

## Memory and superposition in a spin glass

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Nonequilibrium dynamics in a Ag(Mn) spin glass are investigated by measurements of the temperature dependence of the remanent magnetization. Using specific cooling protocols before recording the thermo- or isothermal remanent magnetizations on reheating, it is found that the measured curves effectively disclose nonequilibrium spin glass characteristics such as aging and memory phenomena as well as an extended validity of the superposition principle for the relaxation. The usefulness of this ‘‘simple’’ dc method is discussed, as well as its applicability to other disordered magnetic systems.

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### I. INTRODUCTION

The nonequilibrium nature of spin glasses and other glassy systems at low temperature implies that a comprehensive study of the dynamics usually requires sophisticated dc or ac relaxation experiments.<sup>1,2</sup> Dc relaxation is typically used to investigate the aging properties in spin glasses. In such experiments, the sample is first cooled from above the transition temperature  $T_g$  to a constant temperature. After a waiting time  $t_w$ , a small dc field is applied (or cut off), and the magnetization is recorded versus time. The relaxation occurs in a characteristic way, and shows a clear dependence on  $t_w$ .<sup>3,1,2</sup> Relaxation of the low-frequency ac susceptibility<sup>4-6</sup> is a prime tool to investigate memory effects in glassy magnets. When cooling from above  $T_g$ , a halt at constant temperature  $T_s < T_g$  is made during  $t_s$ , allowing the system to relax towards its equilibrium state at  $T_s$ ; both components of the ac susceptibility then decay in magnitude. This equilibrated state becomes frozen in on further lowering the temperature, and is retrieved on reheating. The weak low-frequency ac field employed in this kind of experiments does not affect the nonequilibrium processes intrinsic to the sample, but only works as a nonperturbing probe of the system.

In the present paper, an alternative method to investigate nonequilibrium dynamics is used; ‘‘simple’’ remanent magnetization measurements on a Ag(Mn) spin glass sample. The magnetization curves are recorded on heating after specific cooling protocols. The comparison of the different curves yields information on dynamic properties such as aging and memory phenomena.

### II. MEMORY AND SUPERPOSITION

The sample used in the experiments is a dilute magnetic alloy: the archetypical three-dimensional spin glass Ag(11 at % Mn). This material is chosen as an established reference system to certify the validity of the dc method presented here.

The sample was prepared by melting pure Ag and Mn together at  $T=1000^\circ\text{C}$  in an evacuated atmosphere. After annealing the sample at  $850^\circ\text{C}$  for 72 h it was quenched to room temperature. The experiments were performed in a noncommercial low-field superconducting quantum interfer-

ence device (SQUID) magnetometer.<sup>7</sup> The dc magnetic field is generated by a small superconductive solenoid coil always working in persistent mode during measurements. The background field was less than 1 mOe.

As seen in Fig. 1, the zero-field cooled (ZFC) magnetization exhibits a characteristic cusp at  $T=33\text{ K}$ , which roughly defines the spin glass temperature  $T_g$  of our sample; below this temperature, the field cooled (FC) magnetization remains constant. Fig. 2 presents the temperature dependence of (a) the thermo-remnant (TRM) and (b) the isothermal remanent (IRM) magnetization. Both were measured on heating in zero field from a low temperature. The different curves have been recorded after cooling the sample using the protocol described as follows: (1) The sample is cooled in constant field  $H_o=0.1\text{ Oe}$  (TRM) or zero field (IRM) from a reference temperature above  $T_g$  to a stop temperature  $T_s$  ( $T_s < T_g$ ), where the sample is kept a stop time  $t_{s1}$  without changing the field. (2) A field change is made: In the TRM case the field is either cut to zero (zero-field stop, ZFS) or kept constant (field stop, FS); for the IRM,  $H_o$  is applied. The magnetic field value is kept during a time  $t_{s2}$ . (3) The field is then shifted back to its initial value and the sample is immediately cooled to a lowest temperature  $T_{min}$ , where (4) the field is cut to zero (TRM) [or kept at zero (IRM)] and the

