Magnetic dilution in the strongly frustrated *kagome* antiferromagnet $SrGa_{12-x}Cr_xO_{19}$

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A study of the structural and magnetic characteristics of the strongly frustrated *kagome* system $SrGa_{12-x}Cr_xO_{19}$ ($1 < X < 9$) is reported with focus on the effects of the magnetic ion dilution. High-resolution powder neutron diffraction shows no evidence of long-range magnetic ordering down to 1.5 K and a nearly random substitution of Cr by Ga atoms within the octahedral sites. An intrinsic quasibidimensional structure integrated by *kagome* blocks is inferred. ac and dc magnetic susceptibility measurements down to 1.6 K make evident the existence of a freezing phenomenon similar to that of spin glasses with the freezing temperature T_f, decreasing as the degree of dilution of the Cr^{3+} ions increases. The extrapolated value of T_f for the fully occupied lattice is 4.15 K, indicating that the nondiluted oxide could be considered as a prototype of geometrically frustrated magnetic systems. The magnetic frustration parameter $\Theta/T_f \approx 135 \pm 5$ is independent of the degree of dilution, giving further support to the idea of an intrinsic structural origin of the frustration in this system. Both magnetic susceptibility measurements and ESR studies show a Curie-Weiss-like behavior in the high-temperature range with magnetic correlations starting around 100 K.

I. INTRODUCTION

The concept of magnetic frustration has long been attracting the attention of physicists. Frustration exists in a wide variety of magnetic systems, and may lead to different phenomena such as, for example, the spin-glass syndrome,[1] or the appearance of noncollinear states.[2]

The existence of frustration effects in antiferromagnetic systems was first reported by Anderson[3] in his studies of spinel lattices. Frustrated antiferromagnets are very interesting from the point of view of their unusual low-temperature magnetic ground state, that may show disorder[4] or a kind of pseudo-order that is not of the Néel type.[5] The discovery by Obradors *et al.*[6] of the magnetoplumbite-like insulator compound $SrGa_4Cr_8O_{19}$ with an extreme degree of magnetic frustration ($\theta/T_f \geq 117$), has generated a considerable amount of experimental and theoretical analysis that tries to clarify the properties of this singular compound,[7–9] and to analyze the features of geometrically frustrated systems.[10–12]

The special magnetic properties of this compound arise from antiferromagnetic coupled Cr^{3+} ions, with $S = \frac{3}{2}$, that are arranged in a geometrically frustrated *kagome* lattice.[6] The crystal structure of $SrGa_4Cr_8O_{19}$ may be represented by a five-layer *kagome* lattice ($4f_{VI}$-$12k$-$2a$-$12k$-$4f_{VI}$ stacked layers) joined together in a three-dimensional structure through the $4f_{VI}$-$4f_{VI}$ magnetic interactions, which are not geometrically frustrated (see Fig. 1). A full description of the magnetoplumbite crystal structure (space group $P6_3/mmc$) may be found in Ref. 13. In spite of the very high Curie-Weiss temperature of the $SrGa_4Cr_8O_{19}$ compound, $\theta \approx -500$ K, Obradors *et al.* did not find any evidence of long-range magnetic ordering down to $T = 4.2$ K. Later on, Ramirez, Espinosa and Cooper[7] extended the susceptibility measurements to lower temperatures, discovering a transition from paramagnetic to spin-glass-like ordering at $T = 3.3$ K and also an increase of the ordering temperature when the magnetic sites were further diluted with Ga^{3+} ions. It was suggested by Obradors *et al.*[6] that the

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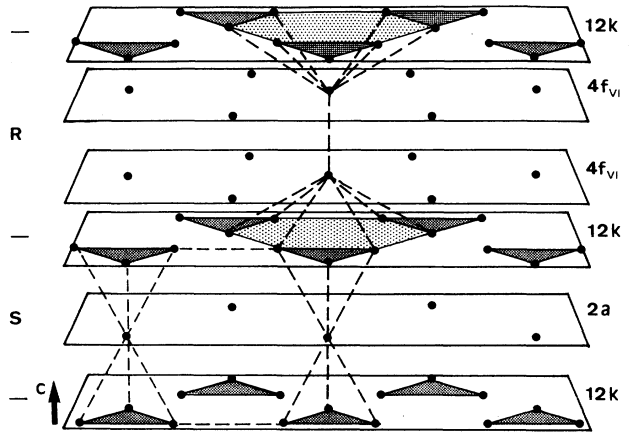

