

## Noncritical behavior and remanent magnetization in magnetically frustrated FeSbO<sub>4</sub>

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Low-field dc-magnetization and thermoremanent-magnetization measurements have been recorded from the rutile-related iron antimonate of composition FeSbO<sub>4</sub>. The zero-field-cooled susceptibility curve at low magnetic fields ( $H = 20$  Oe) shows a small peak at  $\sim 72$  K and a much broader peak at lower temperatures of  $\sim 25$  K. The temperature dependence of the remanent magnetization below  $\sim 72$  K indicates the existence of two different relaxation regimes which depend on the strength of the applied field in which the sample was cooled. The thermoremanent-magnetization measurements as a function of time show aging effects and the characteristic parameters are quite similar to those found in canonical spin glasses. The temperature dependence of the first nonlinear coefficient  $b_3$  in the expansion of the magnetization in odd powers of  $(a_1 H)$  shows no divergence around the peak at 72 K, which is indicative of the absence of a true spin-glass transition. The moderate degree of frustration associated with an inhibited antiferromagnetic transition at  $\sim 72$  K, is nevertheless insufficient to produce a true spin-glass transition where the magnetic correlation length is even much shorter.

### I. INTRODUCTION

Insulating spin glasses and dilute magnetic systems have attracted considerable interest as model systems for the examination of random magnetic phenomena.<sup>1,2</sup> In this respect it is pertinent to note that recent works have shown that simple mixed metal oxides with rutile-related structures,<sup>3,4</sup> and some compounds with similar structures in which there is short-range atomic order,<sup>5,6</sup> offer attractive possibilities for the elucidation of the processes of magnetic freezing and of fundamental phenomena such as percolation or random-field distribution. Our recent studies of rutile-related iron antimonate by neutron diffraction<sup>7</sup> have shown that a temperature-independent finite spin correlation of the order of 30 Å in length exists in the (001) plane at temperatures below  $\sim 70$  K. The observation has been associated with the development of a moderate-frustrated antiferromagnetic ordering below  $\sim 70$  K.

In this paper we report new low-field magnetic susceptibility and remanent-magnetization measurements, which we have recorded from iron antimonate and which demonstrate that its magnetic behavior below  $\sim 70$  K is different from that expected in canonical spin glasses, in spite of the fact that some relaxation features are quite similar to those observed in spin-glass systems. However, the static magnetization properties define a general picture close to that found in spin-glass-like systems such as ZnCr<sub>2-x</sub>Ga<sub>x</sub>O<sub>4</sub> spinel<sup>8</sup> or Pb<sub>1-x</sub>Mn<sub>x</sub>Te semiconductor.<sup>9</sup> From these results we conclude that the anomalies in the magnetic behavior below  $\sim 70$  K are not associated with

a true phase transition (spin glass or antiferromagnetic) and we propose an explanation based on a freezing process which involves uncompensated cluster formation of antiferromagnetic ordering. This nonequilibrium state may be achieved through a mechanism of dynamic inhibition of magnetic correlations in disordered systems which involves a fast divergence of the characteristic times.<sup>10</sup>

### II. EXPERIMENT

Iron antimonate of formula FeSbO<sub>4</sub> was prepared by the calcination of the precipitate as previously described.<sup>11</sup> The dc magnetic susceptibility and remanent magnetization measurements were carried out with a SQUID magnetometer equipped with second-order gradiometer pickup coils at temperatures between 150 and 4.2 K in applied magnetic fields between 40 KOe and 20 Oe. The accuracy of the magnetization measurements is  $\sim 10^{-7}$  emu and the temperature oscillations during long-time periods were less than 0.01 K for  $T < 100$  K. The magnetic field was generated using a superconducting coil and it was calibrated with a lead sample of high purity.

Thermoremanent-magnetization data ( $M_{TR}$ ) as a function of temperature were collected while temperature was increasing at a constant rate of 0.2 K per min after switching off the applied magnetic field in which the sample had been previously field cooled (fc) from room temperature.

The time dependence of the thermoremanence was

measured by the following procedure: After the SQUID detector is reset and the baseline voltage  $V_1$  is measured, the sample is moved from one of the two positions which correspond to a minimum value of the gradiometer signal to the center of the pickup coil system (maximum gradiometer signal); then the SQUID voltage  $V_2$  is measured again and the sample is pulled to the second position of minimum gradiometer signal while the SQUID voltage  $V_3$  is measured. The magnetization of the sample is proportional to  $(V_2 - V_1) - (V_3 - V_1)/2$ . This method corrects, as a first approximation, the spurious drift of the SQUID baseline during the time that the sample is moving. Long-time shifts are also avoided because the total magnetization is determined at each point. As the elapsed time from the beginning of the measurement increases, up to eight measurements are averaged to improve accuracy of the points.

