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Features of antiferromagnetic ordering in the garnet $Mn_3Cr_2Ge_3O_{12}$

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The specific heat and the magnetic properties are investigated in the garnet $Mn_3Cr_2Ge_3O_{12}$, in which the weakly interacting *a* and *c* sublattices are completely filled by magnetic ions. Independent antiferromagnetic ordering of the manganese and chromium sublattices is observed at temperatures 3.0 and 5.1 K, respectively. It is shown that the magnetic symmetry of this garnet is responsible for the peculiarities of its magnetic properties, in particular the nonlinear field dependence of the magnetization below T_N. A calculation carried out in the molecular-field approximation, with use of the exchange-interaction parameters of the corresponding "single-sublattice" garnets, gives good agreement with experimental data.

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1. INTRODUCTION

In compounds with the garnet structure, the magnetic ions may occupy three types of crystallographic sites (sublattices): dodecahedral $\{c\}$, octahedral $[a]$, and tetrahedral (d). The character of the magnetic ordering of garnets is determined to a significant degree by the relation between the magnitudes of the antiferromagnetic intra- and intersublattice exchange interactions: in iron garnets, the strong intersublattice a–d interaction suppresses the considerably weaker intrasublattice interactions and causes ferrimagnetism of these compounds below $T_c \approx 550$ K; by diamagnetic substitution, it is possible to weaken or even completely eliminate the a–d interaction, and then there is observed in the garnet, at sufficiently low temperatures ($T_N \sim 10$ K), antiferromagnetic ordering of the remaining magnetic sublattice. The properties of such antiferromagnetic or "single-sublattice" garnets, in which the magnetic $3d$ or $4f$ ions completely occupy one of the three types of crystallographic sites, have been investigated quite thoroughly (see, for example, the review by Belov and Sokolov[1]).

In order to obtain information about the complex mechanism of indirect exchange interactions, which occur in garnets through one, two, or even three intermediate links (two oxygen ions and a nonmagnetic cation), it is of interest to study magnetic phase transitions in garnets in which the intra- and intersublattice exchange interactions have approximately equal magnitudes. Such a situation occurs, for example, in yttrium iron garnets with a sufficiently high concentration of diamagnetic ions in the a and d sublattices. In a paper of Plakhtiĭ *et al.*[2] it was shown that in such substituted garnets, the transition from ferri- to antiferromagnetism occurs over a quite appreciable range of concentrations with a cluster type of magnetic ordering.

Another variant of systems that have comparable values of the intra- and intersublattice exchange interactions is garnets in which the weakly interacting c and a sublattices are *completely* filled by magnetic ions. The features of the magnetic ordering of one of these compouds, $\{Mn_3\}[Fe_2]Ge_3O_{12}$, have already been discussed.[3,4] The present paper reports the results of measurements of the specific heat and magnetic properties of the garnet $\{Mn_3\}[Cr_2]Ge_3O_{12}$ (MnCrG), in which the antiferromagnetic ordering has an unusual character.[5,6] We shall also consider a simple model, which enables us to give a qualitative interpretation of the

TABLE I. Lattice parameters (a_0), Néel temperatures (T_N), paramagnetic Curie points (Θ_P), values of the susceptibility (χ) at $T \leq T_N$, spin-flip fields (H_E), and exchange-interaction integrals for first and second nearest neighbors (J_1, J_2), for single-sublattice chromium and manganese garnets.

Garnet	a_0, Å	T_N, K	$-\Theta_P$, K	χ, cgs emu/mol	$H_E(T)$, kOe	$-J_1$, K	$-J_2$, K
CdCrG	12.205	12.55	21.5	0.11	260 (4.2)	0.81	0.28
MnAlG	11.894	6.65	28	0.38	210 (3.0)	0.58	0.12

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magnetic properties of MnCrG on the basis of the available experimental data for the corresponding single-sublattice garnets $\{Mn_3\}Al_2Ge_3O_{12}$ (MnAlG) and $Cd_3[Cr_2]Ge_3O_{12}$ (CdCrG). The basic characteristics of these compounds[7,8] are given in Table I.

2. METHOD OF MEASUREMENT

An x-ray single-phase specimen of MnCrG (a_0 = 12.026 Å) was prepared by B. V. Mill' in the Problem-Solving Laboratory of Magnetism of Moscow State University, by a ceramic technology, with a double anneal in air at T = 1160°C.

