ORDERING AND FLUCTUATION OF QUANTUM MULTIPOLES IN CeB_6 *

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The effect of multipolar fluctuations on the quadrupolar phase transition in CeB_6 is investigated theoretically. It is shown that the fluctuations become strong and field-dependent, reflecting the competition of coupled multipolar interactions. Some unusual phenomena around the transition in CeB_6 are shown to be reasonably explained within the RKKY model.

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The antiferro-quadrupolar (AFQ) ordering in CeB_6 has been attracting a renewed interest. Recent experimental and theoretical efforts have revealed gradually its mysterious nature in magnetic fields. A key factor of the microscopic origin is shown to be hidden orbital degrees of freedom of f-electrons, which should be described as the multipolar moments.

Within the degenerate Γ_8 crystal-field groundstate there exist fifteen independent multipolar moments: three dipoles, five quadrupoles and seven octupoles [1]. The recent theoretical work shows that the primary order parameter in AFQ phase is of the Γ_5-type (O_{yz}, O_{zx}, O_{xy}), whose component depends on the direction of the magnetic field [2, 3]. A remarkable consequence of the theory is the importance of the field-induced octupolar order parameter T_{xyz} [2–5], which had been overlooked for a long time. In fact the existence of the octupole resolves the inconsistency between NMR and neutron diffraction, and at the same time it explains the anomalous field-dependence of the AFQ transition temperature ($T_{\rm Q}$).

In this way the mean-field (MF) description of the multipolar ordering state of CeB_6 is well-established. However it should be stressed at this stage that the MF theory is not successful to reproduce the experiments quantitatively. One of the possible origins of this disagreement is the strong fluctuation effect of coupled multipoles. Thus the quantitative analysis beyond

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the MF theory is necessary to understand whether and how the fluctuation correction can improve the agreement with experiments. It also enables us to clarify the mechanism of unconventional multipolar fluctuations in CeB_6.

In the present paper we will study the fluctuation effects on the AFQ phase transition by introducing two effective models for the low- and high-field regions. The dependence of the fluctuation correction on the magnetic field and its mechanism are studied in detail. A part of the results are already presented in Ref. [6, 7].

We begin with defining the multipolar RKKY model for CeB_6 by using two pseudo-spins $\boldsymbol{\tau}$ and $\boldsymbol{\sigma}$. See Ref. [1] for details of the relation between the pseudo-spins and the multipoles. The RKKY model is decomposed in the following way: [1, 3, 5]

$$H_{\mathrm{I}} = H_0 + \delta H_{\mathrm{Q}} + \delta H_{\mathrm{O}} \,. \tag{1}$$

Here H_0 is the SU(4) symmetric part, which is expressed by

$$H_0 = D \sum_{(ij)} \Big(\boldsymbol{\tau}_i \cdot \boldsymbol{\tau}_j + \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j + 4(\boldsymbol{\tau}_i \cdot \boldsymbol{\tau}_j)(\boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j) \Big) \,. \tag{2}$$

The deviations of the interaction strength of the quadrupoles (O_{yz}, O_{zx}, O_{xy}) and the octupole T_{xyz} from the SU(4) limit are introduced by

$$\delta H_{\mathrm{Q}} = D\varepsilon_{\mathrm{Q}} \sum_{(ij)} 4\tau_i^y \tau_j^y (\boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j) \,, \quad \delta H_{\mathrm{O}} = D\varepsilon_{\mathrm{O}} \sum_{(ij)} \tau_i^y \tau_j^y \,. \tag{3}$$

The Zeeman term can be represented by pseudo-spins as [1]

$$H_{\mathrm{Z}} = \tfrac{7}{3} g\mu_{\mathrm{B}} \sum_i (\boldsymbol{\sigma}_i + c\boldsymbol{\eta}_i) \cdot \boldsymbol{H} \,, \tag{4}$$

$$\boldsymbol{\eta} = (-\tau^z\sigma^x + \sqrt{3}\tau^x\sigma^x, -\tau^z\sigma^y - \sqrt{3}\tau^x\sigma^y, 2\tau^z\sigma^z) \,. \tag{5}$$

Although c should be 4/7 for the real Γ_8 states, it is shown to be irrelevant for low fields [6]. Thus the analysis is much simplified in the low-field region by setting $c = 0$. This model corresponds to the isotropic limit for the field direction.