The susceptibility measurements which were used for critical analysis were performed after cooling the sample from room temperature to 4.2 K in a constant magnetic field (FC process). Then the temperature was increased while magnetization data were recorded in a regular manner.

### III. RESULTS AND DISCUSSION

#### A. ZFC-FC susceptibility

The zero-field-cooled (ZFC) and field-cooled magnetic susceptibility data recorded in fields of 500, 100, 50, and 20 Oe and in the temperature range 150–4.2 K are shown in Fig. 1. The temperature dependence of the zero-field-cooled susceptibility data below  $\sim 80$  K showed similar features over the whole range of investigated magnetic fields: Two peaks which were superposed to a practically constant background were observed. The peak at the lower temperature may be considered as a broad maximum centered at  $\sim 25$  K with an intensity that increases with the magnetic field strength. In contrast, the peak at  $\sim 72$  K was found to be very sensitive to the strength of the magnetic field and was not observed in relatively high fields (1 KOe).<sup>5</sup> No significant shifts in the temperature at which the maxima of both peaks occur were identified over the studied range of fields used in these investigations. The results therefore contrast with the  $H^{2/3}$  dependence of the freezing temperature predicted for Ising spin glasses and cluster models<sup>12</sup> or the  $H^2$  law<sup>13</sup> expected in the vicinity of  $H=0$ .

The temperature at which the FC and ZFC susceptibility curves branch was strongly dependent on the magnitude of the applied magnetic field and for  $H < 500$  Oe was higher than 72 K. As the magnetic field was decreased, the point of branching shifted to higher temperatures and even for  $H=20$  Oe the splitting between the FC and ZFC curves is so large that it appears at temperatures well above those at which magnetic hyperfine broadening is observed in Mössbauer spectra.<sup>14</sup> It is also notable that the branching point for low fields is in good agreement with the temperature at which the spin-correlation length, as determined by neutron diffraction, starts a noticeable increase.<sup>7</sup> The existence of irreversibilities above

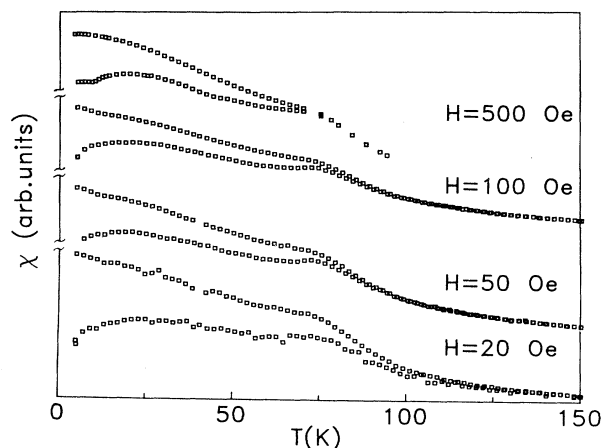


FIG. 1. ZFC and FC susceptibilities  $M/H$  as a function of temperature at low magnetic fields.

the freezing temperature is a common phenomenon and has previously been identified in other spin-glass-like systems such as some diluted spinels,<sup>8</sup> semiconductors,<sup>9,15</sup> or metallic glasses<sup>16</sup> and may be interpreted as a precursor effect associated with cluster size distribution and, consequently, with the relaxation time constants.

#### B. Thermoremanent magnetization as a function of temperature

The thermal dependence of the remanent magnetization recorded for different cooling fields is shown in Fig. 2. Three different regimes of thermal variation were observed depending on the magnitude of the cooling field with the corresponding crossover regions between them.

The first regime is identified at low temperatures where the remanent magnetization decreases according to an expression of the form  $M_{TR}(T) = M_{TR}(0)T^{-\delta}$ , where the exponent  $\delta$  is close to 0.5 and practically field independent. An example of this power-law relaxation behavior,

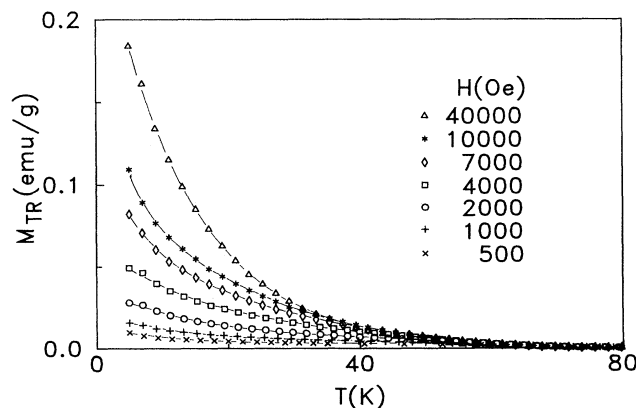


FIG. 2. Variation of the remanent magnetization ( $M_{TR}$ ) with temperature collected after cooling the sample in different magnetic fields between 500 and 40 000 Oe.

recorded after a field-cooled process at  $H=200$  Oe, is shown in Fig. 3. It is important to note that the range of temperatures at which this power-law decay is accomplished is drastically reduced by increasing the magnitude of the cooling field and it is only observed in relatively small fields,  $H < 2000$  Oe. These results contrast with the linear temperature variation observed at low fields in spin-glass systems<sup>15,17</sup> and are indicative of a slower relaxation mechanism.

