is possible that for type-II samples the range of temperatures in which these plots are linear is below 4.2 K. For unsaturated remanent magnetization, however, it was possible to observe at 4.2 K a

FIG. 4. Dependence of $\ln M_{RS}$ (50 s) on α_{RS} for samples of type I. The figure indicates the values of the temperatures; $\ln M_0 = 4.8$, ($M_0 = 120$ G), $\ln$ $(50/\tau_0) = 20.5$ $(\tau_0 \sim 10^{-7}$ s).
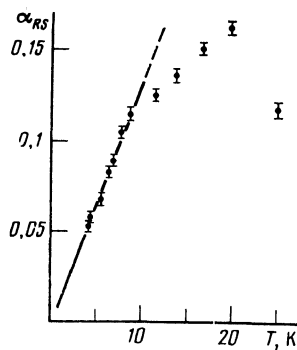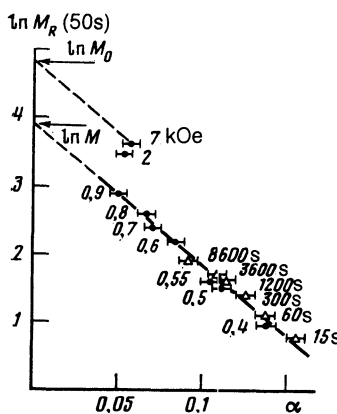

FIG. 6. Dependence of $\ln M_R$ (50 s) on α for samples of type I: (● — IRM(4.2 K, $t_H = 30$ s), ∆—IRM(4.2 K, $H = 400$ Oe); $M_0 \approx 120$ G, $M \approx 50$ G, $\ln$ $(50/\tau) = \tau \sim 10^{-7}$ s).
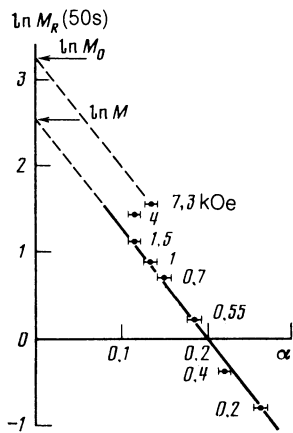
FIG. 7. Dependence of $\ln M_R$ (50 s) on α for samples of type II, obtained for IRM (4.2 K, $t_H = 30$ s) at different values of H; $M_0 \approx 50$ G; $m \approx 12 G$; $\ln 50/\tau \approx 12.5(\tau \sim 10^{-4}$ s).

linear section in the plot of $\ln M_R$ (50 s) vs α, as shown in Fig. 7. The parameters determined from this dependence are $\tau \sim 10^{-4}$ s and $M \approx 12$ G. Assuming that $\tau_0 \approx \tau$ (by analogy with the results for type-I samples), we can obtain $M_0 \approx 30$ G. Note that for type-II samples we have a ratio $M_0/M \approx 2.5$, close to the analogous ratio for type-I samples.

3. Discussion of results

Experiments[8,11] have shown that anisotropy influences the properties of spin glasses of the type CuMn and AgMn. In addition, it was also demonstrated in theoretical papers[4,5] that it is important to take the anisotropy into account when the state of spin glass is considered. The energy spectrum of spin glass comprises a set of energy barriers of varying heights, which separate a set of metastable states of approximately equal energy. This is confirmed for Ising spins by results of numerical experiments of the "random-bond" model with the aid of the Monte Carlo method.[10] For a system of Heisenberg spins, however, the absence of anisotropic interaction causes any configuration of the spins to be degenerate in the angle of rotation around the crystallographic axes, i.e., if each spin is rotated through the same angle around an arbitrarily chosen axis the energy of the system remains unchanged, since the Heisenberg interaction is isotropic. This degeneracy is lifted in the presence of anisotropy The energy barriers between the metastable states depend on the anisotropy energy. Consequently, the quantitative characteristics of spin glasses (for example, the "freezing" tem-

perature and others) should also depend on the anisotropy energy.

The experimental results of the present study favor this assumption. At practically equal paramagnetic Curie points, strongly anisotropic spin glasses (of type I) have a "freezing" temperature almost double that of the weakly anisotropic ones. The influence of the anisotropy can explain also the change of the exponent δ in the law governing the shift of the "freezing" temperature in a constant field H. In addition, strongly anisotropic spin glasses (of type I) are more magnetically viscous in the sense that under identical conditions the value of their remanent magnetization is substantially larger, and it relaxes more slowly than the remanent magnetization of weakly anisotropic spin glasses. The parameter M of strongly anisotropic samples is larger, while the parameter α, which characterizes the relaxation rate of the remanent magnetization, is smaller in strongly anisotropic samples.

We note one more result of the present study. It was found that the parameter M in the law governing the relaxation of unsaturated IRM does not depend on the magnetic field and on the magnetization duration (only the exponent α depends on these conditions). This result cannot be explained in the framework of the model of noninteracting clusters (in this model M increases with H in fields weaker than the IRM saturation field). This result can be regarded as direct evidence that the spin-glass state is a cooperative state of all the spins of the system.

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