Mean-field phase diagrams of $AT_2X_2$ compounds

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Abstract

Magnetic-field – temperature phase diagrams of the axial next-nearest-neighbor Ising model are calculated within the framework of a Landau-type expansion of the free energy derived from molecular-field theory. Good qualitative agreement is found with recently reported results on body-centered-tetragonal $UPd_2Si_2$. This work is expected to also be relevant for related compounds.

75.10.Hk, 75.30.Kz, 75.50.Ee
There exists a large class of compounds with the body-centered-tetragonal (bct) structure of the generic formula \( AT_2X_2 \), where \( A \) represents either \( U \) or a rare-earth element, \( T \) is a transition metal, and \( X \) is either \( Si \) or \( Ge \). The materials of interest here have strong \( c \)-axial anisotropy, with the possibility of long-range Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions. It may thus be expected that the axial next-nearest-neighbor Ising (ANNNI) model and its extensions are relevant in cases where in-plane interactions are ferromagnetic.

It appears to be an empirical rule that such models yield a succession of \textit{principal} phases characterized by increasing periodicities as the temperature is increased. A counter example is found in the compound \( UNi_2Si_2 \), where a more complicated model was required to explain its observed sequence of magnetically ordered states (Ref. 1, hereafter referred to as I). Mean-field theory proved inadequate in this instance. All other relevant \( AT_2X_2 \) compounds, however, appear to exhibit magnetic structures consistent with expectations based on a mean-field approximation (see I). In the present work, the magnetic-field – temperature phase diagram of the ANNNI model is considered within the framework of a Landau-type free energy developed from molecular-field theory. Good qualitative agreement is found with recently reported results on \( UPd_2Si_2 \).

Magnetic interactions are assumed to be well described by a Heisenberg-type Hamiltonian of the form

\[
\mathcal{H} = -\frac{1}{2} \sum_{ij} J(r_i - r_j) \mathbf{s}(r_i) \cdot \mathbf{s}(r_j) - \mathbf{H} \cdot \sum_i \mathbf{s}(r_i),
\]

where the spin density \( \mathbf{s}(r_i) \), as well as the applied magnetic field \( \mathbf{H} \), are assumed to lie along the bct \( c \)-axis (\( \| \hat{z} \)). With ferromagnetic in-plane interactions \( J_0 > 0 \), the problem effectively becomes one-dimensional within mean-field theory. It is the magnetic ordering between the ferromagnetic planes separated by \( c' = \frac{1}{2}c \) which is of interest. A Landau-type free energy functional may be derived from molecular-field theory, based on the analysis of Bak and von Boehm (also see references in I), with results to sixth-order written as

\[
F = \frac{1}{2V} \int dzdz' A(\tau)s(z)s(z') + \frac{B}{4V} \int dz [s(z)]^4
+ \frac{C}{6V} \int dz [s(z)]^6 + \cdots - H \int dz s(z),
\]

(2)
where \( \tau = z - z' \) and

\[
A(z) = aT \delta(z) - j^2 J(z)/V, \quad (3)
\]

\( B = bT, \ C = cT \), with the coefficients \( a, b, c \) given in terms of the angular momentum \( j \) through the Brillouin function (see I). For simplicity, we take here the classical limit \( j \to \infty \) (with energies divided by \( j^2 \)) so that \( a = 3, b = \frac{9}{5}, c \simeq 1.697 \).

The ANNNI model with antiferromagnetic next-nearest-neighbor coupling, \( J_2 < 0 \), is frustrated. Numerous higher-order commensurate phases, as well an incommensurate state (IC), result from mean-field analyses of a model with near-neighbor interactions also antiferromagnetic, \( J_1 < 0 \), and with an additional small ferromagnetic third-neighbor coupling, \( J_3 > 0 \). Monte-Carlo simulations seem to suggest, however, that only a few principal commensurate phases survive the effects of critical fluctuations. For values of \( J_2/J_1 \) not too large, only two commensurate states appear: The period-2 \( \langle 1 \rangle \) state and the period-3 \( \langle 12 \rangle \) state (see Ref. for an explanation of the notation). It is precisely these two commensurate states which occur in the axial \( AT_2X_2 \) compounds (see I).

