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On magnetic phase transitions in CeBi

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Résumé. — On peut interpréter les différentes phases magnétiques de CeBi à l'aide d'un modèle d'Ising anisotrope avec des interactions qui se contrarient entre les plans. Le diagramme de phase ainsi obtenu est comparé avec l'expérience.

Abstract. — We demonstrate how the numerous observed phases of CeBi may be interpreted by the anisotropic Ising model with competing interlayer interactions. The relevant phase diagram is found and compared to the experimental data.

During the last few years the magnetic properties of cerium monopnictide compounds (CeN, CeP, CeAs, CeBi, CeSb) have attracted a great deal of interest, both experimental and theoretical. All of these possess the rocksalt crystallographic structure, but they exhibit different magnetic behaviour. The experimental investigations have revealed the very complicated phase diagrams in the T - h plot (T -temperature, h -magnetic field) with 14 different magnetic phases for CeSb [1-4] and with 8 phases for CeBi [5, 6]. In all the phases these substances consist of Ising ferromagnetic planes with the moments normal to the planes and forming a one-dimensional periodical structure. Now the commonly accepted theoretical interpretation of the experimental data for CeSb makes use of an anisotropic Ising model [7-12]. In this model a modulated ground state is due to competing interactions of nearest and next-nearest neighbouring cerium ions. The analogous explanation holds for CeBi too, but the character of the competition differs from that for the case of CeSb.

The experimentally observed phase diagram for CeSb has been theoretically interpreted [11, 12] in the framework of the ANNNI (axial next-nearest-neighbours Ising) model with a strong anisotropy. Namely the intralayer ferromagnetic exchange integral ($I_0 < 0$) has been assumed much larger than the competing interlayer exchange constants I_1 and I_2 of nearest and next-nearest planes.

The main peculiarity of the phase diagram for CeBi is in the presence of the $\langle 3, 1 \rangle$ ⁽¹⁾ phase occupying a large domain of the diagram. Hence the interlayer bond I_3 enters the problem on an equal footing with I_1 and I_2 . A strong anisotropy is assumed for CeBi too ($|I_0| \gg I_1, I_2, I_3$). It enables the interlayer bonds to be treated using the high-temperature expansion in almost the whole region of existence of the ordered magnetic phases.

The Hamiltonian of the layered system has a form

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1, \quad \mathcal{H}_1 = \sum_{\mathbf{r}, j, l \neq 0} I_l s_j(\mathbf{r}) s_{j+l}(\mathbf{r}) \quad (1)$$

where $\mathcal{H}_0$ is the Hamiltonian of N uncoupled Ising planes, j labels the planes, $\mathbf{r}$ is the coordinate of the site in the plane.

The free energy of this model may be expanded as :

$$F = F_0 + \sum_{n=1}^{\infty} (-1)^{n-1} \langle\langle \mathcal{H}_1^n \rangle\rangle / T^{n-1} n! \quad (2)$$

Here the double angular brackets denote the irreducible average over the Gibbs ensemble of uncoupled Ising layers. Below the Onsager transition temperature, this results in a spontaneous magnetization which is of the same magnitude for each plane but may differ in sign :

$$\overline{s_j(\mathbf{r})} = s_j = s(T) \sigma_j, \quad \sigma_j = \pm 1. \quad (3)$$

Hence the problem reduces to the construction of the ground state of the one-dimensional Ising-like model with the « Hamiltonian » (2) parametrically depending on temperature. The thermal fluctuations generate the long-range interaction between planes.

Let us consider the case of interlayer bonds I_1, I_2, I_3 being positive-valued and of the same order of magnitude. The other « direct » ⁽²⁾ exchange constants are assumed to be rapidly decreasing with distance ($I_l \ll I_1$ for $l \geq 4$).

