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Character of phase transitions to a helical or sinusoidal state in magnetic materials

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It is emphasized that the character of the transition from the paramagnetic state to a sinusoidal or spiral state depends significantly on the form of the magnetic interactions. Thus if there are only exchange interactions, of if they significantly exceed the spin-orbit or dipole-dipole forces, then only a first-order transition is possible. A transition of the second-order is possible only for a definite finite intensity of the relativisitic interactions. In this latter case, there are two tricritical points on the phase diagram.

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1. INTRODUCTION

Phase transitions in magnetic materials, especially transitions from the paramagnetic to the ferro- or antiferromagnetic state, are excellent examples of transitions of second order. But a large number of cases are now known in which magnetic phase transitions that, according to the symmetry considerations characteristic of Landau's theory (see, for example, Ref. 1), could be transitions of second order are actually found to be transitions of first order. A large number of examples of this can be found in the review by Grazhdankina.[2] New facts not mentioned in this review are reported in the articles[3–6] cited below. Until recently, such first-order transitions were explained within the framework of molecular-field theory, by the action of specific forces of rather large (in comparison with ordinary exchange) intensity: magnetostriction, biquadratic exchange, the Jahn-Teller effect, etc. (for details, see Ref. 2). Recently, however, it has been clarified that fluctuations of the short-range order, which increase with approach to a point of (in principle possible) second-order transition, act in the same direction as the just mentioned magnetostriction, biquadratic exchange, etc., tending to convert the continuous transition to a discontinuous one. Furthermore, it has been shown in papers of Brazovskiĭ, Kukharenko, and the author,[3,4] of Bak, Krinsky, and Mukamel,[5] and of Alessandrini, Cracknell, and Przystawa[6] that in certain cases the fluctuations make second-order transitions altogether impossible; that is, the transition is discontinuous even for arbitrarily weak interactions of the biquadratic-exchange type. Transitions of this form include those in MnO, UO_2, TbAs, etc.

From the point of view of molecular-field theory, what causes the transition to become discontinuous is a sufficiently strong nonlinear coupling between the effective field H_{eff} and the order parameter M:

$$H_{eff}=JM+jM^3+\dots$$

For weak nonlinearity, $j \ll J$, the variation of M with temperature T is described by the usual Langevin-Brillouin curve (Curve 1 in Fig. 1). For sufficiently large j, M as a function of T ceases to be single-valued (Curve 3 in Fig. 1); this obviously implies a transition of the first order at some temperature T_0. From a physical point of view, the nonlinearity j originates from the already mentioned biquadratic exchange, magnetostriction, etc. A characteristic feature of such first-order transitions is the presence of a tricritical point T_{c0}, at which a line of first-order transitions is converted to a line of second-order transitions.

The effect of fluctuations on the character of the transition is as follows. In all cases analyzed, the magnetic phase that originates as a result of the transition is characterized by the presence of a large number of equivalent domains. This, for example, is 8 so-called T-domains in MnO, TbAs, etc. (see Refs. 3–5) or 6 domains in UO_2 (see Refs. 3–6). On approaching the transition point from above, the system of course cannot "know" into just which one of the 8 or 6 domains (or

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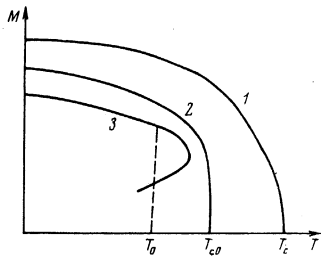


FIG. 1.

into a state described by a "linear combination" of them) it will actually be converted. The system, as it were, becomes confused and as a result is unable to reach a state with an infinite correlation radius. From the point of view of the modern theory of phase transitions (scaling), all stationary points of the renormalization group (RG) that correspond to transitions of the second order are unstable, and the system "wanders" until it ultimately reaches the stability boundary of the paramagnetic phase (see Refs. 3 and 4).

In this article, we shall consider the effect of fluctuations on the character of the phase transitions to a helical or sinusoidal state, restricting ourselves principally to the simplest case, in which there is only one spiral or sinusoidal wave, with vector **Q** (directed, of course, along the crystal axis in tetrangonal, rhombohedral, or hexagonal systems or along any one of the three crystallographic axes in systems of lower symmetry). It turns out that if we completely neglect all interactions except exchange, then continuous transitions are again impossible. The reason is that above the transition the system knows only one thing about the state to which it will change: the wave vector **Q**. The "virtual transitions" (fluctuations) to any one of the three possible sinusoidal waves are equally probable, and the virtual transitions to the spiral states have, in order of magnitude, the same probability.

The character of the transition may change when spin-orbit and dipole-dipole forces are included. In the "easy axis" variant, all virtual states except one sinusoidal wave are suppressed. In this case, only a transition of the second order is possible. In the "easy plane" variant, two virtual sinusoidal waves and one spiral survive. The character of the transition in this case is determined by the relation between the exchange and the relativistic forces. Tuus there is overlap (crossover) between transitions of the first order (corresponding to the maximum number of fluctuating fields) and transitions of the second order (corresponding to a smaller number of fields). Such a crossover point is of course an ordinary tricritical point.

The situation is in principle no different when there are several waves with different vectors $\mathbf{Q}_1$, $\mathbf{Q}_2$, ... (Sec. 3).

The effect of fluctuations on transitions to sinusoidal and spiral states was first considered in the papers, already cited, by Bak *et al.*[5] They studied the transition in Cr and Eu in the strong-anisotropy case and showed that in these materials a transition of the second order is impossible.

2. A SINGLE SPIN-DENSITY WAVE

This includes a large number of magnetic structures observed, for example, in Tb, Dy, Ho, MnO_2, etc.; that is, in uniaxial crystals with hexagonal or tetragonal symmetry, with the wave vector **Q** directed along the crystal axis z. The magnetic structure of such materials is described by a single complex vector

$$\mathbf{S}=\mathbf{S}_0 e^{iQz}$$

or by its real and imaginary parts,

$$\mathbf{S}=\mathbf{s}_+ + i\mathbf{s}_-.$$

A sinusoidal wave corresponds to $\mathbf{s}_+ \parallel \mathbf{s}_-$, a spiral to $s_+ = s_-$ and $\mathbf{s}_+ \perp \mathbf{s}_-$.

