Lattice Distortion and Octupole Ordering Model in Ce$_x$La$_{1-x}$B$_6$

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Possible order parameters of the phase IV in Ce$_x$La$_{1-x}$B$_6$ are discussed with special attention to the lattice distortion recently observed. A $\Gamma_{5u}$-type octupole order with finite wave number is proposed as the origin of the distortion along the [111] direction. The $\Gamma_8$ crystalline electric field (CEF) level splits into three levels by a mean field with the $\Gamma_{5u}$ symmetry. The ground and highest singlets have the same quadrupole moment, while the intermediate doublet has an opposite sign. It is shown that any collinear order of $\Gamma_{5u}$-type octupole moment accompanies the $\Gamma_{5u}$-type ferro-quadrupole order, and the coupling of the quadrupole moment with the lattice induces the distortion. The cusp in the magnetization at the phase transition is reproduced, but the internal magnetic field due to the octupole moment is smaller than the observed one by an order of magnitude.

KEYWORDS: rare-earth hexaboride, Ce$_x$La$_{1-x}$B$_6$, NdB$_6$, NpO$_2$, octupole ordering, thermal expansion, internal field

The multipolar orderings in cubic rare-earth hexaborides have been studied extensively. Especially, CeB$_6$ is a typical material which shows multipolar orderings. In CeB$_6$, there are three phases: the paramagnetic phase (called the phase I), the antiferro-quadrupole phase (II) and the antiferromagnetic phase (III). In Ce$_2$La$_{1-x}$B$_6$, another phase, so called phase IV, was found at $x \simeq 0.75$.\(^1\) This phase has attracted much attention, but the order parameter has not yet been established. In the phase IV, a large softening of the elastic constant $C_{11}$ was observed.\(^2\) Furthermore the magnetic susceptibility shows a cusp at the transition temperature $T_{1IV}$ from the phase I, and the magnetization is almost isotropic at ambient pressure.\(^3\)

Recently, a small lattice distortion along the [111] direction was observed in the phase IV.\(^4\)\(^5\) It is probable that the softening of $C_{11}$ is associated with this distortion. It may be tempting to ascribe the distortion to the ferro $\Gamma_{5u}$-type quadrupole order. However, the quadrupole moment is not necessarily the primary order parameter. Large change of the internal field at $T_{1IV}$, as probed by NMR\(^6\) and $\mu$SR,\(^7\) suggests strongly that the time reversal symmetry is broken in the phase IV, which is incompatible with a pure quadrupole order. Besides, neutron diffraction experiment found no magnetic reflection in the phase IV.\(^8\) Thus dipole moments are unlikely to be the primary order parameter either, although the time reversal symmetry is broken. Therefore, octupole moments, which break the time reversal symmetry, become a candidate for the order parameter in phase IV.\(^9\)\(^10\)

Since the cubic symmetry is broken, and since the anisotropy develops in the magnetization under uniaxial pressure,\(^11\)\(^12\) the order parameter should have an anisotropic nature. Thus the $\Gamma_{5u}$-type, among all octupole moments, is the most plausible candidate for the order parameter in the phase IV.

Kusunose and Kuramoto\(^10\) have already pointed out using the Ginzburg-Landau (GL) theory that the $\Gamma_{5u}$ octupole order with finite wave number should accompany a ferro-quadrupole moment, and have suggested a possible lattice distortion. However, evaluation of the magnitude of the distortion is beyond their GL theory. In this paper, we explore in much greater detail the consequence of the $\Gamma_{5u}$ octupole order by the mean field theory, and propose that the lattice distortion is due to the order of the $\Gamma_{5u}$ octupole moment.

