Heisenberg model in a magnetic field, and metamagnetism of Jahn-Teller systems

K. I. Kugel'[1)] and B. I. Khomskiĭ

P. N. Lebedev Physics Institute, USSR Academy of Sciences
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The possibility is demonstrated of metamagnetic behavior of systems with isotropic spin interaction, characterized by two order parameters.

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Metamagnetism (a nonlinear increase, sometimes even jumplike, of the magnetization of an antiferromagnet with increasing field) is usually attributed to anisotropy (one-ion or exchange) of the magnetic properties.[1] The aim of the present paper is to show that metamagnetic behavior is possible also in substances with an isotropic spin system owing to its interaction with another "degree of freedom."[2)] We shall show that such a mechanism can be realized in systems with orbital degeneracy (Jahn-Teller systems).

In substances with Jahn—Teller ions (for example Mn^{3+}, Cr^{2+} or Cu^{2+} in an octahedral surrounding, whose orbital ground state is doubly degenerate) the exchange interaction, as shown in[4,5], is described by the Hamiltonian of the type of the Heisenberg double Hamiltonian, in the form (with allowance for the external magnetic field h)

$$H = \sum_{i,j} \{ J_1 s_i s_j + J_2 \tau_i \tau_j + J_3 (s_i s_j)(\tau_i \tau_j) \} - h \sum_i s_i . \quad (1)$$

Here s_i is the spin of the ith site ($s^z = \pm 1$), and τ_i is the pseudospin characterizing the orbital state: for example, for the e_g orbitals $\tau^z = 1$ corresponds to the orbital $d_{2z^2-x^2-y^2}$, while $\tau^z = -1$ corresponds to $d_{x^2-y^2}$.

A Hamiltonian of the type (1) is obtained by starting with the degenerate Hubbard model. When account is taken of the concrete form of the l_g orbitals it has, of course, a much more complicated form, but all the qualitative features can be easily explained with the aid of (1). The corresponding interactions in 1 are of the form $(s_i \cdot s_j)$ and $(\vec{\tau}_i \cdot \vec{\tau}_j)$ (the double Heisenberg model), or else s and τ can be Ising variables. In the latter case, our model (1) coincides in essence with the Ashkin-Teller model,[6] interest in which has increased recently.[7,8]

In the considered model, the orbital and spin structures turn out to be closely related. Thus, the spin interaction is described by the Hamiltonian

$$H^s = \sum_{i,j} J^s_{ij} s_i s_j + h \sum_i s_i , \quad J^s_{i,j} = J_1 + J_3 < \tau_i \tau_j > . \quad (2)$$

Analogously, the interaction in the τ system is determined by the quantity $J^{\tau}_{ij} = J_2 + J_3 \langle s_i s_j \rangle$. By varying the spin structure, say with an external magnetic field, we change by the same token also J^{τ}_{ij}, and accordingly the orbital structure. But then, according to (2), the exchange integral J^s_{ij} also becomes different; as a result, the response of the spin system to an external magnetic field will be nonlinear, and this is indeed the cause of the metamagnetic behavior.

Let us consider for the sake of argument the ratio of the constants $J_1 > J_3 > J_2 > 0$. The ground state at $h=1$ will be antiferromagnetic in spin, $\langle s_i s_{i+1}\rangle = -1$, and "ferromagnetic" in the orbitals, with $\langle \tau_i \tau_{i+1}\rangle = 1$; in this case actually $J^s = J_1 + J_3 > 0$ and $J^\tau = J_2 - J_3 < 0$. With increasing h, the spin correlator $\langle \mathbf{s}_i \cdot \mathbf{s}_{i+1}\rangle$ will increase and reach in a field h_{c_2} a value $\langle \mathbf{s}_i \cdot \mathbf{s}_{i+1}\rangle = -J_2/J_3$. The field h_{c_2} can be easily obtained in the classical case by writing down $\langle \mathbf{s}_i \cdot \mathbf{s}_{i+1}\rangle = \cos 2\theta$ and $hs = h\cos\theta$, and minimizing the energy with respect to the angle θ; this yields $h_{c_2}^2 = 8(J_1 + J_3)^2(1 - J_2/J_3)$. J^τ reverses sign at $h > h_{c2}$, and the orbital ordering becomes antiferromagnetic with $\langle \tau_i \tau_{i+1}\rangle = -1$. But then, in accordance with (2), the spin exchange also decreases jumpwise (to $J^s = J_1 - J_3$), leading to a jumplike increase of the magnetization. In fact, h_{c_2} is the "overheat field"; on the other hand, going from the side of single fields, the phase with $\langle \tau_i \tau_{i+1}\rangle = -1$ can be preserved down to the field h_{c_1}, ($h_{c_1}^2 = 8(J_1 - J_3)^2(1 - J_2/J_3)$). We