We shall construct a theory of phase transitions, with allowance for fluctuations, using Wilson's ε-expansion method in the specific form employed in our previous papers.[3,4] For this purpose it is necessary to write the Landau free energy including terms of the fourth order in **S**. We shall take into account two second-order invariants,

$$\mathbf{S}\mathbf{S}^*, \quad S_z S_z^*$$

and two fourth-order exchange invariants,

$$(\mathbf{S}\mathbf{S}^*)^2, \quad S^2 S^{2*}.$$

We shall neglect nonexchange invariants of the type $S_z^2 S_z^{2*}$, supposing that the relativistic interactions are small.

The free energy, expressed in terms of $\mathbf{s}_+$ and $\mathbf{s}_-$, has the form

$$\begin{aligned} F=&{}^1/_2\tau\mathcal{T}(\mathbf{s}_+^2+\mathbf{s}_-^2)+{}^1/_2 a(s_{+z}^2+s_{-z}^2)\\ &+{}^1/_8\Gamma_1(s_+^4+s_-^4)+{}^1/_4(\Gamma_1-2\Gamma_3)s_+^2 s_-^2+{}^1/_2\Gamma_3(\mathbf{s}_+\mathbf{s}_-)^2, \qquad (1)\\ &\tau=(T-T_c)/T_c. \end{aligned}$$

In Wilson's method, $\mathcal{T}$, Γ_1, and Γ_3 are themselves functions of $s_0^2=s_+^2+s_-^2$ and of τ (see Refs. 3 and 4):

$$\begin{aligned} &\Gamma, \mathcal{T}=\Gamma(\xi), \mathcal{T}(\xi),\\ &\xi=\frac{1}{\varepsilon}\left[\left(\frac{\Lambda}{\max(\tau, s_0^2)}\right)^{\varepsilon}-1\right], \quad \varepsilon\to 0. \qquad (2) \end{aligned}$$

The dependence of Γ on ξ is given by the equations of the renormalization group (RG) or, what amounts to the same thing, by the so-called parquet equations. The method of parquet equations, in its application to the the theory of phase transitions, may be found in the papers of Larkin and Khmel'nitskiĭ and of Abrahams *et al.*[7] The equations are determined by the graphs in Fig. 2, where the lines denote functions of the spin correlation

$$G_{ik}^{++}=-\langle s_{+i}s_{+k}\rangle=G_{ik}^{--}=-\langle s_{-i}s_{-k}\rangle=G_{ik};$$
$$i, k=x, y, z.$$

For anisotropy of the "easy plane" type (that is, $a>0$),

$$\begin{aligned} &G_{zz}=\frac{1}{\tau+a+bq^2}, \quad \tau\to 0;\\ &G_{\alpha\beta}=\frac{1}{\tau+bq^2}\delta_{\alpha\beta}, \quad \alpha, \beta=x, y, \end{aligned} \qquad (3)$$

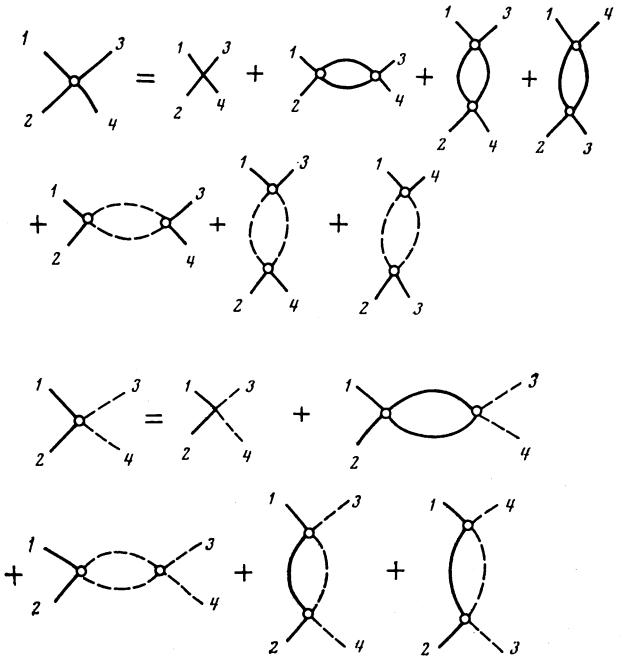


FIG. 2.

For anisotropy of the "easy axis" type ($a<0$),

$$G_{zz}=\frac{1}{\tau'+bq^2}, \quad \tau'=\tau+a\to 0;$$
$$G_{\alpha\beta}=\frac{1}{\tau'-a+bq^2}\delta_{\alpha\beta}. \tag{4}$$

The solid lines in Fig. 2 denote components of $G_{\alpha\beta}$, the broken lines components of G_{zz}.

Sufficiently far from the transition point, when τ and $\tau' \gg |a|$, the contributions of all the loops in the diagrams of Fig. 2 are the same, regardless of whether they are made up of broken or of solid lines, and are proportional to ξ of (2). The problem in this range is equivalent to a phase transition described by the two three-dimensional vector fields $\mathbf{s}_+$ and $\mathbf{s}_-$ and by the energy (1) with $a=0$. This problem was considered by us earlier (Ref. 4, Sec. 3), and in fact for a more general expression for the energy (see (11) in Ref. 4), with an arbitrary coefficient Γ_2 in the $s_+^2 s_-^2$ term instead of $\Gamma_2 = \Gamma_1 - 2\Gamma_3$. The RG equations in this case have the form (12) of Ref. 4. It is easily seen that from this system one can derive the equation

$$-\frac{d}{d\xi}(-\Gamma_1+\Gamma_2+2\Gamma_3)=(-\Gamma_1+\Gamma_2+2\Gamma_3)(11\Gamma_1+\Gamma_2+10\Gamma_3), \tag{5}$$

whence it follows that the system has the particular integral

$$-\Gamma_1+\Gamma_2+2\Gamma_3=0. \tag{6}$$

The existence of the integral (6) obviously implies that the expression (1) for the energy remains invariant under the action of the RG.

Knowing the solutions of the parquet equations for Γ, we can easily find the expression (1) for the free energy. The following method leads most rapidly to the desired result (it was once applied by Larkin and Khmel'nitskiĭ[7] to the case of a single charge).