The second regime is well described by the canonical exponential temperature dependence for the saturated remanent magnetization of the type

$$M_{\text{TR}}(T) = A \exp(-\beta T).$$

This has been previously predicted by theoretical treatments<sup>18</sup> and experimentally verified at high magnetic fields in a great number of spin-glass and spin-glass-like systems.<sup>19,20</sup> A semilogarithmic plot of the remanent-magnetization data collected after a FC process at  $H=40\,000$  Oe is shown in Fig. 4. It is evident from Fig. 4 that only exponential decay occurs if the cooling field is as large as  $H=40\,000$  Oe, even at low temperatures. The inset of Fig. 4 shows that the exponential coefficient  $\beta$  increases with the magnetic field and tends slowly to a saturation value corresponding to magnetic fields higher than  $40\,000$  Oe. The values of the  $\beta$  coefficient when normalized to the freezing temperature of  $\sim 72$  K are comparable with those observed in spin-glass-like systems in which a blocking process of magnetic clusters is involved.<sup>21</sup> The  $A$  parameter is also field dependent, showing a field variation well fitted by the empirical law

$$A(H) = H / (57\,900 \text{ Oe} + 2.18H) \text{ emu/g}.$$

It is also notable that, between a temperature varying from  $\sim 53$  to  $\sim 45$  K, when the field increases from 100 to  $40\,000$  Oe, and the freezing temperature at  $\sim 72$  K, a crossover region separating two different exponential relaxation behaviors occurs, in which the range of temperature is very large compared with that observed in other

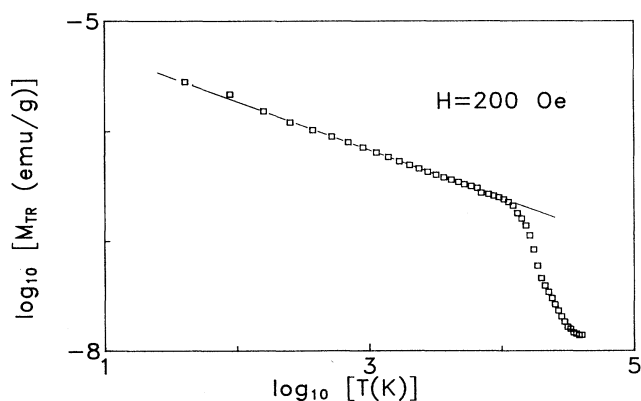


FIG. 3. Plot of the logarithm of the remanent magnetization ( $M_{\text{TR}}$ ) corresponding to a cooling field of  $H=200$  Oe as a function of the logarithm of the temperature. The straight line indicates the regime in which a potential decay is accomplished.

spin-glass-like systems.<sup>8</sup> The remanent magnetization persists above the freezing temperature at  $\sim 72$  K following another exponential behavior with a lower  $\beta$  coefficient. Considering that the region of temperatures in which these persistent relaxation phenomena are present corresponds to the observation of irreversibility features in ZFC-FC curves and with the rapid decrease of the antiferromagnetic correlation length,<sup>7</sup> these high-temperature relaxation phenomena may be interpreted in terms of the blocking of some remaining clusters during the cluster disintegration process as the temperature increases.

From the thermoremanent-magnetization data, we have deduced the field dependence of the remanent magnetization at a constant temperature. Some curves corresponding to selected temperatures below  $T_f$  are displayed in Fig. 5. For temperatures lower than  $\sim 20$  K,  $M_{\text{TR}}(H)$  increases progressively with the strength of the cooling field without achieving the saturation value. This is probably related to some kind of dynamic crossover occurring at temperatures ( $\sim 25$  K) similar to that at which a broad peak in the ZFC susceptibility curves was observed. Above  $\sim 20$  K, a maximum in  $M_{\text{TR}}(H)$  is observed at a value of the field which decreases when the temperature increases and, in higher cooling fields,  $M_{\text{TR}}(H)$  becomes practically field independent. The general shape of these curves is similar to those reported for canonical spin glasses.<sup>1,2</sup> It is remarkable that, for the range of temperatures and cooling fields in which a linear dependence of  $M_{\text{TR}}(H)$  versus field is accomplished, the thermal variation of the remanent magnetization follows the previously reported power-law decay.

