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Hidden octupole order in URu_2Si_2

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Abstract

We propose that the hidden order in URu_2Si_2 is an incommensurate octupole order, which is derived from a spin-1 XXZ model with XX octupole and Z dipole interactions based on the singlet–doublet crystal-field level scheme. The octupole moments break time-reversal invariance and give rise to finite hyperfine fields on the nuclei of the ligand Si and Ru ions. In order for the hyperfine fields to be observed as a nuclear magnetic resonance (NMR) linewidth, the order must be incommensurate, with the ordering vector determined so as to account for the ^{29}Si NMR linewidth data quantitatively.

In spite of numerous efforts over two decades to reveal the nature of the *hidden* order of a heavy-fermion compound URu_2Si_2 [1, 2], its order parameter is still controversial, and so is even the electronic structure of U ions. An order state usually exhibits broken symmetry, and a main subject is to determine what symmetry is broken [3, 4]. In the hidden order phase of URu_2Si_2, time-reversal symmetry is suggested to be broken from the ^{29}Si NMR data, whose linewidth remains finite ($\approx$10 G) for the magnetic field to vanish [5], as pointed out by Chandra *et al* [6]. As an order parameter which breaks time-reversal symmetry, we have proposed an octupole moment of 5f electrons of U ions, by means of accounting for the heavy-fermion behaviour by a model calculation [7]. Here we analyse the ^{29}Si NMR linewidth data [5], and the ^{99}Ru NMR data [8], using the XXZ model with XX octupole and Z dipole interactions proposed in [7]. We show that an order of the octupole moments of $x(y^2 - z^2)$ and $y(z^2 - x^2)$ symmetry is a strong candidate for the hidden order of URu_2Si_2. We find that the order must be incommensurate for the hyperfine fields to be observed as an NMR linewidth, similarly to the argument by Chandra *et al* [6], who have proposed an incommensurate orbital antiferromagnetism, associated with circulating currents between U ions. We further show that the octupole moments are nothing but the spatial distribution of magnetization by illustrating the angular dependence of spin densities in the ordered state.

Above the transition temperature $T_0 = 17.5$ K to the hidden order, URu_2Si_2 exhibits heavy-fermion behaviour, for example in the magnetic susceptibility χ_c for the magnetic field H applied parallel to the c-axis ($H \parallel c$) [1], the resistivity ρ and the electronic part C_e of specific heat [9]. These quantities show large values characteristic of heavy fermions, accompanied by

the peaks at ≈50 K (χ_c), ≈60 K (ρ) and ≈30 K (C_e). Itineracy of the quasiparticles in a heavy-fermion system is certainly significant, and several novel proposals for the hidden order have been made on the basis of itinerant models, such as an orbital antiferromagnetism (a density wave) [6, 10], an unconventional spin density wave [11] and a helicity order [12]. Most of these theories are formulated regardless of the practical electronic structure of 5f shell of U ions. However, it is necessary to consider the electronic structure of 5f electrons in order to account for the magnetic susceptibility χ, as first pointed out by Nieuwenhuys [13]. The practical electronic structure must be closely related to the nature of the hidden order parameter as well.

The main electronic configuration of U ions in URu_2Si_2 is considered to be $(5f)^2$, whose ground-state multiplet $^3H_4$ splits into five singlets and two doublets in a tetragonal crystal field. If we let $|M\rangle$ denote the wavefunctions $|5f^2514M\rangle$ $(=|5f^2LSJM_J\rangle)$ of $^3H_4$, the crystal-field eigenstates are expressed as: $|\Gamma_1\rangle = \alpha(|4\rangle + |-4\rangle) + \beta|0\rangle$ (singlet); $|\Gamma_2\rangle = (|4\rangle - |-4\rangle)/\sqrt{2}$ (singlet); $|\Gamma_3\rangle = (|2\rangle + |-2\rangle)/\sqrt{2}$ (singlet); $|\Gamma_4\rangle = (|2\rangle - |-2\rangle)/\sqrt{2}$ (singlet); and $|\Gamma_5\pm\rangle = \gamma|\pm3\rangle + \delta|\mp1\rangle$ (doublet), where $2\alpha^2 + \beta^2 = 1$ and $\gamma^2 + \delta^2 = 1$. The influence of the crystal-field level splitting is clearly seen in the anisotropy of the magnetic susceptibility χ with respect to the direction of the applied magnetic field H as χ_a for $H \parallel a$ $(\perp c)$ shows a Van Vleck (or Pauli) paramagnetism, in contrast to the heavy-fermion behaviour of χ_c for $H \parallel c$ accompanying a peak structure at ≈50 K [1].