The specific heat was measured over the interval 2–14 K on apparatus described earlier.[9] Below 3.5 K some peculiarities were observed in the calorimetric behavior of MnCrG: the temperature behavior after a period of heating had on the whole a nonlinear character, indicating considerable overheating of the thermometer with respect to the specimen and a long (~30 min) time for establishment of equilibrium in the calorimeter + specimen system. In our view, this was due to the large value of the specific heat of MnCrG in the interval 2–3.5 K and to intense absorption of the heat-exchange helium in the calorimeter, caused by the considerable porosity of the specimen. Under such conditions, the measured value of the specific heat depends on the time interval between successive heating periods and on the method of analyzing the temperature variations in order to determine the change of temperature resulting from the heating. Apparently these reasons are responsible for some difference between our data and the temperature variation of the specific heat of MnCrG obtained by Belov *et al.*[10] We measured the temperature variation after a heat pulse until such time (40–50 min) as the temperature drift (nearly linear) was the same as before the heating, and then made a linear extrapolation of the temperature variations to the middle of the heating period.

For $T \gtrsim 3.5$ K, the temperature behavior before and after heating formed two parallel straight lines. The error in determination of the specific heat at these temperatures did not exceed 3%. The magnetic susceptibility of MnCrG over the interval 1.6–100 K was determined from magnetization isotherms, which were measured by the induction method in a superconducting solenoid, on specimens of cylindrical form from the same production batch as for the specific heat.

3. EXPERIMENTAL RESULTS

Figure 1 shows the results of measurement of the specific heat of MnCrG. The lattice specific heat (dotted line) was calculated from the specific heat of the diamagnetic garnet $Cd_3Ga_2Ge_3O_{12}$.[7] It is seen that $C(T)$ has two maxima; this indicates independent ordering of the a and c sublattices in MnCrG. Because the exchange interaction of Cr^{3+} ions on a sites is about one and a half times as large as the interaction of Mn^{2+} ions on c sites (see Table I), and also in view of the values of the spins of Mn^{2+} and Cr^{3+} (for $S_c = 5/2$, a λ anomaly of greater magnitude should be observed in the magnetic specific heat), it is natural to suppose that the maximum at 5.1 K corresponds to antiferromagnetic ordering of Cr^{3+} on the a sublattice, and that at 3 K ordering of Mn^{2+} on the c sublattice occurs.

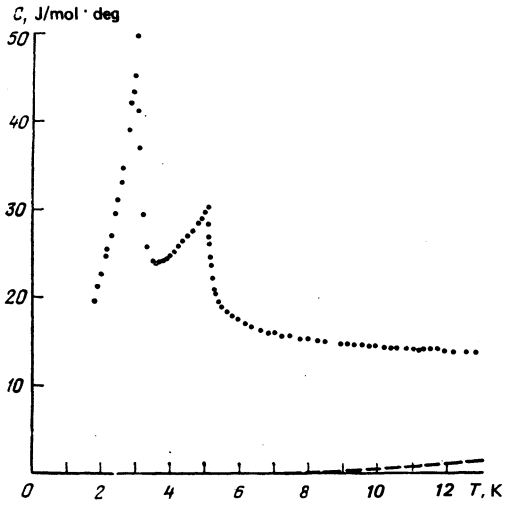

FIG. 1. Temperature dependence of the specific heat of MnCrG (points) and lattice contribution to the specific heat (dotted curve).

This agrees with neutron-diffraction investigations made by Plakhtiĭ and coworkers on the same specimen of MnCrG that was used for the specific heat.[6] According to neutron-diffraction data, the magnetic structures of the a and c sublattices below the Néel points, which are 5.1 and 3.9 K respectively, remain the same as in single-sublattice CaCrG and MnAlG.[11,12] The difference in T_N for the c sublattice as determined by the calorimetric and neutron diffraction methods is apparently due to the different thermometry of these experiments.