Suppose that we know an exact expression for the free energy as a function of the main values of the components $\eta_1, \eta_2, \ldots$ of the order parameter. Then the relation

$$\frac{\partial^4 F}{\partial\eta_a\partial\eta_b\partial\eta_c\partial\eta_d}=\Gamma_{abcd}(k=0,\eta,\tau), \tag{7}$$

holds, where the right member is the exact vertex part for zero momentum. The relation (7) can be derived, for example, by differentiation of the diagram series for F. Conversely, if, as in our case, the Γ_{abcd} are known, the relations (7) may be regarded as differential equations for finding the free energy. They must be supplemented by two boundary conditions:

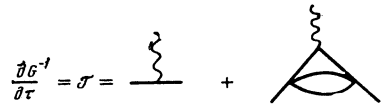
$$\frac{\partial^2 F}{\partial\eta_a\partial\eta_b}\Big|_{\eta=0}=G_{ab}^{-1}(k=0,\eta=0,\tau),$$
$$\frac{\partial^3 F}{\partial\eta_a\partial\eta_b\partial\eta_c}\Big|_{\eta=0}=\frac{\partial F}{\partial\eta_a}\Big|_{\eta=0}=F(\eta=0)=0. \tag{8}$$

The second is essentially the definition of the η_a-dependent part of the free energy, while the first is again obtained by differentiation of the diagrams.

Finally, in order to calculate $G_{ab}^{-1}(\eta=0, \tau)$ it is convenient to use Ward's identity

$$\frac{\partial G_{ab}^{-1}(k=0,\eta=0)}{\partial\tau}=\delta_{ab}\mathcal{T}(\tau), \tag{9}$$

where the three-point vertex $\mathcal{T}$ is expressed as follows:



Summation of these diagrams in the parquet approximation gives the differential equation for $\mathcal{T}(\xi)$:

$$d\mathcal{T}(\xi)/d\xi=-\mathcal{T}(\xi)\tilde{\Gamma}(\xi), \quad \mathcal{T}(0)=1, \tag{10}$$

where $\tilde{\Gamma}$ is a combination of the components Γ_{abcd} formed in a definite manner:

$$\tilde{\Gamma}\delta_{ab}=\sum_c \Gamma_{ccab}. \tag{11}$$

The solution of (9) is obviously given by the formula

$$\mathcal{T}(\xi)=\exp\Big\{-\int_0^\xi \tilde{\Gamma}(\eta)\,d\eta\Big\}. \tag{12}$$

In the parquet approximation, Γ and $\mathcal{T}$ are slowly varying functions of τ and η: $\Gamma\equiv\Gamma(\xi)$, $\mathcal{T}\equiv\mathcal{T}(\xi)$ with ξ from (2); therefore they may be considered constant during the integration over the "fast" variables τ and η in (7), (8), and (9). Then from (9)

$$G_{ab}^{-1}(\eta=0)=\delta_{ab}\tau\mathcal{T}(\xi),$$

and from (7) and (8) we get the expression (1) for the free energy.

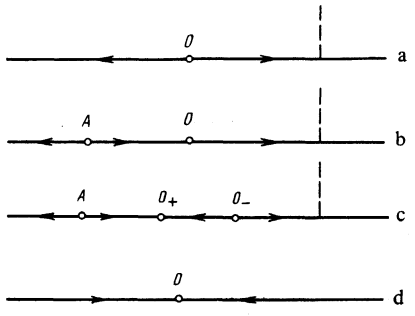


FIG. 3.

We return to the parquet equations given in a previous paper[4] (formula (12)). With allowance for the condition (6) they take the form

$$-\Gamma_1'=11\Gamma_1^2-8\Gamma_1\Gamma_3+8\Gamma_3^2,\quad -\Gamma_3'=6\Gamma_3(2\Gamma_1-\Gamma_3). \tag{13}$$

This system has only one projective stationary point O: $\Gamma_1=1/14\xi$, $\Gamma_3=0$. On introducing the new variable $U=\Gamma_3/\Gamma_1$ and dividing one of equations (13) by the other, we get

$$\Gamma_1\frac{dU}{d\Gamma_1}=-U\frac{1-U+4U^2}{7-4U+4U^2}. \tag{14}$$

From the first equation (13) it follows that Γ_1 is a decreasing function of ξ. The "trajectory" of the system (14) in one-dimensional U space is shown in Fig. 3a. The only stationary point of (14), the point $U=0$, is unstable with increase (decrease) of Γ_1. Therefore a transition of second order in such a system is impossible (see Refs. 3–6). The system will move until it reaches a boundary of the region of positive definiteness of the fourth-order terms in (1), where a transition of first order will occur (see Refs. 3 and 4). These boundaries were found earlier (see Ref. 4, Sec. 3). One of them (with allowance for the fact that $\Gamma_2=-\Gamma_1+2\Gamma_3$) is

$$\Gamma_1\geqslant 0. \tag{I}$$

On it, a transition occurs to a state with $s_+\neq 0$, $s_-=0$ or $s_+=0$, $s_-\neq 0$ or with $\mathbf{s}_+=\pm\mathbf{s}_-$. Both of these states are obviously states with a sinusoidal wave. In Fig. 3a, this transition corresponds to movement of the point U leftward to infinity.

A secondary boundary is

$$\Gamma_3\leqslant\Gamma_1,\quad \Gamma_1>0. \tag{II}$$

On it, a transition occurs to a state with

$$s_+=s_-,\quad \mathbf{s}_+\perp\mathbf{s}_-,$$

that is, to a helical wave. This transition is described by movement of the point U rightward to the stability boundary (region II) $U=1$, marked in Fig. 3a by the broken line.

Thus the character of the transition depends on the "initial conditions" $\Gamma_1(0)$ and $\Gamma_3(0)$ for the RG equations (13); that is, on the values of the coefficients in the Landau energy (1) far from the transition point ($\xi\to 0$), where molecular-field theory is valid. When $\Gamma_1(0)>\Gamma_3(0)>0$ ($0<U_0<1$), we have a first-order transition to a spiral structure; and when $\Gamma_3(0)<0$ ($U_0<0$), a first-order transition to a sinusoidal structure. The transition temperatures are determined by the RG equations (13) (see Refs. 3 and 4). Their solution has the form

$$\begin{aligned}&\frac{\Gamma_1(\xi)}{\Gamma_1(0)}=\left(\frac{U_0}{U}\right)^7\left(\frac{4U^2-U+1}{4U_0^2-U_0+1}\right)^3,\\ &2\Gamma_1(0)\xi=\frac{(4U_0^2-U_0+1)^3}{U_0^7}I(U,U_0);\\ &I(U,U_0)=\int_{U_0}^{U}\frac{U^6\,dU}{(4U^2-U+1)^4}.\end{aligned} \tag{15}$$