The CEF ground state of Ce$^{3+}$ ($J = 5/2$) in Ce$_x$La$_{1-x}$B$_6$ is the $\Gamma_8$ quartet.\(^13\)\(^15\) The excited level $\Gamma_7$ lies about 500K from the $\Gamma_8$ level and is neglected. The $\Gamma_8$ states are represented in terms of eigenstates of $J_z$ as

$$|\uparrow\rangle = \sqrt{5/6}|5/2\rangle + \sqrt{1/6}|3/2\rangle, \quad (1)$$
$$|\downarrow\rangle = |1/2\rangle, \quad (2)$$

where $\uparrow$ and $\downarrow$ denote orbital indices, and their Kramers partners $|\uparrow\rangle$, $|\downarrow\rangle$ are obtained by reversing the signs of $J_z$ in eqs. (1) and (2), respectively. Within the $\Gamma_8$ quartet, the number of independent multipolar moments is 15, and active octupole moments have either of $\Gamma_{2u}$, $\Gamma_{4u}$ or $\Gamma_{5u}$-type symmetry.\(^16\) The $\Gamma_{4u}$-type octupole moments accompany dipole moments,\(^16\) and are unlikely to be the order parameter in the phase IV. The $\Gamma_{2u}$-type octupole moment, as proposed by ref.9, does not accompany quadrupole moments. Therefore it seems difficult to explain the lattice distortion in the phase IV by the $\Gamma_{2u}$ octupole orderings.

Let us introduce pseudospins $\sigma$ and $\tau$ to describe the $\Gamma_8$ quartet:

$$\tau^z = |\pm\rangle = |\pm\rangle = |\pm\rangle = \pm \sigma^z, \quad (3)$$

$$\sigma^z |\tau^\uparrow\rangle = + |\tau^\uparrow\rangle, \quad \sigma^z |\tau^\downarrow\rangle = - |\tau^\downarrow\rangle, \quad (4)$$

and the transverse components which flip the pseudospins. The $\Gamma_{5u}$ octupole, $\Gamma_{3g}$ and $\Gamma_{5g}$ quadrupole moments are given with the notation $(\alpha, \beta, \gamma) = (x, y, z)$, $(y, z, x)$ or $(z, x, y)$ by

$$T^{\alpha}_{\beta\gamma} = (J_{\alpha}J_{\beta}^2 - J_{\beta}J_{\alpha}^2)/(2\sqrt{3}) = (\zeta^+ \sigma^x \zeta^- \sigma^y \tau^z), \quad (5)$$

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Thus, ferro, antiferro and other collinearly ordered states the transition from the phase I to IV, and little from state is two-fold degenerate as shown in the left part of the energy level splits into two levels, and the ground it hard for the $\Gamma_5$ quadrupole moment is only the $\Gamma_5$ quadrupole induces quadrupole moments, but which accompanies no dipole moment, is likely that the degeneracy of the $\Gamma_5$ quadrupole. We find that the ‘easy axis’ of $\Gamma_5$ quadrupole moment $\langle \zeta \rangle$ is an easy axis (left).

$O_2 = (2J_z^2 - J_y^2 - J_x^2)/\sqrt{3} = (8/\sqrt{3})r^2$, \hspace{1cm} (6)

$O_2 = J_x^2 - J_y^2 = (8/\sqrt{3})r^2$, \hspace{1cm} (7)

$O_{\beta\gamma} = 2J_\beta J_\gamma = (2/\sqrt{3})\tau^\beta\tau^\gamma$, \hspace{1cm} (8)

where bars on the products represent symmetrization, e.g., $J_x J_y = (J_x J_y + J_y J_x + J_x J_x)/3$, and we have introduced the notation $\zeta = -(r^x \pm \sqrt{3}r^z)/2$.

We find that the ‘easy axis’ of $\Gamma_5$ octupole moment is along the [111] direction and equivalent ones, i.e., $\langle |O_{x}^\alpha|, |O_{y}^\alpha|, |O_{z}^\alpha| \rangle \parallel (1, 1, 1)$ provided the intersite interaction is isotropic. Then we consider the following single-site Hamiltonian:

$$\mathcal{H}_{\text{single-site}} = A_5u (T_x^5u + T_y^5u + T_z^5u),$$ \hspace{1cm} (9)

where $A_5u$ denotes the octupolar mean field. The energy levels of this Hamiltonian are shown in the right part of Fig. 1. The $\Gamma_5$ level splits into three levels. Not only the time reversal symmetry, but also the cubic symmetry are broken. The multipolar moments in the ground and highest states are $\langle T_x^5u \rangle = \langle T_y^5u \rangle = \langle T_z^5u \rangle = +\sqrt{2}/3$, $\langle O_{zy} \rangle = \langle O_{zx} \rangle = \langle O_{xy} \rangle = -2/3$ and the others are zero.

