Antiferromagnets with frustrated intrasublattice interaction in a magnetic field

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(Submitted 21 September 1989)
Zh. Eksp. Teor. Fiz. **97**, 696–706 (February 1990)

The magnetic properties of disordered Ising antiferromagnets with frustrated intrasublattice exchange interaction are studied. The theory satisfactorily describes the experimental results for the layer antiferromagnets $Fe_xMn_{1-x}TiO_3$ (Refs. 5–7) and $Fe_{1-x}Mg_xCl_2$ (Ref. 2).

INTRODUCTION

A considerable number of experiments, devoted to the study of the properties of disordered Ising antiferromagnets, have now been performed.[1–7] It has been found that below the temperature T_g, the introduction of strong disorder into an antiferromagnet results in the appearance of a re-entrant transition of a new state of matter, in which long-range antiferromagnetic order and nonergodicity—states of an antiferromagnetic spin glass (AFSG)—coexist.

A theoretical study of frustrated Ising antiferromagnets carried out by Korenblit *et al.*[8–12] has shown that the uniqueness of frustrated antiferromagnets is manifested in their behavior in an external magnetic field. Specifically, in a transition from a ferromagnetic phase into a ferromagnetic spin glass the magnetic field, while increasing the magnetization, suppresses the glass state, so that the temperature of the re-entrant transition decreases monotonically as the field is increased, while in an AFSG a magnetic field suppresses both antiferromagnetic and spin-glass order. For this reason, in a wide range of parameters the transition temperature $T_g(H)$ in AFSG increases monotonically as a function of the field, until it intersects the Néel line $T_N(H)$ at the field H_0. In fields stronger than H_0 the temperature T_g decreases monotonically as H is increased, and in this region a transition from the ergodic paramagnetic state into a "pure" spin glass without long-range magnetic order occurs on the line $T_g(H)$. Experimental studies performed for a laminar Ising metamagnet $Fe_{1-x}Mg_xCl_2$ reveal that T_g is almost doubled as the field is increased.[2] In addition, it was found in Ref. 2 that the curve $T_g(H)$ has a wide maximum near the intersection with the Néel line, but there were no reliable experimental data in this region.

A detailed study of a different frustrated Ising antiferromagnet $Fe_xMn_{1-x}TiO_3$ was performed and recently published.[6,7] the results obtained from samples with $x > 0.57$ have a number of important features compared both with studies of $Fe_{1-x}Mg_xCl_2$ and with the theoretical predictions made in Refs. 8–12.

1. It is observed that the temperature $T_g(H)$ increases strongly, compared with $Fe_{1-x}Mg_xCl_2$, as the field is increased (by a factor of 6.3 for $T_N(0)/T_g(0) \approx 6.6$) up to the field H_m at which the temperature $T_g(H)$ is equal to the Néel temperature $T_N(H)$.

2. In the field H_m the line $T_g(H)$ has a distinct and sharp maximum, after which $T_g(H)$ drops rapidly as the field is increased; in addition, the line $T_g(H)$ for $H \gtrsim H_m$ passes inside the antiferromagnetic (AF) phase.

3. The temperature dependence of the magnetic susceptibility $\chi(T)$ in the ergodic AF phase has a minimum whose depth increases with the degree of frustration.

In Refs. 8–12 attention is devoted primarily to antiferromagnets in which frustrations of the intersublattice interaction are stronger than frustrations of the intrasublattice interaction. In the alloys $Fe_xMn_{1-x}TiO_3$ the intrasublattice interaction is mainly frustrated. The point is that both $FeTiO_3$ and $MnTiO_3$ are layer antiferromagnets with antiferromagnetic interlayer interaction, but the intralayer interaction is ferromagnetic in $FeTiO_3$ and antiferromagnetic in $MnTiO_3$. For this reason if $MnTiO_3$ is added to $FeTiO_3$, then frustrations appear within the sublattice layers, while the intersublattice interaction, though its magnitude is random, always carried an antiferromagnetic sign.

In this paper it is shown that the behavior of a frustrated antiferromagnet in an external magnetic field in the case when the frustrations of the intrasublattice interaction are much stronger than the frustrations of the intersublattice interaction is different from that studied previously in Refs. 8–12. It turns out that the obtained theoretical phase diagrams and the temperature dependence of the magnetic susceptibility are qualitatively in agreement with the experimental results mentioned above.[6,7]

1. DERIVATION OF THE BASIC RELATIONS

We study an Ising magnet in which the spins belong the two different sublattices and interact according to the Hamiltonian

$$\mathcal{H} = \sum_{ij} J_{ij} S_{1i} S_{2j} - \sum_{ij,p} V_{ij} S_{pi} S_{pj} - H \sum_{i,p} S_{pi}. \tag{1}$$

The index $p = 1$ and 2 labels the sublattices, while the intrasublattice interaction V_{ij} and the intersublattice interaction J_{ij} do not depend on the distance r_{ij} and are distributed normally[13] with means V_0/N and J_0/N and variances V^2/N and J^2/N, respectively, where N is the number of spins in one sublattice and H is the external magnetic field.