FIG. 1. Schematic representation of the lattice of octahedral sites of the magnetoplumbite-type structure. Two sets of *kagome* lattices may be distinguished: one perpendicular to the *c* axis formed by 12*k* sites, and another one parallel to the *c* axis. The latter is also present in the spinel-type structure with the cubic ⟨111⟩ direction viewed along the hexagonal *c* axis.

origin of the enhanced magnetic frustration in this *kagome*-lattice compound may arise from an important nonmagnetic dilution in the $4f_{VI}$ sublattice leading to a quasi-two-dimensional model compound. This low dimensional magnetic behavior, together with the noncomittant quantum fluctuation effects, was also suggested later by Broholm *et al.*[8,9] in an inelastic neutron-scattering investigation of a polycrystalline sample. Further anomalous characteristics of the $SrGa_{12-x}Cr_xO_{19}$ system were reported by Ramirez and co-workers.[7,11] It was shown that the low-temperature specific heat is proportional to T^2, instead of the usual linear T dependence of spin glasses. This anomalous behavior suggested that new types of spin excitations[10,11] exist in this *kagome* lattice. Finally, it was claimed that an increase of the effective dimensionality of the magnetic sublattice, due to a preferential occupancy of the 12*k* sites by the Ga ions, might be the origin of the observed increase of the spin-glass freezing temperature with dilution.[7]

With the aim to check these effects we have performed a complete structural and magnetic study of the $SrGa_{12-x}Cr_xO_{19}$ $(1<x<9)$ system by using neutron powder diffraction, ac and dc magnetic susceptibility measurements, and electron spin resonance (ESR).

In this work we show that the dilution of magnetic ions leads indeed to a decrease of the freezing temperature, as usually occurs in all the magnetic systems, but the effective magnetic frustration is not decreased by nonmagnetic dilution. From the composition dependence of the freezing temperature, we infer the transition temperature of the hypothetical compound with full Cr occupation ($SrGa_3Cr_9O_{19}$). Some preliminary results concerning the present work have been reported recently.[14]

II. EXPERIMENTAL

Samples of $SrGa_{12-x}Cr_xO_{19}$ with the nominal composition (x) between 2 and 9 were prepared by the ceramic method, using mixtures of $SrCO_3+zCr_2O_3+(6-z)Ga_2O_3$ with $z=x/2$ (pure Merck $SrCO_3$, 99.999% Cr_2O_3, and 99.99+% Ga_2O_3 from Aldrich, were used). The mixtures were thoroughly homogenized in agate mortars, then placed in platinum crucibles and heated at 1573 K for one week. During this period, the samples were further homogenized and the reaction products were checked by conventional x-ray-diffraction procedures. After the process, the sample with nominal composition $x_n=9$ could not be obtained in pure form. This indicates that actually a full occupancy of the octahedral sites by Cr ions cannot be reached. In consequence, an impurity identified as Cr_2O_3 sesquioxide was observed, with a concentration of 10.12(8) wt % (calculated from the refined scale factors of the sesquioxide and the magnetoplumbite-like phases). The composition of the magnetoplumbite-like phase was found to be $x=8.78(4)$, as calculated from the refined site occupation factors.

Neutron powder-diffraction patterns were collected in the high-resolution D2B diffractometer at the Institute Max von Laue–Paul Langevin (ILL), Grenoble. The spectra were recorded using a wavelength of 1.5945 Å, in the angular range $0°\leq 2\theta\leq 150°$, using a step scan mode with $\Delta(2\theta)=0.05°$, at different temperatures in the interval $1.5\leq T\leq 298$ K. The diffraction profiles were refined with the STRAP program package.[15]

dc magnetic measurements were performed by using a Quantum Design Superconducting Quantum Interference Device (SQUID) magnetometer in the temperature range from 1.6 K to room temperature. ac magnetic susceptibility was measured in the temperature range from 2–300 K in a Lakeshore susceptometer. Temperatures lower than 4.2 K were reached by pumping over the He bath. A Varian spectrometer operating at X band (9 GHz) was used to perform ESR measurements between 15 and 500 K.

III. RESULTS

A. Neutron-diffraction measurements

Because of the similarity of the Ga and Cr atomic numbers, x-ray diffraction cannot be used for the determination of the cation distribution. However, the values of their neutron-scattering lengths, $b(Ga)=0.7288\times10^{-12}$ cm and $b(Cr)=0.3635\times10^{-12}$ cm, are different enough to allow this study using neutron-diffraction data. It is important to note that no magnetic contributions to Bragg reflections were detected at any temperature, indicating that no long-range magnetic ordering develops in $SrGa_{12-x}Cr_xO_{19}$ $(2<x<9)$ between 1.5 K and room temperature, in agreement with previous investigations.[6,8,9] The calculated site-occupation factors (SOF's) did not display any variation with temperature, within their standard deviations.