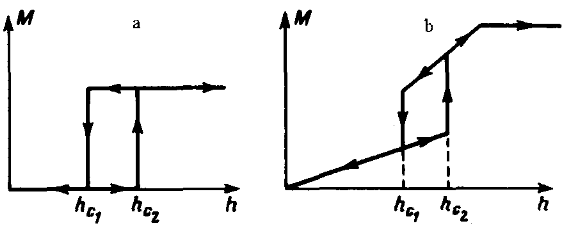

FIG. 1. Dependence of the magnetic moment on the field in the model (1): a—Ising variant, b—Heisenberg variant.

thus have a first order transition. The behavior of the moment M at $T=1$ is a function of h is shown schematically in Fig. 1.

It can be assumed that the jumplike character of the transition is preserved also at $T=0$ in a certain phase-diagram region. In the absence of a field, generally speaking, there are two phase transitions (ordering in s and in τ), which occur at different temperatures and are, at least in the self-consistent-field approximation,[9] transitions of second order (see also[8]). A tricritical point

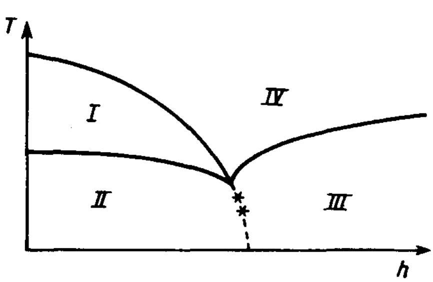

FIG. 2. Schematic form of the phase diagram at $J_1 > J_3 > J_2 > 0$: I—phase antiferromagnetic in s and paramagnetic in τ; II—phase antiferromagnetic in s and ferromagnetic in τ; III—phase with partially (Heisenberg case) or fully (Ising case) collapsed sublattices, antiferromagnetic in τ; IV—phase paramagnetic in s and τ. The asterisk marks the possible position of the tricritical point. Solid curves—second-order transitions, dashed—first-order transitions.

should therefore exist on the $(T-h)$ phase diagram (its possible form is shown in Fig. 2).

In addition to this general analysis, we obtain an exact solution of the corresponding one-dimensional problem for an Ising interaction. The free energy is given in this case by

$$F = -TN\ln\left[2\left\{e^{-\beta J_1}\operatorname{ch}\beta(J_2+J_3)\operatorname{ch}\beta h + e^{\beta J_1}[\operatorname{ch}^2\beta(J_2-J_3) + \operatorname{ch}^2\beta(J_2+J_3)\operatorname{sh}^2\beta h]^{1/2}\right\}\right], \qquad \beta = 1/kT. \tag{3}$$

An analysis of (3) confirms our conclusions in general outline, particularly the form of the phase diagram (Fig. 2), although, of course, there are no true phase transitions in the one-dimensional case. The corresponding lines on the phase diagram were determined from the maxima of the heat capacity.

We have also carried out, in the self-consistent-field approximation a concrete calculation of the influence of the magnetic field on substances with perovskite structure, of the $KCuF_3$ type, [10] using the exchange Hamiltonian obtained in[5]. Qualitatively, the behavior of the magnetization is close to that shown in Fig. 1b. At h_c, a transition takes place from a spin structure of the type of antiferromagnetically coupled ferromagnetic planes to a pure two-sublattice antiferromagnetism.

We note in conclusion certain existing experimental data can be connected with the described metamagnetism mechanism. Thus, in[11], in an investigation of the garnet $Ca_3Mn_2Ge_3O_{12}$ ($T_N \approx 14\,^\circ$K) with Mn^{3+} ions in octahedral interstices, a metamagnetic transition was observed at $h_c \sim 15$ kOe. It is difficult to associate this behavior with anisotropy, for it would be necessary to assume that the anisotropy field is of the order of the exchange field, an unlikely assumption for Mn^{3+}; nor does this agree with the fact that at $h \gtrsim h_c$ the magnetization is $\sim \frac{1}{3}$ of the saturation magnetization.

Among substances with cooperative Jahn-Teller ordering of the orbitals, the tricitical point was observed in DySb [12] and in a number of systems. It would be of interest to investigate substances of the $KCuF_3$ type in a field. The metamagnetic behavior field should then be observed when an important role is played by the superexchange mechanism of orbital ordering. On the other hand, if the orbital ordering is determined experimentally by the electron-vibrational (Jahn-Teller) interaction, then these effects, generally speaking, may also not be realized.

1)Institute of High Temperatures, USSR Academy of Sciences.
2)Metamagnetism is possible also in systems with isotropic biquadratic exchange. [2,3]

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