Antiferromagnetic garnets with fluctuationally interacting sublattices

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Magnets are considered in which it is possible to separate two antiferromagnetic sublattices so placed relative to each other that the molecular field of the spins of the first sublattices at the second, and vice versa, is zero. In an analysis beyond the framework of the molecular-field theory, when fluctuations are taken into account, an exchange interaction appears between the spins of one sublattice via the other and couples the sublattices. Effects that can result from this interaction are considered. Experimental data are discussed on the garnet $Mn_3Cr_2Ge_3O_{12}$, which is a substance of the type considered.

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1. INTRODUCTION

The magnetic ordering of the manganese spins (c sublattice) and the chromium spins (a sublattice) in the garnet $Mn_3Cr_2Ge_3O_{12}$ is antiferromagnetic.[1,2] The symmetry relative to the placements of the a and c sublattices of the garnet is such that the molecular field produced by the manganese at the chromium, and vice versa, is zero. Therefore, notwithstanding the fact that the exchange interaction between Mn and Cr is not small,[2] the manganese and chromium sublattices are not coupled in the molecular-field approximation, and the interaction between them does not influence the magnetic properties of the substance. Neutron-diffraction data show that the magnetic structures of the sublattices are in fact the same as in the single-sublattice garnets $Mn_3Al_2Ge_3O_{12}$ and $Cd_3Cr_2Ge_3O_{12}$; both calorimetric and neutron-diffraction data show the presence of two distinctly pronounced phase transitions at $T_{1N}=5.1$ K(Cr) and $T_{2N}=3.9$ K (Mn) (Refs. 1 and 2).

There are, however, certain data that contradict the undistorted-sublattice picture. Thus, for example, T_{1N} and t_{2N} are approximately half the Néel temperatures of the single-sublattice garnets. Since the intrasublattice interactions changes extremely little when magnetic atoms are replaced by nonmagnetic ones in the second sublattice,[2] it is clear that the intersublattice interaction is responsible for the lowering of the Néel temperature.

The above garnets are not the only example of antiferromagnetism with sublattices in a zero molecular field. This situation obtains on all so-called antiferromagnets of the second kind. In this case the sublattice contain the same atoms. Our analysis pertains also to this case.

The interaction between sublattices comes into play in the theory once we go outside the framework of the molecular-field theory, when account is taken of the fluctuations of the sublattice magnetization. The atoms of one sublattice can interact with one another not only directly (we define as direct the interaction via the oxygen ions), but also indirectly, by polarizing the second sublattices (this indirect exchange was first considered for magnetic impurities in the interstices of a ferromagnet in Ref. 3). It turns out that this interaction can become significant. It causes, in particular, the system energy to depend on the relative directions of the sublattice antiferromagnetism vectors even in the pure exchange approximation, i.e., a magnetic anisotropy of sorts appears; the Néel temperatures become lower than in single-sublattice substances. In addition, when certain relations are satisfied between the parameters, a specific unique magnetic structure with noncollinear ferromagnetic ordering can arise if certain relations between the parameters are satisfied. The main results and the possible methods for their experimental verification are given in the Appendix.

Neutron-diffraction data show that the chromium spins are collinearly arranged, and those of manganese in a more complicated form. Unless otherwise stipulated, the actual type of the magnetic structure is of no importance to us and we assume the simplest model with collinear ordering in both sublattices. The Hamiltonian of the system is

$$\begin{gathered}\mathscr{H}=\mathscr{H}_1+\mathscr{H}_2+\mathscr{H}_{int},\\ \mathscr{H}_1=\sum_{i,j} V_{ij}(\mathbf{S}_{fi}\mathbf{S}_{gj}),\quad \mathscr{H}_2=\sum_{i,j} U_{ij}(\mathbf{I}_{fi}\mathbf{I}_{gj}),\\ \mathscr{H}_{int}=\sum_{i,j} J_{ij}(\mathbf{S}_i\mathbf{I}_j).\end{gathered}\tag{1}$$

Here $\mathbf{S}_i$ and $\mathbf{I}_i$ are the spins of the first and second sublattices at the sites $\mathbf{r}_i$. The subscripts f and g mark the upward and downward spin directions in the ground state. As seen from (1), we neglect the intrasublattice interaction of the spins having the same direction in the ground state. This simplifies the equations without changing anything qualitatively. Naturally, $V_{ij}>0$ and $U_{ij}>0$; in garnets usually $J_{ij}>0$. The atoms of both sublattices are assumed located in sites of primitive cubic lattices displaced relative to one another by the vector $(a/2, a/2, a/2)$, where a is the lattice constant. This model describes quite accurately the body-centered antiferromagnet of second kind $Ca_3Fe_2Ge_3O_{12}$ and describes qualitatively more complicated structures.

If J were larger than V and U, the situation would be that typical of iron garnets: the intersublattice interaction would suppress the intrasublattice interaction and a collinear ferrimagnetic order would set in (the magnetic atoms in iron garnets are usually in a and d sublattices, and the a–d interaction is several times stron-

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ger than the remaining ones). This means that we must assume that $J<J_0 \sim \max(V, U)$. It should be emphasized that this is the only restriction we impose on the exchange-interaction parameters. It is therefore not excluded that garnets of this type can be obtained only using manganese and chromium. It is also possible that symmetrically placed sublattices are contained not only in garnet structure. Therefore, assuming the condition $J<J_0$ satisfied, we consider various possible relations between the parameters of the Hamiltonian.