Using the method of replicas we obtain the free energy spin:

$$f = -T \lim_{\substack{n\to 0 \\ N\to\infty}} \frac{1}{2nN} \Big\{ \exp\Big(\frac{J^2+V^2}{2T^2} Nn \Big) \times \int \prod_{\alpha,p} \Big(\frac{N}{2\pi}\Big)^{1/2} dx_p^{\alpha} \prod_{\alpha} dx_3^{\alpha} \Big(\frac{N}{2\pi}\Big)^{1/2} \prod_{(\alpha\beta),p} \Big(\frac{N}{2\pi}\Big)^{1/2} dy_p^{\alpha\beta} \times \prod_{(\alpha\beta)} \Big(\frac{N}{2\pi}\Big)^{1/2} dy_3^{\alpha\beta} \exp\left(N\Phi\left(x_p^{\alpha}, x_3^{\alpha}, y_p^{\alpha\beta}, y_3^{\alpha\beta}\right)\right) - 1 \Big\}, \tag{2}$$

 0038-5646/90/020388-07$03.00

$$\Phi(x_p^\alpha, x_3^\alpha, y_p^{\alpha\beta}, y_3^{\alpha\beta})$$

$$= -\Big[\sum_{\alpha,p}\frac{1}{2}(x_p^\alpha)^2+\sum_{\alpha}\frac{1}{2}(x_3^\alpha)^2+\frac{1}{2}\sum_{(\alpha\beta),p}(y_p^{\alpha\beta})^2 + \frac{1}{2}\sum_{(\alpha\beta)}(y_3^{\alpha\beta})^2-\ln \mathrm{Tr}\exp L\Big],$$

$$L=\frac{H}{T}\sum_{\alpha,p}S_p^\alpha+i\Big(\frac{J_0}{T}\Big)^{1/2}\sum_{\alpha,p}x_3^\alpha S_p^\alpha+\Big(\frac{J_0+V_0}{T}\Big)^{1/2}\sum_{\alpha,p}S_p^\alpha x_p^\alpha + \frac{J}{T}\sum_{(\alpha,\beta)}S_p^\alpha S_p^\beta y_3^{\alpha\beta}+\frac{(V^2-J^2)^{1/2}}{T}\sum_{(\alpha\beta),p}S_p^\alpha S_p^\beta y_p^{\alpha\beta}. \tag{3}$$

Here n is the number of replicas and α and β are the replica indices. The free energy is calculated by calculating the integral in (2) by the saddle-point method, which gives

$$f=-\frac{J^2+V^2}{4T}-\frac{T}{2}\lim_{n\to 0}\frac{1}{n}\Phi[(x_t^\alpha)_0,(y_t^{\alpha\beta})_0], \quad t=1,2,3, \tag{4}$$

while the saddle-point values $(x_t^\alpha)_0$, $(y_t^{\alpha\beta})_0$ satisfy the equations

$$\begin{aligned}
(x_p^\alpha)_0&=\Big(\frac{J_0+V_0}{T}\Big)^{1/2}m_p^\alpha=\Big(\frac{J_0+V_0}{T}\Big)^{1/2}\langle S_p^\alpha\rangle,\\
(y_p^{\alpha\beta})_0&=\Big(\frac{V^2-J^2}{T^2}\Big)^{1/2}q_p^{\alpha\beta}=\Big(\frac{V^2-J^2}{T^2}\Big)^{1/2}\langle S_p^\alpha S_p^\beta\rangle,\\
(x_3^\alpha)_0&=2im_3^\alpha\Big(\frac{J_0}{T}\Big)^{1/2}=(\langle S_1^\alpha\rangle+\langle S_2^\alpha\rangle)i\Big(\frac{J_0}{T}\Big)^{1/2},\\
(y_3^{\alpha\beta})_0&=\frac{2J}{T}q_3^{\alpha\beta}=\frac{J}{T}\sum_{p=1,2}\langle S_p^\alpha S_p^\beta\rangle.
\end{aligned} \tag{5}$$

The brackets indicate an average weighted with the exponential of the function (3).

Using Parisi's parameterization for the matrices $q_p^{\alpha\beta}$,[14,15] determining the spin-glass order parameter, and passing to the limit $n\to 0$ by Duplantier's method,[16] we obtain the following expression for the free energy f and the equation of state, which are correct in both the ergodic and nonergodic phases:

$$f=-\frac{J^2+V^2}{4T}[1-q_1(1)-q_2(1)]+\frac{V_0}{4}(m_1^2+m_2^2)-\frac{J_0}{2}m_1m_2 - \frac{1}{4}\Big\{\frac{V^2}{2}\int_0^1[q_1^2(x)+q_2^2(x)]dx+J^2\int_0^1 q_1(x)q_2(x)dx\Big\}-f_1-f_2, \tag{6}$$

$$f_p=\frac{1}{2}\int_{-\infty}^{\infty}dy\varphi_p(0,y)P_p(0,y),$$

where the functions $\varphi_p(x,y)$ and $P_p(x,y)$ satisfy the system of equations

$$\frac{\partial\varphi_p}{\partial x}=-\frac{1}{2}\Big[\frac{\partial^2\varphi_p}{\partial y^2}+\frac{x}{T}\Big(\frac{\partial\varphi_p}{\partial y}\Big)^2\Big]\frac{d}{dx}[V^2q_p(x)+J^2q_{\bar p}(x)], \tag{7}$$