It was shown earlier[3,4] that the first-order transition temperature τ_0 is determined by the value

$$\xi_0=\frac{1}{\varepsilon}\left[\left(\frac{\Lambda}{\tau_0}\right)^\varepsilon-1\right],$$

at which the stability of the paramagnetic phase is lost; that is, by the root of the equation $\Gamma_1(\xi_{0s})=0$ for a sinusoidal transition and by the root of $\Gamma_1(\xi_{0h})-\Gamma_3(\xi_{0h})=0$ for a helical transition. We have

$$\begin{aligned}&2\lambda\Gamma_1^2(0)\xi_{0s}=I(-\infty,U_0),\ U_0<0;\\ &2\lambda\Gamma_1^2(0)\xi_{0h}=I(1,U_0),\quad U_0>0;\\ &\lambda=U_0^7/(4U_0^2-U_0+1)^3.\end{aligned}$$

When the initial points lie close to the unstable stationary point $U=0$ ($\Gamma_3=0$) of the RG equations, the transition temperature $\tau_0\to 0$. The system moves "very slowly" (in time ξ) to the transition point. In the limit,

$$\xi_{0s,h}\cong 1.24\cdot 10^{-3}\Gamma_1^6(0)/|\Gamma_3(0)|^7.$$

In accordance with the previous work,[3,4] the main value of the spins at the transition point, s_0, is determined by the behavior of the vertices Γ_1 and $\Gamma_1-\Gamma_3$ close to their zeros ($\Gamma_1\cong\mu(\xi_{0s}-\xi)$) and by the value of $\mathcal{F}$ at the transition temperature, $\mathcal{F}(\xi_0)$, by means of the relation

$$s_0\sim(\tau_0\mathcal{F}(\xi_0)/\mu)^{1/4}.$$

For Γ_1 and $\Gamma_1-\Gamma_3$ near the transition we have

$$\Gamma_1\cong 2^{15}\Gamma_1^2(0)\lambda^2(\xi_{0s}-\xi), \tag{I}$$

$$\Gamma_1-\Gamma_3\cong 2^{15}\Gamma_1^2(0)\lambda^2(\xi_{0h}-\xi). \tag{II}$$

The quantity $\tilde{\Gamma}$ of (11) in our case is equal to

$$\tilde{\Gamma}=4(2\Gamma_1-\Gamma_3).$$

By use of formula (15) we find, in accordance with (12),

$$\mathcal{F}(\xi)=\left(\frac{4U^2-U+1}{4U_0^2-U_0+1}\right)^2\frac{U_0^4}{U^4}$$

whence

$$\mathcal{F}(\xi_0)=\left(\frac{4U_0^2}{4U_0^2-U_0+1}\right)^2.$$

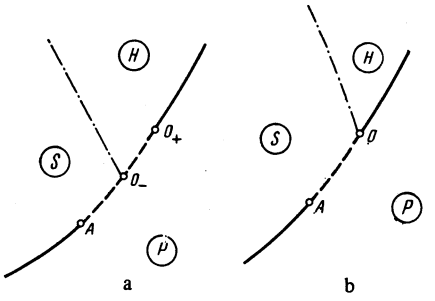


FIG. 4.

Finally, we have

$$s_0^2 \cong \tau_0[4\Gamma_3^2(0)-\Gamma_3(0)\Gamma_1(0)+\Gamma_1^2(0)]^2/16\Gamma_3^5(0).$$

This treatment is valid only with neglect of anisotropy (the term $a(s_{+z}^2+s_{-z}^2)$ in (1)). This means that the temperature τ must not be too small:

$$\tau \geqslant \tau_0 > a,$$

or in terms of ξ,

$$\xi \leqslant \xi_0 < \xi_a = \frac{1}{\varepsilon}\left[\left(\frac{\Lambda}{a}\right)^{\varepsilon}-1\right].$$

If the transition temperature τ_0 is smaller than a, then different loops in Fig. 2 will have different values. With "easy plane" symmetry, loops formed by solid lines will again be proportional to $\xi > \xi_a$, while loops containing at least one broken line will be smaller, $\sim \xi_a$. Thus crossover will occur from the free-component vectors $\mathbf{s}_+$, $\mathbf{s}_-$ to two-component vectors σ_+, σ_- lying in the "easy plane."

The system of RG equations, in the leading-term approximation with respect to $\xi > \xi_a$, will be determined formally by the same energy (1) with $\mathbf{s}_+$ and $\mathbf{s}_-$ replaced by σ_+ and σ_- (the coefficients Γ_1, $\Gamma_2 \to \gamma_1$, γ_2). The RG equations for γ_1 and γ_2 must now be solved with the "initial conditions," at $\xi = \xi_a$,

$$\gamma_{1,3}(\xi_a) = \Gamma_{1,3}(\xi_a),$$

where $\Gamma(\xi_a)$ are determined by formulas (15).

The equations for γ_1, γ_2 (corresponding to Γ_2), and γ_3 have the form

$$\begin{aligned} -\gamma_1' &= 10\gamma_1^2+2\gamma_2^2+4\gamma_2\gamma_3+4\gamma_3^2, \\ -\gamma_2' &= 8\gamma_1\gamma_2+4\gamma_1\gamma_3+4\gamma_2^2+4\gamma_3^2, \\ -\gamma_3' &= 4\gamma_3(\gamma_1+2\gamma_2+2\gamma_3). \end{aligned}$$

This system again has the integral $-\gamma_1+\gamma_2+2\gamma_3 = 0$:

$$-(-\gamma_1+\gamma_2+2\gamma_3)' = (-\gamma_1+\gamma_2+2\gamma_3)(10\gamma_1+2\gamma_2+8\gamma_3).$$

The RG equations for γ_1 and γ_3 are

$$-\gamma_1' = 12\gamma_1^2-4\gamma_1\gamma_3+4\gamma_3^2, \quad -\gamma_3' = 4\gamma_3(3\gamma_1-2\gamma_3), \tag{16}$$

whence we find

$$\gamma_1\frac{du}{d\gamma_1} = -u^2\frac{1+u}{3-u+u^2}, \quad u = \frac{\gamma_3}{\gamma_1}.$$