Thus, ferro, antiferro and other collinearly ordered states with $\langle (T_x^{5u}), (T_y^{5u}), (T_z^{5u}) \rangle \parallel (1, 1, 1)$ have a homogeneous $\Gamma_5$ octupole moment and the crystal distorts along [111].

Obviously a $\Gamma_5$-type ferro-quadrupole interaction can also lead to $\langle O_{zy} \rangle = \langle O_{zx} \rangle = \langle O_{xy} \rangle = 0$. In this case the energy level splits into two levels, and the ground state is two-fold degenerate as shown in the left part of Fig. 1. Experimentally, the entropy changes much at the transition from the phase I to IV, and little from the phase IV to III in $\mathrm{Cu}_{0.75}\mathrm{La}_{0.25}\mathrm{B}_6$. Thus it is likely that the degeneracy of the each $f$-electron state is already lifted in the phase IV. This situation also makes it hard for the $\Gamma_5$ quadrupoles to be the order parameter in the phase IV.

From the above discussion, we find that the order parameter which breaks the time reversal symmetry and induces quadrupole moments, but which accompanies no dipole moment is only the $\Gamma_5$ octupole moment. However, we cannot determine the periodicity of the $\Gamma_5u$ ordered state in the phase IV only from the above consideration, because any collinearly ordered state with $\langle (T_x^{5u}), (T_y^{5u}), (T_z^{5u}) \rangle \parallel (1, 1, 1)$ has a uniform $\Gamma_5$ moment. To carry out the mean field theory explicitly, we consider a G-type antiferro-octupole order as the simplest example of $\Gamma_5u$ orders. The importance of $\Gamma_5u$ nearest-neighbor interaction in causing the change from the phase III to III’, even with weak magnetic field, was pointed out in ref.10, and we consider only this nearest-neighbor interaction. We take the following model:

$$\mathcal{H} = I_{5u} \sum_{(i,j)} \sum_{\alpha=x,y,z} \frac{5}{3} T_{\alpha i}^5 T_{\alpha j}^5,$$ \hspace{1cm} (10)

where $(i,j)$ denotes a nearest-neighbor pair. We study this Hamiltonian by the mean field theory, and choose the value of $I_{5u}$ so as to reproduce the transition temperature $T_{\text{IV}}$ in $\mathrm{Cu}_{0.75}\mathrm{La}_{0.25}\mathrm{B}_6$, i.e., $T_{5u} = 6I_{5u} = 1.7\mathrm{K}$.

The temperature dependence of $\langle T_x^{5u} \rangle$ and $\langle O_{zy} \rangle$ are shown in Figs. 2 and 3. The solutions obtained has the antiferro-octupole order with $\langle T_x^{5u} \rangle = \langle T_y^{5u} \rangle = \langle T_z^{5u} \rangle$, accompanying ferro-quadrupole moment $\langle O_{zy} \rangle = \langle O_{zx} \rangle = \langle O_{xy} \rangle$. We also find other equivalent solutions: $\langle (T_x^{5u}), (T_y^{5u}), (T_z^{5u}), (O_{zy}), (O_{zx}), (O_{xy}) \rangle = \pm B, \pm B, \pm B, \pm C, \pm C, -C$ where quantities $B$ and $C$ depend on temperature.

In Fig. 4, we show the magnetization in magnetic field

Fig. 1. The level scheme in the $\Gamma_5u$ interaction (right) and in the $\Gamma_5g$ interaction (left).

Fig. 2. Temperature dependence of the antiferro-octupole moment $\langle T_x^{5u} \rangle = \langle T_y^{5u} \rangle = \langle T_z^{5u} \rangle$.

