

# Oscillations in the residual magnetization of spin glasses

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Oscillations in the residual magnetization have been detected in the spin glass  $\text{Zn}_{0.4}\text{Cd}_{0.6}\text{Cr}_2\text{Se}_4$  after successive magnetization of the specimen in fields of different magnitude and direction. The specimen was magnetized in a field  $H_1 = 7$  kOe and then in a field  $H_2 = 400$  Oe pointing in the opposite direction. It was found that, after the magnetizing fields were turned off, the residual magnetization consisted of two oppositely directed components, one of which relaxed slowly and was parallel to  $H_1$  and the other relaxed rapidly and was parallel to  $H_2$ . This suggests that the spin glass contains magnetic clusters with different energy and temporal characteristics.

## 1. INTRODUCTION

The characteristic property of spin glasses is their magnetic viscosity. Magnetization and the relaxation of magnetization do not occur in spin glasses at the exponential rate observed for other magnets. It has been shown<sup>1-3</sup> that the magnetization of spin glasses (for historical reasons, it is referred to as the residual magnetization  $M_R$ ) relaxes slowly in accordance with the expression  $M_R \propto (t/\tau)^{-\alpha}$  for  $t \gg \tau$ , where  $t$  is the time that has elapsed since the external magnetic field was turned off. The constant  $\tau$  is usually  $10^{-13}$ – $10^{-4}$  s and  $\alpha \ll 1$ . The rate of remagnetization in spin glasses is a function of  $\tau$  and  $\alpha$ . It has been shown<sup>2</sup> that the magnitude of  $\alpha$  for a given specimen can be varied within wide limits by varying the temperature, magnetizing field, and duration of magnetization. The corresponding mean duration of relaxation processes varies from fractions of a second to 10 hours or more. The magnetic viscosity of spin glasses can be explained in terms of the cluster model.<sup>3</sup> The essential point of this model is that the magnetic subsystem of a spin glass is microscopically inhomogeneous and has an enormous number of metastable states which may be looked upon as the perturbed states of individual clusters. It is assumed that the parameters of these clusters are distributed between wide limits. This approach does not exclude the possibility of an interaction between them or of the existence of an infinite cluster, i.e., long-range order in some particular parameter.

The cluster model of spin glasses has one interesting consequence. Let us consider small perturbations in a spin glass during magnetization in low enough field  $H$  and at a low enough temperature. A sufficiently low external field will produce a realignment of the magnetic moments of clusters that are weakly bound to the environment and are well separated from one another, i.e., do not interact. The analysis is then much easier. For simplicity, we shall assume that the  $i$ -th cluster has only two stable states, namely, magnetic moment up or down. One of these states is the ground state and the other a metastable state. The states are separated by an energy barrier  $E_i$ . The quantities  $E_i$  are distributed between wide limits. The relaxation time  $\tau_i$  from the metastable state to the ground state of a cluster is described by the Arrhenius law  $\tau_i = \tau_0 \exp(E/k_B T)$ , where  $k_B$  and  $T$  are, re-

spectively, the Boltzmann constant and temperature. Let  $h_i = E_i/m_i$  be the field sufficient to realign the magnetic moment  $m_i$  of the  $i$ th cluster. We shall arrange the clusters in the order of increasing  $h_i$ , i.e.,  $h_{i+1} > h_i$ . We shall assume that we then also have  $\tau_{i+1} > \tau_i$ . This is possible, for example, when  $E_i \propto n_i$  and  $m_i \propto n_i^{1/2}$ , where  $n_i$  is the number of magnetic ions per cluster. The quantities  $h$  and  $\tau$  will then monotonically increase with increasing  $n$ . It turns out that this system exhibits a relatively unusual phenomenon, namely, damped oscillations of residual magnetization  $M_R(t)$  if the system is first successively magnetized in fields  $H_1^+ > H_2^- > H_3^+ \dots > H_n^-$  (the superscripts  $+$  and  $-$  indicate the up and down field directions, respectively), provided the entire process of successive magnetization can be performed in a sufficiently short time  $\Delta t < \tau_k$ , where  $k$  is defined by the condition  $h_k < H_n$ . One can then observe  $n/2$  oscillations. In fact, when  $H_1^+$  is turned on, the magnetic moments of all clusters with  $h_i \leq H_1^+$  will point up, the field  $H_2^-$  will realign in the downward direction the magnetic moments of a fraction of these clusters with  $H_i \leq H_2^-$ , and so on. The resultant magnetic moment of the system will therefore be

$$\mathfrak{M} = - \sum_1^a m_i + \sum_{a+1}^b m_i - \sum_{b+1}^c m_i + \dots,$$

where  $a, b, c, \dots$  are determined from the conditions  $h_a = H_n^-, h_b = H_{n-1}^+, h_c = H_{n-2}^-, \dots$