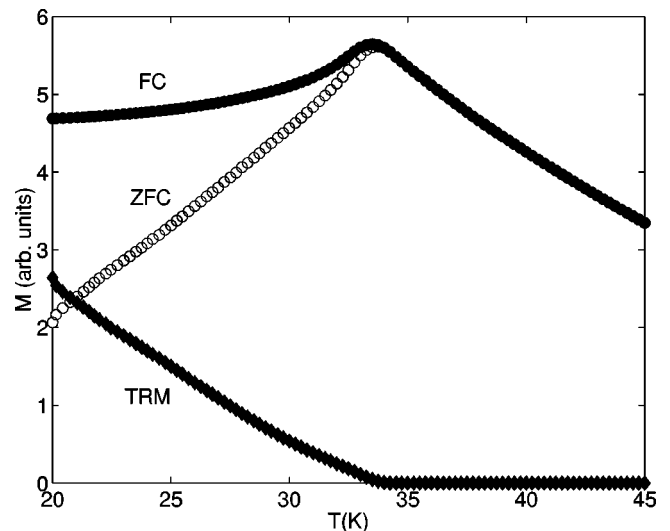


FIG. 1. ZFC, FC, and TRM magnetizations for  $H=0.1\text{ Oe}$ .

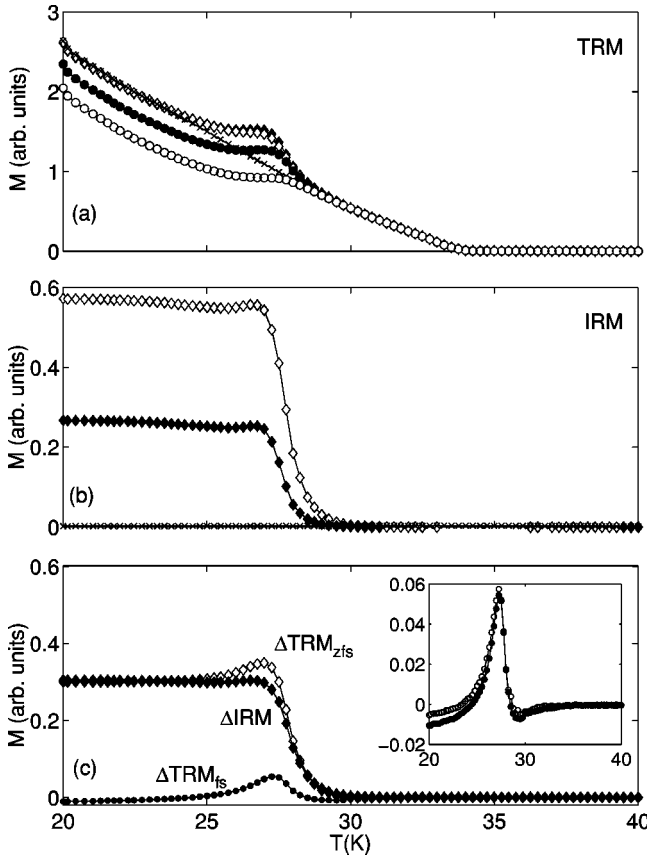


FIG. 2. TRM (a) and IRM (b) magnetizations measured after using different cooling protocols;  $T_s=27$  K,  $H=0.1$  Oe. (c) displays the difference between pairs of curves shown in (a) and (b):  $\Delta\text{IRM}$ ,  $\Delta\text{TRM}_{\text{ZFS}}$ , and  $\Delta\text{TRM}_{\text{FS}}$ . The inset shows the difference ( $\Delta\text{TRM}_{\text{ZFS}}-\Delta\text{IRM}$ ) and  $\Delta\text{TRM}_{\text{FS}}$  (which is marked by the same symbol as in the main frame).

remanent magnetization  $M_{\text{TRM}}(T)$  (denoted  $\text{TRM}_{\text{ZFS}}$  and  $\text{TRM}_{\text{FS}}$  for the case of ZFS and FS, respectively) or  $M_{\text{IRM}}(T)$  (denoted IRM) is measured on heating the sample at a constant heating rate.

The different curves in Fig. 2 are measured for two different values of  $t_{s1}$ : 0 s and 10 000 s, respectively, and in all cases  $t_{s2}=10\,000$  s;  $T_s=27$  K;  $T_{\text{min}}=20$  K. The reference curves, without any field stops, are added. Figures 2(a) and 2(b) show that the 0 s  $\text{TRM}_{\text{ZFS}}$  curve lies significantly below the 10 000 s curve whereas the 0 s IRM curve lies well above the 10 000 s one. These curves reflect the paramount influence that aging has on the magnetic relaxation in spin glasses. For comparison, Fig. 3 presents parallel dc-relaxation experiments (ZFC, FC, and TRM), performed at the temperature  $T_s$ , using the same small magnetic field, for waiting times of  $t_w=0$  s and  $t_w=10\,000$  s; the results for  $t_w=1000$  s are added to show the continuity of the waiting time dependence. The observation time,  $t$ , is defined as the time elapsed after the field application (cut off).