$$\frac{\partial P_p}{\partial x}=\frac{1}{2}\Big[\frac{\partial^2P_p}{\partial y^2}-\frac{2x}{T}\frac{\partial}{\partial y}\Big(P_p\frac{\partial\varphi_p}{\partial y}\Big)\Big]\frac{d}{dx}[V^2q_p(x)+J^2q_{\bar p}(x)],$$

$$(\bar p_{1,2}\neq p),$$

with the boundary conditions

$$P_p(0,y)=\{2\pi[V^2q_p(0)+J^2q_{\bar p}(0)]\}^{-1/2} \times\exp\Big\{-\frac{[y-(H+V_0m_p-J_0m_{\bar p})]^2}{2(V^2q_p(0)+J^2q_{\bar p}(0))}\Big\}$$

$$\varphi_p(1,y)=T\ln 2\,\mathrm{ch}\,y/T. \tag{8}$$

The system is closed by the condition of self-consistency for the magnetizations $m_{1,2}$ of the sublattices and spin-glass parameters $q_{1,2}(x)$:

$$\begin{aligned}
m_p&=\int P_p(x,y)M_p(x,y)\,dy,\\
q_p(x)&=\int P_p(x,y)M_p^2(x,y)\,dy,
\end{aligned} \tag{9}$$

where we introduced the function

$$M_p(x,y)\equiv\partial\varphi_p/\partial y,\ M_p(1,y)=\mathrm{th}\,y/T.$$

The equations (7)–(9) were first derived in Ref. 10. Analogous expressions for a magnet with different concentrations of atoms in the sublattices were derived in Ref. 17.

Writing $\dot q_p=dq_p/dx$ we obtain from the expressions (9), using (7),

$$\begin{aligned}
\dot q_p&=\int dy\Big\{P_p(x,y)2M_p(x,y)\frac{\partial M_p(x,y)}{\partial x} + \frac{1}{2}M_p^2(V^2\dot q_p+J^2\dot q_{\bar p}) \times\Big[\frac{\partial^2P_p(x,y)}{\partial y^2}-\frac{2x}{T}\frac{\partial}{\partial y}\Big(P_p(x,y)\frac{\partial M_p(x,y)}{\partial y}\Big)\Big]\Big\}\\
&=\int dyP_p(x,y)\hat\Omega_pM_p^2(x,y),
\end{aligned}$$

where we introduced the operator[9,18]

$$\hat\Omega_p=\frac{\partial}{\partial x}+(V^2\dot q_p+J^2\dot q_{\bar p})\Big[\frac{\partial^2}{\partial y^2}+\frac{x}{T}M_p(x,y)\frac{\partial}{\partial y}\Big];$$

$$p=1,2,\quad \bar p\neq p. \tag{10}$$

Using the obvious properties

$$\hat\Omega_pM_p=0,$$

$$\hat\Omega_p(fg)=(\hat\Omega_pf)g+f(\hat\Omega_pg)+(V^2\dot q_p+J^2\dot q_{\bar p})\frac{\partial f}{\partial y}\frac{\partial g}{\partial y}, \tag{11}$$

we find

$$\hat\Omega_pM_p^2=(\dot q_pV^2+\dot q_{\bar p}J^2)\Big(\frac{\partial M_p}{\partial y}\Big)^2,$$

and therefore we obtain for $\dot q_{1,2}$ the system of equations

$$\begin{aligned}
&\begin{cases}\dot q_1=\dot q_1V^2I_1+\dot q_2J^2I_2,\\ \dot q_2=\dot q_1J^2I_1+\dot q_2V^2I_2,\end{cases}\\
&I_p=\int dy\,P_p(x,y)\Big[\frac{\partial M_p(x,y)}{\partial y}\Big]^2.
\end{aligned} \tag{12}$$

From here it is obvious that if $\dot{q}_p \neq 0$, then the following relation must hold:

$$[1-V^2I_1][1-V^2I_2]-J^4I_1I_2=0, \tag{13}$$

which is the condition under which Parisi's parameterization is marginally stable.

Above the temperature of the transition into the nonergodic state the relation (13) cannot be satisfied, and for this reason it follows from (12) that $\dot{q}_p = 0$ and hence $\varphi_p(x,y)$, $P_p(x,y)$, and $q_p(x)$ do not depend on x. For this reason the expressions for the free energy and the equation of state assume the following forms:[12]

$$f=-\frac{V^2}{8T}[(1-q_1)^2+(1-q_2)^2]-\frac{J^2}{4T}(1-q_1)(1-q_2)$$
$$-\frac{J_0}{2}m_1m_2$$
$$+\frac{V_0}{4}(m_1^2+m_2^2)-\frac{T}{2}\langle \ln 4\operatorname{ch}E_1\operatorname{ch}E_2\rangle_c,$$
$$m_p=\langle \operatorname{th}E_p\rangle_c, \quad q_p=\langle \operatorname{th}^2E_p\rangle_c, \tag{14}$$

$$E_p=\frac{1}{T}[H+V_0m_p-J_0m_{\bar{p}}+z(V^2q_p+J^2q_{\bar{p}})^{1/2}], \quad p\neq\bar{p}, \tag{15}$$

where the brackets $\langle\ldots\rangle_c$ denote Gaussian averaging over z:

$$\langle u(z)\rangle_c=\int e^{-z^2/2}u(z)\frac{dz}{(2\pi)^{1/2}}.$$

The relation (13) transforms, in the process, into the equation for the temperature of the transition into the nonergodic state $T_g(H)$:

$$(T_g^2-V^2\langle \operatorname{ch}^{-4}E_1\rangle_c)(T_g^2-V^2\langle \operatorname{ch}^{-4}E_2\rangle_c)$$
$$-J^4\langle \operatorname{ch}^{-4}E_1\rangle_c\langle \operatorname{ch}^{-4}E_2\rangle_c=0. \tag{16}$$

