Fluctuation Effects on the Quadrupolar Ordering in Magnetic Field

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Effects of magnetic field on the quadrupolar ordering are investigated with inclusion of fluctuation of order parameters. For the simplest model with the nearest-neighbor quadrupolar interaction, the transition temperature and the specific heat are derived by the use of recently proposed effective medium theory. It is shown that magnetic field $H$ has two competing effects on the quadrupolar ordering: one is to encourage the ordering by suppressing the fluctuation among different components of order parameters, and the other is to block the ordering as in antiferromagnets. The former is found to be of order $H^2$ and the latter of order $H^4$. Hence the fluctuation is suppressed for weak fields, and the transition temperature increases with magnetic field. The fluctuation effect is so strong that only a small entropy released at the quadrupolar ordering is about half of the full value $\ln 4$ even without the Kondo effect.

KEYWORDS: quadrupole moment, CeB$_6$, TmTe, spherical model, effective medium, phase diagram, specific heat, Heisenberg model

§1. Introduction

Some compounds with orbital degeneracy show ordering of quadrupole moments without magnetic order. A typical example is CeB$_6$ which also shows the Kondo effect with the Kondo temperature $T_K \sim 1K$. The Ce$^{3+}$ multiplet ($4f^2$, $J = 5/2, S = 1/2, L = 3$) is split under the cubic crystal field into the ground-state quartet $\Gamma_8$ and an excited-state doublet $\Gamma_7$ separated by about 540K. It has a transition from the phase I (paramagnetic phase) to the phase II at 3.3K under zero field. Neutron and NMR experiments show that electric quadrupole moments are ordered with a periodic pattern in the phase II. Hence it is also called the antiferro-quadrupolar phase (AFQ). With further decrease of temperature, another transition occurs at 2.4K to the phase III, which accompanies the magnetic ordering with a complicated periodic pattern. The phase boundary between the phases I and II shows unusual dependence on magnetic field; the transition temperature $T_Q(H)$ increases with magnetic field $H$. Namely, while a magnetic field will ordinarily destroy an ordered phase, it assists the quadrupolar ordering in CeB$_6$. The phase boundary shifts to lower temperatures in the dilute alloy Ce$_{1-x}$La$_x$B$_6$. The AFQ is found also in TmTe which is insulating. Though its phase diagram has something similar to that of CeB$_6$, $T_Q(H)$ shows a maximum as a function of magnetic field in contrast to that of CeB$_6$. In the latter, $T_Q(H)$ continues to increase up to the maximum field measurable at present.

One of the keys to understanding the transition between the phases I and II should be the result of specific-heat measurements, which indicates large fluctuation there. Although the peak at the AFQ transition is small in zero magnetic field, it grows larger with increasing magnetic field. The small peak suggests that only a small change of entropy is involved at the transition, and that a large amount of short-range order should exist even above $T_Q$. In other words, the long-range order is disturbed by some large fluctuation. Therefore, the experimental result of specific heat suggests that the large fluctuation should be suppressed by a magnetic field. The purpose of this paper is to clarify how a magnetic field affects the fluctuation at the quadrupolar ordering. We take account of fluctuation in our calculation with use of an effective medium theory for quantum spins and multipoles.

Several models for the quadrupolar ordering in CeB$_6$ have been proposed. Hanzawa and Kasuya assumed that the ground state is the doublet $\Gamma_7$, and considered the quadrupolar interaction among Ce ions. In this model, large off-diagonal elements of quadrupole-moment operators between $\Gamma_7$ and $\Gamma_8$ become important as magnetic field increases. Thus, $T_Q(H)$ increases with $H$ in the mean-field approximation. Although this model should be effective for the system with the $\Gamma_7$ ground state, it was shown later by experiment that $\Gamma_8$ is the ground state and $\Gamma_7$ is an excited state. Another model proposed by Ohkawa and Ohyama has the symmetric form between the spin doublet and the orbital doublet represented by pseudo spins. The Hamiltonian includes an intersite interaction which is biquadratic in spin and quadrupole moments. This interaction enhances the quadrupolar interaction with increasing $H$. Hence $T_Q(H)$ increases in the mean-field approximation also in this model. Shina et. al. recently rewrote Ohkawa’s model in the form including magnetic octupoles and dipoles, and solved the resultant model by the mean-field approximation. Finally, Uimin and the present authors treated the effect of fluctuation. They approximated the pseudo-spins by classical spins and applied the spherical model which can include fluctuation effects on the quadrupolar ordering; one is to encourage the ordering by suppressing the fluctuation and an excited-state doublet $\Gamma_8$ is split un

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tuation in a simple manner. As a result, the transition temperature increased with magnetic field in contrast to the result of the mean-field approximation.

In this paper we accomplish substantial improvement over the theory of ref.13. The fluctuation emphasized there is due to the competition among order parameters with different wave numbers \( k \). The intersite interaction was taken to be the electrostatic one between the \( \Gamma_3 \)-type quadrupole moments. Strength of the quadrupolar interaction at \( k = (\pi\pi\pi) \) is almost the same as that at \( k = (\pi\pi0) \) in the case of the electrostatic interaction. Competing fluctuations are suppressed by a magnetic field. However, as will be discussed in detail later, what is essential of the magnetic field is to suppress competition among components of fluctuating order parameters, namely, \( O_2^0 \) and \( O_2^2 \). The presence of competition among order parameters with different wave numbers helps the suppression effect mentioned above. The value of \( dT_Q(H) / dH \) for the electrostatic interaction is about twice as large as that for the nearest-neighbor one. Although this enhancement is substantial quantitatively, we regard that the specific form of interaction is not essential for qualitative understanding. Hence we adopt the nearest neighbor interaction for simplicity of calculation in this paper. Further, we do not restrict the quadrupolar interaction to that of the \( \Gamma_3 \)-type moments.

\section{Model for Quadrupolar Interaction}

\subsection{Model}

Our model corresponds to a localized electron system which has interaction between quadrupole moments only. Possible targets of the model are not only to \( \text{CeB}_6 \) but also related quadrupolar systems under the conditions to be described below. An insulating compound \( \text{TmTe} \) has a phase diagram similar to \( \text{CeB}_6 \). If the increase of transition temperature in these two materials comes from a common mechanism, the interaction with conduction electrons should not be essential. Hence we do not consider this interaction explicitly.