The magnetic interaction energy is :

$$F_h = -hs \sum_j \sigma_j. \quad (4)$$

Hence, if h satisfies

$$\max(\chi I_1^2, I_4) \ll h \ll T \quad (5)$$

one can keep only the first order terms in the free energy expansion (2) (FEE) along with the magnetic energy :

$$F_1 = s^2 \sum_j (I_1 \sigma_j \sigma_{j+1} + I_2 \sigma_j \sigma_{j+2} + I_3 \sigma_j \sigma_{j+3}). \quad (6)$$

Hereafter χ is the irreducible susceptibility of the plane Ising model. This theoretical description holds provided that :

$$\chi I_1 \ll 1 \quad (7)$$

where χI_1 is a perturbation series parameter.

⁽¹⁾ Here $\langle m_1^{n_1}, m_2^{n_2}, \dots \rangle$ denotes the elementary cell of a periodic one-dimensional spin structure. It is made of groups of m_i aligned spins, which make a chain of n_i groups with alternating projections, and so forth. For instance, $\langle 3, 1 \rangle$ has elementary cells either $+++-$ or $---+$, where $+$ or $-$ refers to the projection of the magnetic moment.

⁽²⁾ The « direct » exchange in rare-earth compounds is really indirect interaction of localized moments via conduction electrons. The word « direct » is used to distinguish this type of interactions from that due to thermal fluctuations.

The second order terms of FEE should be kept along with the first order terms only near the phase degeneration line (PDL). Along this line the contribution of fluctuations should be compared with the « direct » exchange constants of the relevant order. At not too low temperatures we put

$$I_4 \gg \chi I_1^2 \gg I_5, I_6, \dots \quad (8)$$

This gives a regular procedure to construct the « effective » Hamiltonian starting with FEE. In n 'th order ($n \geq 2$) we get :

$$F_n = s^2 (-\chi I_3)^{n-2} \sum_j (n-1) I_4 \sigma_j \sigma_{j+3n-2} \quad (9)$$

$$\tilde{F}_n = s^2 (-\chi I_3)^{n-1} \sum_j (n I_2 \sigma_j \sigma_{j+3n-1} + I_3 \sigma_j \sigma_{j+3n}). \quad (10)$$

The construction of the ground state of the effective Hamiltonian (4), (6), (9), (10) goes step by step and is connected with the solution of a certain simple linear programming problem. Analogous problems have been analysed in [9-12]. The proper variables for this method are the probabilities of different configurations of neighbouring spins in a chain. A convenient notation for them is like $(+ - + + \dots -)$. There exist sum rules : $(+) + (-) = I$, $(++) + (+-) = (+) + (-+) = (+)$, etc. The free energy is a linear function of these variables. The main part (4), (6) of the effective Hamiltonian leads to the phase diagrams depicted in figures 1, 2.

Fig. 1. — The phase diagram for the case $h = 3 I_3$ s. The temperature dependence of the I_1, I_2 couplings is depicted by the dashed line. The point A corresponds to low temperatures.

If the magnetic field changes the PDL shifts. Not only the long-range thermal fluctuation constants depend on temperature, but so also do the bonds I_1, I_2, I_3

$$I_1(T) = I_1 - \chi I_2(2 I_1 + 2 I_3) \quad (11)$$

$$I_2(T) = I_2 - \chi I_1(I_1 + 2 I_3) \quad (12)$$

$$I_3(T) = I_3 - 2 \chi I_1 I_2. \quad (13)$$

On the I_1 - I_2 plot there exists a contour (dashed lines in figures 1, 2) parametrically depending on temperature and corresponding to the initial set of the exchange constants (I_1, I_2, I_3) . It intersects the domain of phases $\langle 2, 2 \rangle$, $\langle 2, 1 \rangle$, $\langle 3, 1 \rangle$, AF, F.

Fig. 2. — The case $h = 5 I_3$ s. The notation is the same as in figure 1.

