# Magnetic phase diagram of the garnet $Mn_3Cr_2Ge_3O_{12}$

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The magnetic susceptibility has been measured at 2 mHz to study the effect of diamagnetic dilution and an external magnetic field on the independent antiferromagnetic ordering of the octahedral and dodecahedral sublattices of the garnet $Mn_3Cr_2Ge_3O_{12}$ (MnCrG). A magnetic phase diagram is constructed for this compound.

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An interesting effect observed in the garnet $Mn_3Cr_2Ge_3O_{12}$ (MnCrG), in which the dodecahedral ($c$) and octahedral ($a$) sites are occupied by $Mn^{2+}$ and $Cr^{3+}$ ions, respectively, is an independent antiferromagnetic ordering of the $a$ and $c$ sublattices at $T_N^a = 5.1$ K and $T_N^c = 3.0$ K. Evidence for this independent ordering comes from specific-heat measurements[1] and data on neutron diffraction.[2] According to the neutron-diffraction study by Golosovskiĭ and Plakhtiĭ,[2] the magnetic structure at the Cr atoms, as in the garnet $Ca_3Cr_2Ge_3O_{12}$ (CaCrG), consits of two ferromagnetic sublattices which are set in each other in an antiferromagnetic fashion; the ordering of the Mn atoms is the same as the triangular antiferromagnetic structure of the garnet $Mn_3Al_2Ge_3O_{12}$ [the spins of the Mn atoms lie in the (111) plane and are oriented in a ⟨211⟩ direction].

In the ordered state, the magnetic symmetry of MnCrG is such that the molecular fields $\mathbf{H}_{ca}$ and $\mathbf{H}_{ac}$ created by the spins of one crystal sublattice at any atom of the other sublattice are zero[1], i.e.,

$$\mathbf{H}_{ca} = \gamma_{ca}(\mathbf{M}_1^a + \mathbf{M}_2^a) = 0.$$

Here $\mathbf{M}_1^a$ and $\mathbf{M}_2^a$ are the oppositely directed magnetic moments of the $Cr^{3+}$ antiferromagnetic sublattices, and $\gamma_{ca}$ is the parameter of the exchange interaction between the ions at lattice sites $c$ and $a$. There is a corresponding situation for the field $\mathbf{H}_{ac}$. We may therefore except that an external magnetic field strong enough to disrupt this cancellation of $\mathbf{M}_1^a$ and $\mathbf{M}_2^a$ will lead to the appearance of a field $\mathbf{H}_{ca}$ (and a field $\mathbf{H}_{ac}$) which will transform the antiferromagnetic structures of MnCrG. A partial diamagnetic substitution for the $Cr^{3+}$ or $Mn^{2+}$ ions would evidently have the same result.

In this letter we are reporting an attempt to observe these effects by measuring the magnetic susceptibility ($\chi'$) at the frequency 2 MHz in magnetic fields up to 40 kOe at temperatures in the range 2.5-20 K. Our apparatus could plot, on an $x$-$y$ chart recorder, the field dependence of a signal proportional to the change in the

  

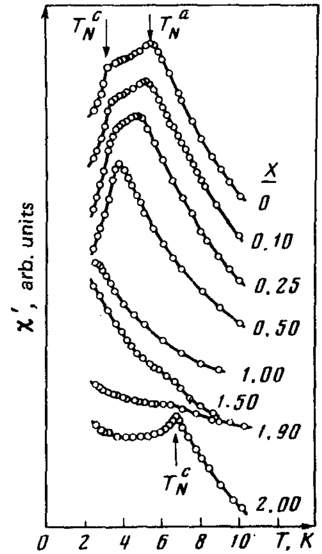


FIG. 1. Temperature dependence of the magnetic susceptibility of the garnets $Mn_3Ga_xCr_{2-x}Ge_3O_{12}$.

frequency of a highly stable cryogenic field-effect-transistor oscillator. For improved sensitivity to the details in the behavior of $\partial M/\partial H$, the output signal could be partially canceled by a special circuit, so that $\Delta\chi'(H)$ could be recorded. The temperature was measured with a carbon thermometer and regulated within ±0.05 K with a DTS-2 thermostat.

For the measurements we used cylindrical polycrystalline samples of $Mn_3Ga_xCr_{2-x}Ge_3O_{12}$ with $0 \leqslant x \leqslant 2$. An x-ray structural analysis showed that all the samples consisted of a single phase and that the lattice constant varied from 12.025 ± 0.02 Å for MnCrG ($x = 0$) to 12.019 ± 0.02 Å in $Mn_3Ga_2Ge_3O_{12}$ (MnGaG).

Figure 1 shows curves of $\chi'(T)$ for the garnets $Mn_3Ga_xCr_{2-x}Ge_3O_{12}$; these curves illustrate a transition from an antiferromagnet with two Neel temperatures ($x = 0$) to the ordinary antiferromagnet MnGaG ($x = 2$). There is a significant concentration interval ($0.5 < x < 1.5$) in which there is apparently no long-range magnetic order. It seems unlikely that 30% of the $Cr^{3+}$ ions ($s = 3/2$) (30% of the total number of magnetic cations in unit cell) in the paramagnetic state could "paint over" the $\chi'(T)$ anomaly caused by the antiferromagnetic ordering of the dodecahedral $Mn^{2+}$ ions ($s = 5/2$).