Cerium sites in \( \text{CeB}_6 \) constitute the simple cubic lattice. At temperatures near \( T_Q \), the excited CEF states, which are separated by about 540K in \( \text{CeB}_6 \),\(^{6}\) have little influence. Therefore we deal with the ground-state quartet \( \Gamma_8 \) only. We represent here the basis set of the \( \Gamma_8 \) states with use of the \( |J_z\rangle \) basis,

\[
|\psi_{1\pm}\rangle = \sqrt{\frac{5}{6}} |\pm\frac{5}{2}\rangle + \sqrt{\frac{1}{6}} |\pm\frac{3}{2}\rangle, \quad |\psi_{2\pm}\rangle = |\pm\frac{1}{2}\rangle. \quad (2.1)
\]

The Hamiltonian for our model is composed of the quadrupolar interaction term \( \mathcal{H}_{\text{int}} \) and the Zeeman term \( \mathcal{H}_Z \):

\[
\mathcal{H} = \mathcal{H}_{\text{int}} + \sum_i \mathcal{H}_Z(i), \quad (2.2)
\]

\[
\mathcal{H}_{\text{int}} = J_{\Gamma_3} \sum_{\langle ij \rangle} \left[ O_2^0(i)O_2^0(j) + O_2^2(i)O_2^2(j) \right]
+ J_{\Gamma_5} \sum_{\langle i \rangle} \left[ O_{yz}(i)O_{yz}(j) + O_{zx}(i)O_{zx}(j) + O_{xy}(i)O_{xy}(j) \right], \quad (2.3)
\]

where \( 6/7 \) in the Zeeman term is the \( g \) factor. The summation in \( \mathcal{H}_{\text{int}} \) is performed over all the nearest neighbor pairs. Quadrupole-moment operators at each site are given as

\[
O_2^0 = \frac{1}{\sqrt{3}} (3J^z)^2 - J(J + 1),
O_2^2 = (J^z)^2 - (J^y)^2,
O_{yz} = J^yJ^z + J^zJ^y,
O_{zx} = J^xJ^z + J^zJ^x,
O_{xy} = J^xJ^y + J^yJ^x. \quad (2.5)
\]

The interaction constant \( J_{\Gamma_3} \) is different from \( J_{\Gamma_5} \) in general. We take three types of interaction constants: (i) \( \Gamma_3 \)-type interaction \( J_{\Gamma_3} > 0, J_{\Gamma_5} = 0 \); (ii) \( \Gamma_5 \)-type interaction \( J_{\Gamma_3} = 0, J_{\Gamma_5} > 0 \); (iii) \( (\Gamma_3 + \Gamma_5) \)-type interaction \( 16J_{\Gamma_3} = J_{\Gamma_5} > 0 \). In the case (iii), interaction constants are taken so that every ordering of five types of quadrupole moments has the same transition temperature in zero magnetic field. This becomes clear in the pseudo-spin representation described in the next subsection. It is expected that the effect of fluctuation is the largest in the case (iii).

This model is the simplest one for the quadrupolar ordering in \( \text{CeB}_6 \). However, in the literature the increase of the transition temperature has never been discussed within this model. This is probably because the result of the mean-field approximation does not show the increasing transition temperature. We treat this model beyond the mean-field approximation to discuss effects of the fluctuation.

\subsection{Pseudo-spin representation}

It is convenient for calculation of \( T_Q(H) \) to use the pseudo-spin representation. When the Hilbert space is limited to the \( \Gamma_8 \) states, all operators are represented by these pseudo-spins because all the matrix elements can be represented by those of pseudo-spins. The pseudo-spin operators are defined by

\[
\tau^z |\psi_{1\pm}\rangle = |\psi_{1\pm}\rangle, \quad \tau^z |\psi_{2\pm}\rangle = -|\psi_{2\pm}\rangle,
\tau^+ |\psi_{1\pm}\rangle = |\psi_{1\pm}\rangle, \quad \tau^- |\psi_{1\pm}\rangle = |\psi_{2\pm}\rangle,
\sigma^z |\psi_{\alpha+}\rangle = |\psi_{\alpha+}\rangle, \quad \sigma^z |\psi_{\alpha-}\rangle = -|\psi_{\alpha-}\rangle,
\sigma^+ |\psi_{\alpha-}\rangle = |\psi_{\alpha+}\rangle, \quad \sigma^- |\psi_{\alpha+}\rangle = |\psi_{\alpha-}\rangle, \quad (\alpha = 1, 2). \quad (2.7)
\]

Note that this definition is different from that in refs.12, 14 and 15 where the positive eigenvalue of the pseudo-spins is set to 1/2. It is set to unity here to remove the constant 1/2 from various expressions.

In this representation, quadrupole-moment operators are given by

\[
O_2^0 = \frac{8}{\sqrt{3}} \tau^z, \quad O_2^2 = \frac{8}{\sqrt{3}} \tau^x,
O_{yz} = \frac{2}{\sqrt{3}} \tau^y \sigma^z, \quad O_{zx} = \frac{2}{\sqrt{3}} \tau^x \sigma^y, \quad O_{xy} = \frac{2}{\sqrt{3}} \tau^y \sigma^x. \quad (2.9)
\]
The interaction Hamiltonian is rewritten as

$$\mathcal{H}_{\text{int}} = J_{13} \sum_{(i,j)} \left[ \tau^x(i) \tau^x(j) + \tau^y(i) \tau^y(j) \right] + J_{15} \sum_{(i,j)} \tau^z(i) \tau^z(j) - \mu_B \left( 1 + \frac{4}{7} T^\alpha(i) \right) \sigma^\alpha(i) H^\alpha, \quad (2.10)$$

with $J_{13} = J_{13} \times 64/3$ and $J_{15} = J_{15} \times 4/3$. Hence $J_{13}$ is satisfied for the $(\Gamma_3 + \Gamma_5)$-type interaction. The Zeeman term is rewritten as

$$\mathcal{H}_Z(i) = - \sum_{\alpha=x,y,z} \mu_B \left( 1 + \frac{4}{7} T^\alpha(i) \right) \sigma^\alpha(i) H^\alpha, \quad (2.11)$$

where $T^\alpha = \frac{1}{\sqrt{3}} \left( 3J^\alpha i - J(i + 1) \right) \times \frac{\tau^\alpha}{2}$ is given in the pseudo-spin representation by

$$T^x = -\frac{1}{2} \tau^x + \frac{\sqrt{3}}{2} \tau^y, \quad T^y = -\frac{1}{2} \tau^y + \frac{\sqrt{3}}{2} \tau^x, \quad T^z = \tau^z. \quad (2.12)$$

