Inhomogeneous magnetic structure of the spinel $Zn_{0.8}Fe_{0.2}Cr_2Se_4$

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Neutron-diffraction and magnetic studies of powdered $Zn_{0.8}Fe_{0.2}Cr_2Se_4$ show that at low temperatures ($T \leqslant 50$ K) this compound has an inhomogeneous magnetic structure. This structure is formed by small ferrimagnetic clusters ($L \leqslant 80$ Å) whose size depends on the temperature.

The statistical substitution of ions in magnetic compounds gives rise to a random competition among superexchange interactions varying in strength. The type of magnetic ordering generally remains an open question.

We have now studied a powdered $Zn_{0.8}Fe_{0.2}Cr_2Se_4$ sample with the normal

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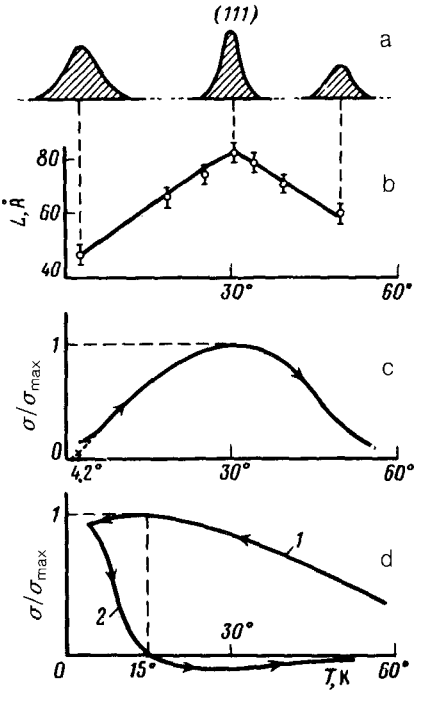

FIG. 1. Temperature dependence of (a) the 111 peak, (b) the cluster size L, and (c,d) the magnetization σ. c—$H = 100$ Oe; d1—$H = 3$ kOe; d2—no field.

spinel structure, in which nonmagnetic Zn^{2+} ions are replaced by magnetic Fe^{2+} ions only at A sites. The Cr^{3+} ions occupy B sites in the spinel structure. The original spinel, $ZnCr_2Se_4$, is known[1] to have a simple spiral structure, formed by the magnetic moments of the Cr^{3+} ions, below $T_N = 21$ K. The pitch of the spiral along the [111] direction at $T = 4.2$ K is 22.5 Å and corresponds to a spin rotation angle $\varphi = 42°$.

The magnetic measurements are carried out on a vibration magnetometer in external magnetic fields of various strengths. It can be seen from Fig. 1(c) that the magnetization σ varies smoothly with the temperature and that its value at $T = 4.2$ K is considerably lower than σ_{max} at $T = 31.5$ K. The imposition of an external magnetic field gives rise to a remanent magnetization. At 4.2 K and $H = 100$ Oe, for example, σ is 14% of σ_{max}, while at $H = 0$ Oe it is only 6%. In a strong field (3 kOe), the $\sigma(T)$ curve becomes smoother; a rounded peak forms at $T = 15$ K; and a large magnetic moment arises at $T = 4.2$ K [curve 1 in Fig. 1(d)]. Turning this field off and heating the sample further showed that the magnetic moment relaxes slowly to its original value, measured at $T = 4.2$ K and $H = 0$ Oe. This temperature dependence of the magnetization also has a rounded peak at $T = 31.5$ K [curve 2 in Fig. 1(d)], as in the case of the external field $H = 100$ Oe. At $T = 4.2$ K and $H = 0$ Oe, the magnetic moment of the sample is thus nonzero, although small.

To determine the type of magnetic order, we measured neutron-diffraction patterns of the sample at various temperatures and without a magnetic field. Calculations based on a diffraction pattern measured at $T = 300$ K showed that Fe replaces Zn only

at A sites. The diffraction pattern measured at $T = 4.2$ K shows, along with structural reflections, some reflections which are allowed for a ferrimagnetic order. An important distinction between this diffraction pattern and that of a ferrimagnet with a "good" long-range magnetic order is that the magnetic reflections are substantially broader than the instrumental reflections. This broadening may be a consequence of a superposition of satellites of a spiral with a pitch longer than that of the original $ZnCr_2Se_4$. However, measurements of the 111 reflection, at a small diffraction angle, showed that the broadening decreases as the diffracted beam is collimated further, but we observed no such fine structure or change in the shape in this peak. These results mean that the broadening of the 111 peak was caused by small regions of coherent magnetic scattering of neutrons, i.e., clusters. Measurements of series of neutron-diffraction patterns in the temperature interval 4.2 K$\leqslant T \leqslant$50 K also showed that the broadening, the amplitude, and the integrated intensity of the magnetic reflections depend on the temperature [Fig. 1(a)]. It can be seen from Fig. 1, (b) and (c), that the changes in the average cluster size and in the magnetization with increasing temperature are similar. The cluster size was determined from the broadening of the 111 magnetic reflection. Consequently, small inhomogeneities from first; the value of σ is correspondingly low. The clusters then become larger, reaching $L \sim 80$ Å at $T = 31.5$ K; this situation corresponds to the maximum of the magnetization. As the temperature is lowered further, the clusters again break up (they decrease in size), and there is a significant decrease in the magnetization. The apparent reason for this significant decrease in magnetization is that, in addition to the breaking up of the clusters, there are random deviations of their magnetic moments from the selected direction. In addition to these deviations of the cluster magnetic moments, there are undoubtedly local deviations of the magnetic moments from the collinear ferrimagnetic structure inside the clusters also. The change in the degree of homogeneity of the magnetic structure with decreasing temperature stems from the increased role played by various random weak competing exchange interactions in comparison with thermal vibrations.

We believe that the magnetic structure of $Zn_{0.8}Fe_{0.2}Cr_2Se_4$ in the temperature interval 4.2 K$\leqslant T \leqslant$32 K has all the features of a cluster spin glass, and the significant decreases in the magnetic moment and size of the clusters are evidence that this structure may undergo a transition to the structure of an "ideal" spin glass at $T \leqslant 3$ K [an extrapolation of the $\sigma(T)$ curve in Fig. 1(c)].

[1]R. Plumier, J. Phys. **27**, 213 (1966).

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