2. WEAK INTERACTION IN ONE OF THE SUBLATTICES

We consider first the case when the direct interaction in one of the sublattices (to be definite, the second) can be neglected; an exact criterion will be indicated below. At $T \ll T_{1N}$ the spins of the second sublattices therefore interact and polarize the first. i.e., via exchange of virtual spin waves. The effective Hamiltonian of the second sublattice can be obtained in the usual manner in second order perturbation theory in $J/V \ll 1$, by averaging over the state of the first sublattice. We obtain

$$\mathscr{H}_{\text{eff}}=-\sum_{\mathbf{q},i,j,\alpha,\beta} J^2(\mathbf{q}) K^{\alpha\beta}(0,\mathbf{q}) \exp\{i\mathbf{q}(\mathbf{r}_i-\mathbf{r}_j)\} I_i^\alpha I_j^\beta. \tag{2}$$

Here $K^{\alpha,\beta}(0, \mathbf{q})$ is the static correlator of the components α and β of the spins of the first sublattice, and $J(\mathbf{q})$ is the Fourier transform of J_{ij}.

At $T \leqslant T_{1N}$ we can neglect $K^{zz}(0, \mathbf{q})$ and, leaving out an inessential constant in $\mathscr{H}_{\text{eff}}$, we have

$$\mathscr{H}_{\text{eff}}=\sum_{\mathbf{q},i} J^2(\mathbf{q}) K(0,\mathbf{q}) (I_i^z)^2 - \sum_{\mathbf{q},i,j\neq i} J^2(\mathbf{q}) K(0,\mathbf{q}) \exp\{i\mathbf{q}(\mathbf{r}_i-\mathbf{r}_j)\} (I_i^x I_j^x + I_i^y I_j^y), \tag{3}$$

the static correlator of the transverse spin component being (this expression can be easily obtained, for example, from the equations of Ref. 4)

$$K(0, \mathbf{q})=2S^2[V(0)-V(\mathbf{q})]/\Omega_1^2(\mathbf{q}), \quad \Omega_1^2=S^2[V^2(0)-V^2(\mathbf{q})]. \tag{4}$$

The first term of (3) (the effective single-ion anisotropy) was first obtained by Ivanov,[5] and the spin interaction was obtained in Ref. 3, where the effects of this interaction on impurities in metamagnets were considered. We note that the equations of Ref. 3 for the effective potential are outwardly different from (3), but can be recast in this form. The Hamiltonian (3) was also obtained in a recent paper.[6]

The magnetic structure of the second sublattice is determined by the value of the vector q, for which the effective potential $V_{\text{eff}}(\mathbf{q})=J^2(\mathbf{q})K(0,\mathbf{q})$ is a maximum. It is easy to verify that for the lattice considered by us this value is zero, i.e., a collinear ferromagnetic ordering in the xy plane is established in the second sublattice. The resultant molecular field magnetizes the first sublattice, whose average spin is

$$\langle S\rangle=IJ(0)/V(0). \tag{5}$$

Thus, at the temperature

$$T_{2N}={}^1/_3 I(I+1)J^2(0)K(0,0)$$

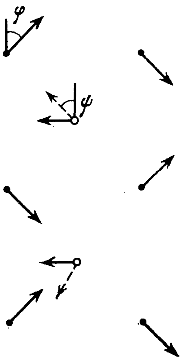

FIG. 1. Magnetic structure for a square lattice at $U<U_{\text{cr}}$. The spins I are at the sites marked by light circles. The solid and dashed arrows show their orientations at $U=0$ and at $J_0>J(0)>(U(0)V(0))^{1/2}$ respectively.

a ferrimagnetic order of sort is produced (since $J>0$). The corresponding magnetic structure for a square lattice is shown in Fig. 1.

This phenomenon can be qualitative understood as follows. If the spins of the first sublattice are deflected by an angle φ from the z axis and a molecular field at the spin I_i is produced, then the change in energy is

$$\Delta E=A_1 V\varphi^2-A_2 J\varphi \tag{6}$$

A_1 and A_2 are constants of the order of unity).

The maximum energy gain $\Delta E_{\max} \sim J^2/V$ is reached at $\varphi \sim J/V$. Since the azimuthal angle is arbitrary here, such a "polaron" can rotate freely about the z axis, meaning anisotropy of the "easy plane" type. The overlap of the polarized clouds leads to indirect interaction and, as shown above, to ferromagnetism of the spins I. The dependence of the angle φ and of the angular momentum on the temperature is given by the usual formulas of the molecular-field theory.

The conclusion that the ferromagnetic ordering in the second sublattice is ordered is not connected, in our opinion, with the concrete type of lattice. The point is that this is the only type of ordering for which the rotations of the spins of the first sublattice are such that the molecular fields produced at each of its spins I_i and their neighbors from the first sublattice are additive (see Fig. 1). Naturally, the energy gain is then a maximum.

