Mixing of spin and quadrupole subsystems in a magnetic field in connection with the antiferroquadrupole transition in CeB_6

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A mixing of antiferromagnetic and antiferroquadrupole subsystems occurs in a magnetic field. As a result, the highest of the transition temperatures increases with increasing magnetic field. This effect explains the increase in the temperature of the antiferroquadrupole transition with increasing magnetic field in CeB_6. © *1995 American Institute of Physics.*

1. According to Refs. 1–3, two phase transitions occur in CeB_6 in a zero magnetic field, to an antiferroquadrupole (AFQ) state and to an antiferromagnetic (AF) one, at temperatures $T_{Q0}=3.2$ and $T_{N0}=2.4$ K, respectively. The temperature of the AFQ transition, T_Q, increases with increasing magnetic field. This behavior of the phase transition is quite surprising: A magnetic field tends to orient the spins in a common direction, so it should suppress both AFQ and AF fluctuations. We would thus expect the temperature of the AFQ transition to *decrease* with increasing magnetic field. On this basis we can say that the phase transition to the AFQ state of CeB_6 is "anomalous."

Here is the gist of the present letter. In a zero magnetic field, in the random-phase approximation (the mean-field approximation), the spin and quadrupole subsystems are not independent, since the correlation functions $\langle J^{\alpha},Q^{\beta\gamma}\rangle$ are zero. Here $\mathbf{J}$ is the total angular momentum of the ion, and the operator $Q^{\alpha\beta}$ represents the quadrupole moment:

$$Q^{\alpha\beta}=\tfrac{1}{2}(J^{\alpha}J^{\beta}+J^{\beta}J^{\alpha})-\frac{J(J+1)}{3}. \tag{1}$$

When a field is applied, these correlation functions become nonzero, and there is a mixing of the AFQ and AF subsystems. As a result, the highest of the transition temperatures rises, while the lowest falls. The transition to the AFQ state is a first-order transition. As is shown below, however, for CeB_6 this transition is nearly a second-order transition, so our qualitative analysis is valid.

In CeB_6, the cerium ions form a simple cubic lattice. Their total angular momentum is $J=5/2$. In the crystal field, the ground state is the Γ_8 quartet. The energy of the Γ_7 doublet is 46 meV, and it has essentially no effect on the properties of low-temperature phase transitions.[1–3]

In a cubic field, the quadrupole exchange interaction of neighboring ions can be represented by two terms, corresponding to components of the quadrupole moment which are transformed by the representations Γ_3 $[O_2^{(0)}=\sqrt{3/2}Q^{zz};\ O_2^{(2)}=\sqrt{1/2}(Q^{xx}-Q^{yy})]$

and Γ_5 (Q^{xy}, Q^{xz}, Q^{yz}). In a previous study[4] of magnetoelastic properties of CeB_6, it was concluded that the interaction corresponding to representation Γ_5 is weak. We will accordingly ignore it.

As a result, we write the Hamiltonian of the system in the form

$$\mathscr{H}=\tfrac{1}{2}\Sigma_{l\neq l'}V_{ll'}(O_{2,l}^{(0)}O_{2,l'}^{(0)}+O_{2,l}^{(2)}O_{2,l'}^{(2)})+\tfrac{1}{2}\Sigma_{l\neq l'}I_{ll'}\mathbf{J}_l\cdot\mathbf{J}_{l'}-g_J\mu H\Sigma_l J_l^z, \tag{2}$$

where $V_{ll'}$ and $I_{ll'}$ are the exchange integrals of the quadrupole–quadrupole and spin–spin interactions, respectively, and the operators $\mathbf{J}$ and Q act in the space of the Γ_8 quadruplet. For $V_{l,l'}$ and $I_{l,l'}$ we use the nearest-neighbor approximation. In momentum space, we can then write them as

$$V_{\mathbf{k}}=2V(\cos k_x+\cos k_y+\cos k_z),$$
$$I_{\mathbf{k}}=2I(\cos k_x+\cos k_y+\cos k_z),$$

where $V>0$ and $I>0$.

We introduce static-approximation Green's functions in accordance with Refs. 5–7:

$$G_{ll'}=\int_0^{1/T}\langle J_l^z(\tau)J_{l'}^z(0)\rangle d\tau-\frac{1}{T}\langle J_l^z\rangle\langle J_{l'}^z\rangle,$$

$$F_{ll'}^{\lambda}=\int_0^{1/T}\langle O_{2,l}^{\lambda}(\tau)J_{l'}^z(0)\rangle d\tau-\frac{1}{T}\langle O_{2,l}^{\lambda}\rangle\langle J_{l'}^z\rangle,$$

$$K_{2,ll'}^{\lambda}=\int_0^{1/T}\langle O_{2,l}^{\lambda}(\tau)\rangle\langle O_{2,l'}^{\lambda}(0)\rangle d\tau-\frac{1}{T}\langle O_{2,l}^{\lambda}\rangle\langle O_{2,l'}^{\lambda}\rangle, \tag{3}$$

where $\lambda=0, 2$. The function $F^{(0)}$ is evidently zero except in a nonzero magnetic field ($F^{(0)}\sim h/T$, $h=g_J\mu H$ at $h\ll T$), while $F^{(2)}$ is identically zero.