Intuitively, $(1 + 4/7 T^\alpha)$ represents the magnitude $|m|$ of the magnetic moment with $3/7 \leq |m| \leq 11/7$ and $\sigma^\alpha$ represents the orientation of it.

**§3. Static Approximation for the Effective Medium**

We recently formulated a dynamical effective medium theory for quantum spins and multipoles.\[1\] The theory is a generalization of the spherical model approximation for the Ising model. It takes account of fluctuation up to $O(1/t_n)$, where $t_n$ is the number of interacting neighbors. We use the static approximation (SA) which neglects the dynamical nature of fluctuations. Although it is difficult to estimate the accuracy of the SA in the temperature range of the quadrupole order, we use the SA as our first step to study the importance of fluctuations. Since derivation of this method is given in ref.\[17\], we explain here some characteristics of this method and details of the calculation.

In the mean-field approximation (MFA), one solves an effective single-site problem, where an operator for interactions. Since derivation of this method is given in ref.\[17\], we explain here some characteristics of this method and details of the calculation.

In order to explain the method concisely, we represent quadrupole-moment operators as $O^\alpha$ symbolically. We use the irreducible tensor operators for $O^\alpha$ to obtain scalar equations from a matrix equation.\[1\] For $H^\parallel(001)$, $O^\alpha$ refers to one of the operators in eq.\[2\] and/or eq.\[3\]. For $H^\parallel(111)$, eq.\[2\] and/or $(-O_{yz} - O_{zx} + 2O_{xy})/\sqrt{6}$, $(O_{yz} - O_{zx})/\sqrt{2}$ and $(O_{yz} + O_{zx} + O_{xy})/\sqrt{3}$. Then, the Hamiltonian for our model can be expressed as

$$\mathcal{H} = -\frac{1}{2} \sum_{ij} \sum_{n=1}^n J_{ij}^n O^n(i) O^n(j) + \sum_i \mathcal{H}_Z(i), \quad (3.1)$$

where $n$ refers to the number of fluctuating components and is equal to 2, 3 and 5 for the $\Gamma_3$, the $\Gamma_5$ and the $(\Gamma_3 + \Gamma_5)$-type interactions respectively.

We write the mean field as $a^\alpha(i) = \sum_j J_{ij}^n \langle O^n(j) \rangle$ and the fluctuating field around it as $\zeta^\alpha(i)$. The effective single-site Hamiltonian $\mathcal{H}_1$ and the partition function $Z_1$ is given by

$$\mathcal{H}_1 = \sum_{\alpha=1}^n \left\{ -a^\alpha O^\alpha - \zeta^\alpha (O^\alpha - \langle O^\alpha \rangle) \right\} + \mathcal{H}_Z, \quad (3.2)$$

$$Z_1 = \prod_{\alpha=1}^n \left\{ \int_{-\infty}^{\infty} \frac{d\zeta^\alpha}{\sqrt{2\pi} \beta \zeta^\alpha} \exp\left[ -\beta \zeta^\alpha \right] \right\} \exp\left[ -\beta \mathcal{H}_1 \right], \quad (3.3)$$

where the site index $i$ is omitted. The term $\zeta^\alpha \langle O^\alpha \rangle$ in the Hamiltonian plays a role to enforce $\langle \zeta^\alpha \rangle = 0$. The variance $\tilde{\zeta}^\alpha$ is determined by the self-consistency condition

$$\chi_L^\alpha = \int \frac{dD_q}{(2\pi)^D} \frac{\chi_L^\alpha}{\tilde{\zeta}^\alpha}, \quad (3.4)$$

where $D$ is the spatial dimensions ($D = 3$ for the present case), $\chi_L$ is the local (strain) susceptibility calculated with $\mathcal{H}_1$, and $\zeta^\alpha$ is the Fourier transform of the interaction: $\zeta^\alpha = \sum_j J_{ij}^n \exp[-i \mathbf{q} \cdot (\mathbf{R}_j - \mathbf{R}_i)]$. The lattice constant is set to unity. This equation requires the consistency between the local susceptibility and its another expression using the inverse Fourier transform: $\chi_L = N^{-1} \sum_q \chi_q$. Note that this consistency is not satisfied in the MFA for a finite dimensional system. The integral can be performed analytically for the nearest neighbor interaction $J_{ij}^n = -2J^\alpha \sum_{l=1}^D \cos q_l$ even in three dimensions.\[3\]

The SA still keeps the noncommuting character of quantum operators. In contrast, our previous theory for CeB$_6$\[4\] replaces the pseudo-spin vectors composed of the Pauli matrices by classical vectors. Further improvement of the present theory is that the renormalization is performed differently for each component $J^\alpha$ when the model has a low symmetry. The spherical model for classical spins used in ref.\[13\] cannot deal with the anisotropy in $J^\alpha$ even if the symmetry is lowered by a magnetic field. These theories coincide with each other in the limit of infinite temperature.

At high temperatures, the variance $\tilde{\zeta}^\alpha$ in Gaussian distribution of the effective field is almost zero, and therefore it has little deviation from the MFA. As the temperature decreases, the short-range order makes $\tilde{\zeta}^\alpha$ larger. In other words, the distribution of fluctuations gets broader. Finally it reaches the critical variance $\tilde{\zeta}^{cr}$ at the critical temperature, and the transition occurs; the susceptibility for a certain component $\lambda$ (or for multiple components simultaneously) diverges with the wave number $q = Q$ at which $\tilde{\zeta}^\alpha$ takes the maximum $J_Q^\alpha$. Hence, we obtain

$$\chi_L^{\lambda^{cr}} = \int \frac{dD_q}{(2\pi)^D} \frac{1}{J_Q^\alpha - J_q^\alpha}, \quad (3.5)$$

and $J_q^{\lambda^{cr}} = J_Q^\alpha - (\chi_L^{\lambda^{cr}})^{-1}$.