To study the high-field phase diagram, however, it is necessary to take into account the effect of finite c. The simplest starting point for high fields is to study the effective model for τ-spins under ferromagnetically-frozen σ-spins [7]. Consider the direction of the field

$$\boldsymbol{H} = H \left(\frac{1}{\sqrt{2}} \sin\theta, \frac{1}{\sqrt{2}} \sin\theta, \cos\theta \right) ,$$

where θ is the angle of the field direction measured from the (001) axis [3]. We define the parallel spin component as $\sigma^{||} = \boldsymbol{\sigma} \cdot \boldsymbol{H} / H$. Then, substituting $\sigma^{||} = 1/2$, the RKKY model in high fields is mapped to the following effective orbital Hamiltonian,

$$\tilde{H}_{\rm I} = 2D \sum_{(ij)} \left(\tau_i^x \tau_j^x + q \tau_i^y \tau_j^y + \tau_i^z \tau_j^z \right) - \sum_i h_\theta \tau_i^z \,, \tag{6}$$

where the anisotropy parameter is given by $q = 1 + \frac{1}{2}(\varepsilon_{\rm Q} + \varepsilon_{\rm O})$. The effective field depends on both the strength and the direction of the real magnetic field as $h_\theta = h_0 c \left(\cos^2\theta - \frac{1}{2}\sin^2\theta \right)$ with $h_0 = \frac{7}{3} g \mu_{\rm B} H$.

It is shown that one can expect the AFQ phase diagram for the entire region of magnetic field from the analysis on the two limiting cases ($c = 0$ for low fields and $\sigma = 1/2$ for high fields), even without analyzing the original complicated model. The analysis can be done for both limiting models by the d^{-1}-expansion method, where d is the spatial dimension. Then one can obtain the generalized Landau free energy with the fluctuation correction, and it is used to calculate various thermodynamic quantities near the AFQ transition temperature. The detailed formulation is given in Ref. [6–8].

Here we show the results of the AFQ transition temperatures $T_{\rm Q}$ with d^{-1}-correction for the low- and high-field models. They are plotted in Fig. 1 as a function of H. See Refs. [3, 6] for details on the MF results. As expected, $T_{\rm Q}$ is more or less suppressed from the MF value for all the parameters. It should be pointed out that the fluctuation effect exhibits a strong H-dependence in the low-field region: $T_{\rm Q}$ is much suppressed for low fields while the correction becomes small as the field is increased. As a result, $T_{\rm Q}$ shows a large enhancement with H in the low-field region. This can be interpreted by the fact that the competition of coupled multipoles produces large fluctuation near the SU(4)-symmetric limit. These results should be compared with experiments [9] plotted in Fig. 1. It is shown that the agreement with the experiments is much improved in comparison with the previous MF result. We have shown also that other unusual behaviors of CeB_6 near the transition are well-explained by the strong fluctuation within the present theory [6]. These facts clearly demonstrate that there exists a large and field-dependent fluctuation of multipoles in CeB_6.

From the analysis of the high-field model one can expect the critical field of $H_{\rm c} \sim$ 60–80 T at $T = 0$ K even for the (001) field, which is not renormalized by the fluctuation. Therefore the ratio $H_{\rm c}(T = 0)/T_{\rm Q}(H = 0)$ becomes very large, reflecting the strong suppression of the zero-field $T_{\rm Q}$. We have also examined the dependence of $T_{\rm Q}$ on the field direction for the high-field model. [7] Then it has been shown that the fluctuation effect on the anisotropy is not large although there are considerable suppressions of

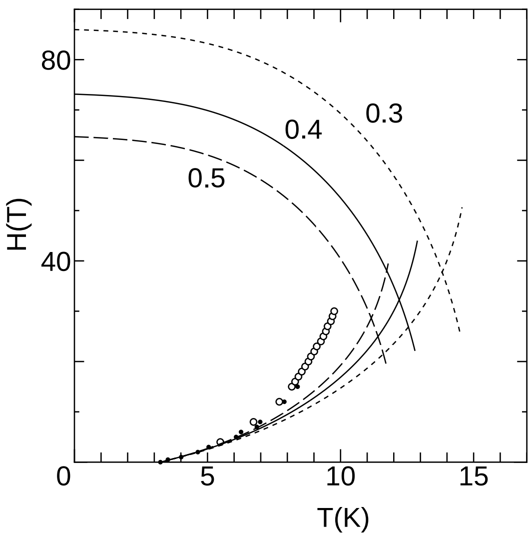

Fig. 1. Comparison of AFQ transition temperatures with experiments for the (001) field. The open circles and the closed dots are the experimental results. [9] The dotted, solid and broken lines are the theoretical results with d^{-1}-correction for $\varepsilon_{\mathrm{Q}}/D = \varepsilon_{\mathrm{O}}/D = 0.3$, 0.4 and 0.5 respectively. The results of both the low- and high-field models are plotted for each parameter and they should be combined to expect the phase diagram corresponding to the experiments.

the absolute values of T_{Q}. These high-field properties are still open experimentally, and future experimental checks are highly required.

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