

The summation is carried out over all the two-dimensional reciprocal-lattice vectors  $\mathbf{G} = (G_x, G_y)$ , while  $B_{\mathbf{G}}$  are arbitrary coefficients. We introduce an arbitrary periodic function

$$v(\rho) = \sum_{\mathbf{G}} B_{\mathbf{G}} e^{i\mathbf{G}\rho}. \quad (\text{A.4})$$

Taking into account the notation of (A.4), we can rewrite (A.3) in more compact form<sup>[12]</sup>:

$$\Psi_{\text{vac}}(z, E, r) = \exp \left\{ z \left[ \frac{2m_0}{\hbar^2} \left( V_0 - E + \frac{\hat{p}_z^2}{2m_0} \right) \right]^{1/2} \right\} e^{iz\rho v(\rho)}. \quad (\text{A.5})$$

Substituting (A.5) in the matching equation (A.2) and eliminating  $v(\rho)$  from them, we obtain the boundary condition in the form (7), where in the given particular case

$$\hat{F}(\pm) = \delta(z - z_0) \left\{ \pm \left[ \frac{2m_0}{\hbar^2} \left( V_0 - E + \frac{\hat{p}_z^2}{2m_0} \right) \right]^{1/2} - \frac{\partial}{\partial z} \right\}. \quad (\text{A.6})$$

It is necessary to take the plus sign in (A.6) if the crystal occupies in accordance with (A.1) the right-hand half of the space. The opposite case corresponds to the choice of the minus sign in (A.6).

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Translated by J. G. Adashko

## Antiferromagnetism of the garnet $Mn_3Al_2Ge_3O_{12}$

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(Submitted December 30, 1975)

Zh. Eksp. Teor. Fiz. 70, 2279–2285 (June 1976)

Possible types of magnetic ordering in dodecahedral sites of the garnet are analyzed within the framework of the generalized molecular field theory. The real magnetic structure of the garnet  $Mn_3Al_2Ge_3O_{12}$ , whose antiferromagnetic ordering at  $T_N = 6.65 \pm 0.05^\circ\text{K}$  is due to exchange interaction between the  $Mn^{2+}$  dodecahedral ions, is considered by taking into account the features of the exchange bonds. The magnetic properties ( $1.6$ – $80^\circ\text{K}$ ) and heat capacity ( $2$ – $20^\circ\text{K}$ ) of polycrystalline  $Mn_3Al_2Ge_3O_{12}$  are measured. The exchange field is determined,  $H_E(3^\circ\text{K}) = 210$  kOe, as well as the exchange interaction integrals  $J_1 = -0.57^\circ\text{K}$ ,  $J_2' = -0.12^\circ\text{K}$ . It is shown that the experimental data can be satisfactorily described by the molecular field theory.

PACS numbers: 75.50.Ee, 75.30.Et, 75.10.Dg

### 1. INTRODUCTION

The properties of cubic antiferromagnetic garnets in which the dodecahedral sites ( $c$ ) are occupied by rare-earth ions have by now been investigated quite in detail. Most of these compounds—gallates ( $R_3Ga_5O_{12}$ ) and aluminates ( $R_3Al_5O_{12}$ )—become antiferromagnetically ordered at  $T_N < 1^\circ\text{K}$ . The singularities of the magnetic properties of rare-earth gallates and aluminates are due to the competition between the exchange, anisotropy, dipole, and hyperfine interactions, which are comparable in size. Capel<sup>[1]</sup> has demonstrated, using

the molecular-field method with allowance for the dipole forces and the anisotropic exchange, that three types of magnetic order of rare-earth ions are possible, two antiferromagnetic and one ferrimagnetic. The realized type of order is determined mainly by the character of the one-ion anisotropy.

It is of interest to study the magnetic ordering of the  $S$  ions in the dodecahedral sublattice of the garnet, since we can expect here a great diversity of magnetic structures, owing to the isotropic exchange. This calculation, in the molecular-field approximation with allow-

ance for an exchange of the Heisenberg type, was carried out by Bertaut.<sup>[2]</sup> He considered, however, nearest-neighbor approximations, so that the resultant antiferromagnetic configurations of the spins turned out to be energywise unfavored, and only ferromagnetic ordering turned out to be possible. The compound  $Mn_3Al_2Ge_3O_{12}$  ( $MnAlG$ ) is in essence the only antiferromagnetic garnet in which only  $c$ -sites are occupied by magnetic S ions ( $Mn^{2+}$ ) (the garnet  $Gd_3Ga_5O_{12}$  remains paramagnetic down to  $0.35$  °K<sup>[3]</sup>). The antiferromagnetism of  $MnAlG$  was simultaneously established by Prandl<sup>[4]</sup> and Plumier<sup>[5]</sup> from experiments on neutron diffraction at  $4.2$  °K. To describe the magnetic order in  $MnAlG$ , they have proposed<sup>[4, 5]</sup> three magnetic structures, of which only one agrees with those predicted by Capel.<sup>[1]</sup>