As the number $D$ of spatial dimensions goes to infinity, the critical variance $\tilde{\zeta}^{cr}$ zero. In short, the SA approaches the MFA. This is consistent with the nature of the theory; the original theory for the SA is the
next leading order theory with respect to the inverse of dimensions, while the MFA is the leading order theory. Small $J^{α}$ leads to a large transition temperature because the effect of fluctuation is small. On the contrary, when the number $n$ of fluctuating components increases, the critical temperature becomes smaller. The reason is that the local susceptibility $\chi_L$ decreases with increasing $n$ as shown below.

To perform the multiple Gaussian integral, the polar coordinate in the $ζ$ space is convenient. The reason is that in zero field some integrands of angular integral are constants by symmetry and remain smooth even in weak magnetic fields. For example, in terms of the polar coordinate for the $(Γ_3 + Γ_5)$-type interaction, we obtain

$$
\prod_{α=1}^{5} \int_{-∞}^{∞} \frac{β\, dζ^α}{\sqrt{2πβJ^{α}}} \exp\left[-β\frac{(ζ^{α})^2}{2J^{α}}\right] = \frac{1}{(2π)^5} \int dΩ_θ \int_{0}^{∞} ζ^4 dζ_r \exp\left[-\frac{ζ^2}{2}\right],
$$

where $ζ_r = \sqrt{\sum_α β(ζ^{α})^2/J^{α}}$ and $Ω_θ$ refers to the generalized solid angle. To perform the integral on the unit hypersphere in five dimensions, we introduce the variables $θ_a, θ_b, \cdot \cdot \cdot$ as follows:

$$
ζ_1 = \sqrt{\frac{J_1}{β} ζ_r \sin θ_a \sin θ_b \sin θ_c \sin θ_d} , \quad ζ_2 = \sqrt{\frac{J_2}{β} ζ_r \sin θ_a \sin θ_b \sin θ_c \cos θ_d} , \quad ζ_3 = \sqrt{\frac{J_3}{β} ζ_r \sin θ_a \sin θ_b \cos θ_c \cos θ_d} , \quad ζ_4 = \sqrt{\frac{J_4}{β} ζ_r \sin θ_a \cos θ_b \cos θ_c \cos θ_d} , \quad ζ_5 = \sqrt{\frac{J_5}{β} ζ_r \cos θ_a} ,
$$

$$
dΩ_θ = \sin^4 θ_a \sin^2 θ_b \sin θ_c dθ_a dθ_b dθ_c dθ_d , \quad (3.8)
$$

with $θ_a, θ_b, \cdot \cdot \cdot \in [0, π]$ and $θ_d \in [0, 2π]$.

In zero field, our model has a high symmetry so that $χ_L$ and $J$ do not depend on $α$. Then, $\sum_{α=1}^{n} ζ^4_α/n$ has the spherical symmetry in the $ζ$ space and is equal to $χ_L$. We obtain

$$
χ_L = \frac{β}{n} \left(1 + (n - 1) \frac{\langle \sinh(βζ)/βζ\rangle_ζ}{\cosh(βζ)}\right) , \quad (3.9)
$$

$$
χ_L = \frac{β}{n} \left(1 + (n - 1) \frac{F(n/2, 3/2; βζ/2)}{F(n/2, 1/2; βζ/2)}\right) , \quad (3.10)
$$

where $\langle \cdot \cdot \cdot \rangle_ζ$ stands for $\int_0^∞ dζ \cdot \cdot \cdot ζ^{n-1} \exp[-βζ^2/(2J)]$ and $F(α, γ; z)$ is Kummer’s confluent hypergeometric function. In fact, this equation is common to the results of the SA for the Heisenberg model $(n = 3)$, the XY model $(n = 2)$ and the Ising model $(n = 1)$ in zero field. Let us take such spin models to see the dependence on $n$. The first term in the bracket of eq.(3.10) represents contribution from a fluctuating field parallel to the spin and the second term with factor $n - 1$ perpendicular to it. We note that the quotient $F(\cdot \cdot \cdot )/F(\cdot \cdot \cdot )$ in the second term of eq.(3.10) is less than unity except for $βζ = 0$. The quotient is reduced to a simple form $(1 + βζ)^{-1}$ for $n = 3$, and $(3 + βζ)(3 + 6βζ + β^2ζ^2)^{-1}$ for $n = 5$. For $n = 2$, it takes a rather complicated form including the incomplete gamma function. As $n$ becomes larger, the number of perpendicular components increases and thus the perpendicular part in eq.(3.10) becomes dominant. Hence it leads to the decrease of $χ_L$. It can be shown by more detailed investigation that $χ_L$ is a monotonically decreasing function of $n$ at a fixed temperature. Physically, the short-range order of the $z$ component disturbs orderings of $x$ and $y$ components. This corresponds in the SA to the fact that the distribution of $ζ^2$ makes $χ_L^x$ and $χ_L^y$ decrease and the orderings of $x$ and $y$ components become less favorable. In §4, we investigate the effect in more detail. This effect becomes large as the number of fluctuating components increases. We note that the Ising model with $n = 1$ does not have such fluctuation effect.