On the line $T_g(H)$, as shown in Ref. 19, the replica-symmetric solution of the saddle-point equations (5) becomes unstable with respect to small fluctuations which break this symmetry. By studying the expression (3) for stability relative to small fluctuations of this solution which do not break the replica symmetry it is possible to find the stability boundary of the paramagnetic $T_{par}(H)$ and antiferromagnetic $T_{af}(H)$ phases in the ergodic state ($T>T_g(H)$).[12] Above some temperature T_c (triple critical point) the lines $T_{af}(H)$ and $T_{par}(H)$ merge and determine the second-order transition line $T_N(H)$. The first-order phase transition line, starting from the triple critical point (T_c,H_c), can be found from the condition that the free energies of the antiferromagnetic and paramagnetic phases, determined by the expression (14), are equal. As regards the triple critical point itself (T_c,H_c) it is not convenient to find it from the condition $T_{af}(H)=T_{par}(H)$. It is simpler to determine it from the condition that the coefficient B in the expansion of the free energy (14) in powers of the antiferromagnetic order parameter $l=m_1-m_2$ vanish:

$$\frac{f}{V_0+J_0}=\frac{f_0}{V_0+J_0}+\frac{1}{2!}Al^2+\frac{1}{4!}Bl^4+\ldots, \tag{17}$$

$$A=1-\beta(1-q)-{}^1/_2\beta w\tilde{g}_2q', \tag{18}$$

$$B=\beta(3q''+2\tilde{g}_3-{}^3/_2\theta w\tilde{g}_3m''q'+{}^3/_2w\tilde{g}_4q'-{}^3/_4uw\tilde{g}_4q'q''$$
$$-{}^1/_8w\tilde{g}_6(q')^3-{}^1/_2w\tilde{g}_2q'''), \tag{19}$$

where we have introduced the following notation:

$$\beta=\frac{J_0+V_0}{T}, \quad u=\frac{V^2+J^2}{(V_0+J_0)^2},$$
$$w=\frac{V^2-J^2}{(V_0+J_0)^2}, \quad \theta=\frac{V_0-J_0}{V_0+J_0},$$
$$\tilde{g}_k=\beta^{k-1}\left\langle \frac{d^k\operatorname{th}E_p}{dE_p^k}\right\rangle_c\Bigg|_{l=0},$$

and the quantities m'', q', q'', and q''' are found from the relations

$$q'=-\tilde{g}_2(1+{}^1/_2w\tilde{g}_3)^{-1},$$
$$\begin{cases} m''[1-\beta\theta(1-q)]^{-1}/_2\beta u\tilde{g}_2q''=\beta[\tilde{g}_2+w\tilde{g}_3q'+{}^1/_4w^2\tilde{g}_4(q')^2], \\ m''\theta\tilde{g}_2+[1+{}^1/_2u\tilde{g}_3]q''=-[\tilde{g}_3+w\tilde{g}_4q'+{}^1/_4w^2\tilde{g}_5(q')^2], \end{cases} \tag{20}$$
$$q'''=-[1+{}^1/_2w\tilde{g}_3]^{-1}\{3\theta\tilde{g}_3m''+\tilde{g}_4+{}^3/_2\theta uw\tilde{g}_4m''q'+{}^3/_2w\tilde{g}_5q'$$
$$+{}^3/_2u\tilde{g}_4q''+{}^3/_4w^2\tilde{g}_6(q')^2+{}^3/_4uw\tilde{g}_5q'q''+{}^1/_8w^3\tilde{g}_7(q')^3\}.$$

As should be the case the equation $A=0$ is identical to the equation for the line $T_{par}(H)$ (Ref. 12). The additional condition $B=0$ determines the position of the tricritical point (T_c,H_c). Thus we have derived equations which make it possible to construct the complete phase diagram in the ergodic region.

2. PHASE DIAGRAM OF ANTIFERROMAGNETS WITH SMALL J

In the limiting case $J/V\to 0$ Eq. (16) for T_g decomposes into two independent equations

$$T_{g_1}^2=V^2\langle \operatorname{ch}^{-4}E_1\rangle_c \tag{21}$$

and

$$T_{g_2}^2=V^2\langle \operatorname{ch}^{-4}E_2\rangle_c. \tag{21'}$$

When there is no external field we have $m_1=-m_2$ and $q_1=q_2$, and for this reason $T_{g_1}=T_{g_2}$. For $H>0$ this is no longer the case.