The system (16) has two unstable stationary points O ($u_0 = 0$) and A ($u_A = -1$) on the u axis (Fig. 3b). The point O is actually the result of fusion of two simple stationary points: a stable one O_- and an unstable O_+ (Fig. 3c). The fusion occurs only in the linear approximation with respect to ε. The stationary points of the RG determined by the energy (1) with the two-component vectors σ_+, σ_- in the ε^2 approximation have been investigated by Mukamel[8] (see also Ref. 5). The stable point O_- corresponds to his point 8, with coordinate $u = -\varepsilon/2$; the unstable point O_+ corresponds to point 5 of Mukamel's paper and has the coordinate $u_+ = 0$. In our further calculations we shall restrict ourselves to the linear approximation in ε and shall suppose that the points O_+ and O_- have fused. The character of the transition is now determined by the location of the initial point

$$u_0 \equiv U_a = \Gamma_3(\xi_a)/\Gamma_1(\xi_a).$$

In the range $u_0 < u_A = -1$, the system moves to $-\infty$, where, as before, a first-order transition occurs to the sinusoidal state. In the range $u_A < u_0 < u_+ = 0$, all trajectories converge to the stable point O_- ($u_- = -\varepsilon/2$), where a second-order transition occurs to the same sinusoidal state. The indices at this point were determined by Mukamel.[8] Finally, in the range $u_0 > u_+ = 0$, the system will again move to the stability boundary of the paramagnetic phase (again it is $\gamma_1 - \gamma_3 = 0$, $u = 1$), denoted in Fig. 3b and c by a broken line, where a first-order transition occurs to the helical state.

It is obvious that the points A and O_+ (or O in the linear approximation in ε) are tricritical points. Coexistence diagrams of the three phases—paramagnetic (P), sinusoidal (S), and helical (H)—are represented schematically in Fig. 4 (the solid lines are transitions of the first kind, the broken lines of the second). We have not studied the character of the transitions between the helical and sinusoidal states.

The coordinates of the tricritical points in our approximation are given by the following relations: for point A,

$$U_a = -1,$$

for point O,

$$\Gamma_3(0) = 0;$$

or (for point A)

$$2\lambda\Gamma_1^2(0)\xi_a = I(-1, \Gamma_3(0)/\Gamma_1(0)).$$

The first-order transition temperatures are now determined by the solutions of equations (16). We have

$$\frac{\gamma_1(\xi)}{\Gamma_1(\xi_a)} = \left(\frac{u}{U_a}\right)^4\left(\frac{1+U_a}{1+u}\right)^5\exp\left(\frac{3}{u}-\frac{3}{U_a}\right),$$
$$4\mu\Gamma_1(\xi_a)(\xi-\xi_a) = j(u, U_a); \tag{17}$$

$$j(u, U_a)=\int_{U_a}^{u}\frac{(1+u)^4}{u^6}e^{-3/u}\frac{3-u+u^2}{3-4u+4u^2}du,$$

$$\mu=\frac{(1+U_a)^5}{U_a^4}\exp\left(-\frac{3}{U_a}\right).$$

The temperature of transition to the sinusoidal state, ξ_{0s}, is given by the formula

$$4\mu\Gamma_1(\xi_a)(\xi_{0s}-\xi_a)=j(-\infty, U_a),\quad U_a<-1,$$

and to the helical state

$$4\mu\Gamma_1(\xi_a)(\xi_{0h}-\xi_a)=j(1, U_a),\ U_a>0.$$

In the vicinity of the tricritical points, the transition temperatures have the following form: near point A,

$$\xi_{0s}=1.3\cdot10^{-3}\frac{\Gamma_1^4(\xi_a)}{|\Gamma_1(\xi_a)+\Gamma_3(\xi_a)|^5},$$

near point O,

$$\xi_{0h}=1.7\cdot10^{-2}\frac{\Gamma_3^4(\xi_a)}{\Gamma_1^5(\xi_a)}\exp\left[\frac{3\Gamma_1(\xi_a)}{\Gamma_3(\xi_a)}\right].$$

We note also that the function $\tilde{\Gamma}$ of (11) now has the form

$$\tilde{\gamma}=2(3\gamma_1-\gamma_3).$$

Therefore

$$\mathcal{T}(\xi)=\mathcal{T}(\xi_a)\exp\left\{-\int_{\xi_a}^{\xi}(3\gamma_1-\gamma_3)d\xi\right\},$$

where $\mathcal{T}(\xi_a)$ is given by the formula presented above for $\xi<\xi_a$.

Finally, in regard to "easy axis" symmetry: in this case the situation is remarkably simple. When $\tau<a$, only fluctuations of the z components of the vectors s_+ and s_- are important. The corresponding energy has the form

$$F=\tfrac{1}{2}\tau\mathcal{T}(s_{+z}^2+s_{-z}^2)+\tfrac{1}{8}\gamma(s_{+z}^2+s_{-z}^2)^2, \tag{18}$$

and the RG equation for the only charge in this case, γ, can be written

$$-\gamma'=9\gamma^2.$$

It is to be solved with the initial condition

$$\gamma(\xi_a)=\Gamma_1(\xi_a).$$

The solution has the form

$$\gamma(\xi)=\frac{\Gamma_1(\xi_a)}{1+9\Gamma_1(\xi_a)(\xi-\xi_a)}.$$

It obviously describes a unique stable stationary point (Fig. 3d): a phase transition of second order.

The state diagram in the case of "easy axis" symmetry again has the form of Fig. 2b. The coordinates of both tricritical points are determined by the following relations (corresponding to equality of the transition temperatures τ_0 calculated in the exchange approximation and of the anisotropy energy): coordinates of the point O,

$$\xi_{0h}=\xi_a,$$

coordinates of the point A,

$$\xi_{0s}=\xi_a.$$

3. SEVERAL SPIN-DENSITY WAVES[1)]

The maximum number of such waves occurs for an arbitrarily directed vector **Q** in crystals with high symmetry. For simplicity, we shall restrict ourselves to the case of two and three waves, with vectors directed along symmetry axes.

