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Time dependence of the magnetization and ESR in *Ag*Mn spin-glass

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We report measurements for *Ag*Mn (1.1 and 4 at.%) of the zero-field-cooled (ZFC) and field-cooled (FC) magnetization (M), ESR linewidth (ΔH), and shift of the resonance field (δH) as a function of time and temperature. We found that M, ΔH, and δH present a similar time evolution for the FC and ZFC states.

In the last few years the existence of spin-glass (SG) regimes in dilute solutions of transition ions in metallic matrices has resulted in many experimental and theoretical investigations.[1,2] Measurements of the time dependence of the magnetic properties are necessary for a better understanding of the nature of the magnetic ordering occurring in SG systems.[3] These data may provide some further insight into whether or not the temperature of the cusp observed in the susceptibility marks the presence of a true phase transition.

Electron-spin resonance (ESR) has shown itself to be a convenient technique to study SG.[4] However, to the best of our knowledge, no attempt has been made to study the evolution with time of the ESR data. The purpose of this report is to present measurements on the time dependence of the ESR linewidth (ΔH) and the shift of the resonance field $\delta H = H_0 - H_R$ (where H_0 is the field corresponding to a gyromagnetic factor $g = 2$ and H_R the resonance field), for zero-field-cooled (ZFC) and field-cooled (FC) samples of *Ag*Mn (1.1 and 4 at.%) and compare these results with the time-dependent ZFC magnetization (M_{ZFC}) and the remanent magnetization (M_R) data on the same samples.

The samples were prepared by repeated arc melting in an Ar atmosphere. Compositions were obtained from the weight of the elements. Samples were prepared as cylinders and thin foils. After the desired shape was obtained, the criterion used to assure homogeneity was the following. The samples were annealed at different sets of temperature (T) and time (t) until we obtained a minimum in the T difference between the maxima of the M_{ZFC} and FC magnetization (M_{FC}) curves when measured in fields of ~10 Oe. The above condition is achieved for the *Ag*Mn samples studied after annealing at 800 °C for about 48 h followed by a rapid quench into ice water. All measurements reported here are for samples so treated. Chamberlain *et al.* introduced a reversibility temperature defined as the T above which no remanence is observed.[5] We obtain such a temperature as follows. The sample is FC in a field of ~10 Oe to T well below T_f, which is the temperature where the maximum of M_{FC} vs T occurs. Then the field is turned off and the remanent magnetization monitored while the sample is slowly warmed. The magnetization, of course, is time varying both because of the characteristic $\ln t$ decay and the increasing temperature. However, one finds a T where M suddenly totally decays over a few seconds. We call this T, above which there is no remanence, T_R, and find that it is within 2% of T_f.

In Fig. 1 we present magnetization (M) curves for a sample of *Ag*Mn (1.1 at.%) obtained with an rf-SQUID magnetometer. The M_{ZFC} was obtained by ZFC to *each T*, then applying a 9.6-Oe field, and measuring M as a function of time. We note that there is a measurable shift of the maximum of the $M_{\mathrm{ZFC}}(t)$ towards lower temperatures at larger times, a feature which was common for all the samples. If we identify a frequency ν with each period of measurement, we find that the relative shift in T_f per decade of frequency, $\Delta T_f / T_f \ln \nu$ is $\lesssim 10^{-2}$ in agreement with results obtained from ac susceptibility measurements.[6,7]

Our ESR data are in agreement with those of previous workers insofar as they overlap.[8,9] In addition to studying the time dependence of ΔH and δH, we also measured the dc magnetization of the sample, *in situ*, utilizing the technique of observing the ESR of two *P*-doped Si spin ESR markers glued to the sample, as described in Refs. 4 and 10. All the ESR measurements were actually done in fields $\simeq$3 kOe. We have made measurements for three different time sequences.