It is concluded in recent papers[7,8] that if the interaction between the sublattices is not of the Heinsenberg type but is anisotropic in the spin space $-J_{ij}^{\alpha\beta} I_i^\alpha S_j^\beta$, antiferromagnetic ordering is produced in the second sublattice and the interaction energy is very large, $V_{\text{eff}} \sim (\delta J)^2/d$ (d is the magnetic anisotropy in the first sublattice, δJ is the interaction anisotropy). We cannot agree with this conclusion. It is clear that no indirect exchange can yield a value larger than J, i.e., than the value obtained if all the neighbors of the spins I_i and I_j were to magnetize them in the same direction. Yet the cited V_{eff} of Refs. 7 and 8 yields a value larger than any constant of the Hamiltonian (1). Divergence of V_{eff} as $d \to 0$ is obtained in Refs. 7 and 8 because of the divergence of $K(0, \mathbf{q})$ at $\mathbf{q}=\mathbf{q}_0=(\pi/a, \pi/a, \pi/a)$ and as $d \to 0$. But an infinite correlator means a Hamiltonian that is invariant to homogeneous rotations of the spins of the first sublattices, so that this sublattice is not polarized and there is therefore no indirect exchange. This manifests itself formally in the fact that $J^2(\mathbf{q}_0)=0$, and therefore $V_{\mathrm{eff}}(\mathbf{q}_0)$ is likewise zero. An attempt was made in Refs. 7 and 8 to get around this fact by assuming that the molecular field

$$\sum_j J_{ij}^{\alpha z}\langle S_j^z\rangle=J^{\alpha z}(\mathbf{q}_0)\cdot S$$

is zero (the spins S are directed along the z axis), and the quantities $J^{\alpha x}(\mathbf{q}_0)$ and $J^{\alpha y}(\mathbf{q}_0)$ differ from zero, and it is this which makes the quantity analogous to $J^2(\mathbf{q}_0)$ different from zero. But these assumptions are contradictory, the spins S cannot be directed along the z axis; they will be directed along x or y, for in this case the produce a molecular field at the spins I, and the energy of this state is therefore lower. Naturally, the spins I have here a ferromagnetic order. Thus, the divergence of V_{eff} as $d \to 0$ means an incorrect choice of the ground state in Refs. 7 and 8, and not the presence of an anomalously strong interaction.

The spin-wave spectrum can be obtained with the aid of the classical equations for the oscillations of the magnetization. Leaving out the standard calculations, we present the final answer:

$$\omega_1{}^2=J^2(0)I^2+S^2(V^2(0)-V^2(\mathbf{q})),$$
$$\omega_2{}^2(\mathbf{q})=\frac{J^2(0)}{2V^4(0)}(V(0)-V(\mathbf{q}))[J^2(0)(V(0)+V(\mathbf{q}))-2J^2(\mathbf{q})V(0)], \tag{7}$$

where $\omega_1(\mathbf{q})$ differs from the frequency $\Omega_1(\mathbf{q})$ of the oscillations in a single-sublattice magnet by the presence of the gap $\omega_1(0)=J(0)I$. This is understandable: in the presence of a field H there appears in the spectrum of the antiferromagnet a gap proportional to $(HV(0))^{1/2}$, and since the first sublattice is acted upon by the molecular field $H \sim J^2(0)/V(0)$ produced by the second sublattice, it is quite understandable that $\omega_1(0) \sim J(0)$.

The second branch is activationless, with a quadratic dispersion law at small momenta. This branch is due to oscillations of the spontaneous moment, and therefore the maximum energy $\omega_{\max}$ of these excitations turns out to be of the order of the indirect-exchange energy $J^2(0)/V(0)$ which causes the appearance of the spontaneous moment.

We note that there are no states in the energy interval from $\omega_1(0)$ to $\omega_{\max}$.

We have neglected in this section the direct exchange in the second sublattices. It is now clear that this is permissible if the direct exchange is smaller than the indirect one:

$$U(0) \ll J^2(0)/V(0). \tag{8}$$

In the next section we shall consider the situation when the energy U is of the order of or larger than the indirect-exchange energy.

3. FLUCTUATIONALLY INTERACTING ANTIFERROMAGNETIC SUBLATTICES

As already noted, in $Mn_3Cr_2Ge_3O_{12}$ both sublattices are antiferromagnetically ordered. This means that inequality (8) is not satisfied for this substance, the direct exchange is large enough, and we must consider its influence on the ground state of the magnet. Assume that the interaction of the sublattices distorted their magnetic structure weakly compared with the single-sublattice structure, so that a small moment was prodcued in each of them (Fig. 1 shows such a structure for a square lattice). If φ and ψ are the angles of deviation of the sublattice spins from the z axis, then the energy change per atom is

$$\Delta E=A_1V(0)\varphi^2+A_2U(0)\psi^2-A_3J(0)\varphi\psi \tag{9}$$

(A_1, A_2, A_3 are constants of the order of unity). It is seen from (9) that if $U(0)$ exceeds a certain critical value $U_{\mathrm{cr}} \sim J^2(0)/V(0)$, then $\Delta E>0$, no distortion takes place, and structures of single-sublattice magnets are realized. This is precisely the situation in $Mn_3Cr_2Ge_3O_{12}$. A simple calculation shows that

$$U_{cr}=J^2(0)/V(0). \tag{10}$$

If the intersublattice interaction lies in the interval $J_0>J(0)>(U(0)V(0))^{1/2}$, the structure shown schematically in Fig. 1 is produced, and at $J(0) \geqslant J_0$ we have, as already noted, collinear ferrimagnetism.