The decrease in the $Cr^{3+}$ concentration in MnCrG apparently gives rise to disordered $Cr^{3+}$ ions in the nearest neighborhood of the $Mn^{2+}$ ions; these $Cr^{3+}$ ions in turn give rise to the field $\mathbf{H}_{ca}$ and thus tend to disrupt the long-range magnetic order in the $c$ sublattice.

Figure 2 shows some of the results on $\chi'(H)$ and $\Delta\chi'(H)$ at various temperatures. The results may be summarized as follows: 1) There are two maxima, at fields $H_1$ and $H_2$, for the garnets with $0 \leqslant x \leqslant 0.5$; 2) the temperature dependence $H_1(T)$ is weak, while $H_2$ increases sharply with decreasing temperature; 3) the values of $\chi'$ for samples with $x > 0.5$ are essentially independent of $H$; 4) $\chi'(H)$ for the garnet CdCrG exhibits a single maximum, at $H_1 \approx 4.6$ kOe. We believe that the maximum of $\chi'(H)$ in CdCrG corresponds to the spin-flop field. Since the exchange field in this

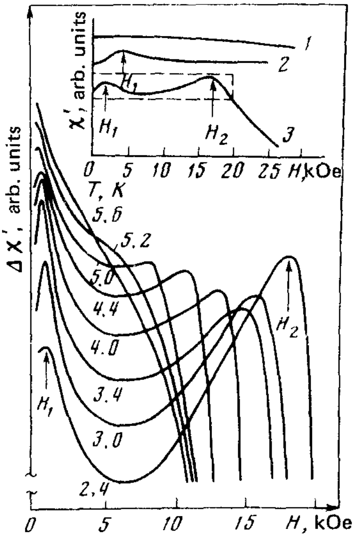


FIG. 2. a–Magnetic-field dependence of $\chi'$ of the garnets MnGaG (1), CdCrG (2), and MnCrG (3); b–Magnetic-field dependence of $\Delta\chi'$ (the part of curve 3 in Fig. 2a in the dashed box has been magnified by a factor of 15) for the garnet MnCrG.

garnet is $H_E = 260$ kOe (Ref. 3), the value found for $H_{sf}$, 4.6 kOe, corresponds to an anisotropy field $H_a = 29.3 \times 10^{-4}$ cm$^{-1}$, in good agreement with the corresponding value found from the antiferromagnetic resonance in Ref. 4. In MnGaG, we have $H_E \approx 200$ kOe (Ref. 5). Judging from our measurements, there is no sublattice flop here, apparently because of the multiple-sublattice magnetic structure of this garnet.

In analyzing the results on MnCrG, we should keep in mind that the values of $T_N^a$ and $T_N^c$ are roughly half the Néel temperatures of the corresponding "single-sublattice" garnets of Cr and Mn. It may therefore be suggested that the field $H_1$ is a measure of $H_{sf}$ of the Cr sublattice. An external magnetic field $H > H_1$ gives rise to an intersublattice exchange interaction ($H_{ca}$) which disrupts the antiferromagnetic ordering of the $Mn^{2+}$ ions in the $c$ sublattice. We believe that the field $H_2$ corresponds to the collapse field ($H_E$) of the $a$ sublattice, although just why this field is so low in comparison with the corresponding field in ClCrG is not completely clear.

From the experimental results on $H_1(T)$ and $H_2(T)$ we can construct a phase

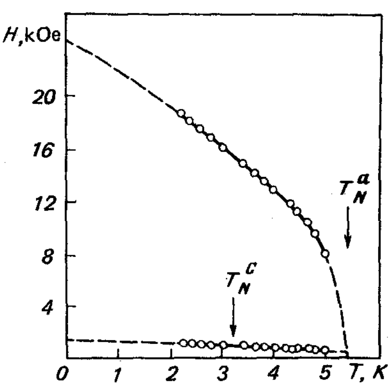


FIG. 3. Magnetic phase diagram of MnCrG.

diagram for MnCrG. It follows from this diagram (Fig. 3) that the "double" antiferromagnetic structure of MnCrG is disrupted by an external field of only ~1 kOe (0 K), at which the antiferromagntic sublattices of the $Cr^{3+}$ ions flop. In fields $H > H_1$, the $c$ sublattice becomes paramagnetic; this circumstance is evidently responsible for the linear increase in the MnCrG magnetization in fields all the way up to 200 kOe, according to observations at liquid-helium temperature in Ref. 1. In the field interval $H_1 < H < H_2$, MnCrG is a mixed phase of paramagnetic $Mn^{2+}$ $c$ ions and antiferromagnetically ordered $Cr^{3+}$ $a$ ions in a spin-flop state. At $H < H_2$, the $a$ sublattice becomes ferromagnetic.

We constructed phase diagrams similar to this one for the garnets $Mn_3Ga_xCr_{2-x}Ge_3O_{12}$ with $x = 0.10, 0.25$, and 0.50. The only differences seen in these other diagrams are in the values of the fields $H_1$ and $H_2$: We find $H_2 \approx 12$ kOe (0 K) at $x = 5$, and $H_1$ increases slightly (~3.5 kOe at 0 K). The latter result can be attributed to an increase in the anisotropy field of the $Cr^{3+}$ sublattice, because the magnetic moment decreases more rapidly than the anisotropy constant.

A neutron-diffraction study of this compound in a magnetic field would be of definite interest for obtaining more information on the magnetic phase diagram of MnCrG.

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