In this paper we use the Bertaut method to analyze the possible spin configurations of magnetic  $c$ -ions with allowance for the interaction of the atoms belonging to different Bravais lattices. To study the singularities of the antiferromagnetic ordering of  $Mn^{2+}$  in the  $c$ -sites of the garnet and to obtain quantitative estimates of the exchange interactions for the chosen magnetic structure, we measured the heat capacity ( $2$ – $20$  °K) and the magnetic properties ( $1.6$ – $80$  °K) of polycrystalline  $MnAlG$ .

## 2. SPIN CONFIGURATIONS AND EXCHANGE BONDS OF MAGNETIC $c$ -IONS

To determine the spin configurations that are allowed by the symmetry of the crystal, we use the Bertaut method, which consists of finding the eigenvectors

$$\xi_{v\mu}^j(\mathbf{k}) = \sum_{i(v)} \mathbf{S}_{i(v)}^j \exp[2\pi i \mathbf{k} \cdot \mathbf{R}_{i(v)}], \quad v, j=1, 2, \dots, n,$$

of the interaction matrix  $\hat{\xi}(\mathbf{k})$ , the elements of which are the Fourier transforms of the exchange integrals

$$\xi_{v\mu}^j(\mathbf{k}) = \sum_{i(v)\mu} J_{i(v)\mu} \exp[2\pi i \mathbf{k} \cdot (\mathbf{R}_{i(v)} - \mathbf{R}_\mu)], \quad v, \mu=1, 2, \dots, n.$$

Here  $\mathbf{S}_{i(v)}^j$  is the spin of the  $i$ -th atom in the  $v$ -th Bravais lattice for the spin configuration  $j$ ,  $\mathbf{R}_{i(v)}$  is the radius vector from the origin of the unit cell to this atom;  $J_{i(v)\mu}$  is the exchange integral between the  $i$ -th lattice and an arbitrarily chosen atom in the  $\mu$ -th lattice with radius vector  $\mathbf{R}_\mu$ ;  $\mathbf{k}$  is propagation vector (it is determined experimentally from the law governing the extinction of the magnetic reflections); the summation is over all the atoms belonging to one and the same Bravais lattice;  $n$  is the number of Bravais lattices in the crystal.

The eigenvectors  $\mathbf{T}_v^j(\mathbf{k})$  are the columns of a unitary matrix  $\hat{T}(\mathbf{k})$ , which diagonalizes  $\hat{\xi}(\mathbf{k})$  via the transformation

$$\hat{T}^{-1} \hat{\xi}(\mathbf{k}) \hat{T} = \hat{\lambda},$$

where  $\hat{\lambda}$  is a diagonal matrix. The spin modes  $\Sigma_j$ , relative to the number of the Bravais lattices are obtained with the aid of the inverse Fourier transformation

$$S_v^j = \sum_i T_v^j(\mathbf{k}) \exp(-2\pi i \mathbf{k} \cdot \mathbf{R}_i).$$

To obtain the matrix  $\hat{\xi}_c(0)$ , we confine ourselves to the case  $\mathbf{k}=0$  (i.e., the magnetic cell coincides with the crystallographic cell), and number the dodecahedral atoms making up the 12 Bravais lattices, in the following sequence:

- 1)  $\frac{1}{4}, \frac{1}{4}, \frac{3}{8}; 2) \frac{1}{4}, \frac{3}{8}, \frac{1}{2}; 3) \frac{3}{8}, \frac{1}{2}, \frac{1}{4};$
- 4)  $0, \frac{1}{4}, \frac{1}{8}; 5) \frac{1}{4}, \frac{1}{8}, 0; 6) \frac{1}{8}, 0, \frac{1}{4};$
- 7)  $0, \frac{1}{4}, \frac{5}{8}; 8) \frac{1}{4}, \frac{5}{8}, 0; 9) \frac{5}{8}, 0, \frac{1}{4};$
- 10)  $\frac{1}{2}, \frac{1}{4}, \frac{7}{8}; 11) \frac{1}{4}, \frac{7}{8}, \frac{1}{2}; 12) \frac{7}{8}, \frac{1}{2}, \frac{1}{4}.$

Each three atoms whose coordinates are written down in the same line are transformed into one another by a threefold axis. The matrix  $\hat{\xi}_c(0)$  should contain in the general case 12 different elements, but allowance for the point symmetry of the  $c$ -sites ( $D_2$ ) makes it possible to decrease the number of independent parameters.