One can see from Eqs. (12) that if in the present case $J/V\to 0$ there is no relation between $\dot{q}_1$ and $\dot{q}_2$, then they can vanish independently. Physically, this means that the transition into the nonergodic state in each sublattice occurs independently at the temperature T_{g_1} and T_{g_2}, respectively. It is obvious that the effects of irreversibility, based on which the transition into the nonergodic state is recorded in an experiment, will be manifested first at the highest of these two temperatures, so that it is the field dependence of this temperature that must be compared with experiment.

It is not difficult to understand that the high transition temperature corresponds to the sublattice whose spins in a weak external field are oriented opposite to this field. Indeed, let the spins of the first sublattice be oriented down. In the entire region of existence of antiferromagnetism in a nonzero field the magnetization of the first sublattice is less in absolute magnitude that the magnetization of the second sublattice. For this reason in this region the line $T_{g_1}(H)$ on the H–T phase diagram will pass above the line $T_{g_2}(H)$. In the region where the antiferromagnetic order is destroyed by an external field (i.e., $m_1=m_2$, $q_1=q_2$) the temperature $T_{g_1}(H)$ is equal to the temperature $T_{g_2}(H)$.

We now present some qualitative considerations which make it possible to understand the field dependence of the

temperature $T_{g_1}(H)$ and $T_{g_2}(H)$. Consider the first sublattice. As the external field grows the magnetization of this sublattice $|m_1|$ will decrease, which will cause T_{g_1} to increase. The temperature $T_{g_1}(H)$ will increase until $|m_1|$ vanishes in some field H.[1)] At $H = H_m$ m_1 changes sign. As the external field increases further m_1 increases, and T_{g_1} correspondingly decreases. Thus the curve $T_{g_1}(H)$ has a maximum at $H = H_m$. Conversely, the magnetization at first increases as the external field is intensified and T_{g_2} decreases. For $H \gtrsim H_m$ the temperature T_{g_2} increases as the field is increased up to levels at which the antiferromagnetism is destroyed.

It follows from the arguments presented above that the maximum value of the temperature $T_{g_1}(H)$ is equal to V. Indeed, on the line $m_1(T,H) = 0$ the parameter q_1 is determined from the equation

$$q_1 = \left\langle \mathrm{th}^2 \frac{Vz}{T_g} q_1^{1/2} \right\rangle_c ,$$

and

$$T_{g_1}(H_m) \equiv T_{g_m} = V^2 \left\langle \mathrm{ch}^{-4} \frac{Vzq_1^{1/2}}{T_{g_m}} \right\rangle_c .$$

These equations have one solution $q_1 = 0$, $T_{g_m} = V$. This result corresponds to the fact that on the line $m_1(T,H) = 0$ on the first sublattice in the field H_m transforms from the paramagnetic state ($m_1 = q_1 = 0$) into the state of a "pure" spin glass ($m_1 = 0, q_1 \neq 0$). For this reason the function $T_{g_1}(H)$ should have a cusp in the field H_m.

Figure 1 shows the H–T phase diagram of a frustrated antiferromagnet with $J = V_0 = 0$ and $T_N(H=0)/T_g(H=0) \approx 9.5$, obtained by means of numerical analysis of Eqs. (15) and (16). It is interesting that the maximum of T_{g_1} lies very close to the Néel line $T_N(H)$. The lines $T_g(H)$ and $T_N(H)$ intersect in the field $H_0 > H_m$, and in addition the slope of the line $T_g(H)$ changes in the field H_0. In weak magnetic fields the change in the temperature satisfies $\Delta T_g = T_g(H) - T_g(0) \propto H$. It can be shown that ΔT_g is a linear function of H in weak fields only for $J = 0$. For $J \neq 0$, then we have $\Delta T_g \propto H^2$.

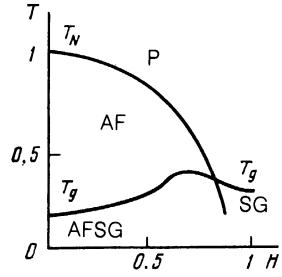

FIG. 2. The phase diagram of a frustrated antiferromagnet in a magnetic field with $V_0 = 0$, $J_0 = 1$, $V^2/J^2 = 5$, $J_0/(V^2 + J^2)^{1/2} = 1.5$ ($T_N/T_g|_{H=0} = 5.5$).

For nonzero but sufficiently small values of J the maximum on the curve $T_{g_1}(H)$ becomes wider and shifts away from the line $T_N(H)$ (Fig. 2). As regards the second solution of Eq. (16) for T_g, strictly speaking, it is physically meaningless for $J \neq 0$, since nonergodicity arises immediately in both sublattices. If, however, we have $J \ll V$, then, as follows from Eqs. (12), in the temperature range $T_{g_1}(H) \gtrsim T > T_{g_2}(H)$ the derivative $\dot{q}_2 \propto J^2 \dot{q}_1 / V^2 \ll \dot{q}_1$. In other words, in this interval of temperatures the nonergodicity of the second sublattice is weak. In this sense the line $T_{g_2}(H)$ now determines not a phase transition, but rather crossover to the regime of strong nonergodicity of the second sublattice.