Fig. 3. Temperature dependence of the ferro-quadrupole moment $\langle O_{zy} \rangle = \langle O_{zx} \rangle = \langle O_{xy} \rangle$. 
$H = 0.2T$ along three high-symmetry directions. The magnetization has a cusp at $T_{Su}$, which is consistent with experimental observation.$^3$ We have also examined the case where the pure $\Gamma_{5g}$ quadrupole moment is the order parameter, and found that the magnetization changes little at the quadrupole transition temperature. The magnetization below $T_{Su}$ is anisotropic. One should not, however, take this anisotropy seriously since other interactions such as quadrupole and dipole interactions also influence the anisotropy. In fact the easy axis in the mean field theory is different from that observed in the phase IV under uniaxial pressure and in the phase III.

We now consider lattice distortion in the antiferro-octupole ordered state. The elastic energy associated with the $\Gamma_{5g}$ moments is given per unit volume by

$$E = \sum_{\alpha\beta=yz,zx,xy} \left( 2\epsilon_{\alpha\beta} C_{44}^{(0)} + g_{rs} \sum_i \epsilon_{\alpha\beta} \langle O_{\alpha\beta i} \rangle \right),$$

where $\epsilon_{\alpha\beta}$ is the strain tensor, $g_{rs}$ is the magneto-elastic coupling constant, and $C_{44}^{(0)}$ is the (bare) elastic constant. The sum runs over Ce sites $i$ in the unit volume. By minimizing the elastic energy, we obtain

$$\epsilon_{\alpha\beta} = -\frac{g_{rs}}{4C_{44}^{(0)}} \sum_i \langle O_{\alpha\beta i} \rangle.$$

We use the following experimental values for Ce$_{0.75}$La$_{0.25}$B$_6$: $g_{rs} = 155K$, $C_{44}^{(0)} \approx 8.2 \times 10^{11}$erg/cm$^3$, and the lattice constant $a = 4.13\AA$. At absolute zero, the magnitude of the quadrupole moments is given by $|\langle O_{yz i} \rangle| = |\langle O_{zx i} \rangle| = |\langle O_{xy i} \rangle| = 2/3$, and we obtain

$$|\epsilon_{yz}| = |\epsilon_{zx}| = |\epsilon_{xy}| = 4.6 \times 10^{-5}.$$

In order to account for the observed lattice contraction along [111], we consider the two possibilities: (i) $g_{rs} < 0$, and (ii) $g_{rs} > 0$. In the case (i), a slight stress accompanying the measurement breaks the equivalence of four octupole domains, and choose a domain for which the contraction becomes the maximum along [111]. Namely we have

$$\Delta l/l = 2\epsilon_{yz} = -9.3 \times 10^{-5},$$

with $\langle O_{yz i} \rangle = \langle O_{zx i} \rangle = \langle O_{xy i} \rangle = -2/3$. Note that along directions [111], [111], and [111] the lattice should expand by

$$\Delta l/l = 2|\epsilon_{yz}|/3 = 3.1 \times 10^{-5}.$$ (15)

On the other hand, in the case (ii), a positive stress along [111] may favor a domain which contracts along this direction. Then the contraction is given by eq.(15) with the minus sign. In the single domain with $-\langle O_{yz} \rangle = \langle O_{zx} \rangle = \langle O_{xy} \rangle$ for example, an expansion along [111] should be present, and its magnitude is three times larger than the contraction along [111].