When the external field is turned off, the magnetic moment of the system will at first rise, largely as a result of the relaxation of the moment

$$- \sum_1^a m_i,$$

which points down (this is the magnetic moment of the group of clusters with  $h_i \leq H_n^-$ ), and will then rise to the value

$$\sum_{a+1}^b m_i - \sum_{b+1}^c m_i + \dots$$

in a time  $\sim \tau_a$ . The moment  $\mathfrak{M}$  will then decrease, mainly as a result of relaxation of the magnetic moment of the group of clusters with  $H_n^- < h_i < H_{n-1}^+$ , down to  $-\sum_{b+1}^c m_i + \dots$

in a time  $\sim \tau_b$ , and so on. Such oscillations of the moment  $\mathfrak{M}$  of the system are possible because the inequality  $h_b > h_a$  leads to  $E_b > E_a$ , i.e.,  $\tau_b \gg \tau_a$  since  $\tau_i \propto \exp(E_i/k_B T)$ . Actually, for each  $E_i$  and  $\tau_i$ , there will be clusters with different magnetic moments  $m_i$ , so it is not possible to arrange all the clusters strictly in the order of increasing  $h$  and  $\tau$ . However, whenever this approach does correspond to reality, at least approximately, it is possible to observe "smeared out" and time-overlapping oscillations in the residual magnetic moment.

To verify these predictions experimentally, we have investigated spin glasses of the form  $\text{Zn}_x \text{Cd}_{1-x} \text{Cr}_2 \text{Se}_4$  with "freezing temperature"  $T_f \approx 21$  K and  $x \approx 0.4$ . The magnetic properties of these spin glasses have been examined in detail in the literature.<sup>2,4-6</sup>

## 2. EXPERIMENTAL METHOD

The residual magnetic moment was measured in a rotating-specimen magnetometer.<sup>7</sup> The axis of rotation of the specimen lay along the  $Z$  axis of the laboratory Cartesian frame. The external field  $H^+$  or  $H^-$  was applied along the  $X$  direction (the superscripts  $+$  and  $-$  indicate that the field had the positive and negative direction, respectively). A synchronous motor, rotating the specimen about the  $Z$  axis at 18 Hz, was turned on after the external field was removed. The specimen was in the form of a disk, placed symmetrically relative to the axis of rotation in the  $XY$  plane (the plane of rotation). The rotating specimen induced an emf in the receiving coils of the magnetometer at a frequency proportional to its residual magnetic moment  $\mathfrak{M}_R$ . The emf was measured using an amplifier with the UPI-1 synchronous detector. The reference signal was generated by a chopper rotating synchronously with the specimen and placed between the photo- and emitting-diodes. The threshold sen-

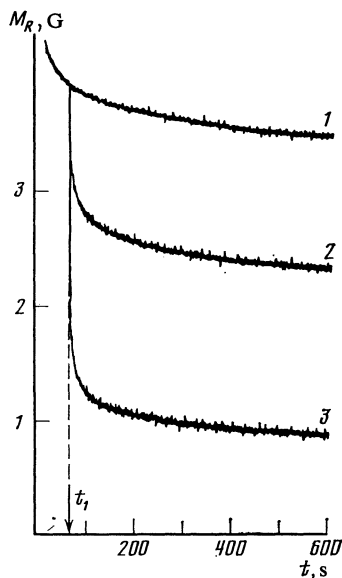


FIG. 1. Relaxation of  $M_R$  at 4.2 K in  $\text{Zn}_{0.4} \text{Cd}_{0.6} \text{Cr}_2 \text{Se}_4$  after magnetization by  $H = 7$  kOe (the field  $H$  was turned on at a time  $t = 0$ ). The following fields were turned on at time  $t_1$ : 1— $H_\omega = 0$ ; 2— $H_\omega = 200$  Oe; 3— $H_\omega = 400$  Oe.