We consider hereafter in this section the case of undistorted structures, and for perturbation theory to be valid we assume that $J(0)/V(0)$, $J(0)/U(0) \ll 1$. In the molecular-field approximation the sublattices are not coupled and the spin orientations in them are arbitrary. However, we shall show presently, the quantum corrections to the ground-state energy depend on the mutual orientations of the spins, and lead to the onset of a sort of magnetic anisotropy. These corrections are made necessary by the fact that the Néel ground state, as is well known, does not correspond to an exact solution of the Hamiltonian H_1 of (1).

We consider now the nonlinear-antiferromagnet model described in the Introduction. The angle between antiferromagnetism axes of the sublattices will be designated γ. For the system-energy correction necessitated by the interaction we have in second-order perturbation theory

$$E_{int}=-T\sum_{\substack{n,\mathbf{q}\\ \alpha,\beta}} J^2(\mathbf{q})K^{\alpha\beta}(\omega_n, \mathbf{q})Q^{\alpha\beta}(\omega_n, \mathbf{q}). \tag{11}$$

Here T is the temperature, and $K^{\alpha\beta}(\omega_n, \mathbf{q})$ and $Q^{\alpha\beta}(\omega_n, \mathbf{q})$ are the correlators of the spins S and I in the temperature diagram technique. For example,

$$K^{\alpha,\beta}(\omega_n, \mathbf{q})=\frac{1}{2}\int_{-1/T}^{1/T}\exp(i\omega_n t)\,dt\sum_{r_2}\langle T(S_{r_2}{}^{\alpha}(t)-\langle S_{r_2}{}^{\alpha}\rangle, S_{r_1}{}^{\beta}(0)-\langle S_{r_1}{}^{\beta}\rangle)\rangle$$
$$\times e^{i\mathbf{q}(\mathbf{r}_2-\mathbf{r}_1)},$$
$$\omega_n=2\pi nT. \tag{12}$$

The tensor $K^{\alpha\beta}$ (as well as $Q^{\alpha\beta}$) is diagonal if the z axis is directed along the sublattice ferromagnetism axis, with $K^{zz}=Q^{zz}=0$, and the transverse components are equal. We therefore have

$$E_{int}=-(1+\cos^2\gamma)T\sum_{n,\mathbf{q}} J^2(\mathbf{q})K(\omega_n,\mathbf{q})Q(\omega_n,\mathbf{q}). \tag{13}$$

Here $K(\omega_n, \mathrm{q})$ is the correlator of the components $S^{\pm}=(S^x \pm iS^y)/\sqrt{2}$, and $Q(\omega_n, \mathrm{q})$ is similarly defined.

At the low temperatures of interest to us $K(\omega_n, \mathrm{q})$ can be obtained from (4) by replacing $\Omega_1^2(\mathrm{q})$ with $\omega_n^2+\Omega_1^2(\mathrm{q})$. Since the correlators K and Q are positive it follows, as seen from (13), that the energy is a minimum if the antiferromagnetism axes are parallel. In this case both transverse spin components interact. On the other hand if the axes are perpendicular, the only interacting component is the one perpendicular to the plane containing the antiferromagnetism axes. It is this which leads to the dependence of the energy on the angle.

Summing over the frequencies in the usual manner, we obtain

$$\begin{gathered} E_{int}=-\Delta(1+\cos^2\gamma), \\ \Delta=2S^2I^2\sum_{\mathbf{q}} J^2(\mathbf{q})\frac{(V(0)-V(\mathbf{q}))(U(0)-U(\mathbf{q}))}{\Omega_1(\mathbf{q})\Omega_2(\mathbf{q})(\Omega_1(\mathbf{q})+\Omega_2(\mathbf{q}))}, \end{gathered} \tag{14}$$

where $\Omega_2^2(\mathrm{q})=I^2(U(0)-U(\mathrm{q}))$.

For the model with a primitive cubic lattice we have

$$\Delta=0.06\frac{J^2(0)SI}{V(0)S+U(0)I}. \tag{15}$$

In a real situation VS^2, UI^2, and JSI are quantities of the same order. As seen from (15), compared with these quantities Δ is a small quantity of the order of $1/S$ or $1/I$, and is natural for the quantum correction; Δ has also a numerical smallness connected with the integration over the momenta. The numerical coefficient depends on the type of lattice. For $Ca_3Fe_2Ge_3O_{12}$ we have $S=I=\frac{5}{2}$, and $J(0)S \sim V(0)S \sim 10$ K, so that Δ is of the order of several tenths of a degree. In other antiferromagnets of the second kind this quantity can be even larger.

The magnetic structure of the Mn sublattice in $Mn_3Cr_2Ge_3O_{12}$ is such that a threefold axis is present. If it is parallel to the z axis, the susceptibility tensor for this sublattice is diagonal, with $Q^{xx}=Q^{yy}=Q^{\perp}$. The properties of the susceptibility tensor of the Cr sublattice are the same as in the model considered above. Taking this into account we obtain

$$\begin{aligned} E_{int}=&-T\sum_{n,\mathbf{q}} J^2(\mathbf{q})K(\omega_n,\mathbf{q})(Q^{\perp}(\omega_n,\mathbf{q})+Q^{zz}(\omega_n,\mathbf{q})) \\ &-\cos^2\gamma\cdot T\sum_{n,\mathbf{q}} J^2(\mathbf{q})K(\omega_n,\mathbf{q})(Q^{\perp}(\omega_n\ \mathbf{q})-Q^{zz}(\omega_n,\mathbf{q})), \end{aligned} \tag{16}$$

where γ is the angle between the threefold axis and the antiferromagnetism axis of the chromium.