It is easy to show that out of the 11 Bravais lattices with which lattice 1 interacts, the lattices 2, 3, 8, 9 ( $\xi_{12} = \xi_{13} = \xi_{18} = \xi_{19} = \xi_1$ ) and 5, 6, 11, 12 ( $\xi_2$ ) will be equivalent. Putting  $\xi_{1-10} = \xi_3$ ,  $\xi_{14} = \xi_{42}$ ,  $\xi_{17} = \xi_5$ ,  $\xi_{11} = \xi_6$ , we can express the matrix  $\hat{\xi}_c(0)$  in the form

$$\begin{aligned} \hat{\xi}_c(0) &= \begin{pmatrix} \hat{C}_1 & \hat{C}_2 & \hat{C}_3 & \hat{C}_4 \\ \hat{C}_2 & \hat{C}_1 & \hat{C}_4 & \hat{C}_3 \\ \hat{C}_3 & \hat{C}_4 & \hat{C}_1 & \hat{C}_2 \\ \hat{C}_4 & \hat{C}_3 & \hat{C}_2 & \hat{C}_1 \end{pmatrix}, \\ \hat{C}_1 &= \xi_1 \begin{pmatrix} \alpha & 1 & 1 \\ 1 & \alpha & 1 \\ 1 & 1 & \alpha \end{pmatrix}, \quad \hat{C}_2 = \xi_2 \begin{pmatrix} \beta & 1 & 1 \\ 1 & \beta & 1 \\ 1 & 1 & \beta \end{pmatrix}, \\ \hat{C}_3 &= \xi_1 \begin{pmatrix} \gamma & 1 & 1 \\ 1 & \gamma & 1 \\ 1 & 1 & \gamma \end{pmatrix}, \quad \hat{C}_4 = \xi_2 \begin{pmatrix} \delta & 1 & 1 \\ 1 & \delta & 1 \\ 1 & 1 & \delta \end{pmatrix}, \\ \alpha &= \xi_8/\xi_1, \quad \beta = \xi_4/\xi_2, \quad \gamma = \xi_5/\xi_1, \quad \delta = \xi_3/\xi_2. \end{aligned}$$

The matrix  $\hat{\xi}_c(0)$  commutes with the matrix in which account is taken of interactions of only the nearest neighbors, and is therefore diagonalized with the aid of the same unitary matrix  $\hat{T}_c(0)$ . According to Bertaut<sup>[2]</sup> we have

$$\hat{T}_c(0) = \frac{1}{\sqrt{12}} \begin{bmatrix} \hat{U} & \hat{U} & \hat{U} & \hat{U} \\ \hat{U} & \hat{U} & -\hat{U} & -\hat{U} \\ \hat{U} & -\hat{U} & \hat{U} & -\hat{U} \\ \hat{U} & -\hat{U} & -\hat{U} & \hat{U} \end{bmatrix}, \quad \hat{U} = \begin{bmatrix} 1 & 1 & 1 \\ 1 & r & r^* \\ 1 & r^* & r \end{bmatrix}, \quad (1)$$

$r = \exp(2\pi i/3)$  and  $r^* = \exp(-2\pi i/3)$ . Since  $\mathbf{k}=0$ , it follows that  $\mathbf{S}_\mu^j(0) = \mathbf{T}_\mu^j(0)$  and the possible spin configurations are given by the columns of the matrix (1).

The modes  $\Sigma_1$ ,  $\Sigma_4$ ,  $\Sigma_7$ ,  $\Sigma_{10}$  are collinear. The remaining modes are triangular configurations, the differences between  $\Sigma_2$  and  $\Sigma_3$ ,  $\Sigma_5$  and  $\Sigma_6$ ,  $\Sigma_8$  and  $\Sigma_9$ ,  $\Sigma_{11}$  and  $\Sigma_{12}$  are only in direction of rotation and therefore not in energy.