Two waves occur when the **Q**'s are directed along the x and y (or [110] and [1$\bar{1}$0]) axes of a tetragonal crystal. Such are the magnetic structures in DyC_2 and $TbAu_2$. They are described by two complex vectors:

$$\mathbf{S}_1\sim\mathbf{S}_{10}e^{iQx},\ \mathbf{S}_2\sim\mathbf{S}_{20}e^{iQy}.$$

There are, correspondingly, three second-order invariants,

$$\mathbf{S}_1\mathbf{S}_1^*+\mathbf{S}_2\mathbf{S}_2^*,\quad S_{1z}S_{1z}^*+S_{2z}S_{2z}^*,$$
$$S_{1x}S_{1x}^*+S_{2y}S_{2y}^*,$$

of which the first is exchange and the other two relativistic, and four fourth-order exchange invariants,

$$(\mathbf{S}_1\mathbf{S}_1^*)^2+(\mathbf{S}_2\mathbf{S}_2^*)^2,\quad S_1^2S_1^{2*}+S_2^2S_2^{2*},$$
$$(\mathbf{S}_1\mathbf{S}_1^*)(\mathbf{S}_2\mathbf{S}_2^*),\quad (\mathbf{S}_1\mathbf{S}_2)(\mathbf{S}_1^*\mathbf{S}_2^*)+(\mathbf{S}_1\mathbf{S}_2^*)(\mathbf{S}_1^*\mathbf{S}_2).$$

The case of three waves occurs, for example, in cubic crystals, when the **Q**'s are directed along the three cubic axes (this includes the structures in TbD_2, Cr, and Eu), and in hexagonal or rhombohedral crystals, when the **Q**'s are directed along the three twofold axes (the structure of Nd). Such structures are described by three complex vectors,

$$\mathbf{S}_1\sim\mathbf{S}_{10}e^{iQx},\quad \mathbf{S}_2\sim\mathbf{S}_{20}e^{iQy},\quad \mathbf{S}_3\sim\mathbf{S}_{30}e^{iQz}$$

(for hexagonal crystals, x, y, and z are to be understood as coordinates along the three twofold axes); and, correspondingly, there are again four fourth-order exchange invariants,

$$(\mathbf{S}_1\mathbf{S}_1^*)^2+(\mathbf{S}_2\mathbf{S}_2^*)^2+(\mathbf{S}_3\mathbf{S}_3^*)^2,$$
$$S_1^2S_1^{2*}+S_2^2S_2^{2*}+S_3^2S_3^{2*},$$
$$(\mathbf{S}_1\mathbf{S}_1^*)(\mathbf{S}_2\mathbf{S}_2^*)+(\mathbf{S}_1\mathbf{S}_1^*)(\mathbf{S}_3\mathbf{S}_3^*)+(\mathbf{S}_2\mathbf{S}_2^*)(\mathbf{S}_3\mathbf{S}_3^*), \tag{19}$$
$$(\mathbf{S}_1\mathbf{S}_2^*)(\mathbf{S}_1^*\mathbf{S}_2)+(\mathbf{S}_1\mathbf{S}_2)(\mathbf{S}_1^*\mathbf{S}_2^*)+\ldots$$

For cubic crystals there are two quadratic invariants

$$\mathbf{S}_1\mathbf{S}_1^*+\mathbf{S}_2\mathbf{S}_2^*+\mathbf{S}_3\mathbf{S}_3^*,\quad S_{1x}S_{1x}^*+S_{2y}S_{2y}^*+S_{3z}S_{3z}^*,$$

an exchange and a relativistic; for hexagonal crystals, in addition to the exchange, there are two relativistic,

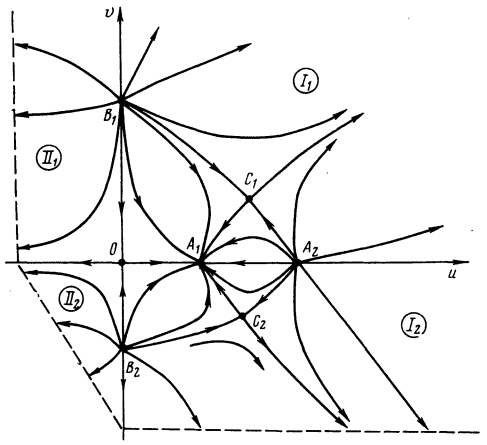


FIG. 5.

$$S_{1\xi}S_{1\xi}^{*}+S_{2\xi}S_{2\xi}^{*}+S_{3\xi}S_{3\xi}^{*},$$

$$S_{1\zeta}S_{1\zeta}^{*}+S_{2\eta}S_{2\eta}^{*}+S_{3\zeta}S_{3\zeta}^{*}, \tag{20}$$

where the coordinates ξ, η, and ζ are measured along the twofold axes.

In the case of a larger number of waves, nothing new appears. There are four fourth-order exchange invariants, of the same structure as (19), and two quadratic relativistic invariants, as in (20).

In the exchange approximation, the Landau energy is described by the single formula ($\mathbf{S}_a = \mathbf{s}_{a+} + i\mathbf{s}_{a-}$)

$$\begin{aligned}F=&{}^1/_2\tau\mathcal{T}(s_{1+}^2+s_{1-}^2+\ldots)+{}^1/_8\Gamma_1(s_{1+}^4+s_{1-}^4+\ldots)\\&+{}^1/_4(\Gamma_1-2\Gamma_3)(s_{1+}^2s_{1-}^2+\ldots)+{}^1/_2\Gamma_3[(\mathbf{s}_{1+}\mathbf{s}_{1-})^2+\ldots]\\&+{}^1/_2\Gamma_4[s_{1+}^2s_{2+}^2+s_{1+}^2s_{2-}^2+s_{1-}^2s_{2+}^2+s_{1-}^2s_{2-}^2+\ldots]\\&+{}^1/_2\Gamma_5[(\mathbf{s}_{1+}\mathbf{s}_{2+})^2+(\mathbf{s}_{1+}\mathbf{s}_{2-})^2+(\mathbf{s}_{1-}\mathbf{s}_{2+})^2+(\mathbf{s}_{1-}\mathbf{s}_{2-})^2+\ldots].\end{aligned} \tag{21}$$

To this must be added the second-order relativistic energy. For cubic crystals this is

$${}^1/_2a(s_{1+x}^2+s_{1-x}^2+s_{2+y}^2+s_{2-y}^2+s_{3+z}^2+s_{3-z}^2), \tag{22a}$$

for hexagonal and tetragonal

$${}^1/_2a_1(s_{1+z}^2+s_{1-z}^2+s_{2+z}^2+s_{2-z}^2+s_{3+z}^2+s_{3-z}^2)+{}^1/_2a_2(s_{1+x}^2+s_{1-x}^2+s_{2+y}^2+s_{2-y}^2+\ldots). \tag{22b}$$

It is clear that for hexagonal and tetragonal crystals, and also for a large number of waves, we always have to do with "easy axis" symmetry; that is (for large anisotropy), only with sinusoidal structures. It is only for cubic crystals with positive a in (22a) that the "easy plane" situation arises and spiral structures become possible.