Depending on the sign of the sum in (16), the anisotropy is of the "easy axis" or "easy plane" type. To determine the sign of this sum we need a calculation that takes into account the rather complicated structure of the Mn sublattice (24 atoms in the unit cell, 12 sublattices). This question deserves a special treatment.

In $Mn_3Cr_2Ge_3O_{12}$ the value of Δ is of the order of several hundredths of a degree, i.e., it is equal to or somewhat larger than the one-ion anisotropy energy. Therefore the quantum effect considered by us should influence substantially the orientation of the antiferromagnetism vectors of the sublattices and make this orientation different from that in single-sublattice garnets. In garnets with large J the anisotropy can increase substantially, since it is proportional to J^2.

The anisotropy Δ should lead to a gap in that spin-wave spectrum branch which corresponds to relative oscillations of spins of different sublattices. To obtain the spin-wave dispersion law we use the diagram technique of Vaks, Larkin, and Pikin (Ref. 9).[1)] We denote the parts of $K(\omega_n, \mathrm{q})$ and $Q(\omega_n, \mathrm{q})$ that are irreducible in the interaction J by $K(\omega_n, \mathrm{q})$ and $Q(\omega_n, \mathrm{q})$. The relation between them is

$$K(\omega_n,\mathbf{q})=\frac{\tilde{K}(\omega_n,\mathbf{q})}{1-J^2(\mathbf{q})\tilde{K}(\omega_n,\mathbf{q})\tilde{Q}(\omega_n,\mathbf{q})}, \tag{17}$$

and the same holds for $Q(\omega_n, \mathrm{q})$. The spin-wave spectrum is given by the equation

$$1-J^2(\mathbf{q})\tilde{K}(\omega,\ \mathbf{q})\tilde{Q}(\omega,\ \mathbf{q})=0, \tag{18}$$

where $\tilde{K}(\omega, \mathrm{q})$ and $\tilde{Q}(\omega, \mathrm{q})$ are the analytic continuations of the correlation functions to real frequencies.

The expressions used above for $K(\omega_n, \mathrm{q})$ and $Q(\omega_n, \mathrm{q})$ agree with the zeroth approximation of the self-consistent-field method for $\tilde{K}$ and $\tilde{Q}$. Substituting them in (8) we obtain expressions for the frequencies in this approximation:

$$\begin{aligned} \omega_{1,2}^2=&{}^1/_2\{\Omega_1^2(\mathbf{q})+\Omega_2^2(\mathbf{q})+[(\Omega_1^2(\mathbf{q})-\Omega_2^2(\mathbf{q}))^2 \\ &+16S^2I^2J^2(\mathbf{q})(U(0)-U(\mathbf{q}))(V(0)-V(\mathbf{q}))]^{1/2}\}. \end{aligned} \tag{19}$$

At $T_{2N} \ll T \ll T_{1N}$ (this interval exists if $V \gg U$) the spin-wave frequency is $\Omega_1(\mathrm{q})$. With decreasing temperature, at $T \ll T_{2N}$, the sublattice interaction causes, as seen from (19), an increase of the velocity of the spin waves in this branch.

To calculate the corrections to the spectrum we must go outside the framework of the zeroth approximation of the self-consistent-field method. Since the intrasublattice interactions couple only oppositely directed spins, it becomes necessary to introduce correlators with lower indices α and β ($\alpha, \beta = f, g$), which determine the equilibrium orientation of the spins $\mathbf{S}_{r_2}$ and $\mathbf{S}_{r_1}$ in (12). Introducing for the correlators $K_{\alpha\beta}$ the parts $\Sigma_{\alpha,\beta}$ that are irreducible in V, we find from Dyson's equation that

$$K_{\alpha,\beta}=\frac{\Sigma_{\alpha\beta}}{1-\Sigma_{ff}\Sigma_{gg}V^2(\mathbf{q})-(\Sigma_{fg}+\Sigma_{gf})V(\mathbf{q})} \tag{20}$$

Since $\Delta \sim J^2$, we discard terms proportional to higher powers of J in the dispersion relation (18), and therefore also in the correlation functions. For example, in the denominator of (20) we have discarded terms containing products of the irroducible correlators of spins of different sublattices.