(1) FC decay measurements. The sample is cooled to T from a temperature $> T_R$ in a field of 3 kOe at which point M, δH, and ΔH are measured. The field is then turned off for a time t_1, at which point, in order to measure the three quantities of interest (ΔH, δH, and M), the field is turned to that needed for

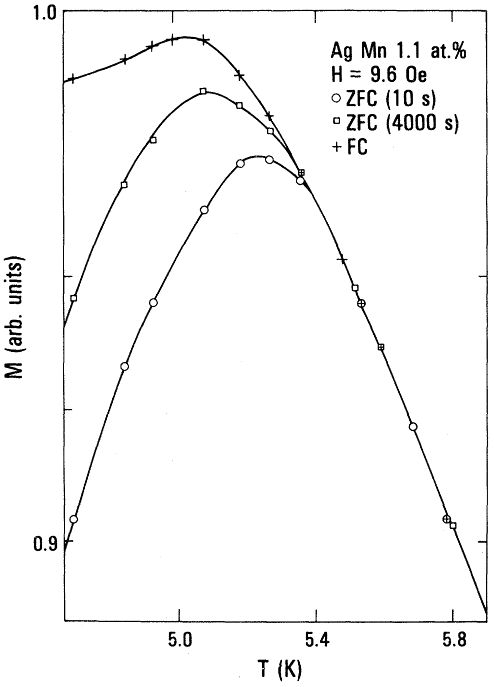

FIG. 1. ZFC and FC dc magnetization data for *Ag*Mn (1.1 at.%) as a function of T near T_f in an $H = 9.6$ Oe field. The ZFC data were obtained by cooling to each T and measuring as a function of time. ZFC data obtained at 10 and 4000 s are shown by ○ and □, respectively.

resonance, and the measurements taken after 25 s. (This time is always small compared to t_1, and represents the shortest time in which we could take a resonance spectrum.)

(2) Recovery after FC decay. Following a FC as described in (1), the field is off for a very long time, $t_1 = 10^4$ s; the field is then turned on and the recovery followed continuously from 25 to 10^4 s.

(3) ZFC measurements. The sample is cooled to T from a temperature $> T_R$ in zero field (< 5 Oe). The field appropriate to resonance is then turned on and the data taken continuously from 25 to 10^4 s.

Preliminary data for the time dependence of M have been reported earlier.[11]

In Fig. 2 we present time-dependent measurements for a cylinder of *Ag*Mn (4 at.%) of M, ΔH, and δH for sequences (1) and (3) described above. The FC decay data were taken at $t_1 = 250$ s. The ZFC data were taken at times of 25 and 250 s. These data are representative of more extensive measurements from which we find that the time dependence of the FC and ZFC data can be well approximated by $A(t) = A(t_0) \pm S \ln t/t_0$, where A represents M, ΔH, or δH. We define a dimensionless slope S/Δ of the logarithmic decay. S was obtained from the change of each quantity between 25 and 250 s, and Δ is the difference between the value measured after FC at 3 kOe and the value obtained at the ZFC 25-s point. In Fig. 3, S/Δ is presented as a function of T/T_R for M, ΔH, and δH. We found similar values of S/Δ for ΔH and δH for a foil sample rolled from the cylinder and subsequently annealed.