2. We write a system of equations for the Green's functions in the random-phase approximation in the paramagnetic region:

$$F^{(0)}=F_0^{(0)}-G_0\frac{I_{\mathbf{k}}}{T}F^{(0)}-F_0^{(0)}\frac{V_{\mathbf{k}}}{T}K^{(0)},$$

$$K^{(0)}=K_0^{(0)}-K_0^{(0)}\frac{V_{\mathbf{k}}}{T}K^{(0)}-F_0^{(0)}\frac{I_{\mathbf{k}}}{T}F^{(0)},$$

$$K^{(2)}=K_0^{(2)}-K_0^{(2)}\frac{V_{\mathbf{k}}}{T}K^{(2)}, \tag{4}$$

where $K_0^{(0)}$, $K_0^{(2)}$, $F_0^{(0)}$, and G_0 are single-site functions.[5,6,8] Since the temperature of the AFQ transition is higher than that of the AF transition, we consider only the equations for the Green's functions $K^{(0)}$ and $K^{(2)}$. The equations for these functions do not contain Green's functions constructed from any components of the quadrupole and the spin other than those incorporated in definitions (3). We also note that all the results in this letter can be extended immediately to the case $T_N > T_Q$, with the appropriate replacement of $K^{(0)}$ and $K^{(2)}$ by the function G and by a function constructed from the spin components J^x, J^y.

A solution of system (4) is

$$K^{(0)} = \frac{K_0^{(0)} - \dfrac{I_{\mathbf{k}}(F_0^{(0)})^2}{T + I_{\mathbf{k}} G_0}}{1 + \dfrac{V_{\mathbf{k}}}{T} K_0^{(0)} - \dfrac{V_{\mathbf{k}} I_{\mathbf{k}}}{T} \dfrac{(F_0^{(0)})^2}{T + I_{\mathbf{k}} G_0}}, \quad K^{(2)} = \frac{K_0^{(2)}}{1 + \dfrac{V_{\mathbf{k}}}{T} K_0^{(2)}}. \tag{5}$$

Here the zeros of the denominator determine points of phase transition. If there are several such points, the temperature of the transition to the AFQ state will evidently be the same as above.

In a zero magnetic field we have $F^{(0)} = 0$, and the AFQ transition occurs at the temperature

$$T_{Q0} = -V_{\mathbf{k}_0} K_0^{(0)} = -V_{\mathbf{k}_0} K_0^{(2)} = 74V,$$

where $k_0 = (1/2, 1/2, 1/2)$. Here $K_0^{(0)}$ and $K_0^{(2)}$ are calculated by taking an average over the Γ_8 quadruplet. Correspondingly, we find an expression for the temperature of the AF transition:

$$T_{N0} = -I_{\mathbf{k}_0} G_0 = \frac{35}{2} I.$$

If we ignore the interaction with the AF subsystem ($I = 0$), and if we assume $h \ll T$, then the phase transitions in subsystems 0 and 2 occur at the temperatures

$$T_Q^{(0)} = -V_{\mathbf{k}_0} K_0^{(0)} = T_{Q0}\left(1 + A\,\frac{h^2}{T_{Q0}^2}\right), \quad T_Q^{(2)} = -V_{\mathbf{k}_0} K_0^{(2)} = T_{Q0}\left(1 - A\,\frac{h^2}{T_{Q0}^2}\right), \tag{6}$$

where the constant

$$A = \frac{\left\langle (O_2^{(0)})^2 \left(J^{z2} - \dfrac{J(J+1)}{3} \right) \right\rangle}{8 K_0^{(0)}}$$

is calculated for the ground state in a zero magnetic field. It can be seen from Eqs. (6) that a magnetic field which disrupts the cubic symmetry of the system leads to different transition temperatures for the two components of the quadrupole moment.

In our case of the Γ_8 ground state, with $J = 5/2$, we have $A = 40/111$, and the quantity T_Q^0, which determines the transition temperature, increases with increasing magnetic field. By way of comparison, in the case of a Γ_8 ground state with $J = 3/2$, we would have $A = 0$, and the field-dependent correction to the temperature would be proportional to $(h/T)^4$.

