Quenching of ferrimagneticlike ordering in $SrCr_8Fe_4O_{19}$ hexagonal ferrite

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We report here measurements of high field ($H \leqslant 150$ kOe) isothermal magnetization, dc and ac susceptibility, and Mössbauer spectroscopy of the exchange frustrated insulator $SrCr_8Fe_4O_{19}$. The isothermal magnetization and Mössbauer spectroscopy results are consistent with a ferrimagneticlike transition at $T_C \approx 250$ K. Low field dc and ac susceptibility measurements reveal a reentrant spin-glass transition at low temperature but they fail to show any anomaly at the nominal Curie temperature T_C. This unusual feature is interpreted as indicative of a transition where long range ferrimagnetic ordering has been replaced by a correlated spin-glass phase with finite spin correlation length.

INTRODUCTION

Crystalline hexagonal ferrites $SrFe_{12-x}Cr_xO_{19}$ ($x \leqslant 8$) are multisublattice ferrimagnetic compounds evolving from a collinear magnetic ordering ($x = 0$) to some kind of noncollinear disordered magnetic structure, as it has been proved from magnetization and Mössbauer spectroscopy measurements.[1,2] The fundamental feature needed to understand the magnetic behavior of this series of insulating compounds is the underlying topological magnetic frustration associated with the Cr ions distributed within the three octahedral sites of the magnetoplumbite structure.[1,3] For instance, we have recently shown that in an isomorphous compound containing only Cr ions, $SrCr_8Ga_4O_{19}$, no magnetic ordering exists down to 4.2 K in spite of the existence of very strong antiferromagnetic interactions.[4] Actually, the topological magnetic frustration of these magnetoplumbite systems is very similar to that existing in octahedral site spinels, such as $ZnCr_{2x}Ga_{2-2x}O_4$, where a complex antiferromagnetic–spin-glass behavior has been detected.[5–7]

In this work we report a magnetic investigation of the end member of the series, $SrCr_8Fe_4O_{19}$, focusing mainly on the ferrimagneticlike ordering observed at $T_C \approx 250$ K. We conclude from high field magnetization, low field dc and ac susceptibilities and zero field Mössbauer spectroscopy measurements that the transition observed at $T_C \approx 250$ K is to a correlated spin-glass state with vanishing small spontaneous magnetization. The existence of this intermediate state among spin-glass and true long-range magnetic ordering has been experimentally characterized[8] and clearly detected from measurements of the spin correlation length by means of small angle neutron scattering.[9]

EXPERIMENT

The synthesis procedure and the basic structural characterization of the $SrCr_8Fe_4O_{19}$ oxide has been previously reported together with an investigation of the cation distribution by means of neutron diffraction and Mössbauer spectroscopy.[1] In summary, it has been shown that the octahedral sites of the magnetoplumbite structure contain about 10% of Fe ions while the tetrahedral and five-fold sublattices are fully occupied by Fe ions.

Isothermal high field ($H \leqslant 150$ kOe) magnetization measurements were carried out in the $4.2\ K \leqslant T \leqslant 300$ K temperature range at the Service National des Champs Intenses, Grenoble. In the same temperature range we performed low field dc ($H = 8$ kOe) and ac magnetic susceptibility measurements. The dc magnetization was measured by means of an extraction magnetometer and ac susceptibility was measured with a mutual inductance bridge in a rms field of 10 Oe and a frequency of 110 Hz. Finally, zero field ^{57}Fe Mössbauer spectra were recorded in a conventional constant acceleration spectrometer in the $78\ K \leqslant T \leqslant 300$ K temperature range.

RESULTS AND DISCUSSION

In Fig. 1 we display the temperature dependence of several isothermal magnetization curves where it may be ob-

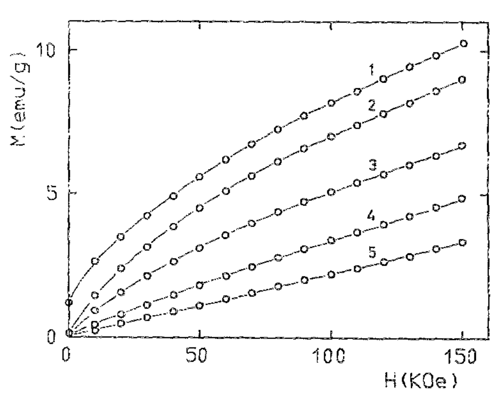

FIG. 1. Isothermal magnetization curves of $SrCr_8Fe_4O_{19}$. (1) $T = 4.2$ K, (2) $T = 68$ K, (3) $T = 151$ K, (4) $T = 220$ K, (5) $T = 297$ K.