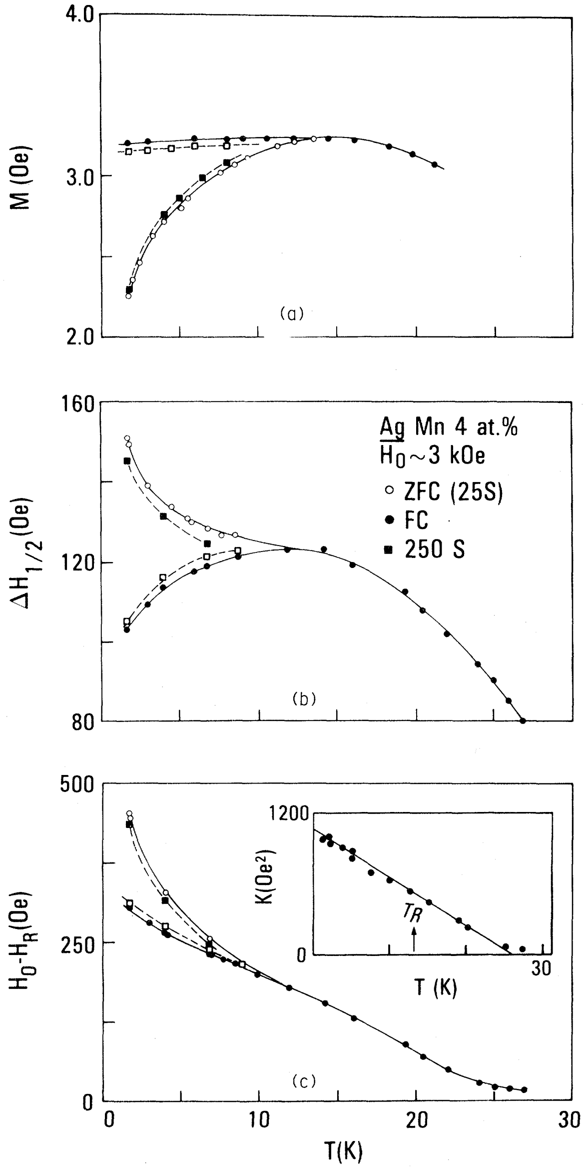

FIG. 2. ZFC and FC data for a *Ag*Mn (4 at.%) cylinder as a function of temperature. (a) the magnetization M, (b) the half-linewidth ($\Delta H^{1/2}$), and (c) the shift of the resonance field from $g = 2$ given by $\delta H = H_0 - H_R$. ○ and ■ were obtained by ZFC to each T and measuring after H_R was applied for 25 and 250 s, respectively. ● were obtained after FC at 3 kOe, and for □ the field was turned off for 250 s from the FC condition, and subsequently H_R was reapplied for 25 s. The insert gives the anisotropy energy K as a function of T (see text).

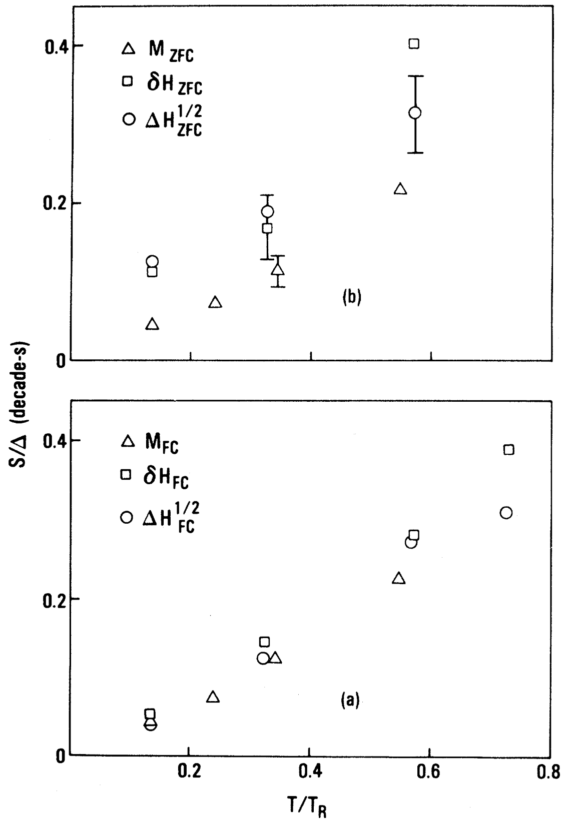

FIG. 3. Variation of the normalized slope S/Δ for (a) FC and (b) ZFC data with a reduced temperature T/T_R for the magnetization M (Δ), shift of the resonance field $\delta H = H_0 - H_R$ (□), and half-linewidth $\Delta H^{1/2}$ (○).

As seen in Fig. 3 the values of S/Δ are nearly the same for the FC decay case for all three quantities measured, and increases as T approaches T_R. Although there are clearly some differences contrasted to the ZFC data, qualitatively they behave very similarly.

When we made measurements on the recovery following a very long decay [i.e., procedure (2)] we found that the magnitude of S/Δ was within a few percent of that of the FC decay. Thus we infer that the characteristic time dependences associated with both a dynamical measurement (ESR) and quasistatic (M) properties are similar.

The shift δH can be interpreted as resulting from a magnetic anisotropy $K = \delta H\, M_0$, where M_0 is the magnetization measured in a field $H_0 = \omega/\gamma$.[4] The anisotropy energy as a function of T/T_R is given in the insert of Fig. 2. Similar values of K were found for both FC and ZFC states. However, it is not clear why H_R starts to shift at T well above T_R. A similar behavior has been observed for CuMn.[4]

For $T < T_R$ ΔH and δH present a similar time and temperature dependence, with the FC values smaller than the ZFC ones. Currently there is no explanation for the ESR linewidth for $T < T_R$. However, it seems reasonable that ΔH is mainly due to an inhomogeneous broadening, being the envelope of a multitude of spin packets to which we can associate a distribution of H_R. Since at each temperature δH is larger for the ZFC state than that for the FC state, it is likely that there is a wider distribution of H_R for the ZFC state which is in agreement with the larger ΔH observed. This would also explain why ΔH and δH follow a similar time evolution.

In summary, we found that the three quantities present a comparable time evolution for both the FC and ZFC states, but we have only a phenomenological explanation for these data.

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