Once a magnetic field is applied, the model has a lower symmetry and each $J^{α}$ can have different values. As temperature decreases, one of them reaches the critical value and satisfies eq.(3.3). Hence one must solve the set of nonlinear equations to determine the other $J^{α}$’s and the critical temperature. We used the Newton method to solve the set of equations. In solving it, we took care of the fact that $⟨χ_L^x⟩− 1 + J^α$ in the integral in eq.(3.4) is always larger than $J_G^α$. At finite magnetic fields, the multiple Gaussian integral could not be performed analytically, and therefore we did numerical integration. For the $Γ_3$- and the $Γ_5$-type interactions, the integral is no more than a double integral by symmetry, and thus executable. However, for the $(Γ_3 + Γ_5)$-type interaction, the integral on the unit hypersphere represented by eq.(3.8) was too time-consuming to use the standard routine of numerical integration. Therefore, we approximated the integral by summation over ten points $(±1, 0, 0, 0, 0), (0, ±1, 0, 0, 0), \cdot \cdot \cdot , (0, 0, 0, ±1)$ on the unit hypersphere in five dimensions. This method is exact in zero field. We have checked that the same approximation for the $Γ_3$- and the $Γ_5$-type interactions gives only a few percent of deviation in the transition temperature from the results with use of the double integral.

For calculation of the specific heat, we use the formula $C = dU/dT$ where the internal energy $U = ⟨H⟩$ is differentiated numerically. In this paper, we restrict the calculation only to the disordered phase. Then, expectation values of quadrupole moments $⟨O^{α}(i)⟩$ and dipole moments $⟨M(i)⟩$ do not depend on the site index $i$. The internal energy is written within the SA as

$$
U = -\frac{T}{2} \sum_{α=1}^{n} J^{α}_q x^{α}_q - \frac{1}{2} \sum_{ij} J^{α}_ij ⟨O^{α}⟩^2 - NH \cdot ⟨M⟩ \quad (3.11)
$$

$$
= -N \left(\frac{T}{2} \sum_{α=1}^{n} J^{α}_q x^{α}_q + \frac{1}{2} \sum_{q=0}^{α} J^{α}_q ⟨O^{α}⟩^2 + H \cdot ⟨M⟩\right) , \quad (3.12)
$$

where $x^{α}_q = x^{α}_q \{1 - (J^{α}_q - J^{α})x^{α}_q\}^{-1}$ and the site index is omitted. We used here the relation $N^{-1} \sum_q J^{α}_q x^{α}_q = J^{α}_q x^{α}_q$, which is derived from eq.(3.4). All of the quantities in eq.(3.12) can be calculated with the effective
4. Numerical Results

4.1 Quadrupolar ordering temperature

In contrast to the MFA, the present effective medium theory with the SA gives the result that $T_Q(H)$ increases with $H$. Figure 1 shows the phase diagrams obtained by the present theory and those by the MFA for comparison. The low temperature phase is the antiferro-quadrupolar phase. The magnetic field is oriented along either (001) or (111). As magnetic field increases, numerical integration of Gaussian distribution becomes difficult. Hence the phase diagrams are plotted up to the maximum field we can calculate.

We take the interaction constant $J'$ as the unit of energy, where $J'$ refers to either $J'_{\text{3}}$ or $J'_{\text{5}}$ in eq.(2.10). Accordingly $J'/k_B$ is taken to be the unit of temperature and $J'/\mu_B$ the unit of magnetic field. In this unit, $T_Q(H=0)$ in the MFA is 6 ($=z_n$) irrespective of the number of competing order parameters. With inclusion of fluctuations we have the following relation:

$$T_Q^{3+5} < T_Q^{5} < T_Q^{3}.$$  

At finite $H$, $T_Q(H)$ shows some increase except for the $\Gamma_3$-type with $H\|\langle 111 \rangle$. The rate of increase $R \equiv \{\max[T_Q(H)] - T_Q(0)\}/T_Q(0)$ satisfies $R^{3+5} < R^{5} < R^{3+5+18}$. That is to say, the rate becomes larger as the number of components of interaction increases.

The types of order parameters on the phase boundary are summarized in Table I. A cusp in the phase diagram means the change from $(O_2^0)$ to $(O_2^2)$ with increasing $H$. The magnetic field $H_e$ at the cusp is also shown in Table I. The phase boundary for the $(\Gamma_3 + \Gamma_5)$-type interaction in weak fields has very small dependence on the orientation of magnetic field. Although $T_Q(H)$ in weak fields seems to have almost linear field-dependence in Figure 1, it is in fact perpendicular to the $T$-axis at $H=0$. Since we are interested mainly in the vicinity of the phase boundary between the phases I and II, study of the order parameters at $T < T_Q(H)$ is left for future study.

4.2 Specific heat and entropy

Figure 2 shows the specific heat for the case of the $\Gamma_3$-type interaction with $H\|\langle 111 \rangle$ above the transition temperature $T_Q(H)$. We notice that the MFA gives zero specific heat in zero field above $T_Q(H)$. It is known in the SA for the Ising model that the specific heat has a cusp at the transition without a jump. We expect a similar behavior in our model although we have not calculated the specific heat below the transition temperature. At weak magnetic field the specific heat decreases monotonically as temperature increases above $T_Q(H)$. It is evident that the specific heat near $T_Q(H)$ becomes sharper and larger with increasing magnetic field. With further increase of the field beyond $\mu_B H \sim 5 J'$, the peak becomes smaller and the Schottky type anomaly appears. This is due to the Zeeman splitting of the $\Gamma_8$ levels. We discuss implication of the result to experiment in § 5.

We calculate the entropy relative to the value at the transition temperature by numerical integration of the specific heat. Figure 3 shows the result. In the high temperature limit the entropy should tend to $\ln 4$. Hence we can roughly estimate the value at $T_Q(H=0)$ since it almost saturates in the high temperature end of the calculated range. It is remarkable that the entropy at the transition temperature is much less than the full value $\ln 4$ even in the absence of Zeeman splitting. We will report the details of thermodynamic quantities including magnetization for various direction of $H$ and different types of interactions in a separate paper.

5. Mechanism of Increased Transition Temperature by Magnetic Field

5.1 Decreasing number of fluctuating components

Our results show pronounced difference of the phase diagram and entropy between the MFA and the SA. The simplest model we take proves to be a convenient model to investigate the effect of the fluctuations. In this section we discuss how a magnetic field suppresses the fluctuation.