When mixing between AFQ and AF subsystems in a magnetic field is taken into account, we find

$$T_Q^0 = -V_{\mathbf{k}_0}K_0^{(0)} + (F^{(0)})^2 \frac{V_{\mathbf{k}_0} I_{\mathbf{k}_0}}{T + I_{\mathbf{k}_0} G_0}, \quad T_Q^2 = -V_{\mathbf{k}_0} K_0^{(2)}. \tag{7}$$

Expanding expressions (7) in a series in h/T, we find

$$T_Q^0 = T_{Q0}\left[1 + \left(A + \frac{K^{(0)}}{G_0}\frac{T_{N0}}{T_{Q0}-T_{N0}}\right)\right] = T_{Q0}\left[1 + \left(\frac{40}{111} + \frac{296}{105}\frac{T_{N0}}{T_{Q0}-T_{N0}}\right)\frac{h^2}{T_{Q0}^2}\right]$$

$$= T_{Q0}\left[1 + \left(0.36 + \frac{2.82 T_{N0}}{T_{Q0}-T_{N0}}\right)\frac{h^2}{T_{Q0}^2}\right],$$

$$T_Q^2 = T_{Q0}\left[1 - A\frac{h^2}{T_{Q0}^2}\right] = T_{Q0}\left(1 - \frac{40}{111}\frac{h^2}{T^2}\right) = T_{Q0}\left(1 - 0.36\frac{h^2}{T_{Q0}}\right). \tag{8}$$

We thus see that incorporating the mixing of AFQ and AF fluctuations has substantially intensified the increase in the transition temperature with increasing magnetic field.

In reality, the AF transition in CeB_6 is observed not at $\mathbf{k}=\mathbf{k}_0$; it corresponds to the star of vectors (0, 1/4, 1/4), (1/2, 1/4, 1/4), (0, 1/4, -1/4) (1/2, 1/4, -1/4) (Ref. 1). The circumstance means that (first) the AF interaction is more complex than the interaction of nearest neighbors and (second) below T_Q we need to allow for the presence of a low-range AFQ order. These questions will be analyzed in a separate publication.[8] When these complicating factors are taken into account, the T_{N0} in (8) is replaced by an effective $\tilde{T}_{N0}$, and the ratio $\tilde{T}_{N0}/(T_{Q0}-\tilde{T}_{N0})$ turns out to be on the order of unity. Nevertheless, the channel-mixing effect remains the governing effect, because of the relatively large numerical coefficient.

As we have already mentioned, one can show that the mixing of the AFQ and AF subsystems in a magnetic field in the case $T_Q < T_N$ leads to an increase in the temperature of the AF transition with increasing magnetic field.

3. Our analysis of the mixing of subsystems is correct at a qualitative level if the transition to the AFQ state is a first-order transition which is nearly a second-order transition. We will now demonstrate that this is indeed the case. Using a Stratonovich–Hubbard transformation, we find an effective Landau–Ginzburg Hamiltonian for the AFQ subsystem in the case of a zero magnetic field:

$$H = \tfrac{1}{2}\sum_k \left(\frac{3T}{37V_{\mathbf{k}}} + 1\right)(h^{\mathbf{k}_0}h_0^{-\mathbf{k}} + h_2^{\mathbf{k}}h_2^{-\mathbf{k}}) + \sum_{\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3=0} \frac{40}{333}\sqrt{\frac{3}{2}}(h_0^{\mathbf{k}_1}h_0^{\mathbf{k}_2}h_0^{\mathbf{k}_3} - 3h_0^{\mathbf{k}_1}h_2^{\mathbf{k}_2}h_2^{\mathbf{k}_3})$$

$$+ \frac{6263}{2592}\left(\frac{3}{37}\right)^{\frac{3}{2}} \sum_{\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4=0} (h_0^{\mathbf{k}_1}h_0^{\mathbf{k}_2}h_0^{\mathbf{k}_3}h_0^{\mathbf{k}_4} + 2h_0^{\mathbf{k}_1}h_0^{\mathbf{k}_2}h_2^{\mathbf{k}_3}h_2^{\mathbf{k}_4} + h_2^{\mathbf{k}_1}h_2^{\mathbf{k}_2}h_2^{\mathbf{k}_3}h_2^{\mathbf{k}_4}). \tag{9}$$

Here h_0 and h_2 are auxiliary fields, normalized in such a way that their Green's function g_0 is equal to $\tau^{-1} + k^2/6$, where $\tau = (T - T_Q)/T_Q$. In the case of the Γ_8 quartet with $J = 3/2$, the factor in the third-degree term is zero, and we have a second-order transition. In our case, straightforward calculations lead to the following expression for the Green's function g, in which the first three-particle correction has been incorporated:

$$g^{-1}=g_0^{-1}+\Sigma=\tau+\frac{k^2}{6}+\frac{4800}{50653}\frac{\sqrt{6}}{\pi}\frac{1}{\sqrt{\tau}}=\tau+\frac{k^2}{6}+\frac{0.074}{\sqrt{\tau}}. \tag{10}$$

We see from this result that this correction can be ignored if $\tau>0.18$; i.e., our transition is indeed a first-order transition which is approximately a second-order transition.

4. Let us summarize the basic results of this letter. The anomalous behavior of the antiferromagnetic quadrupole phase transition of CeB_6 in a magnetic field can be explained at a qualitative level on the basis of a mixing of the spin and quadrupole subsystems in the magnetic field. The antiferroquadrupole transition turns out to be a first-order transition which is nearly a second-order transition.

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