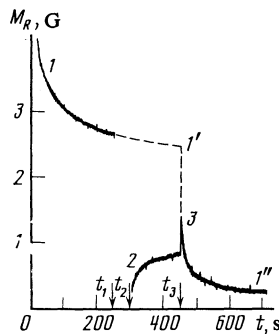


FIG. 2. Relaxation of  $M_R$  at 4.2 K in  $\text{Zn}_{0.4} \text{Cd}_{0.6} \text{Cr}_2 \text{Se}_4$  after magnetization in the field  $H_{1^+} = 7$  kOe (the field  $H_{1^+}$  was turned on at time  $t = 0$ ). Curve 1-1'-1'': the rotating field  $H_\omega = 400$  Oe was turned on at time  $t_3$ . Curve 1-2-3-1'': the specimen was magnetized by the constant field  $H_2^- = 400$  Oe opposite in direction to  $H_{1^+}$  between  $t_1$  and  $t_2$ ; the rotating field  $H_\omega = 400$  Oe was turned on at time  $t_3$ .

sitivity of the system for the residual magnetic moment was  $\sim 10^{-5}$  G·cm<sup>3</sup>. The standard volume of the specimens was 0.001–0.01 cm<sup>3</sup>. We note that the rotating-specimen magnetometer can be used to determine the residual magnetic moment even in the presence of an external magnetic field. The residual magnetic moment rotates together with the specimen, whereas the magnetic moment induced by the external field is parallel to  $H$  or oscillates relative to  $H$  at the even harmonics of the rotation frequency if the specimen is anisotropic. When  $\mathfrak{M}_R = 0$ , the parasitic signal at the rotation frequency does not exceed  $10^{-5}$  G·cm<sup>3</sup> in a field of about 100 Oe. Thus, by rotating the specimen with the residual magnetic moment in the external magnetic field, it was possible to investigate the relaxation of its residual magnetic moment. In other words, we were able to investigate the influence of the rotating magnetic field  $H_\omega$  on the residual magnetic moment.

Figure 1 shows the effect of the rotating magnetic field  $H_\omega$  on the relaxation of the residual magnetization  $M_R$  at 4.2 K in the case of the  $\text{Zn}_{0.4} \text{Cd}_{0.6} \text{Cr}_2 \text{Se}_4$  specimen magnetized isothermally in a field of 7 kOe. Curve 1 corresponds to relaxation in zero field (the initial time  $t = 0$  corresponds to the time at which the magnetizing field was turned off). Curves 2 and 3 correspond to the rotation of the specimen in fields  $H_\omega = 200$  and 400 Oe (the field  $H_\omega$  was turned off at time  $t_1$ ). We found that the relaxation process could be described by  $M_R \sim t^{-\alpha}$  for different  $H_\omega$ , including  $H_\omega = 0$  (Ref. 2). The exponent  $\alpha$  is proportional to  $H_\omega^2$ . Thus, the rate of relaxation of  $M_R$  can be substantially increased by introducing the rotating field  $H_\omega$ .

## 3. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 2 shows the relaxation of residual magnetization  $M_R$  at 4.2 K for the  $\text{Zn}_{0.4} \text{Cd}_{0.6} \text{Cr}_2 \text{Se}_4$  specimen magnetized by the field  $H_{1^+} = 7$  kOe (curve 1-1', the origin of time corresponds to the point at which the field  $H_{1^+}$  was turned off). The rate of relaxation of  $M_R$  can be substantially increased by turning on the rotating magnetic field  $H_\omega = 400$  Oe (curve 1' - 1'').

The same figure shows the behavior of  $M_R$  for a speci-

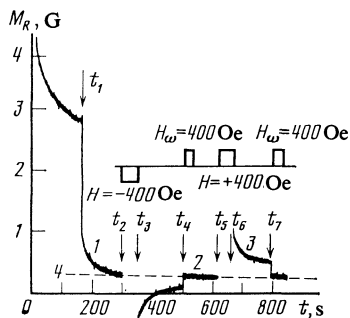


FIG. 3. Relaxation of  $M_R$  at 4.2 K in  $\text{Zn}_{0.4}\text{Cd}_{0.6}\text{Cr}_2\text{Se}_4$  after magnetization in the field  $H_1^+ = 7$  kOe (the field  $H_1^+$  was turned off at time  $t = 0$ ). The rotating field  $H_\omega = 400$  Oe was applied to the specimen between  $t_1$  and  $t_2$ ; a constant field  $H_2^- = 400$  Oe was applied between  $t_2$  and  $t_3$ ; the rotating field  $H_\omega = 400$  Oe was introduced for 30 s at  $t_4$ ; the constant field  $H_3^+ = 400$  Oe was present between  $t_5$  and  $t_6$ , and the rotating field  $H_\omega = 400$  Oe was applied at  $t_7$ .