At high temperatures, $\tau > a$, as in the preceding section, the RG equations for Γ_1, Γ_3, Γ_4, and Γ_5 in the exchange approximation are valid. We shall write them for the case of an arbitrary number n of waves and of an arbitrary dimensionality m of the spin ($m = 3$ for exchange forces, $m = 2$ for an "easy plane," $m = 1$ for an "easy axis"). We have

$$\begin{aligned}-\Gamma_1'&=(m+8)\Gamma_1^2+m\Gamma_2^2+4\Gamma_2\Gamma_3+4\Gamma_3^2\\&\quad+2(n-1)(m\Gamma_4^2+4\Gamma_4\Gamma_5+4\Gamma_5^2),\\-\Gamma_2'&=2(m+2)\Gamma_1\Gamma_2+4\Gamma_1\Gamma_3+4\Gamma_2^2+4\Gamma_3^2+2(n-1)(m\Gamma_4^2+4\Gamma_4\Gamma_5),\\-\Gamma_3'&=4\Gamma_1\Gamma_3+8\Gamma_2\Gamma_3+2(m+2)\Gamma_3^2+4(n-1)\Gamma_5^2,\\-\Gamma_4'&=2(m+2)\Gamma_1\Gamma_4+4\Gamma_1\Gamma_5+2m\Gamma_2\Gamma_4+4\Gamma_2\Gamma_5\\&\quad+4\Gamma_3\Gamma_4+2(mn-2m+2)\Gamma_4^2+8(n-2)\Gamma_4\Gamma_5+4\Gamma_5^2,\\-\Gamma_5'&=4\Gamma_1\Gamma_5+4\Gamma_3\Gamma_5+8\Gamma_4\Gamma_5+2(2n+m-2)\Gamma_5^2.\end{aligned} \tag{23}$$

The system of course has the particular integral $-\Gamma_1 + \Gamma_2 + 2\Gamma_3 = 0$, which again leads to invariance of the energy (21) with respect to the RG (23).

We note that the values of $\tilde{\Gamma}$ necessary for calculation of the coefficients $\mathcal{T}$ in the expression for the free energy are now given by the formula

$$\tilde{\Gamma}=2(m+1)\Gamma_1-2(m-1)\Gamma_3+2(n-1)m\Gamma_4+4(n-1)\Gamma_5.$$

Finding the stationary points of (23) is a very troublesome problem; it can be verified, in particular, that even the number of points depends on n. A previous test,[3 6] however, has shown that in all problems in which the dimensionality of the RG space was no less than two, i.e., the number of significant charges Γ was no less than three, all RG stationary points proved unstable. I am convinced that this fact is a general consequence of some kind of topological properties of the RG, connected, for example, with the positive definiteness of the energy and the monotonicity of the charge Γ_1. From this point of view, the possibility of transitions of second order is a trivial consequence of the one-dimensionality of the RG space (the number of charges is one or two), in which stable and unstable points always alternate. Therefore, without carrying out numerical calculations, I assume that all stationary points of (23) are unstable and, as a result, transitions of the second kind, as in Sec. 2, are impossible in the exchange region.

In support of this deduction, some formal considerations can be offered. As we have seen, the character the phase transition is determined by the behavior of the trajectories in the projective phase space of the variables Γ_2/Γ_1, Γ_3/Γ_1, $\Gamma_4/\Gamma_1, \ldots$. It can be shown that trajectories to infinity are always "outgoing" (see the figures in Refs. 3 and 4 and Figs. 3a–c and 5 in the present paper). Therefore in one-dimensional space, a necessary and sufficient condition for the occurrence of a stable stationary point is the presence of no fewer than three stationary points (Fig. 3c). In the two-dimensional case, a necessary condition is the presence of at least five such points (see Fig. 5), but this condition is not sufficient. In the case of a three-dimensional space that is of interest to us, the number of necessary points is very large, and this makes the occurrence of a stable point in a system of general form improbable.

The situation mentioned above of an "easy plane" in a cubic crystal is described by the same four charges as is the exchange case. Therefore we suppose that transitions of second order are again impossible. This deduction is supported by experiments on Cr and Eu, where in fact the transitions observed are of first order The same conclusion was reached by Bak, Krinsky, and

Mukamel,[5] who considered transitions of the "easy plane" type in Cr and Eu. They actually demonstrated the instability of all stationary points in a five-dimensional space, including also in their consideration two charges of nonexchange nature.

The "easy axis" situation is described by the energy

$$F=\tfrac{1}{2}\tau\mathcal{T}(s_{1+}^2+s_{1-}^2+\ldots)+\tfrac{1}{8}\gamma_1[(s_{1+}^2+s_{1-}^2)^2+\ldots] \\ +\tfrac{1}{4}\gamma_2[(s_{1+}^2+s_{1-}^2)(s_{2+}^2+s_{2-}^2)+\ldots], \quad (24)$$

where s_{1+}, s_{1-}, ... are the z components of the corresponding vectors. The RG equations can be obtained from (23) by noting that when $m=1$, the system (23) splits into three independent systems, of which the one for the variables

$$\gamma_1=\Gamma_1=\Gamma_2+2\Gamma_3, \quad \gamma_2=\Gamma_4+2\Gamma_5 \quad (25)$$

has the form

$$-\gamma_1'=10\gamma_1^2+2(n-1)\gamma_2^2, \quad -\gamma_2'=8\gamma_1\gamma_2+2n\gamma_2^2. \quad (26)$$

Comparison of the two expressions for the energy shows that the relations (25) correspond exactly to the transition from (21) to (24).

The system (26), as before, is to be solved with the boundary conditions at $\xi=\xi_a$

$$\gamma_{10}=\Gamma_1(\xi_a), \quad \gamma_{20}=\Gamma_4(\xi_a)+2\Gamma_5(\xi_a), \quad (27)$$

which insure a joining with the solutions of the system (23) in the exchange region.

The RG defined by the expressions (24) and (26) was investigated in a somewhat different form by Mukamel and Krinsky[5]; therefore we shall merely enumerate the results briefly. On transforming to the variable $u=\gamma_2/\gamma_1$, we get

$$\gamma_1\frac{du}{d\gamma_1}=-u(u-1)\frac{(n-1)u-1}{5+(n-1)u^2}.$$

This equation has three stationary points:

$$A:\ u=0; \quad O_-:\ u=1/(n-1); \quad O_+:\ u=1.$$

Of these, only the point O_- is stable, so that we again encounter the situation of Fig. 3c. The range of thermodynamic stability extends from $+\infty$ to the point $u_0=-1/(n-1)$, and the points A and O_+ are tricritical. The system (26) can be solved by quadratures, so that we are able to confirm all the calculations of the preceding section and to construct a state diagram of the type of Fig. 4.