In contrast to octahedral and tetrahedral garnet sites, in which the magnetic atoms are bound by exchange chains consisting at least of two oxygen atoms, the nearest neighbors in dodecahedral positions are coupled by an indirect exchange interaction with participation of one intermediate oxygen atom. The bonds with

TABLE I. Radii of the coordination spheres ( $R$ ), numbers of the Bravais lattices ( $N$ ) with which the atom from lattice 1 interacts, and the number of the neighboring atoms  $z$ .

Number of sphere	$R$ (in fractions of the lattice parameter)	$N$	$z$	Exchange parameter $J$
1	0.306	2, 3, 8, 9	1×4	$J_1$
2	0.468	5, 6, 11, 12	2×4	$J_2$
3	0.500	10	2	$J_3$
4	0.559	4	4	$J_4$
5	0.586	7	4	$J_5$
		2, 3, 8, 9	1×4	$J'_1$

the second, third, fourth, and fifth coordination spheres are effected by chains consisting of at least two oxygen atoms. In these bonds, however, it is practically impossible to separate the oxygen atoms, which participate in the exchange interaction to a greater degree than others. Moreover, it is difficult even to distinguish bonds of the types  $M_c-O-O-M_c$  from  $M_c-O-M_d-O-M_c$  ( $M_c$  is a magnetic cation in the  $c$ -position, and  $M_d$  is a nonmagnetic cation in the  $d$ -position). Owing to the high coordination of the oxygen polyhedron ( $Z=8$ ), several approximately equivalent bonds of different type are formed immediately between the magnetic atoms, and this leads to a certain averaging of the exchange parameters  $J$ . In this connection, and also taking the radii of the coordination spheres into account (see Table I), we shall assume that  $J_2=J_3=\bar{J}_2$ ,  $J_4=J_5=J'_1=\bar{J}_3$ . The interactions of the magnetic atoms located at a distance  $R \geq 0.685a_0$  (sixth coordination sphere) will be neglected.

The eigenvalues  $\lambda_i$  of the matrix  $\xi_c(0)$  for the spin configuration  $\Sigma$ , constitute the exchange energy expressed in terms of the exchange parameters. In our case  $\xi_1=J_1+J'_1$ ,

$$\xi_2=2J_2, \xi_3=2J_3, \xi_4=4J_4, \xi_5=4J_5, \xi_6=0$$

and the parameters are given by

$$\begin{aligned} \lambda_1 &= 4J_1 + 10\bar{J}_2 + 12\bar{J}_3, \quad \lambda_{2,3} = -2J_1 - 2\bar{J}_2 + 6\bar{J}_3, \\ \lambda_4 &= \lambda_{5,6} = -2\bar{J}_3, \quad \lambda_7 = 4J_1 + 2\bar{J}_2 - 2\bar{J}_3, \\ \lambda_{8,9} &= -2J_1 + 2\bar{J}_2 - 2\bar{J}_3, \quad \lambda_{10} = \lambda_{11,12} = 2\bar{J}_2 - 8\bar{J}_3. \end{aligned} \quad (2)$$

### 3. HEAT CAPACITY AND MAGNETIC PROPERTIES OF MnAlG

The single-phase (by x-ray structure) sample of MnAlG ( $a_0=11.897 \text{ \AA}$ ) was prepared by a ceramic technology with double annealing in air at  $t=1170 \text{ }^\circ\text{C}$ . The heat capacity and the magnetic properties were measured with the previously described installations.<sup>[6, 7]</sup> Figure 1a shows the experimental temperature dependence of the heat capacity of MnAlG in the interval  $2-20 \text{ }^\circ\text{K}$ . The  $\lambda$  anomaly observed at  $T_N=6.65 \pm 0.05 \text{ }^\circ\text{K}$  corresponds to antiferromagnetic ordering of the  $\text{Mn}^{2+}$ . In the investigated temperature region, the heat capacity of MnAlG is defined by

$$C = C_{\text{latt}} + C_{\text{mag}} + C_{\text{nuc}}.$$

Using the hyperfine interaction constants of  $\text{Mn}^{2+}$  in the garnet matrix,<sup>[8]</sup> we calculated  $C_{\text{nuc}}$ , which amounts to  $\sim 0.3\%$  of the total heat capacity at  $T=2 \text{ }^\circ\text{K}$ . There-