### C. Thermoremanent magnetization as a function of time: Aging effects

Thermoremanent-magnetization data as a function of time were collected after the following procedure: The

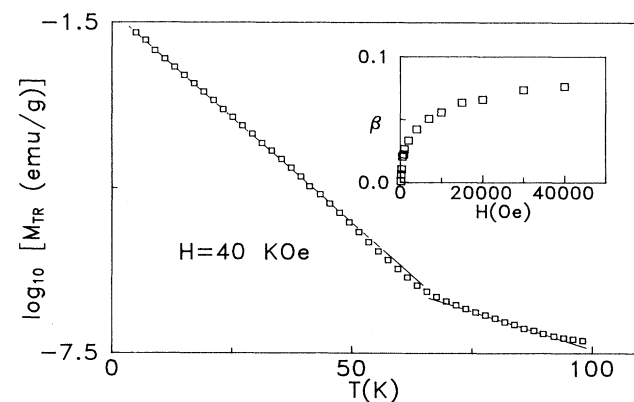


FIG. 4. Plot of the logarithm of the remanent magnetization ( $M_{\text{TR}}$ ) vs temperature, corresponding to a cooling field of  $H=40$  KOe. The straight lines indicate the two regimes of exponential decay. The field dependence of the coefficient  $\beta$  of the low-temperature exponential relaxation regime is displayed in the inset.

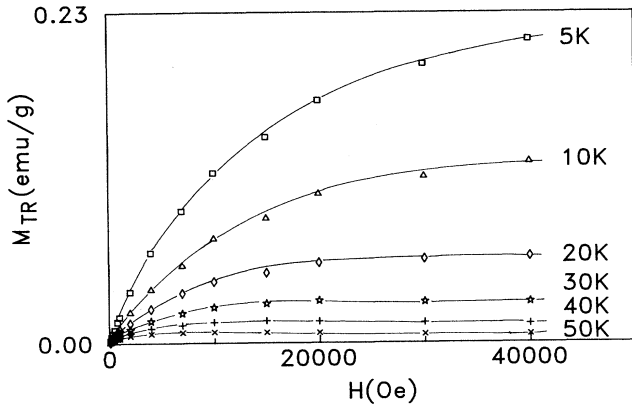


FIG. 5. Field dependence of the remanent magnetization ( $M_{TR}$ ) at selected temperatures between 5 and 50 K.

sample was field cooled from room temperature to a given temperature below  $T_f$  trying to minimize the elapsed time; it was then kept at a constant temperature and field during the waiting time  $t_w$ , and finally the field was switched off and the magnetization data were recorded at a constant rate of 4 s for each measurement. For long-time measurements, several points were averaged to minimize experimental noise.

The measurements were performed at two values of the magnetic field (236.4 and 23.6 Oe) and at temperatures and waiting times ranging from 25 to 70 K and 17 min to 15 h, respectively. All the experimental curves (see Figs. 6 and 7) show aging effects, which are correlated with the time that the sample was kept at a constant temperature and field before the relaxation measurements started. The main feature related with this fact is the existence of an inflection point roughly located at  $t_w$  in a log-log plot of the  $M_{TR}(t)$  data, as has been first reported by Lundgren *et al.*<sup>22</sup> for spin-glass systems. Aging is a com-

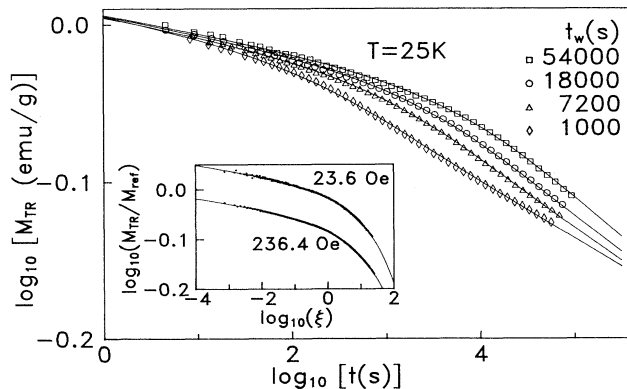


FIG. 6. Logarithm of the thermoremanent magnetization vs the logarithm of the observation time (in s) for different waiting times  $t_w$  at  $T=25$  K. The points are the experimental values and the solid lines are calculated from Eq. (2) using the values of the fitting parameters. The inset shows the master curves for two values of the magnetic field. Solid lines represent the fit to Eq. (2).  $M_{ref}$  is an arbitrary reference value.