Both  $\text{TRM}_{\text{FS}}$  curves in Fig. 2 (a) lie significantly above the reference curve, conveying that a considerable reinforcement of the spin structure occurs during the stop time  $t_{s1}+t_{s2}$ . The effect even overcomes the downward relaxation

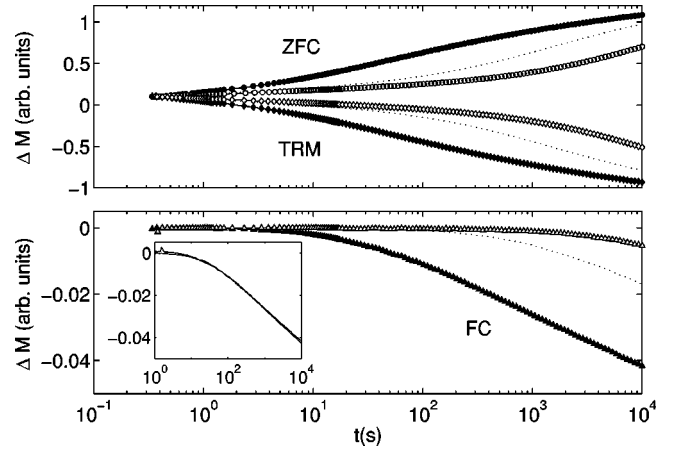


FIG. 3. ZFC, TRM (a) and FC (b) relaxations at  $T=27$ K,  $H=0.1$  Oe for different waiting times:  $t_w=0$  s (filled symbols),  $t_w=1000$  s (dotted lines), and  $t_w=10^4$  s (open symbols). The insert shows the agreement between ZFC+TRM and FC relaxations ( $t_w=0$  s).

of the FC magnetization occurring during the stop at 27 K (cf. Fig. 3). One notices that the difference between stopping 10 000 and 10 000+10 000=20 000 s at  $T_s$  is significant but comparably small. Provided the experiments are made at low enough field, where there is a linear response to a field change, one can relate the results from different relaxation experiments through the principle of superposition.<sup>8,9</sup> This implies, e.g., that:

$$M_{\text{ZFC}}(t_w, t) = M_{\text{FC}}(0, t_w + t) - M_{\text{TRM}}(t_w, t).$$

Figure 2(c) shows difference plots,  $\Delta\text{TRM}_{\text{ZFS}}$  and  $\Delta\text{IRM}$ , of the  $\text{TRM}_{\text{ZFS}}$  and IRM curves shown in Figs. 2(a) and 2(b) giving a direct measure of the frozen in excess magnetization due to aging. Also plotted in the Fig. 2 is a difference plot of the  $\text{TRM}_{\text{FS}}$  curves:  $\Delta\text{TRM}_{\text{FS}}$ . The inset shows a plot of  $\Delta\text{TRM}_{\text{ZFS}}-\Delta\text{IRM}$  and  $\Delta\text{TRM}_{\text{FS}}$ , the two curves look very similar and suggest an extended validity of the superposition principle to apply also to the temperature dependence of the frozen in excess magnetization of a spin glass. (IRM reflects  $M_{\text{ZFC}}$ ,  $\text{TRM}_{\text{ZFS}}$  reflects  $M_{\text{TRM}}$ , and  $M_{\text{FC}}$  corresponds to  $\Delta\text{TRM}_{\text{FS}}$ .) The applicability of the ‘‘ordinary’’ superposition principle for the relaxation of our spin glass is illustrated in Fig. 3; the insert shows the sum of the ZFC and TRM relaxations obtained for  $t_w=0$  s and the FC relaxation for the same waiting time. The curves are plotted without markers to be able to visually distinguish that there are two of them; as expected, they are closely equal to each other.