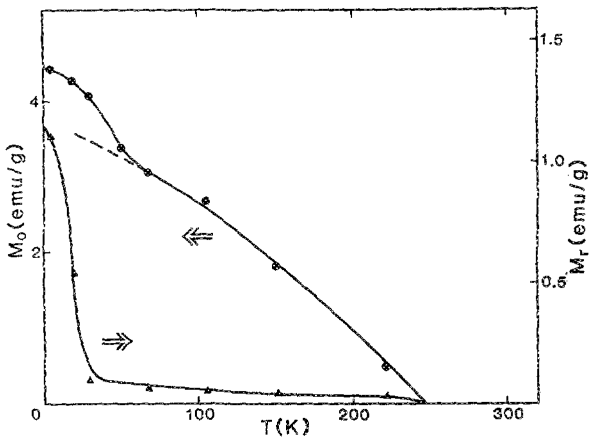

FIG. 2. Temperature dependence of spontaneous magnetization M_0 and magnetic remanence M_r.

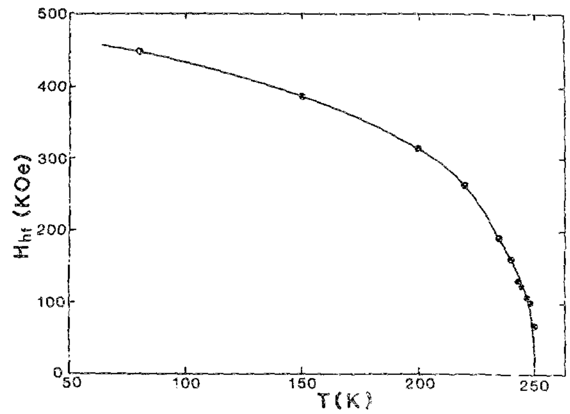

FIG. 4. Temperature dependence of the ^{57}Fe hyperfine field as measured by Mössbauer spectroscopy.

served that the room-temperature curve has a linear M-H relationship, indicative of pure paramagnetic behavior, while in the remaining curves the magnetization is very far from saturation, having a very important differential susceptibility even at the maximum field of 150 kOe. This behavior is consistent with the previous conclusion of a noncollinear spin ordering in the $SrFe_{12-x}Cr_xO_{19}$ series, as indicated by the composition dependence of the 0 K saturation magnetization.[1] In Fig. 2 we plot the temperature dependence of the spontaneous magnetization M_0 while the high field differential susceptibility X, corresponding to the $M(H) = M_0 + XH$ law followed in the high field region is represented in the inset of Fig. 3. In the Fig. 2 we plot as well the temperature dependence of the magnetic remanence M_r measured after the application of the maximum magnetic field of 150 kOe.

From the results reported in Fig. 2 we infer that a ferrimagneticlike ordering is established at $T_C \approx 250$ K with some kind of anomaly or transition occurring at a lower temperature. It is noteworthy that the setting of the low-temperature anomaly in M_0 goes in parallel to a step increase of M_r. We remark as well that the spontaneous magnetization at $T = 0$ K corresponds to about $M_0 \approx 0.9\mu_B$/f.u. which is much smaller than the value expected from a collinear ferrimagnetic ordering ($M_0 \approx 10.8\mu_B$/f.u.), thus indicating a very strongly perturbed ferrimagnetic spin structure.

The existence of a well-defined transition at $T_C \approx 250$ K has been deduced as well from the temperature dependence of the ^{57}Fe hyperfine field measured by means of Mössbauer spectroscopy. A complete report of our Mössbauer investigation will be published elsewhere; however, we would like to look here at the temperature dependence of the mean hyperfine field (Fig. 4) which has a behavior typical of magnetically ordered compounds with a Curie temperature in agreement with the isothermal magnetization study.

All the magnetic characteristics of $SrCr_8Fe_4O_{19}$ reported up to now seem to be consistent with a ferrimagneticlike ordering at $T_C \approx 250$ K and indicate that probably a spin-glass reentrance occurs at a lower temperature. The existence of a spin-glass reentrance at low temperature has been indeed verified from low field susceptibility measurements. As it may be observed in Fig. 3 and 5 the dc susceptibility and the in-phase component of the ac-susceptibility X' display