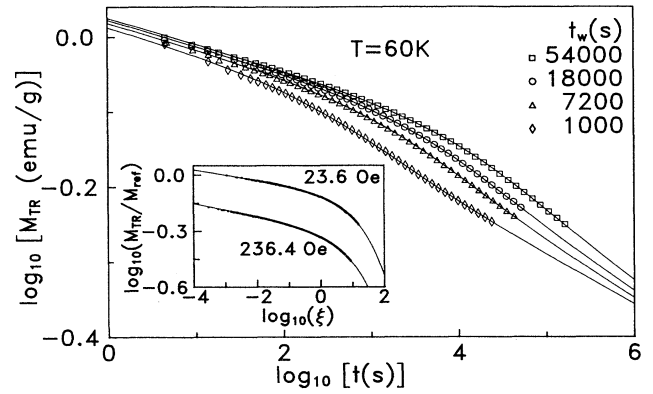


FIG. 7. The same as Fig. 6 for  $T=60$  K.

mon property of the glassy state that has been first observed in small strain creep curves for polymeric glasses and amorphous compounds and more recently has also been found in other systems with some kind of structural disorder such as high- $T_c$  superconductors<sup>23</sup> or charge-density-wave systems.<sup>24</sup>

Recent theoretical approaches to this problem due to Koper and Hilhorst<sup>25</sup> and Hoffmann and Sibani<sup>26</sup> only give a qualitative explanation of the experimental data. To get the best fits of the experimental curves and, consequently, quantitative estimations of the relaxation parameters, the phenomenological theory developed by Struik,<sup>27</sup> and used to explain creep curves in glassy polymers, is best applied. This theory was adapted by Ocio *et al.*<sup>28</sup> to account for aging phenomena in spin glass systems. In this model, thermoremanence relaxation data are plotted as a function of a reduced time defined by

$$\xi = \frac{t_0^{\mu-1}}{1-\mu} [(t+t_w)^{1-\mu} - t_w^{1-\mu}], \quad (1)$$

where the exponent  $\mu$  is a parameter that must be chosen properly to get a good superposition of all the data corresponding to the same temperature and different waiting times  $t_w$  in a unique master curve which can be interpreted as the relaxation function of the system at a given constant age. The parameter  $t_0$  is an arbitrary reference time ( $t_0=1$  s) introduced to build  $\xi$  as a dimensionless variable. The master curves can be well fitted by the empirical relation

$$M_{TR}(\xi) = M_0 \xi^{-\alpha} \exp[-(\xi/\tau_p)^{1-n}], \quad (2)$$

where the stretched exponential accounts for aging effects and dominates relaxation at times of the order of  $t_w$  while the decreasing power law is the equilibrium relaxation of the system which is the major contribution for  $t \gg t_w$  and at short times  $t \ll t_w$ , where the stretched exponential does not vary too much.

In the insets of Figs. 6 and 7, the logarithm of the master curves corresponding to the temperatures 25 and 60 K and magnetic fields  $H=23.6$  and 236.4 Oe are plotted as a function of the logarithm of the reduced time  $\xi$ . The values of  $\mu$  that give a good superposition of the experi-

mental data at a given temperature and different waiting times are  $\mu=0.80\pm 0.03$  for  $T=25$  K and  $\mu=0.88\pm 0.03$  for  $T=60$  K. No significant changes in the values of the  $\mu$  parameter are observed for the two studied magnetic fields showing that aging effects are not affected in this range of field variation. This fact indicates that linear response regime is accomplished even at fields as large as  $\sim 200$  Oe, in contrast with the very low-field ( $\sim 1$  Oe) linear limit found in spin-glass systems.<sup>22,28</sup> The relative large range of the linear regime is also evident from the thermoremanence-magnetization data as a function of the magnetic field, as has already been discussed.

The master curves have been fitted to the empirical relation (2) and the values obtained for the parameters characterizing relaxation are listed in Table I. As in the case of the parameter  $\mu$ , no significant variation in the values of  $\alpha$ ,  $\tau_p$ , and  $n$  is observed for the two values investigated for the strength of the cooling magnetic field. The exponent  $\alpha$  of the power-law decay increases when approaching  $T_f$  and, hence, the time decay of the remanent magnetization is accelerated. On the other hand, the characteristic time  $\tau_p$  decreases when the temperature rises, shifting to shorter times the region where the stretched exponential has a significant contribution. The values of the parameter  $n$  for the two temperatures investigated are roughly constant and close to 0.5, which is in good agreement with the results found in the thio spinel  $\text{CdIn}_{0.3}\text{Cr}_{1.7}\text{S}_4$  (Ref. 29) and other spin-glass systems.<sup>30,31</sup>

#### D. Nonlinear susceptibility

A series of FC susceptibility measurements  $M(T)/H$  as a function of temperature recorded under magnetic fields in the range 20–1000 Oe, has enabled us to establish the isothermal magnetization curves. Some of the experimental FC susceptibility curves covering the range of fields investigated are shown in Fig. 8. One of the deduced isothermal magnetization curves at 74 K is shown in the inset of Fig. 8. As is evident from a consideration of Fig. 8, the contribution to the total susceptibility of the nonlinear terms becomes important around the freezing temperature at  $\sim 72$  K. More direct evidence for the nonlinear effects can be obtained from the deviation from the linear behavior as the field increases in the isothermal magnetization curves, as shown in the inset of Fig. 8.