The Cr^{3+} ions site-occupation factors are displayed in Fig. 2. The most important feature of this figure is that Cr^{3+} cations occupy the three different octahedral sites, namely, 2*a*, 12*k*,and $4f_{VI}$, with an almost random distribution, the 2*a* sites being occupied by Cr^{3+} cations with a slight preference.

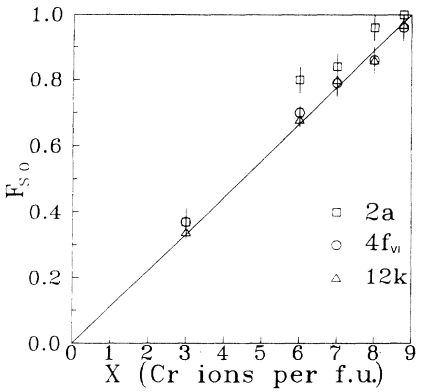

FIG. 2. Plot of the Cr^{3+} site-occupation factors (F_{SO}) vs the Cr content per formula unit, with their $2s$ error bars.

As stated in previous works,[6,7,9] the strong magnetic frustration observed in $SrGa_4Cr_8O_{19}$ arises from the particular arrangement of the octahedral sites. Henceforth, since the magnetic cation Cr^{3+} only enter in the octahedral sites, we will refer to these sites generically as magnetic sublattice. The magnetic sublattice may be regarded as a sequence of c-stacked two-dimensional (2d) *kagome* layers ($12k$ sites) laying on the R-S interphases of the magnetoplumbite-type structure. Two consecutive $12k$ layers are connected either through the face sharing octahedra ($4f_{VI}$ sites) in the R block, or through the octahedral $2a$ sites in the spinel-like S block. This c-axis stacking is that characterizing a *kagome* plane and thus we should look at the crystal structure as a five-layer *kagome* lattice connected through a nonfrustrated bond ($4f_{VI}$-$4f_{VI}$). Actually the crystal lattice is composed of twice this five-layer stacking because of the 6_3 axis which rotates 180° the R and S blocks. It is straightforward to note that each cation located in a $4f_{VI}$ site has, as nearest neighbors, six cations located in the $12k$ adjacent layers, but only one in the next $4f_{VI}$ layer. Such arrangement of the octahedral sites in the R block leads to a sandglass-like configuration which could promote an intrinsic quasi-2D character of the crystal structure. It is straightforward to note that this sandglass configuration will be specially effective in destroying the superexchange path along the c axis when Cr ions are substituted by nonmagnetic Ga ions in the $4f_{VI}$ sites. In fact, as we will show later, even with a random cationic substitution the 2D behavior of this system is enhanced because of the sandglass configuration of the lattice.

In order to provide a quantitative measure of the variation of the magnetic dimensionality with dilution, we have calculated the probability of formation of superexchange paths along the c axis, either by crossing the R block X_R, ($12k$-$4f_{VI}$-$4f_{VI}$-$12k$), or the S block X_S, ($12k$-$2a$-$12k$). In this way, the composition dependence of X_R and X_S gives a measure of the influence of dilution on the probability to find a path for magnetic interactions between two consecutive $12k$ layers. For these calculations, Eqs. (1) and (2) have been used:

$$X_R = p_{4f_{VI}}^2 \left[\sum_{\xi=1}^{6} \begin{bmatrix} 6 \\ 6-\xi \end{bmatrix} (1-p_{12k})^{6-\xi} p_{12k}^{\xi} \right]^2 , \quad (1)$$

$$X_S = p_{2a} \left[\sum_{\xi=1}^{3} \begin{bmatrix} 3 \\ 3-\xi \end{bmatrix} (1-p_{12k})^{3-\xi} p_{12k}^{\xi} \right]^2 , \quad (2)$$

where the p's represent the Cr^{3+} SOF's in the corresponding sites; ξ is the number of Cr^{3+} cations located in $12k$ sites which are first neighbors of a $4f_{VI}$ site, and similarly, ζ is the number of Cr^{3+} cations located in $12k$ sites which are first neighbors of a $2a$ site. In summary, both expressions represent the probability of finding uninterrupted "chromium superexchange paths" between two interfaces through the R and S blocks, respectively. In Fig. 3 the X_R and X_S probabilities calculated with the experimental SOF's are displayed. The solid line indicates the calculated probabilities assuming a random Cr^{3+} distribution within the magnetic sublattice. This figure clearly indicates that $X_S > X_R$ even for the $x=8$ compound, i.e., the magnetic interactions are preferentially destroyed at the R block. Actually this effect is slightly enhanced by the small preference of Cr^{3+} for $2a$ sites in the S block. It is straightforward to note, however, that the difference X_R-X_S remains rather constant all over the composition range, and thus we should expect that the quasibidimensional behavior generated by this nonmagnetic dilution remains constant. As we will see later on, this is indeed what is actually observed through the composition dependence of the frustration parameter θ/T_f.