A magnetic field induces antiferromagnetic moments which tend to be perpendicular to $H$. Hence with $H || [111]$ without external stress, a state where one of $\langle O_{\alpha\beta} \rangle$'s has a sign different from the others is stabilized. Experimentally the contraction is enhanced under $H || [111]$, which alone favors the case (ii) in our model. In the actual system, diagonal components $\epsilon_{\alpha\alpha}$ becomes positive in the phase IV. This comes from the volume strain which is not included in our model. According to experiment,$^5$ the shear strain with the assumption of the trigonal symmetry around [111], i.e., the case (i), is derived as $\epsilon_{\alpha\beta} = -4 \times 10^{-6}$ without magnetic field at 1.3K. Assuming that the phase IV is stable down to zero temperature, $\epsilon_{\alpha\beta}$ extrapolate to $(-6 \sim -10) \times 10^{-6}$ at absolute zero. The absolute value is by an order of magnitude smaller than our estimate in eq.(13). Then the observed reduction of $|\langle O_{\alpha\beta} \rangle|$ should come from quantum fluctuations neglected here. Another possibility is that the case (ii) is realized. In this case three times larger $|\epsilon_{\alpha\beta}|$, i.e., $(2 - 3) \times 10^{-5}$, is obtained from the same experimental result.$^5$ Our model in the case (ii) predicts a larger lattice contraction along the [110] direction with $H || [110]$ than that along the [111] direction with $H || [111]$.

For comparison, we estimate in the same way the magnitude of the lattice distortion of NdB$_6$ in the $O_2^3$ ferro-quadrupole ordered state by using the experimental values: $|g_{rs}| = 220K$, $(C_{11} - C_{12})/2 \approx 20 \times 10^{11}$erg/cm$^3$, $a = 4.12\AA$, and the Lea-Leask-Wolf parameter $x_{LLW} = -0.82$.$^{19}$ We assume that $(C_{11}^{(0)} - C_{12}^{(0)})/2$ is almost the same as the observed $(C_{11} - C_{12})/2$. In a magnetic field $H || [100]$, a state with the antiferromagnetic moment perpendicular to [100] is stabilized. For a domain where the magnetic moment is along [001], we obtain $|\Delta l/l| = 2.8 \times 10^{-4}$ along $H$. This value compares favorably with the experimental one $|\Delta l/l| \approx -2 \times 10^{-4}$ at 2K and $H = 2.1T$.$^{20}$ The distortion in NdB$_6$ is by an order of magnitude larger than that in eqs.(14) and (15). The reason is that the quadrupole moment $|\langle O_{yz}^{(0)} \rangle| = 4.5$ in NdB$_6$ is much larger than the corresponding value $|\langle O_{yz} \rangle| = 2/3$ in the octupole state of Ce$_{0.5}$La$_{0.5}$B$_6$, while the magneto-elastic coupling constants are of the same order.

We now discuss the internal magnetic field associated with the octupole order. The $\mu$SR time spectra in the phase IV consist of a Gaussian component and an exponential component.$^7$ The observation of a Gaussian relaxation indicates that internal fields are randomly distributed, and/or fluctuating. The internal field deduced
from the Gaussian relaxation is the order of 0.1T. We discuss whether the octupole moment can be the origin of the Gaussian relaxation. In μSR measurement, μ^2 locates at (a/2, 0, 0) and equivalent sites^{21-23} with a Ce ion chosen as the origin. As a reference the internal field from a Bohr magneton μ_B is estimated to be
\[ H_{\text{dipole}} = \mu_B/(a/2)^3 \sim 0.1T, \]  with a/2 ≈ 2Å. The internal field from an octupole with the size r is estimated to be
\[ H_{\text{octupole}} = \mu_B r^2/(a/2)^5 \sim 0.01T, \]  with r ≈ a_B = 0.53Å. For a more accurate estimate, we consider the multipole expansion of the vector potential from local electrons as given by^{24}
\[ A(r) = \sum_{k,m} \frac{-i}{k} r^{-(k+1)} \left( IC_{m}^{(k)}(\theta, \phi) \right) \hat{M}_{k,m}^{m}, \]  (16)
where l is the orbital angular momentum operator, \( C_{m}^{(k)}(\theta, \phi) \) is \( 4\pi/(2k+1) \) times the spherical harmonics \( Y_{k,m}(\theta, \phi) \), and \( \hat{M}_{k,m}^{m} \) is the magnetic multipole moment. The multipole moment is determined by the wave function \( \psi_i(r) \) of the i-th electron, the orbital and spin angular momentum operators \( \hat{l}_i \) and \( \hat{s}_i \). Namely we have
\[ M_{k,m}^{m} = \mu_B \sum_i \int \mathrm{d}r \psi_i^*(r) \left( \nabla \psi_i \right)^* C_{m}^{(k)}(\theta, \phi) \left( \hat{l}_i + 2\hat{s}_i \right) \psi_i(r). \]  (17)
Eq.(17) is evaluated with use of the operator equivalents method^{25}. For our purpose it is sufficient to consider only octupole moments. Then we obtain \( M_{k,m}^{m} \) through calculation of the reduced matrix element of the third rank tensor. The result for one electron states with \( J = 5/2, L = 3, S = 1/2 \) is given by
\[ M_{k,m}^{m} = -\frac{2}{35} \mu_B (r^2) \langle J_{m}^{(3)} \rangle, \]  (18)
where the third-rank tensor operators \( J_{m}^{(3)} \) are defined in ref. 16.