The diagrams for Σ_{ff} are obtained from the diagram shown in Fig. 2(a) by connecting two arbitrary vertices S^- and S^+ by effective-interaction lines. The entire notation is the same as in Ref. 9, except for the factor T^{-1}, which we have introduced in (12), so that

$$G_{ff}=G_{gg}^{*}=1/(y-i\omega_n),\quad y=SV(0). \tag{21}$$

The contribution of the diagram in which the interaction line joins simple vertices is cancelled out in (18) by the quantum correction to the sublattice magnetization (a similar cancellation appears in the calculation of the temperature dependence of the excitation spectrum in Ref. 9). The correction of interest to us stems from a diagram in which the interaction line enters a triple vertex. The correction to Σ_{ff} equals

$$\Sigma_{ff}^{(1)}(\omega_n,\mathbf{q})=-G_{ff}^{2}(\omega_n)L_{ff},$$

$$L_{ff}=\sum_{n,\mathbf{k}}\{K_{fg}(\omega_n,\mathbf{k})V(\mathbf{k})+(\bar{K}_{ff}(\omega_n,\mathbf{k})+\bar{K}_{fg}(\omega_n,\mathbf{k}))J^2(\mathbf{k})Q(\omega_n,\mathbf{k})\}. \tag{22}$$

The expression for $\Sigma_{gg}^{(1)}$ is perfectly analogous. The diagram for Σ_{fg} and Σ_{gf} is shown in Fig. 2(b). Since the vertices S_{fi}^{z} and S_{gj}^{z} can be connected only by the interaction V, we have

$$\begin{gathered}\Sigma_{fg}^{(1)}(\omega_n,\mathbf{q})=\Sigma_{gf}^{(1)}(\omega_n,\mathbf{q})=G_{ff}(\omega_n)G_{gg}(\omega_n)L_{fg}(\mathbf{q}),\\ L_{fg}(\mathbf{q})=\sum_{n,\mathbf{k}}V(\mathbf{q}-\mathbf{k})K_{fg}(\omega_n,\mathbf{k}).\end{gathered} \tag{23}$$

Using (20)–(23) we obtain from (18):

$$\begin{aligned}&[\omega^2-\Omega_1^2(\mathbf{q})-2V(0)(L_{ff}-L_{fg}(\mathbf{q}))][\omega^2-\Omega_2^2(\mathbf{q})\\ &-2U(0)(L_{ff}-L_{fg}(\mathbf{q}))]-4V(0)U(0)(L_{ff}-L_{fg}(\mathbf{q}))^2\\ &-4S^2I^2J^2(\mathbf{q})(V(0)-V(\mathbf{q}))(U(0)-U(\mathbf{q}))=0.\end{aligned} \tag{24}$$

From (22) and (23) it can be seen that

$$L_{ff}-L_{fg}(0)=\Delta. \tag{25}$$

One of the branches determined by (24) has a gap equal to

$$\omega_1(0)=[2\Delta(V(0)+U(0))]^{1/2}, \tag{26}$$

and the second is activationless, with a linear dispersion law at small momenta

$$\omega_2(\mathbf{q})=\left[\frac{\Omega_1^2(\mathbf{q})U(0)+\Omega_2^2(\mathbf{q})V(0)}{V(0)+U(0)}\right]^{1/2}. \tag{27}$$

We proceed now to the question of the Nêel temperatures. Since $V_{eff}(q=\pi/a)=0$, the indirect exchange does not influence the Neel temperatures in the molecular-field approximation. On going beyond the framework of this approximation, the situation changes. It is known that correlation effects influence the Nêel temperature.[9] The interaction V_{eff} between the sublattices, causing an additional correlation between the spins, changes the Nêel temperature compared with the values in single-sublattice garnets. Near T_{1N} the spins of the first sublattice interact and magnetize the paramagnetic spins of the second sublattice. The corresponding effective potential

$$V_{\mathrm{eff}}^{\alpha\beta}(\omega_n,\mathbf{q})=\frac{I(I+1)}{3}J^2(\mathbf{q})\delta_{n0}\delta_{\alpha\beta},\quad \alpha,\beta=x,y,z, \tag{28}$$

has the ferromagnetic sign. Near T_{2N} the spins of the second sublattice interact via spin waves with the first:

$$V_{\mathrm{eff}}^{xx}(\omega_n,\mathbf{q})=V_{\mathrm{eff}}^{yy}(\omega_n,\mathbf{q})=J^2(\mathbf{q})K(\omega_n,\mathbf{q}),\ V_{\mathrm{eff}}^{zz}=0. \tag{29}$$

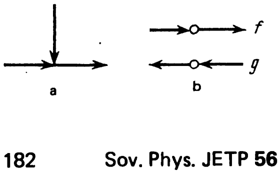

FIG. 2. Diagrams for the corrections to Σ_{ff} (a) and Σ_{fg} (b).

As shown in the preceding section, these correlations also contribute to the ferromagnetism. It is therefore natural that both Nêel temperature are smaller than in single-sublattice substances.

The correlation increments to the Nêel temperatures can be calculated in the manner used for a ferromagnet.[9] The relative change of these temperatures in the lowest order in $J/V \ll J/U \ll 1$ is

$$\frac{\delta(T_{1N})}{T_{1N}}=-\frac{5c}{3a^2}\frac{I(I+1)}{S(S+1)}\sum_{\mathbf{q}}\frac{J^2(\mathbf{q})}{(V(0)+V(\mathbf{q}))^2}, \tag{30}$$

$$\begin{aligned}\frac{\delta(T_{2N})}{T_{2N}}=&-\frac{4c}{3a^2}\sum_{\mathbf{q}}\frac{J^2(\mathbf{q})U(0)}{(V(0)+V(\mathbf{q}))(U(0)+U(\mathbf{q}))^2}\\ &+\frac{1}{3a}\sum_{\mathbf{q}}\frac{J^2(\mathbf{q})(U(0)-U(\mathbf{q}))}{(V(0)+V(\mathbf{q}))U^2(0)}\end{aligned} \tag{31}$$

where

$$a=S(S+1)/3,\quad c={}^3/_5a^2+{}^1/_{10}a. \tag{32}$$

The difference between the coefficients between (30) and the first term in (31) is connected with the fact that in the second case $V_{\mathrm{eff}}^{zz}=0$. The second term in (31) is of pure quantum-mechanical origin. It has no analog in (30), for in this case the interaction, as seen from (28) is purely static.