B. dc and ac susceptibility

In Fig. 4 we show the temperature dependence of the reciprocal susceptibility for the two extreme members of the solid solution $SrGa_{12-x}Cr_xO_{19}$ ($x=2$ and $x=8$). It is evident from the figure that the high-temperature susceptibility follows a Curie-Weiss law $\chi = C/T - \theta$, the

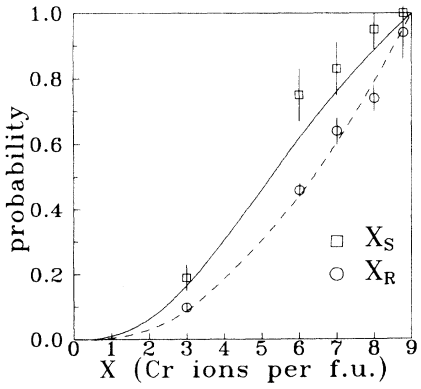

FIG. 3. Dependence on the degree of dilution of the probability to find a path for magnetic interactions between Cr^{3+} ions located in consecutive $12k$ layers, across the $4f_{VI}$ layers (X_R), or across the $2a$ layer (X_S). $2s$ error bars are indicated.

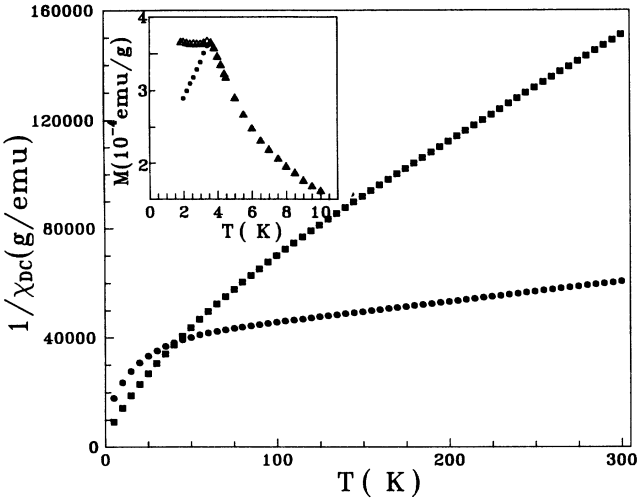

FIG. 4. Reciprocal susceptibility showing the Curie-Weiss behavior in the high-temperature range, and the appearance of magnetic correlations below 100 K approximately. (■$x=2$, ●$x=8$). Inset: Magnetization in the zero-field-cooled (●) and field-cooled (△) processes for sample with $x=8$ showing the onset of irreversibility.

same conclusion is reached when looking at all the other samples. The values obtained for the Curie temperature θ and the effective magnetic moment are summarized in Table I. The effective magnetic moment varies from 3.5 μ_B for $x=2$ to 3.9 μ_B for $x=8$, a typical value corresponding to spin only configuration for Cr^{3+}. The values of the extrapolated Curie temperature range from -90–-537 K making evident the existence of strong antiferromagnetic interactions in all the members of the family.

A low-temperature upturn of the susceptibility is found below temperatures from 70 K for $x=8$ to 120 K for $x=2$. But no signals of long-range antiferromagnetic ordering or spin-glass ordering were observed down to 4.2 K, in conflict with the results in single crystals previously reported by Ramírez, Espinosa, and Cooper.[7] If θ, the expected ordering temperature in absence of frustration, is so high and no signal of long-range ordering is found down to 4.2 K, it is evident that a very high degree of magnetic frustration exists in these compounds as was pointed out by Obradors *et al.*[6] The study of the low-temperature ($1.5 \leq T \leq 4.2$) region was undertaken looking for the spin-glass transition reported by Ramírez, Espinosa, and Cooper.[7]

TABLE I. Values of the effective magnetic moment, extrapolated Curie-Weiss temperature, freezing temperature, frustration parameter, and the extrapolated ESR linewidth for infinite temperature as a function of the Cr^{3+} concentration.

x	μ (μ_B/f.u.)	θ (K)	T_f (K)	θ/T_f	ΔH_{pp}^{∞} (G)
2	3.5(1)	90(5)			465
3	3.5(1)	145(5)			375
4	3.5(1)	177(5)			550
5	3.5(1)	237(5)	1.80	132	730
6	3.6(1)	322(5)	2.65	122	500
7	3.8(1)	446(5)	3.10	144	640
8	3.9(1)	537(5)	3.90	138	800
8.78			4.05		