When different parameters are competing for the stability, the transition temperature should be lowered in general. If there are many competing order parameters, different types of short-range order should occur and disturb each other. Therefore the transition temperature should become lower than that of a system in which only one of them is favored. For our model, the $\Gamma_3$-type interaction has two competing components, the $\Gamma_5$-type has three and the $(\Gamma_3 + \Gamma_5)$-type has five. Then, $T_Q(H=0)$ in the SA decreases with increasing number of components. This is the reason that a model with many components has many types of competing orderings. On the other hand, the MFA cannot deal with this competition and gives the same $T_Q(H=0)$ for all types of interaction.

This type of competition should be suppressed with increasing $H$ in our model. In other words, the ordering of the one (or two) of several components becomes more favorable than the others, and the number of competing orderings decreases. Consequently $T_Q(H)$ should increase. This effect becomes larger as the number of components increases because the situation changes more drastically when one of a larger number of components becomes favorable. Therefore, this mechanism naturally explains the result that the rate $R$ of increase in $T_Q(H)$ satisfies $R^{3} < R^{5} < R^{3+5+18}$. On the contrary, we obtained monotonically decreasing $T_Q(H)$ for the $\Gamma_3$-type interaction with $H\|\langle 111 \rangle$. In this case, such suppression of fluctuations does not occur because the ordering of $O_2^2$ is as favorable as that of $O_2^0$ owing to the trigonal symmetry. In order to confirm this mechanism, we calculated $T_Q(H)$ in another model which has only one component like the Ising model. This model does not have such mechanism of suppression. We indeed find that $T_Q(H)$ does not increase with $H$.

5.2 Effects of fluctuating magnetic field on spin ordering

The SA applied to the Heisenberg model and the XY model does not give increasing transition temperature
with increasing magnetic field. One difference between the Heisenberg model and the $\Gamma_3$-type interaction model is that while only one component among three components becomes favorable at finite magnetic fields in the latter, two components perpendicular to the magnetic field become favorable in the former. As a result, the suppression in the Heisenberg model is rather small. However, this remark cannot be applied in comparing the XY model and the $\Gamma_3$-type interaction. Another difference, which is probably more important, is that a magnetic field is not conjugate to quadrupole moments and is coupled with them in a complex way. To analyze this, it is convenient to use the pseudo-spin representation of the Zeeman term with $H||\langle 001 \rangle$, namely, $\{\sigma^x + (4/7)\tau^z \sigma^z\} \mu_B H$. The quadrupole moment $\sigma^z$ ($O_2^0$) couples to a kind of fluctuating field $\sigma^z$ which has the Ising-type distribution. As shown below, the fluctuating field $\sigma^z$ has a character of suppressing fluctuation of quadrupole moments. However, the Zeeman term plays a dual role of favoring a particular type among competing fluctuating fields, and also destroying the quadrupolar ordering as in antiferromagnets. In the following, we first take a spin model to clarify effects of a fluctuating magnetic field. Then, we investigate how a static magnetic field affects the fluctuation in the $\Gamma_3$-type interaction model. It is shown that the dominant effect of the Zeeman term at weak field is suppressing quadrupolar fluctuations.

First of all, we take a single classical spin coupled with a Gaussian fluctuating field. The Gaussian identity,

$$\int_{-\infty}^{\infty} \frac{d\varphi^z}{\sqrt{2\pi\beta V^z}} \exp[-\beta(\varphi^z)^2/2V^z + \varphi^z S^z] = \exp[\beta V^z/2(S^z)^2],$$

(5.1)

shows that a Gaussian fluctuating field $\varphi^z$ coupled to a classical variable $S^z$ is equivalent to a field $V^z/2$ coupled to $(S^z)^2$. That is to say, $\langle (S^z)^2 \rangle$ increases with increasing $V^z$ and the relation $-2\partial \chi/\partial V^z = \langle (S^z)^2 \rangle$ is satisfied where $\Omega$ is the thermodynamic potential. Therefore, the fluctuating field increases the susceptibility $\chi^z = \beta\langle (S^z)^2 \rangle$, the contrary, $\chi^x$ and $\chi^y$ are decreased because of the identity $\langle (S^z)^2 \rangle + \langle (S^y)^2 \rangle + \langle (S^x)^2 \rangle = S(S + 1)$. Note that the increase of $\chi^z$ and the decrease of $\chi^x$ and $\chi^y$ are of $O(V^z)$ because $V^z$ is a field conjugate to $(S^z)^2$. Even for a quantum spin, the discussion above is still valid provided that $S^z$ is replaced by $\beta^{-1} \int_0^\beta d\tau S^z(\tau)$. Namely, the susceptibility $\chi^z$ is given by

$$\chi^z = \beta \langle (S^z)^2 \rangle = \left\{ \beta^{-1} \int_0^\beta d\tau S^z(\tau) \right\}^2$$

(5.2)

A difference from a classical spin is that the susceptibilities are not directly determined by the identity $\langle (S^z)^2 \rangle + \langle (S^y)^2 \rangle + \langle (S^x)^2 \rangle = S(S + 1)$. For the case of $S = 1/2$, the susceptibilities in the presence of the Gaussian fluctuating field with variance $V^z/\beta$ are calculated as

$$\chi^z = \frac{\beta}{4}, \quad \chi^x = \frac{\beta F(1/2, 3/2; \beta V^z/2)}{4 F(1/2, 1/2; \beta V^z/2)}$$

(5.4)

$\chi^z$ has the same form as the quotient in the second term of eq.(3.10) and is a monotonically decreasing function of $V^z$. Therefore, the fluctuating field decreases $\chi^x$ and $\chi^y$, but does not affect $\chi^z$. The reason for the latter is that the Hamiltonian and $S^z$ are commutative, and that one has $\{\beta^{-1} \int_0^\beta d\tau S^z(\tau)\}^2 = (S^z)^2 = 1/4$ independent of $V^z$. For $S > 1/2$, $(S^z)^2$ is not a constant and thus $\chi^z$ is increased by the fluctuating field. Note that in any case the change of susceptibilities is of first order in $V^z$ by the same reason as in the classical system.