men magnetized in the field  $H_1^+ = 7$  kOe under the following conditions. The rotation of the specimen was stopped at time  $t_1$  (the specimen was aligned in a standard way in the initial position) and the field  $H_2^- = 400$  Oe, opposite to  $H_1^+$ , was turned on. When the former field was turned off (at time  $t_2$ ), the residual magnetization was found to decrease, but then began to increase in the positive direction (curve 2). To accelerate the relaxation process, the rotating field  $H_\omega = 400$  Oe was turned on at time  $t_3$ . The residual magnetization rose rapidly and then relaxed along the curves 3–1". This behavior of  $M_R$  can only be understood in terms of the cluster model of the spin glass. In fact, the magnetic moment of the group of clusters with  $h_i < H_2^-$  will be negative, whereas the magnetic moment of the group with  $H_2^- < h_i < H_1^+$  will be positive. The magnetic moment of the group of clusters with small  $h_i$  relaxes more rapidly, which explains the increase in  $M_R$  after magnetization in the field  $H_2^-$ . When the rotating field is introduced, this produces a still more rapid relaxation of the magnetic moment of the group of clusters (rapid rise in  $M_R$  at time  $t_3$ ), and subsequent relaxation is due to relaxation field of the magnetic moment of the group of clusters with  $H_2^- < h_i < H_1^+$ .

Figure 3 shows the relaxation of  $M_R$  when, after magnetization in  $H_1^+ = 7$  kOe, the specimen is partially demagnetized in the rotating field  $H_\omega = 400$  Oe, and is then magnetized in the field  $H_2^- = 400$  Oe. After the field  $H_2^-$  is turned off (at time  $t_3$ ), the residual magnetization is negative (curve 2). It then relaxes in the absence of the external field to zero, changing sign and rising to level 4, which gradually relaxes to zero as shown by the dashed line in Fig. 3 (the

relaxation process was accelerated by turning on the rotating field  $H_\omega = 400$  Oe at time  $t_4$ ). If, between  $t_5$  and  $t_6$ , the specimen is magnetized by the constant field  $H_3^+ = 400$  Oe, the relaxation of  $M_R$  after this field is turned off occurs along curve 3 (symmetric to curve 2 relative to level 4).

The behavior of  $M_R$ , shown in Fig. 3, can readily be understood in terms of the cluster model. The application of the rotating field  $H_\omega \ll H_1^+$  ensures that only clusters with sufficiently high  $h_i$  become magnetized, and the sum of their magnetic moments determines level 4 and relaxes very slowly even in the rotating field. Subsequent magnetization by fields  $H_2^-$ ,  $H_3^+ \ll H_1^+$  ensures that the group of clusters with  $h_i < H_2^-$ ,  $H_3^+$  acquires a magnetic moment which relaxes at a much greater rate, i.e., relatively rapid relaxation of the magnetic moment of the group of clusters with small  $h_i$  is observed (curves 2 and 3 in Fig. 3) against the background of the slowly relaxing magnetic moment of the group of clusters with large  $h_i$  (level 4).

#### 4. CONCLUSIONS

We have observed oscillations in the residual magnetization of a spin glass after the specimen has been magnetized by fields of different strength and direction. In other words, by magnetizing the specimen in some particular direction by the field  $H_1^+$  and then in the opposite direction by the field  $H_2^- < H_1^+$ , one is able to observe a spontaneous rise in the residual magnetization, followed by its relaxation to zero. Under certain definite conditions, it is even possible to observe a spontaneous variation in the direction of the residual magnetization. This behavior of  $M_R$  can be explained in terms of the cluster model of spin glasses. It is then clear that the temporal and energy characteristics of clusters are distributed between wide limits. An analogous behavior of  $M_R$  was previously seen<sup>8</sup> in the spin glass  $\text{Fe}_{65}\text{Ni}_{20}\text{Cr}_{15}$  at  $T = 4.2$  K ( $T_f \approx 20$  K).

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