We measured several samples of the family following the well-known zero-field-cooled–field-cooled (ZFC-FC) processes at very low fields (≈ 10 Oe). In the inset of Fig. 4 we show the obtained results in the case of the sample with $x=8$, while the results for all the series are summarized in Table I. The typical curve with strong irreversibility between the ZFC and FC branches is observed, with the onset of irreversibility starting at the cusp of the peak as in canonical spin glasses,[1,16] and in disagreement with some data of Ref. 7. Furthermore, the freezing temperature clearly decreases as the dilution of Cr^{3+} ions increases as one should expect, and in strong contrast with the results obtained by Ramírez, Espinosa, and Cooper.[7] These differences may be due to the different preparation techniques and thermal treatment of the samples.

The influence of frustration on the cooperative transition may be estimated by means of the ratio θ/T_f. In our case we observe that his ratio remains almost constant, $\theta/T_f \approx 135 \pm 5$, and independent of x, within the experimental error, for samples with $x \geq 5$, i.e., the sample with the lowest T_f accessible with our equipment. These results suggest at least in the range of compositions measured, that the observed giant magnetic frustration is intrinsic to the crystal structure of nondiluted $SrGa_3Cr_9O_{19}$.

Actually, this independence of magnetic frustration on the degree of dilution is in concordance with the result obtained from neutron diffraction that shows that the differences X_R-X_S, which somehow reflects the influence of dilution on the dimensionality of the magnetic lattice, does not depend on the degree of dilution either.

The ac magnetic susceptibility measurements were done in zero dc field, the ac field was 5 Oe and the ac frequency was 111 Hz. The high-temperature regime shows a Curie-Weiss-like behavior and the values of θ and μ_{eff} are equivalent to those obtained from dc susceptibility. In the low-temperature range both the real and imaginary parts of the susceptibility show a peak that indicates the existence of the spin-glass freezing. In Fig. 5 we show the behavior of the low-temperature real part of the ac susceptibility for the samples with $x=6$, 7, 8, and 8.78, ($H_{ac}=5$ Oe, $\nu=111$ Hz) that are those with transition temperatures accessible with our ac susceptometer. As in the case of dc susceptibility, we observe that the temperature of the peak decreases as the degree of dilution of the Cr^{3+} ions increases (Fig. 6).

C. ESR Measurements

Several samples with Cr concentration between $x=2$ and $x=8$ were measured, and we found that they exhibit a single broad line, with a gyromagnetic factor $g=1.98(2)$, associated to the resonance of Cr^{3+} ions. No temperature dependence of the g factor was observed in the temperature range $150 < T < 400$ K.

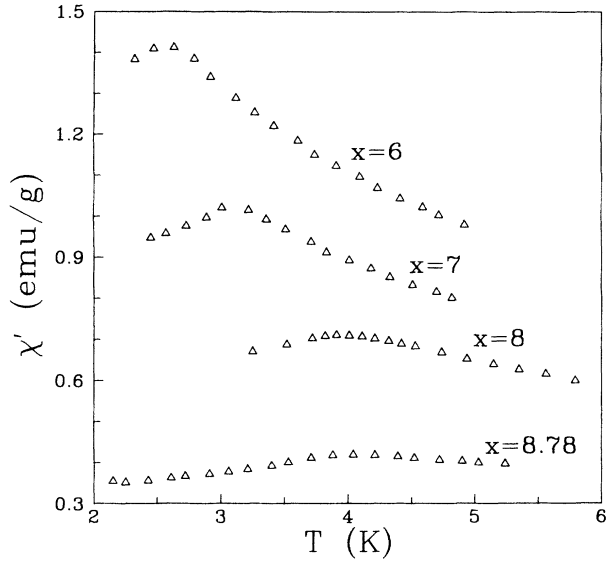

FIG. 5. Real part of the ac susceptibility showing the variation of the peak associated with the freezing temperature as a function of the Cr concentration. Sample with $x=8.78$ is also included.