Figure 3 shows the phase diagram at $J = 0$, $T_N/T_g|_{H=0} = 5.6$, $V_0/T_N = 0.3$. In accordance with the analysis performed in Refs. 10 and 12 the line $T_N(H)$ contains two tricritical points K_1 and K_2, between which lies the region of the first-order phase transition. In this case the phase diagram contains an interval of field $H_A < H < H_{K_2}$, in

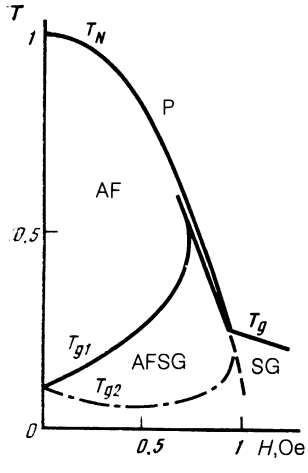

FIG. 1. The phase diagram of a frustrated antiferromagnet in a magnetic field with $J = V_0 = 0$, $J_0/V = 1.7$, $J_0 = 1$ ($T_N/T_g|_{H=0} = 9.5$). The dot-dashed line is the line $T_{g_2}(H)$; the dashed line is the approximate continuation of the Néel line $T_N(H)$ in the nonergodic state. Phases: P—paramagnetic, AF—antiferromagnetic ergodic, AFSG—antiferromagnetic nonergodic, SG—spin glass.

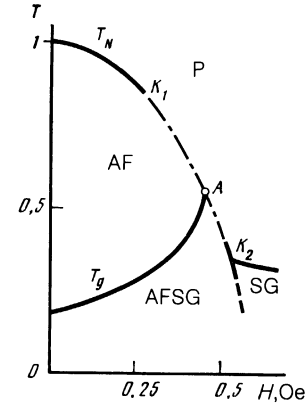

FIG. 3. The phase diagram of a frustrated antiferromagnet in a magnetic field with $J = 0$, $J_0 + V_0 = 1$, $V_0 = 0.3$, $(J_0 + V_0)/V = 1.5$ ($T_N/T_g|_{H=0} = 5.5$). The dot-dashed line is the first-order phase transition line; K_1 and K_2 are the triple critical points; on the section AK_2 the transition from the paramagnetic (P) phase into the antiferromagnetic nonergodic phase (AFSG) occurs in a jump-like fashion; on the section K_2B the difference between the lines $T_{g_1}(H)$ and $T_N(H)$ is so small that it cannot be shown in the figure.

which as the temperature is lowered nonergodicity and antiferromagnetism abruptly appear at the same time. It should be noted that we did not determine the position of the section AK_2 on the line of the first-order phase transition completely rigorously, since in determining it the free energies of the paramagnetic and ergodic antiferromagnetic phases were equated, while the magnetically ordered phase, according to Fig. 3, is nonergodic in this region. It is obvious, however, that this circumstance does not affect the qualitative conclusion that an AFSG can arise discontinuously.

In the present case, when the point A is approached from the AF phase the moment of the first sublattice is oriented opposite to the external field. For this reason T_g increases with the field up to the point A.

We note that in the case $J \neq 0$ and $V_0 \neq 0$ a discontinuous transition into a nonergodic state can be accompanied not by the appearance, but rather by the vanishing of long-range magnetic order (the transition AF→SG).[10,12]

3. MAGNETIC SUSCEPTIBILITY

Differentiating the expression (9) for the magnetizations of the sublattice $m_{1,2}$ and the parameters $q_{1,2}$ ($x = 0$) with respect to the external field and using the boundary condition (8) we obtain an expression for the magnetic susceptibility of an antiferromagnet which is applicable in both the ergodic and nonergodic phases $\chi = \Pi/[1 - (V_0 - J_0)\Pi]$,

$$\Pi = \frac{1}{T}\left[\langle m'(0,y)\rangle_0 - (J^2-V^2) \times \frac{\langle m''(0,y)\rangle_0 \langle m(0,y)m'(0,y)\rangle_0}{T^2+(J^2-V^2)\langle (m(0,y)m'(0,y))'\rangle_0}\right]. \qquad (22)$$

Here we have introduced the notation $u'(0,y) = T\partial u(0,y)/\partial y$; the angular brackets $\langle ... \rangle_0$ denote averages over y with the distribution function $P_{1,2}(0,y)$ from (8) with $H = 0$. An expression for χ was derived in Refs. 9 and 11 in the case $V_0 = V = 0$. In the ergodic phase the expression for χ assumes the form[2)]

$$\Pi = \frac{1}{T}\left[g_1 + \frac{J^2-V^2}{2T^2} g_2^2 \left(1 - \frac{J^2-V^2}{2T^2} g_3\right)^{-1}\right], \qquad (23)$$

$$g_k = \left\langle \frac{d^k \operatorname{th} E_p}{dE_p^k} \right\rangle_c \Big|_{H=0}.$$

It is obvious that the quantities J^2 and V^2 enter into the expressions (22) and (23) for χ with different signs. It is easy to understand why the frustrations of the intra- and intersublattice interaction affect the susceptibility differently.