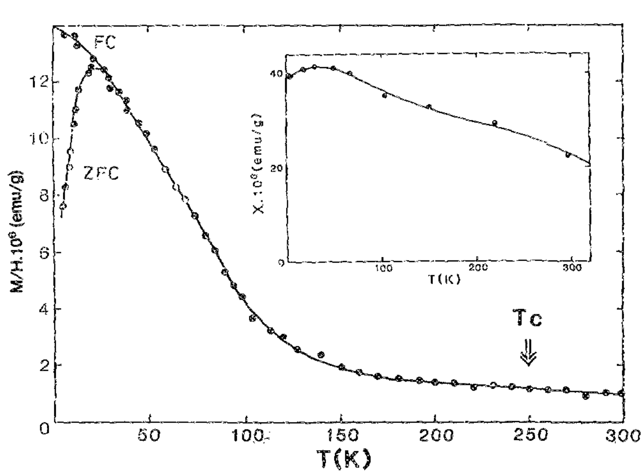

FIG. 3. dc magnetization measured at constant field ($H = 8$ kOe) in field cooled (FC) and zero field cooled (ZFC) processes. Inset: Temperature dependence of the high field differential susceptibility measured in the isothermal magnetization curves.

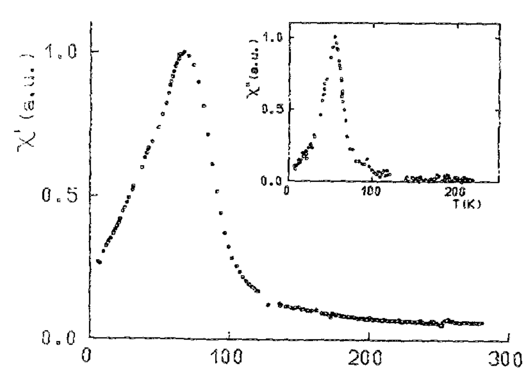

FIG. 5. Temperature dependence of the in-phase ac-susceptibility X'. Inset: Temperature dependence of the out of phase ac-susceptibility X''.

the typical spin-glass cusp at 23 and 68 K, respectively. In the same way, the out of phase component of the ac-susceptibility X'' show a peak at about 55 K, a temperature slightly lower than the cusp in X', as typically occurs in reentrant spin glasses.[9] Furthermore, the dc susceptibility presents the characteristic spin-glass irreversibility among zero field cooled (ZFC) and field cooled (FC) processes.[10] It is clear then that the low-temperature anomaly observed in the spontaneous magnetization corresponds to a phenomenon of spin-glass reentrance.

A full study of the low-temperature spin-glass reentrance in $SrCr_8Fe_4O_{19}$ will be reported elsewhere but we want to remark here that a very strong magnetic field dependence of the dc susceptibility peak temperature has been observed and the $T_g = 68$ K peak temperature of X' corresponds to the zero field limit of the dc susceptibility peaks. This explains then the different peak temperatures observed in Figs. 3 and 5.

However, what is strongly surprising in these low field susceptibility curves is the fact that any anomaly is actually observed at the nominal Curie temperature T_C. This striking anomalous result may only be understood if we assume that the transition observed at T_C is not to a true long-range ferrimagnetic ordering which should display a divergence in the susceptibility at T_C but only a transition to some kind of correlated spin-glass phase have a finite correlation length. In this way if the spin correlation length remains finite and the developed magnetic clusters have vanishing small magnetic moments the divergence of the susceptibility will drop out. Actually, a tendency to this behavior is often observed in reentrant spin glasses where it is found that in the ferromagneticlike transition the ac susceptibilty remains much smaller than that expected from the demagnetizing field limit.[8]

A quenching of the long-range ferromagnetic order has been conclusively observed by Rhyne, in exchange frustrated amorphous systems such as $Fe_{91}Zr_9$, by means of small-angle neutron scattering[9,11] clearly showing that the spin correlation length remains finite and as low as 27 Å.

Characteristic features of this correlated spin-glass phase should be an important spin relaxation, thus leading to a very small magnetic remanence, and a strong polarizibility with external magnetic fields which enhances the apparent ferrimagnetic order. Within this context the low-temperature spin-glass reentrance should mark a further freezing of the magnetic moments below T_g. It remains, however, to be elucidated if the spin freezing occurring at T_g is consistent with a slowing down of the transverse spin components, as predicted by Gabay and Toulouse[12] and experimentally verified by Mössbauer spectroscopy in several systems[13,14] or corresponds mostly to a small clusters blocking phenomena.[15] Actually, our interpretation of the magnetic properties of the exchange frustrated $SrCr_8Fe_4O_{19}$ system in terms of a quenched ferrimagnetic ordering points to the validity of the second model for this system. Further neutron scattering and Mössbauer spectroscopy studies are in progress to clarify this open question.

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