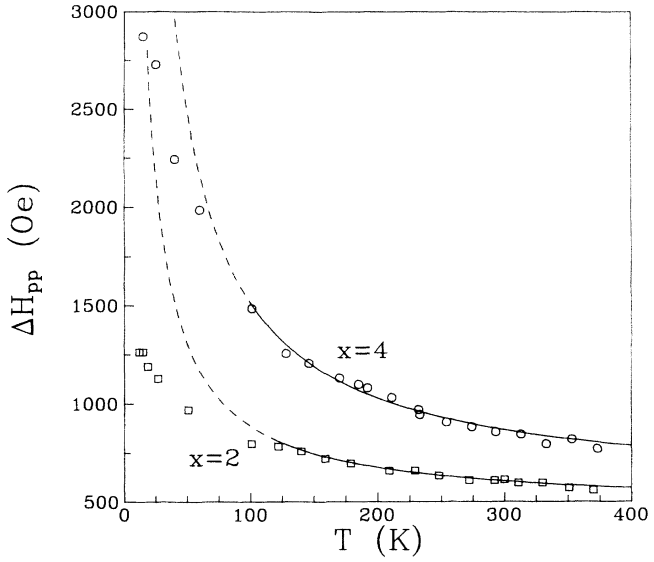

FIG. 7. ESR linewidth vs temperature for $x=2$ and $x=4$. Lines represent the expression (5) of the text, with the values of θ and ΔH_{pp} given in Table I. The continuous parts correspond to the range of temperature for which this expression fits well with experiment, i.e., the region of quasiparamagnetic behavior.

The ESR peak-to-peak linewidth, ΔH_{pp}, is found to be strongly dependent on composition and temperature (see Figs. 7 and 8). In the high-temperature limit, the ESR linewidth is the result of a competition between dipolar coupling, that tends to broaden the line, and exchange interactions, that tend to narrow it. The resulting line has a truncated Lorentzian shape, and a temperature-independent linewidth[17] given by

$$\Delta H_{pp}=(1/\gamma)\omega_p^2/\omega_e \ , \tag{3}$$

where γ is the gyromagnetic ratio, ω_p is the root mean square dipolar perturbation frequency, and ω_e is the exchange frequency. At lower temperatures the linewidth

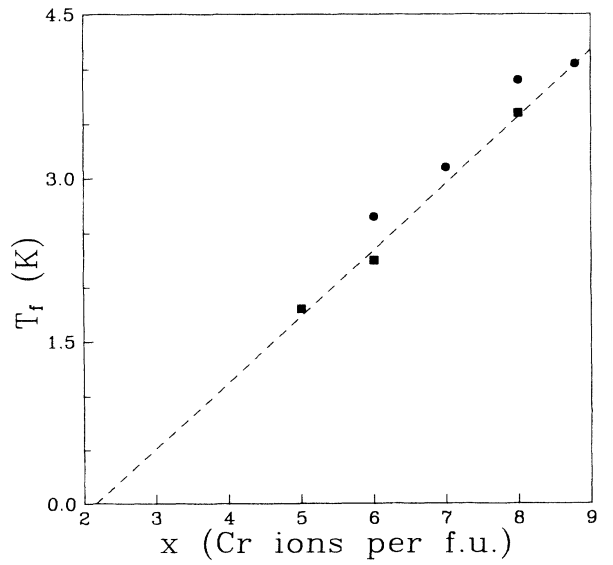

FIG. 6. Variation of the freezing temperature as a function of the Cr concentration showing the extrapolated values for the nondiluted compound ($x=9$) and the percolation threshold ($T_f=0$). Data from ac (●) and dc (■) measurements are included.

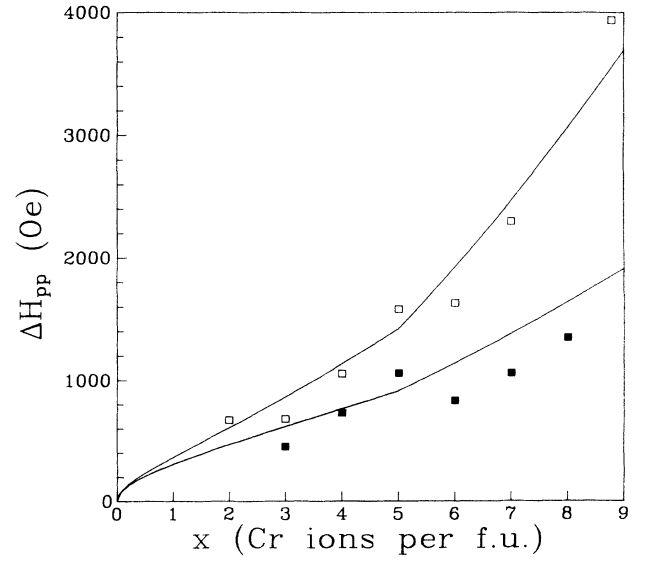

FIG. 8. ESR linewidth vs Cr concentration for $T=200$ K (□) and $T=500$ K (■). The continuous lines correspond to fits of Eq. (6), using the linear fit of θ vs x found in Fig. 9. Note that two different regimes are observed: one for Cr concentrations $x\leq 5$, and another one from $x=5$ to $x=8.78$.