Figure 2 shows the temperature variation of the inverse molar susceptibility, $\chi^{-1}(T)$, of MnCrG. For $T > 5$ K, $\chi^{-1}(T)$ differs from both the Néel and the Weiss types, and the variation is of somewhat different character for $60 < T < 100$ K and for $5 < T < 60$ K: above 60 K, $\chi^{-1}(T)$ is close to the Néel type, i.e., the susceptibility increases with lowering of temperature faster than according to the Curie-Weiss law; below 60 K, this in-

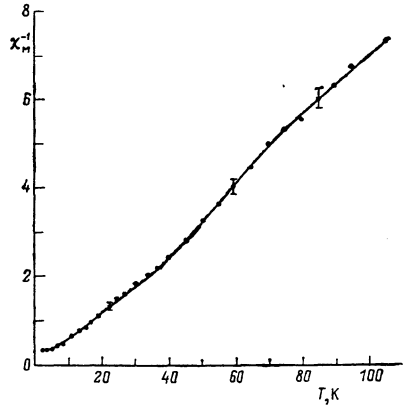

FIG. 2. Temperature dependence of the inverse molar susceptibility of MnCrG.

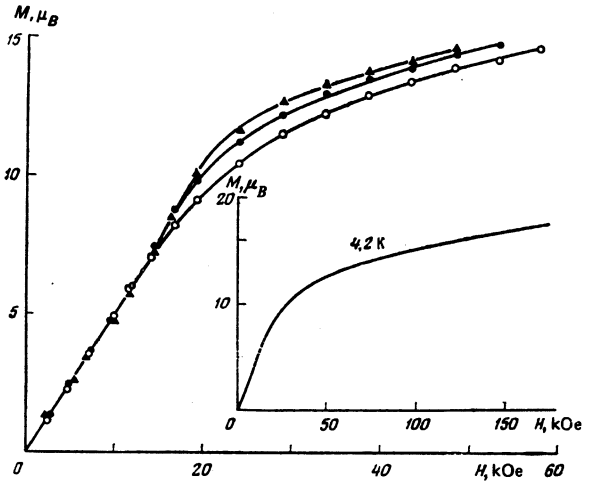

FIG. 3. Magnetization isotherms of MnCrG in fields of a superconducting solenoid: ○, at 4.2 K; ●, at 3.0 K; ▲, at 1.6 K; inset, magnetization of MnCrG at 4.2 K in pulsed magnetic fields.

crease slows down. For $T<5$ K, antiferromagnetic ordering occurs, and χ is independent of temperature, as in antiferromagnetic single-sublattice garnets.[7,8] But the value of the susceptibility of MnCrG below T_N ($\chi = 2.75$ cgs emu/mol) exceeds by a factor of more than five the sum of the susceptibilities of MnAlG and of CdCrG in the antiferromagnetic range (see Table I).

Figure 3 shows isotherms of the magnetism of MnCrG at helium temperatures, measured in static and in pulsed (inset) magnetic fields.[1)] It is seen that the $M(H)$ relation is already nonlinear at fields ~15 kOe, although according to the data given in Table I the values of H_E of the corresponding single-sublattice garnets exceed 200 kOe. In magnetic fields $H \gtrsim 100$ kOe (at 4.2 K), there is a linear increase of the magnetization of MnCrG with slope $\chi = 0.2$ cgs emu/mol. Within the limits of sensitivity of the apparatus, we detected no residual magnetic moment in MnCrG at $H = 0$; that is, this garnet is not ferrimagnetic, as was conjectured by Belov *et al.*[10] on the basis of the nonlinear $M(H)$ relation.

4. DISCUSSION OF RESULTS

According to neutron-diffraction data,[6] the magnetic structure of Cr^{3+} on the octahedral sites of MnCrG forms two ferromagnetic sublattices, one inserted antiferromagnetically within the other; in the twelve-sublattice traiangular magnetic structure of Mn^{2+} on the dodecahedral sites of MnCrG, the magnetic moments lie in the (111) plane and are directed along or opposite to one of the three crystallographic axes $[2\bar{1}\bar{1}], [\bar{1}2\bar{1}], [\bar{1}\bar{1}2]$. The feature of the magnetic structure of MnCrG that obviously is responsible for the independent antiferromagnetic ordering of the a and c sublattices is that in the absence of an external magnetic field, its energy is independent of intersublattice exchange interaction, even when account is taken of interactions in all coordinate spheres.