Several crystal-field level structures to account for χ_c have been proposed, such as $\Gamma_1^{(1)}$–Γ_2–$\Gamma_1^{(2)}$–Γ_5 [13]; Γ_3–Γ_1–Γ_2 (3 singlets) [14]; Γ_1–Γ_4–Γ_5–Γ_2 [15]; and Γ_1–Γ_5 (singlet–doublet) [7]. In these cases, there exist finite matrix elements of J_z for χ_c only or mainly in the excited states, whose feature causes the peak structure of χ_c, or the decrease at low temperature with decreasing T. Even in the case where the itinerant character of 5f electrons is included, this feature is crucial to account for the T dependence of χ_c, as shown in [7]. In the previous letter [7], we have performed a practical calculation taking account of the heavy-fermion band formation to analyse χ_c and C_e above T_0, assuming a variety of the crystal-field level schemes. We have obtained fairly good agreement for Γ_3–Γ_1–Γ_2 and Γ_1–Γ_5, while poor agreement is obtained for the other schemes, including the lowest Γ_5-doublet scheme. This is because the behaviours of χ_c and C_e are governed by the two significant parameters Δ_{CF} (crystal-field splitting) and Δ_{HF}, which characterizes the heavy-fermion band width.

Here we comment on the scheme of Γ_1–Γ_4–Γ_5–Γ_2 [15], in comparison with Γ_1–Γ_5 [7]. In these schemes, the existence of the excited Γ_5 doublet is crucial to account for χ_c. If the Γ_4 singlet is located near Γ_5, χ_a is also expected to behave similarly to χ_c, in contrast to the Van Vleck-like χ_a in the experiment [1]. This results from the fact that $\langle\Gamma_4|J_{x,y}|\Gamma_5\pm\rangle$ is inevitably large enough, the reason for which is as follows. The Γ_4 and $\Gamma_5\pm$ states with $\gamma = \sqrt{7/8}$ $(\delta = -\sqrt{1/8})$ comprise the cubic Γ_5 triplet in a cubic crystal field, for which $\chi_a = \chi_c$. The $\Gamma_5\pm$ states, as well as Γ_1, in both schemes do not deviate substantially from those of the cubic states with $\alpha = \sqrt{5/24}$. We further note that $\langle\Gamma_1|J_{x,y}|\Gamma_5\pm\rangle = 0$ for the cubic states, which is the same as the assumption for the tetragonal Γ_1–Γ_5 scheme to derive our octupole order model. Therefore, the justification of our assumption may be simply reduced to considering the reason why the Γ_4 state goes away from the cubic Γ_1–Γ_5 subspace in the relevant low-energy region.