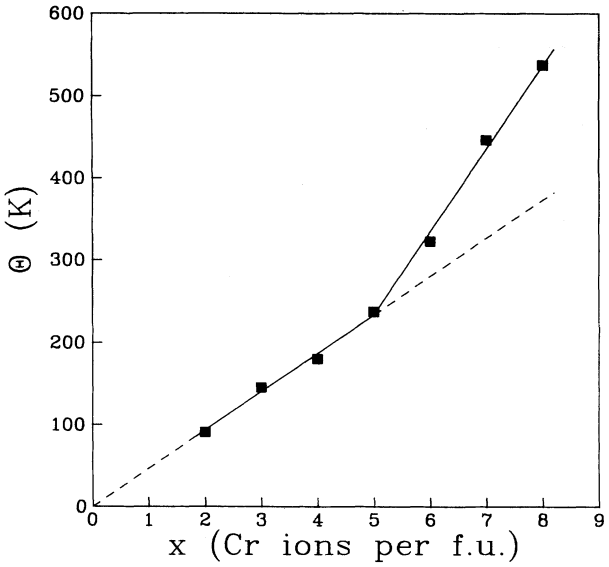

FIG. 9. Plot of the Curie-Weiss temperature, deduced of the dc measurements, vs the Cr concentration. The continued lines are linear fits to the experimental data, extrapolating the low concentration regime to zero.

becomes temperature dependent and its change can be related to the magnetic susceptibility[18] through the expression

$$\Delta H_{pp}(T)=[C/T\chi(T)]\Delta H_{pp}^{\infty} , \quad (4)$$

where $\chi(T)$ is the measured susceptibility, C is the Curie constant, and ΔH_{pp}^{∞} is the peak-to-peak linewidth in the limit of infinite temperature. While the deviation from paramagnetic behavior is not too large, we can use the Curie-Weiss law for susceptibility, and then expression (4) has the form

$$\Delta H_{pp}(T)=(1+\theta/T)\Delta H_{pp}^{\infty} . \quad (5)$$

This equation remains valid only for temperatures above ≈ 100 K, as we can see in Fig. 7. This range of temperature coincides with the high-temperature Curie-Weiss regime observed in dc and ac susceptibility measurements.

The dependence of ΔH_{pp} versus x, at a fixed temperature, is determined by the effects of dilution on the limit ΔH_{pp}^{∞} and on the Curie-Weiss temperature θ (Ref. 19), and takes the form

$$\Delta H_{pp}(x)=[1+(ax+b)/T]x^{1/2}\Delta H_{pp}^{\infty}(x_{max}) . \quad (6)$$

The change in the slope of the Curie-Weiss temperature versus x can explain the two different regimes shown in Fig. 8: One for $x \leq 5$, and another one that extends from $x=5$ to the maximum achieved value of x, i.e., $x=8.78$.

IV. SUMMARY AND CONCLUSIONS

The results obtained from neutron powder-diffraction studies show that no long-range magnetic ordering exists in any of the samples of the solid solution $SrGa_{12-x}Cr_xO_{19}$ down to 1.5 K as was previously stated by Broholm *et al.*[8,9] and hence we confirm that the *kagome* system $SrGa_{12-x}Cr_xO_{19}$ may be considered as a prototypical case of frustration-induced inhibition of long-range order in a solid. Neutron diffraction also shows that Cr^{3+} ions do enter in octahedral sites ($2a$, $12k$, and $4f_{VI}$) with almost random distribution among these sites. The octahedral sites network has a *kagome*-like crystal structure and hence the lattice has a high degree of geometrical frustration. Layers of $12k$ Cr^{3+} ions having a *kagome* structure are connected between them through the more diluted $2a$ and $4f_{VI}$ layers, the later acting as a bottleneck since there are two $4f_{VI}$ layers between two consecutive $12k$ layers (see Fig. 1). This stacking mimics a *kagome* lattice along the c axis and leads, in fact, to a *kagome* block structure made of five stacked layers ($4f_{VI}$-$12k$-$2a$-$12k$-$4f_{VI}$).