We first study the case $V = 0$. Assume that when there is no field we have $m_1 < 0$ and $m_2 > 0$. Then a weak external field decreases the effective mean field acting on the spins of the first sublattice $\overline{H}_{1e} = H + V_0 m_1 - J_0 m_2$ and it increases the variance of this effective field $\delta H_{1e} = J[q_2(0)]^{1/2}$. Both these factors decrease the magnetization of the sublattice m_1. Now let $J = 0$. Then a weak external field decrease $\overline{H}_{1e}$ as happened before, but unlike the preceding case it also decreases the variance of the random field on the first sublattice $V[q_1(0)]^{1/2}$. Thus the changes in the mean effective field and its variance have opposite effects on the magnetization m_1. Analogous arguments can also be made for the second sublattice.

We now study the temperature dependence of the magnetic susceptibility. This is most easily done analytically in the case of a strongly frustrated antiferromagnet, when we have $b = (J_0 + V_0)/(J^2 + V^2)^{1/2} - 1 \ll 1$. Then, as shown in Ref. 8, the temperature satisfies $T_g = T_N(1 - b^{1/2})$ and a perturbation theory in the parameter b can be constructed in the entire ergodic phase.

If we have $V = 0$, then, according to Ref. 8, the susceptibility increases monotonically as the temperature is lowered in the entire interval $T_g < T < T_N$. The situation is different with $J = 0$. In this case it follows from (15) and (23) that

$$m^2 = 2\tau(b - \tau^2/3), \quad q = \tau(1 + {}^4/_3 b - {}^4/_9 \tau^2),$$
$$\chi = \frac{1}{2J_0}\left(1 - \frac{T_N}{J_0}\tau\frac{b - \tau^2/3}{b+\tau}\right), \quad \tau = \frac{T_N - T}{T_N} < b^{1/2} \ll 1. \qquad (24)$$

If follows from Eq. (24) and $\chi(T)$ varies nonmontonically: for $\tau = (3b^2/2)^{1/3} < b^{1/2}$ the susceptibility has a minimum.

This monotonic dependence $\chi(T)$ in an ergodic AF state is preserved, as one can see from Fig. 4, with an arbitrary degree of frustration, if $J = 0$ holds. The ratio J/V for which the function $\chi(T)$ has a minimum in the AF state depends on the degree of frustration b.

To determine the behavior of the magnetic susceptibility in the nonergodic state it is necessary to solve Eqs. (7)–(9). This can be done only near T_g with $\tau_g = (T_g - T)/T_g \ll 1$. The quantity $(d\chi/dT)_{T_g - 0}$ can be calculated using the procedure described in Ref. 9.

Figure 5 shows the dependence of the quantities $D_\pm = (d \ln \chi/d \ln T)_{T_g \pm 0}$ on the ratio $(V^2 - J^2)/(V^2 + J^2)$. One can see that for $J \neq V$ the slope of the susceptibility changes at $T = T_g$ and the dependence $\chi(T)$ becomes weaker in the nonergodic region than in the

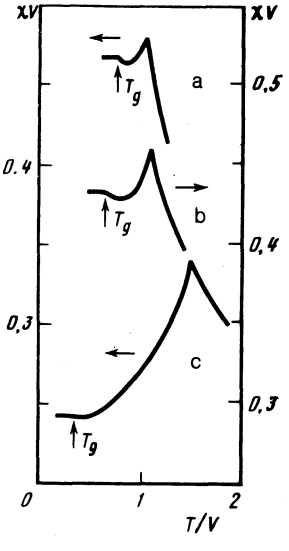

FIG. 4. The temperature dependences of the reduced magnetic susceptibility $\chi = 1/2\, Vd(m_1 + m_2)/dH$ with $J_0 = 1$ and $V_0 = J = 0$ and different degrees of frustration: a—$J_0/V = 1.05$ $T_N/T_g|_{H=0} \approx 1.37$, b—$J_0/V = 1.1$, $T_N/T_g|_{H=0} \approx 1.6$, c—$J_0/V = 1.5$, $T_N/T_g|_{H=0} \approx 5.5$. The broken lines show the behavior of $\chi(T)$ for $T < T_g$.

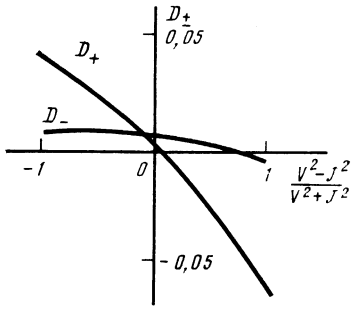

FIG. 5. The dependence of the quantities $D_{\pm} = d \ln \chi / d \ln T|_{T=T_{\pm 0}}$ on the ratio of the variance of the intrasublattice interaction V and the variance of the intersublattice interaction J.

ergodic region. There exists a region of parameters in which we have $D_+ < 0$ and $D_- > 0$, i.e., the susceptibility has a sharp maximum (cusp) at $T = T_g$.