For an "easy axis" of another type, for example for the case of TbD_2, where only the components s_{1x}, s_{2y}, and s_{3z} are nonzero (energy of type (22a)), or for the case of DyC_2 and $TbAu_2$, where in principle the pairs s_{1x}, s_{2y} or s_{1y}, s_{2x} may be nonzero (energy of type (22b)), the energy expression (24) and the RG equations (26) are retained. All that changes is the form of the initial conditions (27); they now read:

$$\gamma_{10}=\Gamma_1(\xi_a), \quad \gamma_{20}=\Gamma_4(\xi_a).$$

Thus with "easy axis" symmetry and with any number of waves, the state diagram of Fig. 4 is realized, with two tricritical points. We recall that with "easy plane" anisotropy in a cubic crystal, as in Cr and Eu, a transition of second order is in general impossible.

4. TRANSITION OF FIRST ORDER IN MnS_2

Recently Hastings and Corliss[9] established that the transition in MnS_2 is a transition of first order. The MnS_2 crystal has the cubic group $Pa3(T_h^6)$. The magnetic structure in it is described by three complex vectors

$$\mathbf{S}_1\sim\mathbf{S}_{10}e^{inx}, \quad \mathbf{S}_2\sim\mathbf{S}_{20}e^{iny}, \quad \mathbf{S}_3\sim\mathbf{S}_{30}e^{inz},$$

that is, it belongs to the case considered in the preceding section. But because the wave vectors $\mathbf{Q}$ occupy the symmetrical positions $\frac{1}{2}00$, $0\frac{1}{2}0$, $00\frac{1}{2}$, there is an additional exchange invariant,

$$S_1^4+S_2^4+S_3^4,$$

in consequence, the second fourth-order term in (21) will have an arbitrary coefficient Γ_2. It can also be shown that for MnS_2 there are three second-order invariants, i.e., "easy axis" anisotropy.

In the exchange approximation, the transition is again described by the system (23) with $m=n=3$. The corresponding projective space is four-dimensional—Γ_2/Γ_1, Γ_3/Γ_1, Γ_4/Γ_1, Γ_5/Γ_1—and therefore (see the note in the preceding section) stable points are certainly absent. The strong-anisotropy case was considered by Bak *et al.*[5] They showed that the corresponding system has a stable stationary point, so that a transition of second order is in principle possible. We shall show, however, that a continuous transition to the state observed experimentally is again impossible.

In the "easy axis" case, when each of the vectors $\mathbf{S}_1$, $\mathbf{S}_2$, and $\mathbf{S}_3$ has only one component, the energy expression (21) and the system (23) simplify greatly. In appropriate variables, they have the form

$$F=\tfrac{1}{2}\tau(s_{1+}^2+s_{1-}^2+\ldots)+\tfrac{1}{8}\gamma_1[(s_{1+}^2+s_{1-}^2)^2+\ldots] \\ +\tfrac{1}{4}\gamma_2(s_{1+}^2s_{1-}^2+\ldots)+\tfrac{1}{4}\gamma_3[(s_{1+}^2+s_{1-}^2)(s_{2+}^2+s_{2-}^2)+\ldots] \quad (28)$$

and

$$-\gamma_1'=10\gamma_1^2+2\gamma_1\gamma_2+\gamma_2^2+4\gamma_3^2, \\ -\gamma_2'=3\gamma_2(4\gamma_1+\gamma_2), \quad -\gamma_3'=2\gamma_3(4\gamma_1+\gamma_2+3\gamma_3). \quad (29)$$

Here

$$\gamma_1=\Gamma_1, \quad \gamma_2=-\Gamma_1+\Gamma_2+2\Gamma_3, \quad \gamma_3=\Gamma_4+2\Gamma_5$$

when all three vectors $\mathbf{S}_1$, $\mathbf{S}_2$, and $\mathbf{S}_2$ are parallel, and

$$\gamma_1=\Gamma_1, \quad \gamma_2=-\Gamma_1+\Gamma_2+2\Gamma_3, \quad \gamma_3=\Gamma_4,$$

when they are mutually perpendicular.

The trajectories of the system (29) in the plane

$$u=\gamma_3/\gamma_1, \quad v=\gamma_2/\gamma_1$$

are shown in Fig. 5. There are 7 stationary points:

$$O(00), A_1(1/20), A_2(10), B_1(02),$$
$$B_2(0-1), C_1(3/4\ 1/2), C_2(3/5-1/2).$$

The point A_1 was found to be stable; therefore, in principle, a continuous transition is possible from the region of initial values u_0v_0 bounded by the curve $OB_2C_2A_2C_1B_1$. The form of the magnetic structure that originates as a result of such a transition can be found by writing the energy (28) in the scaling range, where

$$\gamma_1\cong 2\gamma_3\cong \text{const}/\xi, \quad \gamma_2\cong 0.$$

We have ($\gamma_1>0$)

$$F_{(4)}\sim\gamma_1\{(s_{1+}^2+s_{1-}^2+s_{2+}^2+\ldots)^2-(s_{1+}^2+s_{1-}^2)(s_{2+}^2+s_{2-}^2)-\ldots\}.$$

It is clear this expression is smallest when all three of S_1, S_2, S_3 are simultaneously nonzero.

Other magnetic structures can occur only as a result of a transition of first order. A simple analysis of the energy expression (28) shows that (see the discussion in Ref. 4) loss of positive definiteness, and with it a transitionof first order, occur in the following four states:

I_1: $s_{1+}\neq 0$, $s_{1-}=0$, $s_{2+}=\ldots=0$.
I_2: $s_{1+}=s_{1-}\neq 0$, $s_{2+}=\ldots=0$.
II_1: $s_{1+}=s_{2+}=s_{3+}\neq 0$, $s_{1-}=\ldots=0$.
II_2: $s_{1+}=s_{1-}=\ldots\neq 0$.

A transition to the experimentally observed state I_1 occurs when the trajectory goes out to infinity in the first quadrant in Fig. 5. A transition to I_2 occurs when the trajectory intersects the dashed straight line $v+2=0$. State II_1 appears on the straight line $2u+1=0$, state II_2 on the straight line $4u+v+2=0$.

The considerations presented are of course also applicable to the transition in K_2IrCl_6 considered by Bak *et al.*[5]

[1)] All these cases have already been treated by Bak *et al.*[5] What is new in our investigation is the assertion that in the case of purely exchange forces, the transition is always a transition of the first order. In Ref. 5 can also be found a more detailed description of the magnetic structures and literature citations.

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