In fact, the magnetic symmetry of MnCrG is such that each Mn^{2+} spin interacts with an equal number of oppositely directed Cr^{3+} spins; therefore the effective field H_{ca} exerted on an arbitrarily chosen c ion by the a sublattice is zero:

$$\mathbf{H}_{ca}=\gamma_{ca}(\mathbf{M}_{a1}+\mathbf{M}_{a2})=0. \quad (1)$$

Here $\mathbf{M}_{a1}$ and $\mathbf{M}_{a2}$ are the oppositely directed magnetic moments of the Cr^{3+} sublattices, and γ is the exchange-interaction parameter:

$$\gamma_{ij}=2z_iJ_{ij}/x_iNg_ig_j\mu_B^2, \quad i, j=c, a, \quad (2)$$

where J_{ij} is the exchange-interaction integral, z_{ij} is the number of neighbors, N is Avogadro's number, $x_a = 2, x_c = 3, g_{i,j} = 2$ is the spectroscopic splitting factor, and μ_B is the Bohr magneton. Consequently the intersublattice exchange interaction shows up only in an external magnetic field, when $\mathbf{M}_{a1}$ and $\mathbf{M}_{a2}$ cease to compensate each other. An analogous situation will exist also for the effective field $\mathbf{H}_{ac}$.

On the assumption of such a model, we shall find the susceptibility of MnCrG on the linear section of the $M(H)$ curve at $T = 0$, supposing that the value of the magnetic field exceeds the values of the spin-flop fields for both sublattices. The arrangement of magnetic moments of the a and c sublattices ($\mathbf{M}_{am}$ and $\mathbf{M}_{cn}$ respectively; $m = 1, 2$; $n = 1, \ldots, 12$) with respect to H is represented schematically in Fig. 4a. For the c sublattice, the moments are given only for four of the twelve sublattices in the MnCrG structure (the numbering of the sublattices correspond to that adopted in our earlier paper[8]); the moments of the remaining sublattices can be obtained by rotation about the field direction through ±120°.

The magnetization of MnCrG can be written in the form

$$M(H)=\sum_{m=1}^{2}M_{am}\cos\theta_a+\sum_{n=1}^{12}M_{cn}\cos\theta_c=M_a\cos\theta_a+M_c\cos\theta_c. \quad (3)$$

Here θ_a and θ_c are the angles that $\mathbf{M}_{am}$ and $\mathbf{M}_{cn}$ make with H (Fig. 4a); $M_a = 2Ng\mu_BS_a$, $M_c = 3Ng\mu_BS_c$. To determine the equilibrium values of θ_a and θ_c, we write the energy of MnCrG in an external magnetic field, omitting terms that are independent of the orientation of the magnetic moments:

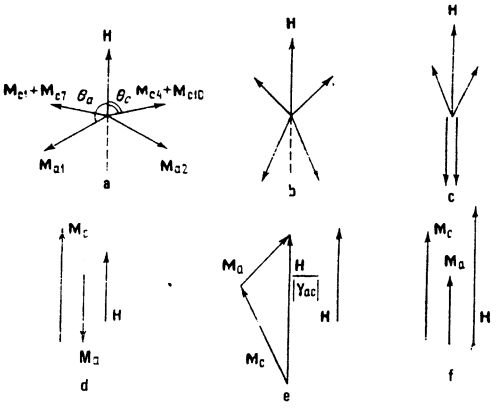

FIG. 4. Sketch of the change of magnetic structure of MnCrG under the influence of an external magnetic field.

$$E(\mathrm{H}) = -H\sum_{i=a,c} M_i \cos\theta_i + \sum_{\substack{i<j \\ i,j=a,c}} E_{\mathrm{exc}}^{ij}(\mathrm{H}), \quad (4)$$

where $E_{\mathrm{exc}}^{ij}(\mathrm{H})$ are the contributions of the exchange energy. On the basis of the magnetic structure of the a sublattice,[13]

$$E_{\mathrm{exc}}^{aa}(\mathrm{H}) = -{}^1/_2\gamma_{aa}M_a^2\cos 2\theta_a. \quad (5)$$

The parameter γ_{aa} is determined by the relation (2) and (in the absence of intersublattice interaction) is connected with the value of the susceptibility of the a sublattice by the relation[14]

$$-1/2\gamma_{aa} = \chi_a(T_N^a) = \chi_a. \quad (6)$$

Thus

$$E_{\mathrm{exc}}^{aa}(\mathrm{H}) = \frac{M_a^2}{4\chi_a}\cos 2\theta_a \quad (7)$$

and analogously for the c sublattice

$$E_{\mathrm{exc}}^{cc}(\mathrm{H}) = \frac{M_c^2}{4\chi_c}\cos 2\theta_c, \quad (8)$$

where χ_c is the susceptibility of the c sublattice at $T \leq T_N^c$ in the absence of a–c interaction.