Next, we treat the lattice systems. Let us take the following model as an example which have both the Heisenberg-type exchange interaction and Gaussian fluctuating fields:

$$\mathcal{H}_{\varphi S} = -\frac{1}{2} \sum_{ij} J_{ij} S_i \cdot S_j - \sum_i \varphi_i S_i^z.$$

(5.5)

For classical spin systems and quantum systems with $S > 1/2$, the local susceptibility $\chi^z_i$ at each site is increased by the Gaussian fluctuating field as well as that of the single spin. Consequently, the transition temperature to the ordered state should increase. In the mean-field approximation, the susceptibility $\chi_Q = \chi_L / (1 - J_Q \chi_L)$ for a certain wave number $Q$ diverges at the phase transition. Hence the transition temperature increases if $\chi^z_i$ is increased by the fluctuating field $\varphi^z_i$. Even in quantum spin systems with $S = 1/2$, the transition temperature should still increase by the fluctuation effect. As $V^z$ increases, ordering of $S^z$ or $S^y$ should become less favorable. Therefore, in the limit of large $V^z$, only the $S^z$ ordering should be possible. This means that the fluctuation among components is suppressed. Certainly, the transition temperature increases with $V^z$ in the SA to this model. Further, the local susceptibility $\chi^z_i$ increases here. The reason is that $V^z$ makes $\chi^z_i$ and $\chi^y_i$ decrease. Then, $J^z$ and $J^y$ decrease and $\chi^z_i$ increases. On the other hand, when we solve the model by the mean-field approximation, the transition temperature $T_c$ for the $S^z$ and $S^y$ orderings decreases with increasing $V^z$, while $T_c$ for the $S^z$ ordering does not change. Namely, in the spin model defined by eq.(5.5) with $S = 1/2$ the transition temperature does increase with field in contrast to the result of the mean-field approximation. This fluctuating field has similar effect even if $\varphi^z$ is not a Gaussian-distributed variable but the Ising-type variable taking values of $\varphi^z = \pm \sqrt{V^z/\beta}$. This field resembles $\hbar z^2$ in the Zeeman term with $H||\langle 001 \rangle$ represented by the pseudo-spins for the quadrupolar model. We discuss it in the next subsection.

5.3 Two competing effects of magnetic field on the quadrupolar ordering

In the pseudo-spin representation, the interaction Hamiltonian for the $\Gamma_3$-type interaction is the same as that of the XY model. The only difference between them is the Zeeman term. Nevertheless, their phase diagrams
by the SA with $H \parallel (001)$ show a remarkable difference; while the transition temperature increases with $H$ in the $Γ_3$-type interaction, it does not in the XY model. In this subsection we discuss the reason for the difference in weak fields.

The Hamiltonian of the $Γ_3$-type interaction with $H \parallel (001)$ is written as

$$\mathcal{H} = -\frac{1}{2} \sum_{ij} J_{ij} (\tau_i^x \tau_j^x + \tau_i^y \tau_j^y) - \sum_i \mu_B H \sigma_i^z \left(1 + \frac{4}{7} \tau_i^\| \right),$$

where $h$ refers to $\mu_B H$. In this expression, the $σ$-spin emerges only as $σ^z$. Hence, one can treat it as a classical variable taking values $±1$. To study the structure of the partition function, we introduce path-integral representation for the $σ$-spin. Since the explicit expression for the Berry phase term is not necessary here, we write it symbolically as $L_B$.

$$Z = \sum_{\{σ_i\}} \int \mathcal{D}τ \exp \left[ -\int_0^β dτ \{L_B(τ) + \mathcal{H}(τ)\} \right],$$

(5.8)

After summation over $σ^z$, we neglect terms of higher order in $β h$ to investigate the behavior in weak fields. We use the approximation $\cosh x \simeq e^{x^2/2}$ for $x \ll 1$. Here we have

$$x = h \int_0^β \left(1 + \frac{4}{7} τ_i^\| (τ) \right) dτ.$$

Then we use the identity eq.(5.11) to introduce the fluctuating field $ϕ_\|$. Finally we turn to the operator representation with $\text{Tr}$ instead of $\int \mathcal{D}τ$. The calculation explained above goes as

$$Z \simeq 2^N \int \mathcal{D}τ \exp \left[ -\int_0^β \{L_B(τ) + \mathcal{H}_{\text{int}}(τ)\} dτ + \frac{(β h)^2}{2} \sum_i \left\{ \frac{1}{β} \int_0^β \left(1 + \frac{4}{7} τ_i^\| (τ) \right) dτ \right\}^2 \right]$$

$$= 2^N \int \mathcal{D}τ \prod_i \int_{-∞}^{∞} \frac{dϕ_i^\|}{\sqrt{2πh^2}} \exp \left[ -\int_0^β \{L_B(τ) + \mathcal{H}_{\text{int}}(τ)\} dτ \right.$$

$$+ \sum_i \left\{ \frac{(ϕ_i^\|)^2}{2h^2} + ϕ_i^\| \int_0^β \left(1 + \frac{4}{7} τ_i^\| (τ) \right) dτ \right\} \right]$$

$$= 2^N \prod_i \int_{-∞}^{∞} \frac{dϕ_i^\|}{\sqrt{2πh^2}} \exp \left[ -\sum_i \frac{(ϕ_i^\|)^2}{2h^2} \right] \text{Tr} \left[ -β \left\{ \mathcal{H}_{\text{int}} - \sum_i ϕ_i^\| \left(1 + \frac{4}{7} τ_i^\| \right) \right\} \right]$$

$$= 2^N \prod_i \int_{-∞}^{∞} \frac{7dϕ_i^\|}{4\sqrt{2πh^2}} \exp \left[ \sum_i \left( -\frac{(ϕ_i^\|)^2}{2(4h/7)^2} + \frac{1}{2} β h^2 \right) \right]$$

$$\times \text{Tr} \left[ -β \left\{ \mathcal{H}_{\text{int}} - \sum_i \left( ϕ_i^\| τ_i^\| + \frac{4}{7} β h^2 τ_i^\| \right) \right\} \right].$$

(5.12)

In the expression (5.11), the only difference from the original expression (5.6) is that the Ising-type distribution $hσ_i^z$ is replaced by $ϕ_i^\|$, which obeys the Gaussian distribution with the variance $h^2$. Then, we replaced $ϕ_i^\|$ by $(7/4)ϕ_i^\| + βh^2$ in deriving eq.(5.12).