With the limited goal of describing these two commensurate, as well as the IC, states, the spin density can be written as

\[
s(z) = m + Se^{iQz} + S^*e^{-iQz}, \quad (4)
\]

where \( m \) is the uniform component, \( S \) is the complex polarization vector, and \( Q \) is the wave vector. Using this expression in (2) yields many types of Umklapp terms, non-zero only for \( nQ = G \), where \( G \) is a reciprocal lattice vector. For the purposes of the this work, contributions with \( n = 1 \) and \( n = 5 \) are omitted. It is instructive to present the many terms which are relevant, and to write these as

\[
F = F_2 + F_4 + F_6 - mH, \quad (5)
\]

with

\[
F_2 = \frac{1}{2}A_0m^2 + A_Q \mid S \mid^2 + \frac{1}{2}A_Q[S^2 + c.c.] \Delta_{2Q,G}, \quad (6)
\]
\[ F_4 = \frac{1}{4} B \{ S_T^4 + 2 | S^2 |^2 + 8 | mS |^2 \]
\[ + 2 [2(mS)^2 + S_T^2 S^2 + c.c.] \Delta_{2Q,G} \]
\[ + 4 [mS^3 + c.c.] \Delta_{3Q,G} + [S^4 + c.c.] \Delta_{4Q,G} \}, \quad (7) \]

\[ F_6 = \frac{1}{6} C \{ S_T^6 + 6 S_T^2 S^2 + 24 S_T^2 | mS |^2 + 12 [m^2 S^2 (S^*)^2 + c.c.] \]
\[ + 3 [4 S_T^2 (mS)^2 + S_T^4 S^2 + 8 | mS |^2 S^2 + | S^2 |^2 S^2 + c.c.] \Delta_{2Q,G} \]
\[ + 2 [6 S_T^2 mS^3 + 4 (mS)^3 + 3 (mS^*) S^4 + c.c.] \Delta_{3Q,G} \]
\[ + 3 [S_T^2 S^4 + 4 m^2 S^4 + c.c.] \Delta_{4Q,G} + [S^6 + c.c.] \Delta_{6Q,G} \}, \quad (8) \]

where \( S_T^2 = m^2 + 2 | S |^2 \). The coefficients in (6) are given by \( A_q = aT - J_q \), where \( J_q \) is the Fourier transform of the exchange integral,

\[ J_q = 4 J_0 + 2 [J_1 \cos q + J_2 \cos(2q) + J_3 \cos(3q)], \quad (9) \]

with \( q = cQ \). The commensurate states \( \langle 1 \rangle \) and \( \langle 12 \rangle \) are represented here by \( q = \pi \) and \( q = \frac{2}{3} \pi \), respectively. The IC phase is determined by the value of \( q \) which maximizes \( J_q \). This is usually the first ordered state which occurs as the temperature is lowered from the paramagnetic (P) phase, with the Néel temperature \( T_{N1} = J_q/a \). Due to the Umklapp terms, the free energies of each of these ordered states must be evaluated separately. None of the Umklapp terms contribute to \( F \) of the IC-phase. All non-Umklapp terms, as well as those with \( n = 2, 4, \) and 6, must be included in the free energy for the \( \langle 1 \rangle \)-phase, whereas only non-Umklapp terms and those with \( n = 3 \) contribute to the \( \langle 12 \rangle \)-phase. These separate free energies must be minimized as a function of the relevant variables in each case: \( F_{IC}(m, | S |, q) \), \( F_{(1)}(m, S) \), and \( F_{(12)}(m, | S |, \phi) \), where \( S = | S | e^{i\phi} \). The stable phase as a function of temperature, magnetic field and exchange parameters, is then determined by comparing the numerical results for these free energies. For the \( \langle 12 \rangle \)-state, the free energy is minimized by a phase angle \( \phi = (2m + 1)\pi/3 \), where \( m \) is an integer.

It is convenient to consider phases which occur as a function of \( J_2/J_1 \), with \( J_3/J_1 \) set to a small value. In the absence of a magnetic field, a number of sequences of phases results
from the present model depending on $J_2$. With increasing temperature, the following can occur (see I) $\langle 1 \rangle \rightarrow P$, $\langle 1 \rangle \rightarrow IC \rightarrow P$, $\langle 1 \rangle \rightarrow \langle 12 \rangle \rightarrow IC \rightarrow P$, $\langle 12 \rangle \rightarrow IC \rightarrow P$. The resulting $J_2/J_1 - T$ phase diagram corresponds qualitatively to the Monte-Carlo simulation results (see Fig. 1 of Ref. [8]). (The sequence $\langle 12 \rangle \rightarrow \langle 1 \rangle \rightarrow IC \rightarrow P$ observed in $UNi_2Si_2$ cannot be explained by mean-field theory.) All transitions except for $IC - P$ are first order.

