Magnetic ordering of Mn^{2+} and Cr^{3+} ions in the garnet $Mn_3Cr_2Ge_3O_{12}$

I. V. Golosovskiĭ, V. P. Plakhiĭ, O. P. Smirnov, Yu. P. Chernenkov, A. V. Kovalev, and M. N. Bedrizova

B. P. Konstantinov Institute of Nuclear Physics, USSR Academy of Sciences
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The magnetic structure of the spins of Mn^{2+} in the dodecahedral sites and the spins of Cr^{3+} in the octahedral sites of the garnet $Mn_3Cr_2Ge_3O_{12}$ is determined by a neutron-diffraction method. The magnetic ordering in each of the sublattices sets in independently at 3.9 and 5.1°K, respectively.

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The magnetic ions in a garnet structure can occupy sites of three types (octahedral, tetrahedral, and dodecahedral); this leads to a great variety of the magnetic structures, depending on the ratio of the interactions between the sublattices and within the individual sublattices. It is of interest in this connection to ascertain the magnetic structure of the garnet $Mn_3Cr_2Ge_3O_{12}$, in which the Mn^{2+} ions are in dodecahedral sites, and the Cr^{3+} ions are in octahedral sites. Measurements of the heat capacity [1] show several anomalies at helium temperatures, and this may offer evidence of the unusual character of the magnetic ordering.

We have carried out a neutron-diffraction investigation of a polycrystalline sample of this compound in the temperature interval 2 – 300°K. The neutron-diffraction pattern measured at 2°K reveals, on top of the results at room temperature, reflections and contributions to certain nuclear reflections, due to long-range antiferromagnetic order. All the reflections are indexed in a chemical cell, and the sum of all the indices is even. Consequently, the magnetic cell remains body centered. The symmetry of the crystallographic positions occupied by the magnetic ions admits within the framework of the Heisenberg Hamiltonian twelve spin-structure modes of the ions in the dodecahedral sublattice and eight in the octahedral one. [2] A careful analysis of the

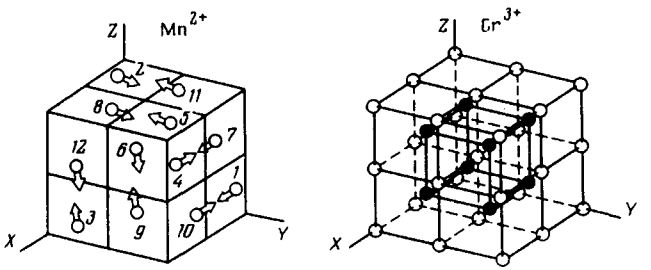

FIG. 1. Antiferromagnetic structure of the spins of Mn^{2+} and Cr^{3+}.

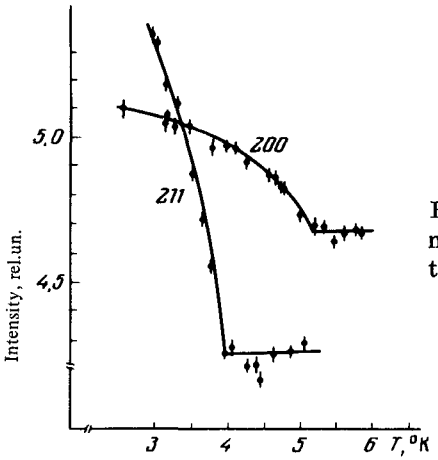

FIG. 2. Temperature dependence of the magnetic reflections 200 and 211 corresponding to the sublattices of Cr^{3+} and Mn^{2+}.

laws governing the extinction of the magnetic reflections corresponding to these configurations shows that the observed system of reflections can be due to a single magnetic mode for the ions in the octahedral sites and to one of four modes (or their linear combination) for the ions in the dodecahedral sites.

The magnetic structure of the Cr^{3+} ions is analogous to the structures observed in garnets with magnetic ions Ni^{2+}, Co^{2+}, Mn^{2+}, and Cr^{3+} [3] only in octahedral sites. This model represents two ferromagnetic lattices imbedded in each other and antiferromagnetically ordered relative to each other (Fig. 1). The orientation of the Cr^{3+} spins relative to the crystallographic axes cannot be determined from the neutron-diffraction data. It follows from the temperature dependence of the reflection 200 (Fig. 2) corresponding to this structure that the magnetic order in the system of the Cr^{3+} ions sets in below $T_{N\,1} = 5.1$ °K, which may correspond to an anomaly in the heat capacity.

A refinement of the possible configurations in the dodecahedral sublattice by least squares leads in only one possible way to a noncollinear triangular mode. All the spins lie in the (11) plane along $[\bar{2}11]$ directions. The spins of the atoms whose coordinates are connected by an inversion center are oppositely directed (Fig. 1). (The numbers of the atoms are the same as those used in [2] and [4]). A similar structure was proposed in [5] for the garnet $Mn_3Al_2Ge_3O_{12}$ with Mn^{2+} ions occupying dodecahedral sites. From the temperature dependence of the 211 reflection corresponding tc this structure we determined the magnetic-ordering temperature $T_{N\,2} = 3.9$ °K of the Mn^{2+} ions, which is close to the maximum of the heat capacity. [1]

The spins of the Cr^{3+} and Mn^{2+} ions are thus ordered independently. The coupling between the octahedral and dodecahedral sublattices is weak and leads only to a decrease of the intrasublattice interaction in comparison with garnets that contain one of these ions ($T_N \approx 12$ °K for the garnet $Ca_3Cr_2Ge_3O_{12}$ [6] and $T_N = 6.68$ °K for the garnet $Mn_3Al_2Ge_3O_{12}$ [4]).

In conclusion, we are grateful to B. V. Mill' for synthesizing the sample.

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