For aprimitive cubic lattice

$$\begin{aligned}\frac{\delta(T_{1N})}{T_{1N}}&=-0.07\frac{I(I+1)}{S(S+1)}\frac{J^2(0)}{V^2(0)},\\ \frac{\delta(T_{2N})}{T_{2N}}&=-\left(0.04-\frac{0.05}{S(S+1)}\right)\frac{J^2(0)}{V(0)U(0)}\end{aligned} \tag{33}$$

It can be seen from (33) that $\delta(T_{1N})$ is always negative, and the sign of δT_{2N} depends on the spin S. At $S>\frac{1}{2}$ we have $\delta T_{2N}<0$. In $Mn_3Cr_2Ge_3O_{12}$ we have $S=\frac{5}{2}$, so that both Nêel temperatures should decrease. The complicated magnetic structure does not change this conclusion. The point is that the coefficient preceding the first sum in (31) is approximately six times larger than the coefficient of the second at $S=5/2$, and the values of the two sums should not differ greatly (for a primitive cubic lattice they differ by only 20%). Therefore $\delta T_{2N}<0$ for the manganese-chromium garnet.

Thus, in accord with experiment, the interaction between the sublattices lowers the transition temperature.

4. ANTIFERROMAGNON SCATTERING BY SPINS IN A ZERO MOLECULAR FIELD

At temperatures $T_{2N} \ll T \ll T_{1N}$ the scattering of the antiferromagnons of the first sublattice by the paramagnetic spins of the second sublattice leads to damping the latter spins. The distinguishing feature of this scattering is that it is effected by degenerate-ground-state spins which, unlike the usually considered substitution impurities, do not participate in the oscillations of the antiferromagnetic sublattices.

In fourth-order perturbation theory the self-energy operator of the correlation function $K(\omega_n,\mathbf{q})$ is

$$\Sigma(\omega_n,\mathbf{q})=T\sum_{m,\mathbf{p}}J^2(\mathbf{q})J^2(\mathbf{p})K_0(\omega_m,\mathbf{p})\Gamma(\omega_n,\omega_m,\omega_m,\omega_n), \tag{34}$$

where

$$K_0(\omega_m, \mathbf{p})=2S^2(V(0)-V(p))/(\omega_m{}^2+\Omega_1{}^2(\mathbf{p})), \tag{35}$$

and the four-spin correlation function is, according to Ref. 9,

$$\Gamma(\omega_1, \omega_2, \omega_3, \omega_4)=-bG_1G_2(G_3+G_4)\delta_{\omega_1+\omega_2-\omega_3-\omega_4} +b'G_1G_2(\delta_{\omega_1-\omega_3}\delta_{\omega_2-\omega_4}+\delta_{\omega_1-\omega_4}\delta_{\omega_2-\omega_3}). \tag{36}$$

Here $G_k=G(\omega_k)$, $b=IB_I(yI/T)$; B_I is the Brillouin function, the quantity y in G and B_I is proportional to the magnetic field, which we set equal to zero at the end of the calculations; b' is the derivative of b with respect to y/T.

The spin-wave damping is proportional to the imaginary part of the analytic continuation of $\Sigma(\omega_n, \mathbf{q})$ to real frequencies, and is determined by the third term in (36). In the case of greatest interest, of low frequencies $\omega \ll SV(0)$, the damping $\gamma=\operatorname{Im}\omega$ turns out to be

$$\gamma=S^2\frac{V(0)-V(q=\omega/u)}{\omega}\operatorname{Im}\Sigma\left(\omega, q=\frac{\omega}{u}\right)=\frac{\Omega_0}{8\pi}\frac{I(I+1)}{3}\frac{J^4(0)}{u^3V(0)}\omega^2, \tag{37}$$

Ω_0 is the volume of the unit cell and $u=\Omega(q)/q$ is the velocity of the waves.

The damping calculated by us was found to be proportional to the square of the frequency, whereas scattering by large magnetic inhomogeneities leads to the classical Rayleigh damping $\gamma \sim \omega^4$. The scattering mechanism considered by us is much more effective at low frequencies. Even a small number of magnetic impurities landing in the interstices of an antiferromagnet and hence located in a zero molecular field can lead to a spin-wave damping that is larger than usual, and to broadening of the magnetic-resonance line. We note that the damping (37) does not contain the small factors such as T/T_{1N} which are typical of magnon-magnon damping.