We have also studied the effect of simple field stops in the ZFC and FC magnetizations. Figure 4 shows the ZFC, FC, and TRM reference curves already presented in Fig. 1, as well as the same curves recorded on reheating after a stop of duration  $t_s=10^4$  s in constant field at  $T_s=27$  K. In the TRM and FC case, the field is thus kept to its  $H=0.1$  Oe value, while for the ZFC, it remains zero. The FC curve is only weakly affected by the stop,<sup>10</sup> whereas the ZFC and TRM curves are considerably affected.<sup>11</sup> The curves reflect the reinforcement of the spin structure that occurs during the

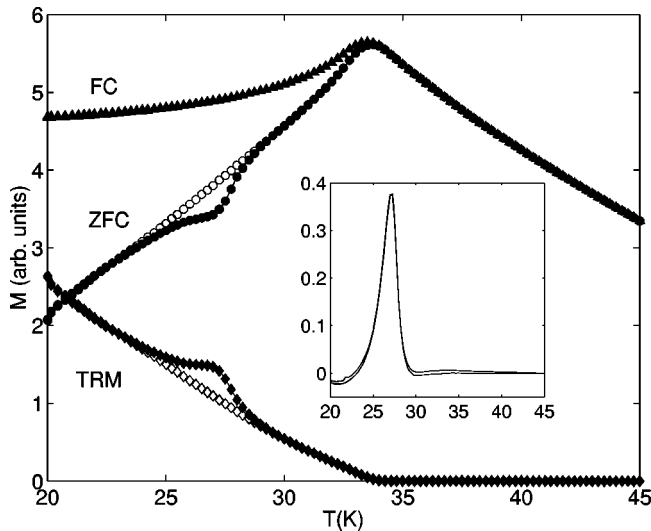


FIG. 4. ZFC, FC, and TRM magnetizations for  $H=0.1$  Oe. Two curves (ZFC and TRM) measured after a  $t=10^4$  s stop at  $T=27$  K while cooling are added; the inset shows the difference with the corresponding reference.

stop at constant temperature and that this reinforcement sustains when the temperature is recovered on reheating. The insert of Fig. 4 shows the difference between the FS and reference curves for ZFC, TRM, and FC, the excess magnetization gained due to the FS in TRM + FC corresponds to the magnetization “lost” in the ZFC. The extended validity of the principal of superposition is further supported by this apparent agreement.

Looking at the bump and dip in the TRM and ZFC magnetization curves of Fig. 4 and the derived excess magnetization in the inset, one observes that the influence of the stop at constant temperature during cooling is limited to a restricted temperature range around  $T_s$ . The width of this region may be assigned to the existence of an overlap between the spin configuration attained at  $T_s$  and the corresponding state at a very neighboring temperature ( $T+\Delta T$ ). The two concepts that explain the width are then a chaotic nature of the spin glass equilibrium configuration and an overlap on short length scales between the equilibrium configurations at  $T$  and  $T+\Delta T$ . These concepts have been elaborated earlier.<sup>4,5</sup> Further information on these matters can be obtained also by extending the current dc method to include two stops when cooling, first at  $T_s$  and then at  $T'_s < T_s$ . The results of this cooling protocol emphasize the chaotic nature of the equilibrium states at the two temperatures: the equilibration at  $T_s$  has no influence on the nonequilibrium processes occurring only a few kelvins below, outside the region of overlap.

The actual spin configuration of a spin glass is controlled by its thermal history. In this paper we have shown that it is possible to extract substantial information on complex non-equilibrium phenomena linked to the evolving spin configuration by simply recording the temperature dependence of the remanent magnetization after employing certain cooling protocols. Introducing an excess magnetization in the spin glass by intermittently applying or removing a magnetic field during a stop at constant temperature it is possible to study memory phenomena and the influence of aging on the relaxation of the magnetization. The magnitude of the excess magnetization is governed by the duration of the field cycle and the wait time at constant temperature before the magnetic field is changed. The logarithmic nature of the relaxation and the aging processes can be explored by systematically altering the duration of the field cycle and/or the wait time. An extended validity of the principal of superposition to apply to frozen in relaxation processes that are thermally activated on heating is disclosed by combining results from TRM, ZFC, and FC experiments.

A note on the experimental procedure: the relaxation of spin glasses extends to eternal times and has a logarithmic character and the relaxation processes are thermally activated. These properties require that the cooling or heating rates in the experiments are controlled and kept the same in all different measurements to achieve comparable experimental data. The heating (and cooling) rate determines, due to the overlap between equilibrium states at different temperatures, an effective age of the system and defines an effective observation time (of order 10 s in our experiments) in the measurement of the remanent magnetization.

### III. CONCLUSION

The present results from the here introduced dc method have been obtained on an archetypical three-dimensional spin-glass. Corresponding remanent magnetization studies can advantageously be performed even using standard SQUID magnetometers. For example, the slow dynamics of the less conventional  $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$  glassy system has been successfully investigated in this way,<sup>12</sup> exposing characteristic memory and superposition features.

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