To elucidate whether the increase of the nonlinear effects around the freezing temperature at  $\sim 72$  K is associated with a spin-glass transition, we have carried out a quantitative study of this contribution from the usual development of the magnetization above  $T_f$  in terms of odd

TABLE I. Values of the relaxation parameters  $\alpha$ ,  $\tau_p$ , and  $n$  obtained by fitting the master curves to Eq. (2).

$T$ (K)	$H$ (Oe)	$\alpha$	$\tau_p$	$n$
25	23.6	$0.011\pm 0.002$	$800\pm 50$	$0.54\pm 0.02$
25	236.4	$0.011\pm 0.002$	$910\pm 50$	$0.57\pm 0.02$
60	23.6	$0.027\pm 0.004$	$120\pm 20$	$0.48\pm 0.02$
60	236.4	$0.033\pm 0.004$	$90\pm 20$	$0.49\pm 0.02$

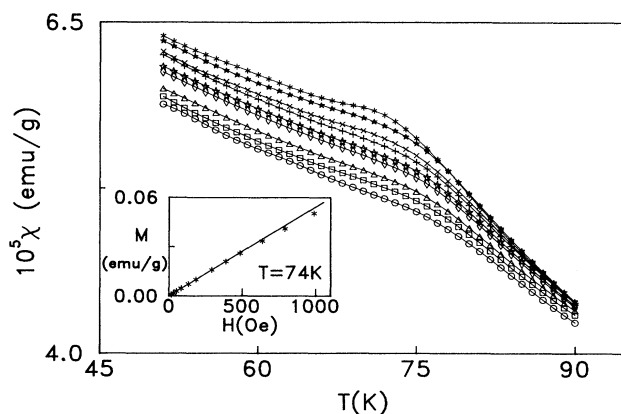


FIG. 8. Thermal dependence of the FC susceptibility  $M/H$  for cooling fields of: (○) 20 Oe, (□) 50 Oe, (△) 100 Oe, (◇) 200 Oe, (☆) 300 Oe, (+) 500 Oe, (×) 650 Oe, (★) 800 Oe, and (∗) 1000 Oe. The solid lines are only aids to the eye. The variation of the isothermal magnetization at 74 K vs magnetic field up to 1000 Oe is shown in the inset. The straight line indicates the deviation from linear behavior.

powers of the field.<sup>32</sup> This can be written in the form

$$M = a_1(T)H - a_3(T)H^3 + a_5(T)H^5 - \dots, \quad (3)$$

where the  $a_1(T)$  coefficient is the linear susceptibility corresponding to the initial slope of the magnetization curves while the nonlinear part is well described by the remaining coefficients  $a_3$ ,  $a_5$ , etc. The least-squares fit to Eq. (3) performed for each of the isothermal magnetization curves enables the evaluation of  $a_1(T)$  and  $a_3(T)$ . It is notable that, at temperatures close to  $T_f$ , though only the first two coefficients of the development have been determined in the present work, up to five coefficients were necessary to reproduce the experimental behavior. It is also remarkable that the preceding expansion (3) also holds (disregarding the problem of the equilibrium magnetization) below the freezing temperature of  $\sim 72$  K, in contrast to canonical spin-glass systems where development in odd and even powers of the field is necessary.

The nonlinear component of the magnetization [ $a_1(T)H - M$ ] is shown in Fig. 9, as a function of  $H$ , in the temperature range 90–65 K, and shows that the nonlinear component has a maximum contribution to the total magnetization at  $\sim 72$  K. The thermal dependence of the reciprocal linear susceptibility [ $a_1(T)^{-1}$ ] plotted in the inset of Fig. 9 follows a Curie-Weiss law above  $\sim 72$  K, with a Curie constant  $C=2.95 \times 10^{-3}$  emu K/g and a Curie temperature  $\Theta=20\pm 5$  K. This value of the Curie temperature is probably due to the existence of weak ferromagnetic interactions among the frozen uncompensated antiferromagnetic clusters.