The linear term with respect to $h$ is absent in eq.(5.12) and $τ^\|$ couples to $h^2$ because of the time reversal symmetry. The term $-(4/7)βh^2 τ_i^\|$ is of the same form as the Zeeman term in the XY model and thus it works to induce $⟨τ_i^\|⟩$ uniformly and to destroy the antiferro-type ordering. Since the lowest order contribution of a magnetic field to the susceptibilities in the XY model is the square of the field, the lowest-order contribution of the term $-(4/7)βh^2 τ_i^\|$ to the strain susceptibilities should be the square of $-(4/7)βh^2$, namely $O[(βh)^2]$.

On the other hand, the Gaussian fluctuating field $ϕ_i^\|$ with variance $(4h/7)^2$ affect the susceptibilities by $O[(βh)^3]$ as described in the previous subsection. Therefore, it should make a dominant contribution at weak fields. This term makes the ordering of $τ^\|$ favorable. After all a weak magnetic field suppresses the fluctuation and increases the transition temperature.

In the previous paper of Uimin and the present authors the pseudo-spins for the quadrupole moments are approximated by classical spins. In the previous subsection, we have shown that the fluctuating field $ϕ_i^\|$ has smaller influence on $χ$ in the quantum spin with $S = 1/2$ than that in the classical spin. Hence the approximation in ref. enhances the effect of the fluctuating field $ϕ_i^\|$ in eq.(5.12). The classical treatment should be the reason why the result of the $Γ_3$-type interaction with $H \parallel (001)$ shows much larger increase of $T_Q(H)$ than that of the present paper.
§6. Discussion and Concluding Remarks

In this paper we have shown for quadrupolar interaction systems that a magnetic field has two contrasting effects. One effect is inducing quadrupole moments uniformly and destroying the antiferro-quadrupolar ordering. The induced quadrupole moments are even functions of magnetic field owing to the time reversal symmetry, and the change of $T_Q(0)$ is very small ($\propto H^4$) at weak fields. The other effect is suppressing the competition among different components of order parameters. Since the resultant change is of $O(H^2)$, the latter effect dominates at weak fields. Hence the transition temperature increases as $H$ increases.

For comparison with experimental results in CeB$_6$, a crude estimate of $J'$ is obtained by comparing the theoretical value of $T_Q(0)$ with the experimental one. With the value of $J'$ so determined, the unit for magnetic field is also fixed. Let us assume for the moment the $\Gamma_5$ order. Since the numerical value of $T_Q(0)$ in units of $J'/k_B$ is the same order as the experimental value in units of Kelvin, one may roughly estimate $J'/k_B \sim 1K$, and the unit for magnetic field as 1 T (tesla). Then the increase of $T_Q(H)$ in Figure 1 turns out not so strong as the experimentally observed one, and the re-entrant field is much smaller than the actual one which is larger than the maximum of available magnetic field.

On the other hand, results of the specific heat at weak fields agree qualitatively with experimental ones both for the magnitude and the temperature dependence. We emphasize that the lowering of the cubic symmetry is not necessary to explain the entropy smaller than ln 4 at the transition temperature, and the large tail of the specific heat experimentally observed. The temperature range of a weak Schottky type anomaly found in Figure 2 for $H \sim 12$ T and larger overlaps actually with the experimental $T_Q(H)$ up to 15 T in CeB$_6$. Hence the anomaly is not seen experimentally. However in the alloy system Ce$_{0.5}$La$_{0.5}$B$_6$, such Schottky anomaly is indeed observed due to the lowered $T_Q(H)$.

For serious comparison with experimental results, we have to consider two directions of development on the basis of the present study. The first is to choose a realistic model to describe the interaction among quadrupole moments at different sites within the SA. Concerning the intersite interaction, the role of the range $J'$ and types including higher multipoles should be reexamined with inclusion of fluctuations. With the intersite octupolar interaction, the effect of magnetic field on the shift of $T_Q(H)$ becomes larger and should compare more favorably with experiment. Although we have neglected the higher crystal field levels other than the ground level, the crystal field splitting is so small in TmTe that the $\Gamma_6$ and the $\Gamma_7$ doublets seem to be within a few meV above the ground $\Gamma_8$ level. Hence, it is interesting to investigate how the crystal field splitting affects the phase diagram of the quadrupolar ordering.

Another direction of development is including dynamical and quantum effects of fluctuations. The most important may be the Kondo effect in the case of CeB$_6$ but not in the case of insulating systems such as TmTe. In the latter case the quantum fluctuation of multipole moments should persist even without the Kondo effect. In order to deal with dynamical effects in the effective medium theory, numerical methods such as the resolvent method, the quantum Monte Carlo or the numerical renormalization group seem promising. We hope to report on these development in the near future.

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Table 1. Order parameters on the phase boundary. Here $H_{c}$ refers to the critical magnetic field at which the type of order parameters changes.

| $\Gamma_{3}$ | $H < H_{c}$ | $H > H_{c}$ | $H || (111)$ | $H_{c}$ |
|-------------|-------------|-------------|-------------|---------|
| $\langle O_{2x} \rangle$ | $\langle O_{2y} \rangle$ or $\langle O_{2z} \rangle$ | $(O_{xx} + O_{xy} + O_{yx})/\sqrt{3}$ | 5.7 |
| $\Gamma_{5}$ | $\langle O_{xy} \rangle$ | $\langle O_{xy} \rangle$ | $\langle O_{xy} \rangle$ | $\langle O_{xy} \rangle$ | 4.8 |
| $\Gamma_{3} + \Gamma_{5}$ | $\langle O_{2x} \rangle$ or $\langle O_{2y} \rangle$ | $\langle O_{xy} \rangle$ | $\langle O_{xy} \rangle$ | $\langle O_{xy} \rangle$ |

Fig. 1. The phase diagrams obtained by the SA(solid lines) and the MFA(broken lines).
Fig. 2. The specific heat above the transition temperature obtained by the present theory for the Γ₅-type interaction with $H || (111)$. Each transition temperature is indicated by an arrow. The unit of the ordinate corresponds to 8.31J/(mol·K).

Fig. 3. The change of entropy relative to the value at the transition temperature.