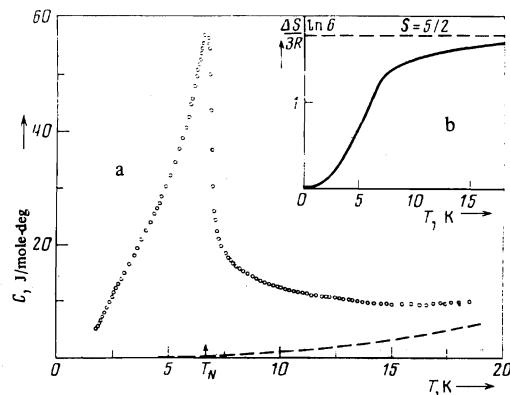


FIG. 1. Temperature dependence of the heat capacity of MnAlG (Fig. a): o—experimental points, dashed—lattice heat capacity; b—temperature dependence of the entropy.

fore the nuclear contribution to the heat capacity of MnAlG was disregarded by us. The value of  $C_{\text{latt}}$  was determined under the assumption that at  $T \gg T_N$  we have

$$C = aT^{-2} + bT^2, \quad (3)$$

where the first term is the magnetic part of the heat capacity and the second is the lattice part.

Relation (3) is satisfied within the limits of experimental accuracy ( $\sim 3\%$ ) for MnAlG in the interval  $13-20 \text{ }^\circ\text{K}$ . From the dependence of  $CT^2$  on  $T^5$  we determine the coefficients  $a=60 \pm 2 \text{ }^\circ\text{K}^2$  and  $b=(3.5 \pm 0.25) \cdot 10^{-5} \text{ }^\circ\text{K}^{-3}$ . The lattice contribution to the specific heat yields a Debye temperature  $\Theta_D=416 \pm 12 \text{ }^\circ\text{K}$ . The lattice specific heat of MnAlG is shown by the dashed line in Fig. 1.

Using the customary relation for the magnetic entropy

$$\Delta S = \int_0^\infty \frac{C_{\text{mag}}}{T} dT,$$

we determine the value of  $\Delta S$ . In the investigated temperature region  $2-20 \text{ }^\circ\text{K}$ , the contribution to the total change of the entropy is 90% (Fig. 1b). At  $T>20 \text{ }^\circ\text{K}$ , the change of the heat capacity was calculated from the relation  $C_{\text{mag}}=a/T^2$ . Extrapolation to the temperature range  $0-2 \text{ }^\circ\text{K}$  was carried out assuming validity of the  $C_{\text{mag}} \propto T^3$  law which follows for the heat capacity of antiferromagnets at  $T \ll T_N$  from spin-wave theory. The so-obtained change of the magnetic entropy,  $\Delta S=1.80R$  (per  $\text{Mn}^{2+}$  ion,  $R$  is the gas constant), is in good agreement with the theoretical value  $\ln(2S+1)=1.79R$  for  $S=\frac{5}{2}$ .

Figure 2 shows the temperature dependence of the

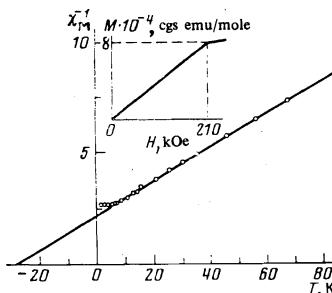


FIG. 2. Temperature dependence of the reciprocal molar susceptibility of MnAlG; the insert shows the magnetization of MnAlG at  $3 \text{ }^\circ\text{K}$  in pulsed magnetic fields.

reciprocal molar susceptibility of MnAlG. Above  $T_N$ , the susceptibility  $\chi_M^{-1}$  obeys the Curie-Weiss law with constants  $\Theta_p = -28 \pm 1$  °K and  $C_M = 13 \pm$  cgs emu/mole. The last quantity gives  $\mu_{eff} = 5.89 \mu_B$ , in good agreement with  $\mu = g[S(S+1)]^{1/2} = 5.92 \mu_B$  for  $S = \frac{5}{2}$  and  $g = 2$ . Below  $T_N$ , the susceptibility is independent of the temperature.