The extraction of the nonlinear part of the susceptibility responsible for the singular behavior in a spin-glass transition is more adequately achieved by expanding the magnetization in odd powers of  $(a_1H)$ , as has been suggested by Omari *et al.*<sup>32</sup> The relation between the coefficients of this last expansion [ $b_n(T)$ ] and those used in Eq. (3) [ $a_n(T)$ ] is of the form

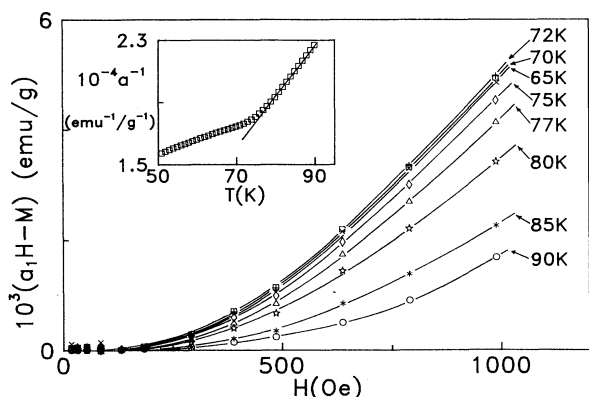


FIG. 9. Nonlinear part of the magnetization ( $a_1H - M$ ) as a function of the field for different temperatures around 72 K. The inset shows the reciprocal linear susceptibility  $[a_1(T)]^{-1}$ , as determined by fitting Eq. (3) to the experimental data. The straight line shows the region in which the Curie-Weiss law is obeyed.

$$b_n(T) = \frac{a_n(T)}{[a_1(T)]^n} \quad (4)$$

The thermal dependence of the first nonlinear coefficient  $b_3$  around the freezing temperature is shown in Fig. 10. The  $b_3$  coefficient varies by less than 1 order of magnitude when the temperature is lowered from 90 to 72 K, showing an intermediate behavior between the temperature-independent coefficient  $b_3$  in superparamagnetic systems<sup>33</sup> and the typical variation of 3 orders of magnitude, which is taken as a proof of a true spin-glass transition in systems such as Cu-Mn.<sup>32</sup> Hence, we conclude that the freezing temperature at  $\sim 72$  K cannot be associated with the transition to a canonical spin-glass phase and the observed nonlinear susceptibility contribution may be interpreted as a consequence of an inhibited antiferromagnetic transition.

The values of the first two coefficients  $a_1(T)$  and  $a_3(T)$  allow the estimation of the order of magnitude of the

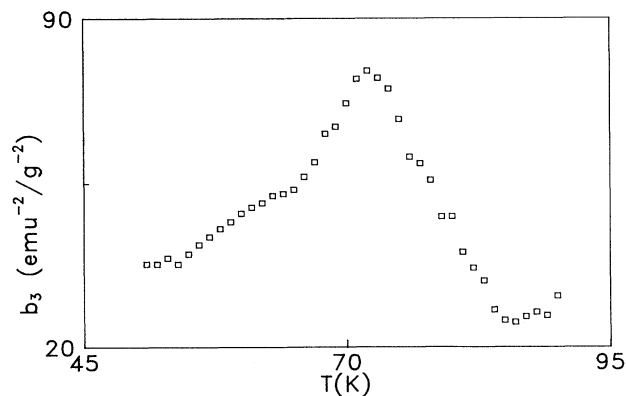


FIG. 10. Plot of the first nonlinear coefficient  $b_3$  of the expansion of the magnetization in odd powers of  $(a_1H)$  vs temperature.

maximum value of the magnetic field where the linear response is accomplished, which is found to be

$$[a_1(T)/a_3(T)]^{1/2} \approx 10^3 - 10^4 \text{ Oe}.$$

This confirms the interpretation given to the relaxation measurements.

A simple model involving rigid antiferromagnetic clusters of imperfect ordering can be used to give a comparative estimation of the first nonlinear coefficient in the expansion of the magnetization [Eq. (3)]. It may be assumed that, above  $T_f$ , the magnetic behavior of the system is dominated by rigid uncompensated antiferromagnetic clusters with an average magnetic moment given by

$$M = [(3k_B m P C)/N_A]^{1/2} = 2.39m^{1/2}\mu_B,$$

where  $k_B$  is the Boltzmann constant,  $m$  is the mean number of spins in such a cluster,  $P$  is the molecular weight,  $C$  is the Curie constant deduced from the Curie-Weiss law followed by the susceptibility above  $T_f$ , and  $N_A$  is Avogadro's number.

The mean size of the clusters can be estimated from the saturated thermoremanent magnetization extrapolated to 0 K, assuming that, at low temperatures, the total magnetic moment of the cluster remains blocked in the orientation of the local anisotropy direction when the field is switched off. This hypothesis leads to the expression

$$m = 0.5N_A M / PM_{\text{TR}}(T=0),$$

where the factor 0.5 is the average value of the cosines of the angles between the total magnetic moment of each cluster and the external magnetic field over one half-space, and  $M_{\text{TR}}(T=0)$  is the extrapolated saturation value of the thermoremanent magnetization at 0 K. In the case considered here,  $M_{\text{TR}}(T=0)$  has been extrapolated from the thermal dependence of the remanence obtained after cooling in a field of 40 KOe (see Fig. 4) leading to the value  $M_{\text{TR}}(T=0) = 0.27 \text{ emu/g}$ . It is clear that this estimation is only a lower limit of the real value because, at low temperatures, a saturation of the thermoremanence cannot be achieved even at the highest cooling field of 40 KOe which was investigated during this work. The values obtained for  $m$  and  $M$  by this procedure are  $m = 10^4$ ,  $M = 240\mu_B$ . If we assume that, in the paramagnetic region, the total magnetic moments of the clusters behave as classical vectors whose spatial distribution is governed by the Langevin function, the coefficient of the first nonlinear part of the susceptibility  $a_3$  can be written in the following form:

$$a_3(T) = \frac{1}{15}a_1(T) \left[ \frac{M}{k_B(T - \Theta)} \right]^2 \quad (5)$$