By use of (1), $E_{\mathrm{exc}}^{ac}(\mathrm{H})$ can be written

$$E_{\mathrm{exc}}^{ac}(\mathrm{H}) = -\mathrm{H}_{ca}\sum_{n=1}^{12}\mathrm{M}_{cn} = -\gamma_{ca}\sum_{m=1}^{2}\mathrm{M}_{am}\sum_{n=1}^{12}\mathrm{M}_{cn} = -\gamma_{ca}M_aM_c\cos\theta_a\cos\theta_c. \quad (9)$$

On substituting (7)–(9) in (4) and minimizing the resulting energy expression with respect to the angles θ_a and θ_c, we get

$$\cos\theta_i = \frac{\chi_i + \gamma_{ca}\chi_a\chi_c}{(1-\gamma_{ca}^2\chi_a\chi_c)M_i}H = k_iH \quad (10)$$

($i = a, c$), and consequently for the magnetization (3)

$$M(H) = \frac{\chi_a+\chi_c+2\gamma_{ca}\chi_a\chi_c}{1-\gamma_{ca}^2\chi_a\chi_c}H = \chi H, \quad (11)$$

that is, the magnetization of MnCrG, at not too large fields ($|k_iH| \leq 1$), actually varies linearly with the field.

On substituting in (11) the experimental values of χ for MnCrG at $H < 15$ kOe and $1.6 \leq T \leq 4.2$ and of χ_a and χ_c for CdCrG and MnAlG (see Table I), we get two solutions for γ_{ca}: a ferromagnetic with $\gamma_{ca}^+ = 4.1$ cgs emu/mol and a ferrimagnetic with $\gamma_{ca}^- = -4.8$ cgs emu/mol. The ferromagnetic solution contradicts the experimental $M(H)$ relations at helium temperatures (Fig. 3), since in this case the magnetization of MnCrG should already at $H \approx 40$ kOe amount to $21\mu_B$ (the sum of the magnetizations of the a and c sublattices), whereas actually at 1.6 K $M(40\ \mathrm{kOe}) \approx 14\ \mu_B$, and at 4.2 K the magnetization in fields ~200 kOe is $M = 17\ \mu_B$. Thus the $\{Mn^{2+}\}$–$[Cr^{3+}]$ exchange interaction in MnCrG is negative and is described by the value $J_{ac} = -1.8$ K (relation (2)); that is, it is not negligibly small in comparison with the intrasublattice interaction, as might be supposed on the basis of the calorimetric data.

For the γ_{ca}^- found, we have $k_a < 0$ and $k_c > 0$; that is, with increase of the magnetic field the magnetization of the c sublattice increases in the direction of the field (the angle θ_c decreases), the magnetization of the a sublattice in the direction opposite to the field (the angle θ_a increases) (Fig. 4b). Consequently an external magnetic field switches on, as it were, the intersublattice exchange interaction, which tends to establish a ferrimagnetic arrangement of the moments of the a and c sublattices. Calculation shows that in fields of 15–20 kOe first the a sublattice flips ($k_aH = -1$) and then the c sublattice ($k_cH = 1$) (Fig. 4c, d); this causes the experimentally observed deviation of the $M(H)$ relation from linearity (Fig. 3). It may be assumed that on further increase of the field, the same magnetic-sublattice flip effects occur as are observed in iron garnets (Fig. 4 e, f). In the noncollinear phase, according to Clark and Callen,[15] the resultant moment will be oriented along the field:

$$\mathrm{M} = \mathrm{M}_a + \mathrm{M}_c = \mathrm{H}/|\gamma_{ac}| \approx 0.2H \ \text{(cgs emu/mol)}.$$

The estimate thus obtained for the susceptibility of MnCrG in a strong magnetic field agrees with the experimental value of χ at 4.2 K for $H \gtrsim 100$ kOe.