The obtained values of  $T_N$  and  $\Theta_p$  make it possible to determine the exchange field  $H_E$  that destroys the anti-ferromagnetic order in MnAlG. It is easy to show that in the molecular-field approximation we have

$$H_E(T) = \frac{3(T_N - \Theta_p)k\sigma(T)}{g\mu_B(S+1)}, \quad (4)$$

$$\sigma(T) = \frac{M_s(T)}{M_s(0)} = B_{5/2} \left[ \frac{3S}{S+1} \frac{T_N}{T} \sigma(T) \right], \quad (5)$$

where  $M_s(0) = 3Ng\mu_B S = 8.37 \cdot 10^4$  cgs emu/mole, and  $B_{5/2}$  is a Brillouin function. Using the experimental values of  $T_N$  and  $\Theta_p$ , we obtain  $H_E(3$  °K) = 207 kOe from (4) and (5). Measurement of the magnetization of MnAlG in pulsed magnetic fields at 3 °K (Fig. 2, insert) yields for the collapse field a value 210 kOe. Thus, the magnetic properties of MnAlG are well accounted for by the molecular-field theory.

#### 4. MAGNETIC STRUCTURE OF MnAlG; EXCHANGE-INTERACTION INTEGRALS

It follows from neutron-diffraction data<sup>[4, 5]</sup> for MnAlG that  $\mathbf{k} = 0$  and consequently the magnetic ordering of this garnet is described by one of the modes represented by the columns of the matrix  $\hat{T}_c(0)$ , or by a linear combination of these modes. Plumier has proposed two models of the magnetic structure of MnAlG, corresponding to the modes  $\Sigma_4$  and  $\Sigma_{5,6}$ .<sup>[5]</sup> The Prandl model is described by a linear combination of  $\Sigma_7$  and  $\Sigma_{8,9}$ .<sup>[4]</sup> We propose that the value of the  $R$ -factor, is equal to 0.14, 0.52, and 0.07, respectively for these three models favors the choice of the last model.

In the Prandl model the admixture of  $\Sigma_7$  barely exceeds the standard experimental error. It is therefore desirable to assume that the magnetic structure of MnAlG is described by the mode  $\Sigma_{8,9}$ . In this case the magnetic moments of all the atoms lie in the (111) plane and are directed parallel or antiparallel to one of the three crystallographic axes: [211], [121], [112].

The eigenvalue  $\lambda_r$  of an actually existing mode should be maximal, i.e., the exchange-interaction parameters in expressions (2) should correspond to

$$\lambda_r > \lambda_s \quad (6)$$

(we note that this condition is not satisfied for  $\Sigma_4$  and  $\Sigma_{5,6}$  under the fully realistic assumption  $J_4 = J_5$ ).

Combining the minimum-energy condition (6) for the

mode  $\Sigma_{8,9}$  with the molecular-field relations

$$\lambda_1 = 3\Theta_p/2S(S+1), \quad (7)$$

$$\lambda_{8,9} = 3T_N/2S(S+1), \quad (8)$$

we obtain for the exchange-interaction parameter of MnAlG

$$0.51 < -J_1 < 0.64 \text{ K}, 0.09 < -J_2 < 0.14 \text{ K}, 0.07 < -J_3 < 0.15 \text{ K}. \quad (9)$$

As expected,  $J_2$  and  $J_3$ , which characterize exchange interactions with participation of not less than two intermediate links, turn out to be close in magnitude. It is interesting that these interactions are only several times weaker than the indirect exchange interaction via one oxygen ion. The relatively small value of  $J_1$  can be attributed apparently to the Mn-O-Mn angle, the value of which, 101.3°, is not favorable for exchange.<sup>[4]</sup>

Putting in accordance with (9)  $\bar{J}_2 = \bar{J}_3 = J'_2$ , we have determined  $J_1$  and  $J'_2$  for MnAlG from relations (7) and (8):  $J_1 = -0.57$  °K,  $J'_2 = -0.12$  °K. Using the obtained  $J_1$  and  $J'_2$ , we can estimate the contributions of the exchange interactions to the heat capacity above  $T_N$ . From the high-temperature expansion<sup>[6]</sup> we have

$$\frac{C_{exch}}{R} = \frac{2S^2(S+1)^2}{3T^2} (z_1 J_1^2 + z_2 J'_2),$$

where  $z'_2 = 22$ . From this we obtain for MnAlG the value  $CT^2/R = 80$  °K<sup>2</sup>. This is 25% higher than the coefficient  $a$  in (2). Taking into account the accuracy with which the lattice contribution is determined at  $T \gg T_N$ , as well as the averaging used in the determination of the exchange parameters, this agreement can be regarded as satisfactory.

In conclusion, the authors take the opportunity to thank K. P. Belov for interest in the work, B. V. Mill' for preparing the sample, and Yu. F. Popov for the measurements in the pulsed magnetic fields.

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Translated by J. G. Adashko