At the lowest temperature at which the Curie-Weiss law is accomplished ( $\sim 77$  K), the value of  $a_3$  deduced from Eq. (5) is  $3.7 \times 10^{-13} \text{ emu g}^{-1} \text{ Oe}^{-2}$ , which is only 1 order of magnitude less than the experimental value at the same temperature. This disagreement indicates that the freezing phenomena responsible for the anomalies observed at  $\sim 72$  K are more complex than the simple noninteracting cluster model considered before, but, in any case, if a

spin-glass transition might occur at  $\sim 72$  K, the nonlinear susceptibility divergence could not be masked by the cluster contribution.

#### IV. CONCLUSIONS

From dc magnetic measurements we have shown that the magnetic behavior of iron antimonate presents some features which differ from those which are characteristic of random diluted antiferromagnetic systems and those of canonical spin glasses. The origin of these differences may be associated with the moderate magnetic frustration arising from imperfect antisite Fe-Sb cationic order as determined by electron beam diffraction.<sup>34</sup> Due to such a cationic order, the nearest-neighbor interactions may become of comparable strength to the next-neighbor interactions preventing the development of long-range antiferromagnetic correlations along the magnetic lattice. Moreover, a recent EPR study of this system and the diluted compounds  $\text{Fe}_{1-x}\text{Ga}_x\text{SbO}_4$  (Ref. 34) suggests that the above-mentioned Fe/Sb atomic ordering leads to a magnetic behavior characteristic of a two-dimensional system. Of course, this weakening of the magnetic exchange interactions along the  $c$  axis would further reduce the magnetic correlations along this direction, thus reducing the chance to achieve a three-dimensional long-range antiferromagnetic ordering.

The critical analysis of the nonlinear part of the susceptibility in iron antimonate reveals that the magnetic frustration associated with antisite cationic order is not sufficient to produce a true spin-glass transition. On the other hand, magnetic frustration is responsible for the inhibited antiferromagnetic transition observed at  $\sim 72$  K by low-field dc susceptibility measurements and neutron diffraction. The absence of a true antiferromagnetic transition may be related to a dynamic inhibition of the magnetic correlation which has been associated with thermally activated relaxation processes which produce a fast divergence of the characteristic relaxation times preventing the development of infinite-range correlations along the system within the finite experimental time scale. In this respect, it is then clear that  $\text{FeSbO}_4$  is a simple model

system of a new category of random magnets.

In the absence of any further theoretical model for these systems, an explanation of the general magnetic behavior of iron antimonate can be attempted, given in terms of uncompensated antiferromagnetic cluster formation with a net magnetic moment. As the temperature decreases, the short-range antiferromagnetic ordering evolves towards a collective blocking at  $\sim 72$  K. The second peak at  $\sim 25$  K may be associated with some kind of dynamic crossover due to residual interactions among rigid clusters, which do not modify the mean antiferromagnetic correlation length. Moreover, recent neutron-diffraction measurements<sup>7</sup> have shown that only the (100) reflection is present in the low-temperature patterns, which is consistent with the development of a highly anisotropic spin structure (quasibidimensional antiferromagnetism). Hence, the peak at  $\sim 72$  K may then be associated with the development of the two-dimensional antiferromagnetic correlations within the (001) plane, whilst the broad peak at  $\sim 25$  K may be related to the freezing of the magnetic moments in the third direction.

The irreversibility effects and the relaxation processes observed above the freezing temperature confirm the presence of remaining uncompensated clusters even at temperatures higher than  $T_f$  and reflect the existence of a broad distribution of cluster relaxation times. The thermal relaxation of the remanent magnetization in the exponential regime (high fields) seems to be consistent with a thermally activated blocking process following the Arrhenius law. It is noticeable that, at low cooling fields, thermoremanence decays following a power law instead of the theoretically predicted linear behavior.

The existence of aging effects in the time relaxation of the thermoremanence demonstrates that this phenomenon is probably a general feature of a great family of systems with some kind of magnetic frustration and not only of the spin-glass systems. Moreover, the values of the parameters which characterize relaxation are similar to those observed in canonical spin glasses indicating that the mechanism that produces aging is not necessarily associated with the spin-glass state.

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