Analysis shows that the model considered above persists also at 4.2 K (when the c sublattice is in the paramagnetic state) and consequently enables us to explain qualitatively the experimentally observed features of the magnetic properties of MnCrG. As regards the quantitative agreement of the calculated and experimental values, that is apparently fortuitous, since the χ_a and χ_b used in the calculation were those for the corresponding single-sublattice garnets; that is, it was assumed that the magnitudes of the intrasublattice exchange interactions do not change on going from CdCrG and MnAlG to MnCrG. But according to calorimetric data, antiferromagnetic ordering of the a and c sublattices occurs in MnCrG at much lower temperatures than in CdCrG and MnAlG. From the point of view of molecular field theory, this may be due solely to decrease (by a factor of about two) of the magnitudes of the intrasublattice exchange interactions, and unrelated to the existence in MnCrG of an intersublattice exchange interaction, since the exchange energy of this garnet is independent of γ_{ca}. But such a large change of the exchange parameters seems improbable, since the geometry of the exchange bonds changes imperceptibly on going from CdCrG and MnAlG (in particular, according to calculations, the changes of the Mn-O distance and of the Mn-O-Mn angle do not exceed 1%). In order to establish the mechanism that leads to lowering of the magnetic ordering temperatures in MnCrG as compared with the corresponding single-sublattice garnets, it is apparently necessary to use models more accurate than the molecular field method, which take account of the fluctuations of the magnetic moments.

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Temperature dependence of magnetic hyperfine fields in the metallic ferromagnets Rh_3FeSn and Rh_2CoSn

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Mössbauer γ spectroscopy is used to measure the temperature dependences of the magnetic hyperfine fields for ^{57}Fe and ^{119}Sn in the ordered metallic ferromagnets Rh_2FeSn and Rh_2CoSn. It is found that for Rh_2FeSn in the range $0.4 < T/T_c < 0.8$ the temperature dependences of the normalized fields $h(T) = H(T)/H(77\ K)$ are substantially different for the Fe and the Sn. To explain this temperature anomaly of the hyperfine field, a model is proposed in which account is taken of the temperature dependences of the local magnetization of the Fe and Rh atoms. The form of the temperature anomaly $h(T)$ for ^{119}Sn can be fully explained if account is taken of the fact that near T_c the temperature dependences of the magnetic moments of the Fe and Rh atoms are the same, but in the $0.4 < T/T_c < 0.8$ they are substantially different.

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1. INTRODUCTION

Among the unsolved problems of the theory of magnetic hyperfine interaction in metallic ferromagnets, a special role is played by the interpretation of the temperature dependence of the magnetic hyperfine fields. The great attention paid to this problem is due primarily to the attempts to obtain objective data on the nature of the hyperfine fields by explaining their temperature dependence. In particular, for nonmagnetic atoms (i.e., for atoms with zero intrinsic magnetic moments), the form of the temperature dependence of the hyperfine field may depend on the peculiarities of the mechanisms whereby the spin density is transferred from the magnetic environment to the nonmagnetic atom.

In a number of cases, nonmagnetic atoms reveal temperature anomalies of the hyperfine field, i.e., substantial deviations of the normalized hyperfine field $h(T) = H(T)/H(0)$ on the normalized magnetization of the system $M(T)/M(0)$. Particularly strong anomalies were observed for Sn impurity atoms in ferromagnetic metallic matrices. It is well known, for example, that Sn has a large anomaly in Fe (Refs. 1–3) and a giant anomaly in Co (Refs. 4 and 5). The reason why the anomalies are large precisely for the Sn atoms is qualitatively understandable. In this case, two large but oppositely directed contributions to the hyperfine field almost cancel each other [6] and therefore the absolute value of H should be very sensitive to small changes of the ratio of these contributions. Up to now, however, no unequivocal reason had been found for the temperature dependence of the relation between the two contributions. Several variants of an explanation of the anomalous behavior of $H(T)$ have been proposed for nonmagnetic atoms in metallic ferromagnets.[1-5,7] A common shortcoming of these explanations is that the initial premises are not very well founded and that the models cannot be quantitatively verified objectively.

As a rule, the experimental data are analyzed without allowance of the dependence of the hyperfine field on the thermal expansion of the crystal lattice; it was assumed that the corresponding corrections are negligible. Recently Möller[8] and Nikolaev *et al.*[9] obtained data on the dependence of H on the external pressure in the case of Sn. It follows from these results that in a number of cases the hyperfine field for Sn depends very strongly on the distance between the atoms. This means that the thermal expansion of the lattice can exert a strong in-

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