The perturbations (9)-(10) not only shift the PDL, but may also split them. As an example let us consider the PDL $\langle 3, 1 \rangle$ -AF. The following probabilities do not vanish along this line :

$$(+++-) = (+-+-) = (-+++) = (-+++) = (I - 2p)/4 \quad (14)$$

$$(+--+)=(-+-+)=p. \quad (15)$$

Here p is a degeneration parameter, $0 \leq p \leq 1/2$. In the vicinity of PDL $\langle 3, 1 \rangle$ -AF the free energy (4), (6) has the form

$$f = \xi p, \quad \xi = 2(I_2 - I_1 - I_3)s^2 + hs. \quad (16)$$

The parameter ξ alters its sign across this PDL. Obviously, f takes the extremal values at the vertex points $p = 0$ or $p = 1/2$.

With the second order terms included in the effective Hamiltonian (9), (10) one is led to consideration of quintets of nearest spins instead of quartets as above.

A new degeneration parameter q arises such that :

$$(-+-+)=q; (+++-)=(-++-)=p-q; (++++)=(I-6p+4q)/4. \quad (17)$$

Since the r.h.s. of equation (17) are probabilities, one has the following inequalities :

$$p \geq q \geq 0 \quad \text{and} \quad 6p - 4q \leq 1. \quad (18)$$

There are three vertex points in the (p, q) -space : $(0, 0)$, $(1/2, 1/2)$, $(1/6, 0)$. Of these the first and the second correspond to the above mentioned phases $\langle 3, 1 \rangle$ and AF. The point $(1/6, 0)$ corresponds to the phase $\langle 3, 1^3 \rangle$.

If $I_4 > 0$, PDL $\langle 3, 1 \rangle$ -AF splits and a strip of the intermediate phase $\langle 3, 1^3 \rangle$ appears in between. If $I_4 < 0$, PDL $\langle 3, 1 \rangle$ -AF does not split neither to this nor to the higher orders of the perturbation theory. The theoretical analysis of the AF-to- $\langle 3, 1 \rangle$ phase transition in the case $I_4 > 0$ predicts the following sequence of phases as the magnetic field increases :

$$\text{AF} \rightarrow \langle 3, 1^3 \rangle \rightarrow \langle 3, 1, 3, 1^3 \rangle \rightarrow \langle 3, 1 \rangle. \quad (19)$$

The sequence (19) is in a perfect agreement with the experimental observations [5, 6]. At low temperature and magnetic field (see Fig. 1) the free energy of the phase $\langle 2, 2 \rangle$ is a minimal one. As h increases, the theoretically predicted sequence of phases is as follows

$$\langle 2, 2 \rangle \rightarrow \langle 2, 1 \rangle \rightarrow \langle 3, 1 \rangle. \quad (20)$$

Fig. 3. — The possible phase diagram in the χ - h plot.

It differs from the experimentally observed sequence [5, 6] :

$$\langle 2, 2 \rangle \rightarrow \langle 2^5, 1 \rangle \rightarrow \langle 2^3, 1, (2, 1)^3 \rangle \rightarrow \langle 3, 1 \rangle. \quad (21)$$

The possible origin of this discrepancy may be due to an oversimplified approximation of the low-temperature behaviour of the real CeBi by the Ising model. If the « direct » interaction does not decrease rapidly, then thermal fluctuations at low temperatures can be neglected, rather in a vicinity of PDL one has to consider « direct » couplings.

The phase diagram of the model discussed here is depicted on the χ - h plot in figure 3. The necessary prerequisite for the construction of the phase diagram of figure 3 is couplings I_1, I_2, I_3 versus the temperature (Eqs. (11), (13)) and the family of phase diagrams on the I_1 - I_2 plot at different fields h (two typical diagrams are presented in figures 1-2). As one changes the temperature, T the scale of χ on the χ - h plot, changes nonuniformly, because $\chi(T)$ increases slowly at low temperatures, $\sim \exp(-\text{const}/T)$, and rather rapidly near the Onsager transition temperature. Therefore the phase diagram on the T - h plot differs from that depicted in figure 3 by a nonuniform deformation. This well agrees with the experimental picture.

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