4. COMPARISON WITH EXPERIMENT

We shall compare the results obtained with experiment for the alloys $Fe_x Mn_{1-x} TiO_3$ and $Fe_{1-x} Mg_x Cl_2$. As we have already pointed out, $FeTiO_3$ and $MnTiO_3$ are laminar antiferromagnets with antiferromagnetic interaction between the layers, while within the layers the interaction is ferromagnetic for $FeTiO_3$ and antiferromagnetic for $MnTiO_3$. Since the interlayer interaction is weaker than the intralayer interaction in pure $MnTiO_3$ antiferromagnetic order exists in each layer, i.e., the magnetic structure of this compound actually consists of four sublattices. When $FeTiO_3$ is added in interlayer interaction remains antiferromagnetic and fluctuates in magnitude, but not in sign, i.e., nonergodicity arises independently in layers with oppositely directed spins. In each layer, however, the iron impurity brings about frustration of interaction between the manganese sublattices.[17] In this sense the manganese-enriched alloy can be regarded as a two-sublattice antiferromagnetic with frustrated intersublattice interaction, the theory of which we constructed previously.[8,9,11]

If, however, $MnTiO_3$ is added to $FeTiO_3$, then a system which, as we have already pointed out, consists of a two-sublattice antiferromagnet with frustrations within the sublattice layers is obtained.

We now show that the theory developed above with $J = 0$ satisfactorily describes all the basic features of the behavior of $Fe_x Mn_{1-x} TiO_3$ with $x > 0.57$. We note first that in Ref. 7 in all the samples long-range magnetic order in a magnetic field vanishes through second-order phase transition. This means that either the parameter V_0 is quite small or the first-order phase transition is suppressed by disorder.[12] In any case, however, since there is no first-order phase transition line, the results are virtually independent of V_0 and for this reason we compare with experiment the results obtained for $V_0 = 0$.

1. For $J = 0$ the maximum increase in T_g in a magnetic field is significantly larger than in the case $V = 0$ which we studied previously:[9,11] for $T_N/T_g|_{H=0} \approx 9.5$ T_g increases approximately by a factor of two if $V = 0$ and by almost a factor of 6 in the case $J = 0$ (Fig. 1). The last value does not differ much from experiment (increase by a factor of 6.3 with $T_N/T_g|_{H=0} \approx 6.6$).

2. Comparison of Figs. 1 and 3 from Ref. 7 shows that the overall character of the behavior of $T_g(H)$ is the same in both theory and experiment. First of all, both in fields $H < H_m$ and in fields $H \gtrsim H_m$ the line $T_g(H)$ passes within the antiferromagnetic phase; second, in a weak magnetic field the experimental behavior of $T_g(H)$ is close to the linear behavior following from the theory. In Ref. 17 he results of the molecular-field theory are also compared with the experimental results from Ref. 5–7. The phase diagram is constructed for $V_0/J_0 = V/J = 5$, when, unlike the experiment, long-range magnetic order vanishes abruptly. For this reason neither a maximum of $T_g(H)$ in the AF region nor linear growth of $T_g(H)$ in weak fields, (since $J \neq 0$) was obtained in Ref. 17.

3. It follows from the theory, in complete qualitative agreement with experiment, that the curve $\chi(T)$ has a minimum for $T_g < T < T_N$. Moreover, in both theory and experiment the minimum almost vanishes in a weakly-frustrated sample. This can be seen by comparing Fig. 1 from Ref. 7 with Fig. 4, which shows the temperature dependence $\chi(T)$ approximately for the same ratios rp $T_N/T_g|_{H=0}$ as in the experiment.

The experimental behavior of the magnetic susceptibility in the nonergodic state differs from the predicted theoretically in one important respect. The theoretical susceptibility has a change in slope at T_g and as the temperature is further lowered the susceptibility remains virtually constant. The experimental susceptibility, however, obtained with the sample cooled in a magnetic field (this value of the susceptibility is customarily regarded as the equilibrium susceptibility) passes through the point of irreversibility without a change in slope and is temperature-independent only appreciably below T_g.

The theory also provides an explanation for the wide maximum on the curve $T_g(H)$ in the alloy $Fe_{0.552} Mg_{0.448} Cl_2$.[22] Pure $FeCl_2$ is a laminar metamagnet, in which the interlayer interaction is antiferromagnetic and is approximately and order of magnitude weaker than the intralayer interaction.[20] When the alloy is diluted with magnesium atoms a re-entrant transition occurs at concentrations close to the percolation threshold in the plane and in our model this situation corresponds to $V \gtrsim J$. As one can see from Fig. 2 in this theory the dependence $T_g(H)$ also has a smooth maximum.

CONCLUSIONS

The foregoing analysis shows that the model of an antiferromagnet with an infinite radius of interaction between spins quite satisfactorily describes the properties of real frustrated antiferromagnets, at least in the ergodic state. it is possible to explain the basic behavior of the line $T_g(H)$. The theory can pick up the difference in the physical properties of disordered antiferromagnets depending on the method of frustration. The theory predicts a number of properties of substances which, in our opinion, it would be interesting to study experimentally. In particular, for $J \ll V$ a transition into a regime where the second sublattice is strongly nonergodic should be observed in the (T,H) plane. This region can apparently be manifested experimentally as a maximum in the temperature dependence of the imaginary part of the magnetic susceptibility.

[1)] We note that such a field may not exist. In the case of a first-order transition the moment m_1 can change in a jump-like fashion from a negative to a positive value, equal to m_1 (see the end of this section).
[2)] In Ref. 12 a factor of $(-1/2)$ in the coefficient of g_3 is omitted in this expression.

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Translated by M. E. Alferieff