dc and ac magnetic measurements display a Curie-Weiss behavior in the high-temperature regime with values of the effective moment very similar to that corresponding to spin only Cr^{3+} ion. For temperatures lower than around 100 K magnetic susceptibility deviates from the Curie-Weiss law, signaling that magnetic correlations start to develop and finally display an important increase below 20 K, in good agreement with previous inelastic neutron-scattering experiments.[8,9] A departure from paramagnetic behavior at the same temperatures is also observed in ESR measurements. The values of the Curie-Weiss temperature ($-537 \leq \theta \leq -90$ K) indicate the existence of strong unsatisfied antiferromagnetic interactions between the Cr^{3+} ions. The dependence of θ on the dilution of Cr^{3+} ions shows two different regimes, also evident through ESR measurements: One for the compounds with $x \leq 5$, and another one for the compounds from $x=5$ to $x=8.78$. The degree of magnetic frustration measured as the ratio $\theta/T_f \approx 135$ remains almost constant, within the experimental error, in the concentration range $5 \leq x \leq 8$. This independence of the θ/T_f ratio on the dilution degree for $x \geq 5$ may be considered as a proof of the intrinsic structural origin of the magnetic frustration. In this way, the study of magnetic dilution allows us to ascertain that the strong magnetic frustration in the *kagome* antiferromagnet $SrGa_{12-x}Cr_xO_{19}$ arises mainly from the geometrical frustration of Cr^{3+} ions arranged in the layered *kagome* lattice, and not from a preferential dilution in the $4f_{VI}$ sites.

From the structural point of view, this last factor could be expected to enhance the two-dimensional behavior but, apparently, a major role should be attributed to the crystal structure itself. A numerical simulation of the low-temperature dc magnetic measurements makes evident the existence of a freezing phenomenon with the freezing temperature decreasing as the degree of dilution increases, going from $T_f=3.8$ K for $x=8$ to $T_f=1.8$ K for $x=5$. ac magnetic susceptibility shows also a peak at temperatures that decrease as the dilution degree increases in agreement with dc results.

The extrapolated value of T_f for the sample with $x=9$ (full occupation of the octahedral sites by the Cr^{3+} ions) is around 4.15 K and there are no signals of any kind to indicate a substantially different behavior of this nondi-

luted sample from the rest of the samples.

Having in mind that the results obtained from neutron diffraction indicate that the relation X_R-H_S, that may be taken as a measure of the variation of the dimensionality of the magnetic sublattice, is independent of the degree of dilution, and the fact that the magnetic frustration is also independent of the Cr^{3+} concentration, we conclude that the giant frustration of this compound is an intrinsic property of the geometric structure and dilution only plays a minor role. In fact, we think that the effect of dilution is just to decrease the freezing temperature as in common spin glasses and that the extrapolation to $T_f=0$ indicates the percolation threshold of the system instead of a two-dimensional behavior.

As we have pointed out, the structure is integrated by *kagome* blocks made of five stacked layers and since the distance between spins within the *kagome* planes is close to that between spins located along the c axis, it is not obvious that the magnetic behavior of the system should exhibit two-dimensional features. Whether or not the system will show bidimensional behavior depends on the relation between the correlation length along the c axis and the thickness of the *kagome* block. If the correlation length is larger than the thickness of the *kagome* block, two-dimensional behavior may be observed.[20,21] On the contrary, even if the system has a marked quasibidimensionality, a three-dimensional magnetic behavior will be observed. Since neutron diffraction shows that static correlations have only very short range,[8,9] a three-dimensional behavior of the spin-glass transition is expected. A complete study of the nonlinear susceptibility of the sample with $x=8$ is underway with the aim to clarify what is the real dimensionality of the magnetic sublattice in this pure geometrically frustrated system.

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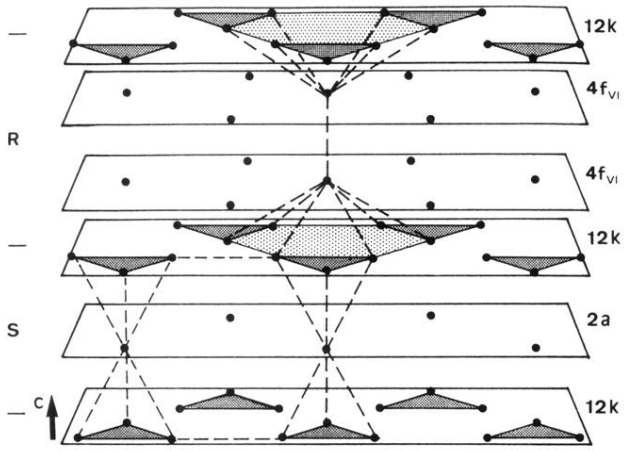

FIG. 1. Schematic representation of the lattice of octahedral sites of the magnetoplumbite-type structure. Two sets of *kagome* lattices may be distinguished: one perpendicular to the c axis formed by $12k$ sites, and another one parallel to the c axis. The latter is also present in the spinel-type structure with the cubic $\langle 111 \rangle$ direction viewed along the hexagonal c axis.