A quadratic dependence of the damping on the frequency is observed also in phonon scattering by degenerate centers.[10,11]

5. LOW DENSITY OF MAGNETIC ATOMS IN ONE OF THE SUBLATTICES

It is known that in structures of the garnet type it is easy to change the composition of the a and c sublattices, by replacing the magnetic atoms with nonmagnetic ones. At low density of the magnetic atoms, the direct interaction can be neglected. The type of magnetic order established in the system of magnetic impurities is determined by the energy of the indirect exchange at distances much larger than the lattice constant a. We express the energy V_{eff} of (3) in the r-representation

$$V_{\text{eff}}(\mathbf{R}_1-\mathbf{R}_2)=\sum_{\mathbf{r}_1,\mathbf{r}_2}J(\mathbf{R}_1-\mathbf{r}_1)K(\mathbf{r}_1-\mathbf{r}_2)J(\mathbf{R}_2-\mathbf{r}_2), \tag{38}$$

where $K(\mathbf{r})$ is the Fourier transform of $K(0, \mathbf{q})$. At $r \gg a$ we have $K(\mathbf{r}) \sim r^{-1}e^{i\mathbf{q}_0\mathbf{r}}$ and

$$V_{\text{eff}}=\frac{\Omega_0S^2V(0)}{\pi u^2}\sum_{\mathbf{r}_1,\mathbf{r}_2}\frac{J(\mathbf{R}_1-\mathbf{r}_1)e^{i\mathbf{q}_0\mathbf{r}_1}J(\mathbf{R}_2-\mathbf{r}_2)e^{-i\mathbf{q}_0\mathbf{r}_2}}{|\mathbf{r}_1-\mathbf{r}_2|}. \tag{39}$$

As can be seen from (39), V_{eff} is proportional to the interaction energy of two systems of charges, distributed with densities $J(\mathbf{R}_1-\mathbf{r}_1)e^{i\mathbf{q}_0\mathbf{r}_1}$ and $J(\mathbf{R}_2-\mathbf{r}_2)e^{-i\mathbf{q}_0\mathbf{r}_2}$ near the points $\mathbf{R}_1$ and $\mathbf{R}_2$. The total charge $\Sigma_{\mathbf{r}}J(\mathbf{R}-\mathbf{r})e^{i\mathbf{q}_0\mathbf{r}}$ near each point is equal to the molecular field acting on the spin I_R, i.e., to zero. The interaction energy of such charges at large distances is determined by the lowest nonzero multipole. The energy averaged over the angles of the vector $\mathbf{R}_1-\mathbf{R}_2$ relative to the multipole axes is zero in all cases. The interaction considered leads therefore to spin-glass ordering in the impurity system.

The dependence of V_{eff} on R is determined by the type of magnetic structure. If it constitutes a layered antiferromagnet with ferromagnetic ordering layers, then the "dipole moment" differs from zero and therefore $V_{\text{eff}} \sim R^{-3}$. This result was obtained in Ref. 3 by direct integration. In our model with "checkerboard" order, the first nonzero order is quadrupole and $V_{\text{eff}} \sim R^{-5}$; for $Mn_3Cr_2Ge_3O_{12}$ we have $V_{\text{eff}} \sim R^{-3}$.

Besides the indirect exchange there is also the dipole-dipole magnetic interaction. In $Mn_3Cr_2Ge_3O_{12}$ it is weaker than the dipole interaction due to indirect exchange. For substances where only the higher multipoles differ from zero the situation is more complicated. If $J^2/V(0)$ exceeds the energy V_{dip} of the dipole-dipole interaction of the nearest neighbors in the lattice, we can neglect, at not too low impurity densities, the dipole-dipole interaction energy of impurities spaced on the average $r_c \sim n^{-1/2}$ apart (n is the density) if it is lower than their indirect-interaction energy (the corresponding criterion for the density can be easily written down). In this case the characteristic spin "quenching" temperature, which determines, for example, the position of the maximum heat capacity, is

$$T_f \sim V_{\text{eff}}(r_c) \sim n^{-(2l+1)/3}$$

(l is the number of first nonzero multipole). With decreasing density, the average dipole interaction density at the average distance becomes larger than $V_{\text{eff}}(r_c)$ (if no layered antiferromagnet is considered). Such systems have been named dipole glasses and have $T_f \sim n$.

In the manganese and chromium garnets the quantity $J^2(0)/V(0)$ is several times larger than V_{dip}, so that a situation is possible in which the dipole interaction is negligible. To be sure, at such not too low densities the direct interaction of the second neighbors in the lattice can become important. A definite answer to this question can be obtained only by experiment.

6. CONCLUSIONS

The sublattice interaction leads thus to the following effects.

1) In substances with antiferromagnetic sublattice the spin-wave spectrum of the sublattice with the higher Néel temperature should change on going from the vapor into the antiferromagnetic phase of the second sublattice, on account of the indirect exchange.

2) In the temperature range the spin-wave damping is anomalously large and depends quadratically on the frequency, and the magnetic-resonance line broadens.

3) Because of the quantum effects considered in this paper, the orientation of the sublattices should not correspond to the direction of their single-ion anisotropy axes. The gap in the spin-wave spectrum also changes.

4) It would be of interest to select the components in the *a* and *c* sublattices such that the canted structures, considered in the second and third sections, are realized. The presence of such structures can be detected by neutron diffraction and by the field dependence of the magnetization.

5) It is of interest to study the thermodynamics of a system of magnetic impurities located in the interstices of an antiferromagnet.

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1) It would be incorrect, generally speaking, to obtain a spin-wave dispersion law with the aid of the classical equation for the angular momentum, by adding E_{int} to the expression for the energy, since the quantum correction to the equations of motion can be of the same order as to the energy. In our case, however, this approach yields the same results as the rigorous analysis that follows.

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