It is important to note that the $n = 3$ Umklapp terms in (7) and (8) are linear in $m$. As a consequence, the spin density of the $\langle 12 \rangle$ phase has a non-zero uniform component even in the absence of an applied field. It is this coupling between $m$ and $S$ which enhances the stability of this state in the presence of a magnetic field.

Magnetic phase diagrams were calculated in the manner described above. Exchange parameters were set to $J_0 = 1$, $J_1 = -1$, and $J_3 = 0.03$. With these values, phases $\langle 1 \rangle$ and $\langle 12 \rangle$ are degenerate in energy at $T = 0$ with $J_2 \lesssim 0.35$ and the ordering wave vector in the $IC$ phase is approximately $\frac{3}{4}\pi$, as observed experimentally in some $AT_2X_2$ compounds (such as $UPd_2Si_2$). There is no temperature or field dependence of the $IC$ wave vector within the present model. This may be included by adding biquadratic exchange (see I).

Typical results shown in Figs. 1 and 2 demonstrate the increasing stability of the $\langle 12 \rangle$-phase with increasing field. (This feature is also observed in $UNi_2Si_2$, see I.) The phase diagram of Fig. 1 corresponds qualitatively to experimental results on $UPd_2Si_2$. Only relatively low field strengths were available in this experiment and the merging of the $\langle 12 \rangle - IC$ and $IC - P$ boundary lines was not observed, although there is clear indication of this tendency. For smaller values of $J_2/J_1$, the $\langle 12 \rangle$-phase may not occur at available field strengths. At even smaller values of this parameter, the $IC$-phase also disappears. For larger $J_2/J_1$, only the $\langle 12 \rangle$-state, and perhaps a small region of the $IC$-state, will occur in the phase diagram. At sufficiently large values of this parameter, a period-4 state is stabilized.

In conclusion, a simple mean-field model based on a Landau-type free energy derived from molecular-field theory has been shown to capture the essential features of the ANNNI model in a magnetic field. It describes many of the sequences of magnetically ordered states which occur in axial $AT_2X_2$ compounds. In particular, the magnetic phase diagram of
$UPd_2Si_2$ is well described by this model. It is hoped that this work will serve to stimulate further experimental investigation of magnetic field effects in this class of materials.

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REFERENCES

1 A. Mailhot, M.L. Plumer, A. Caillé, and P. Azaria, Phys. Rev. B 45, 10399 (1992).

2 W. Selke, Phys. Rep. 170, 213 (1988); J. Yeomans, in Solid State Physics, edited by H. Ehrenreich and D. Turnbull (Academic, New York, 1988), Vol. 41; W. Selke, in Phase Transitions and Critical Phenomena, edited by C. Domb and J.L. Lebowitz (Academic, New York, 1992), Vol. 15.

3 B. Shemirani et al., Phys. Rev. B 47, 8672 (1993).

4 M.F. Collins et al., Phys. Rev. B 48, 16500 (1993).

5 P. Bak and J. von Boehm, Phys. Rev. B 21, 5297 (1980).

6 Y. Yamada and N. Hamaya, J. Phys. Soc. Jpn. 52, 3466 (1983); W. Selke, M. Barreto, and J. Yeomans, J. Phys. C 18, L393 (1985).

7 W. Selke and M.E. Fisher, J. Mag. Mag. Materials 15-18, 403 (1980).

8 G.N. Hassold, J.F. Dreitlein, P.D. Beale, and J.F. Scott, Phys. Rev. B 33, 3581 (1986).

9 L. Rebelsky et al., Physica B 180-181, 43 (1992).
FIGURES

FIG. 1. Magnetic phase diagram with exchange parameters $J_0 = 1$, $J_1 = -1$, $J_2 = -0.30$, and $J_3 = 0.03$. Solid and broken curves represent first-order and continuous transitions, respectively.

FIG. 2. As in Fig. 1, with $J_2 = -0.35$. 