Freeman and Desclaux obtained the estimate \( \langle r^2 \rangle = 1.298 \) in atomic unit by a relativistic Dirac-Fock calculation.^{26} By using this value, we obtain about 40G as the magnitude of internal field at (a/2, 0, 0) from a Ce ion. This value is by an order of magnitude smaller than that derived by the μSR measurement. Thus the static octupolar moment alone cannot account for the Gaussian relaxation of the μSR spectra. In a future work, we plan to study in more detail fluctuations in the octupole ordered state.

We mention that Paixão el al. have proposed for the ordered state of NpO_2 that a triple-\( q \) \( \Gamma_{5u} \)-type octupole ordering is realized and a triple-\( q \) \( \Gamma_{5g} \) quadrupole moment is induced.\(^{18}\) We estimate the internal magnetic field in NpO_2 to be about 80G at 8K from the observed muon spin precessing frequency of 7MHz.\(^{20}\) The internal field is of the same order as our estimate for CeL_1α−\( x \)B_6.\(^{4}\)

In the antiferro-octupolar phase, antiferromagnetism should be induced by uniaxial stress.\(^{9}\) In the following we estimate the magnitude of the induced moment. The uniaxial pressure \( p \) along the [001] direction accompanies the \( \Gamma_3 \) strain
\[ (C_{11} - C_{12})(2\epsilon_{xx} - \epsilon_{yy} - \epsilon_{yy}) = 2p. \]  (19)
We have solved the mean field equation with the finite \( \Gamma_3 \) strain and the corresponding quadrupole-strain interaction \( g_{r_s} \). The antiferromagnetic moment is induced in the \( xy \)-plane since \( (T_{5u}^2) \neq 0 \) together with \( (J_{y}^2 - J_{z}^2) \neq 0 \) gives \( \langle J_{y} \rangle \neq 0 \). The antiferromagnetic moment is along the [110] or [110]. With experimental values: \( (C_{11} - C_{12})/2 \approx 20.4 \times 10^{-11} \text{erg/cm}^3 \) and \( |g_{r,s}| = 120K \), the mean-field solution for \( (T_{5u}^2) \) becomes zero for \( p \geq 0.9GPa \). This implies the octupole and dipole moments both lying in the \( xy \)-plane. With \( p = 1GPa \), the magnetic moment is estimated to be 0.88\( \mu_B \) (0.82\( \mu_B \)), if \( g_{r_s} \) is positive (negative). This value should actually be reduced by quantum fluctuations. However, it is likely that the magnitude remains within experimental access.

To summarize, we have proposed that the \( \Gamma_{5u} \) octupole moment is a plausible candidate for the order parameter of the phase IV. This ordered state is consistent with the lattice distortion along the [111] direction,\(^4,5\) the broken time reversal symmetry,\(^6,7\) no dipole moment found,\(^8\) and the cusp in the magnetization at \( T_{LIV} \).\(^3\) However, the internal field estimated by our mean field theory is much smaller than that suggested by the μSR experiment.\(^7\) We recall that the NMR spectra become broad in the phase IV,\(^6\) and the spectra cannot be explained by the static and staggered octupole moments either. Therefore clarifying dynamical aspects and identifying the periodicity in the phase IV are challenging open problems to be addressed in the near future.

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