In the case where a practical crystal-field level scheme is taken into account, a candidate for the hidden order parameter may be a quadrupole or an octupole moment, as proposed by several authors: quadrupole order models are that of $O_2^2(\propto J_x^2 - J_y^2)$ based on the Γ_3–Γ_1–Γ_2 level scheme [14], O_2^2 or $O_{xy}(\propto J_xJ_y)$ on Γ_5 [16]; octupole order models are that of $T_z^\beta(\propto J_z(J_x^2 - J_y^2))$ or $T_{xyx}(\propto J_xJ_yJ_z)$ on Γ_1–Γ_4–$(\Gamma_5-)\Gamma_2$ [15, 17], $T_x^\beta(\propto J_x(J_y^2 - J_z^2))$ and $T_y^\beta(\propto J_y(J_z^2 - J_x^2))$ on Γ_1–Γ_5 [7]. Among them, the quadrupole order models [14, 16] may be inconsistent with the ^{29}Si NMR measurement [5], which suggests the breaking of time-

reversal invariance by the hidden order [6], since any quadrupole order parameter is invariant under time-reversal symmetry. The octupole order models [7, 15, 17] are consistent with the feature of time-reversal symmetry breaking. Here we adopt the octupole order model based on the Γ_1–Γ_5 scheme [7], which has the potential to account for the characteristics of the hidden order phase, including the ^{29}Si NMR linewidth data and also the feature of the close proximity of the antiferromagnetism to the hidden order. The discussion in [7] is summarized as follows.

In the subspace spanned by $|\Gamma_1\rangle$ and $|\Gamma_5\pm\rangle$ of the 5f state for a U site, we have eight independent matrices except the unit matrix, which usually represent three dipole and five quadrupole moments. We make use of the vector $\{|\Gamma_5+\rangle, |\Gamma_1\rangle, |\Gamma_5-\rangle\}$ as the base to represent the matrices, among which three indispensable ones are given as:

$$\left(\begin{pmatrix} 0 & \frac{1}{\sqrt{2}} & 0 \\ \frac{1}{\sqrt{2}} & 0 & \frac{1}{\sqrt{2}} \\ 0 & \frac{1}{\sqrt{2}} & 0 \end{pmatrix}, \begin{pmatrix} 0 & \frac{-\mathrm{i}}{\sqrt{2}} & 0 \\ \frac{\mathrm{i}}{\sqrt{2}} & 0 & \frac{-\mathrm{i}}{\sqrt{2}} \\ 0 & \frac{\mathrm{i}}{\sqrt{2}} & 0 \end{pmatrix}, \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix} \right).$$

These matrices are nothing but those for the x, y, z components of the spin-1 ($S = 1$) moment (S_x, S_y, S_z), which usually correspond to the dipole moments as $J_x = c_x S_x$, $J_y = c_y S_y$, $J_z = c_z S_z$ with $c_x = 2\alpha\gamma + \sqrt{10}\beta\delta$, $c_y = -c_x$, $c_z = 3\gamma^2 - \delta^2$. However, the Van Vleck nature of χ_a indicates that J_x and J_y are almost inactive in the subspace of Γ_1 and $\Gamma_5\pm$, and hence $c_x = 2\alpha\gamma + \sqrt{10}\beta\delta \approx 0$. From the analysis of $\chi_{a,c}$ and C_e [7], we have estimated the values of the parameters as $\gamma \approx \sqrt{0.8}$, $\delta \approx -\sqrt{0.2}$, $\alpha \approx \sqrt{0.28}$, $\beta \approx \sqrt{0.44}$, and $\Delta_\mathrm{CF} \approx 140$ K. In this case, the degrees of freedom corresponding to inactive J_x and J_y are carried by the octupole moments $T_x^\beta (\equiv (\sqrt{15}/2)(J_x J_y^2 - J_x J_z^2))$ and $T_y^\beta (\equiv (\sqrt{15}/2)(J_y J_z^2 - J_y J_x^2))$, whose matrix representations are given as $T_x^\beta = -dS_x$ and $T_y^\beta = -dS_y$, where $d = (3\sqrt{6}/4)[5\beta(\sqrt{7}\gamma - 3\delta) + \sqrt{10}\alpha(7\gamma + \sqrt{7}\delta)]$. Then, as a minimal interaction model to describe the hidden order, we have a spin-1 XXZ interaction model with a uniaxial anisotropy on the body-centred hexagonal lattice: $\mathcal{H}_\mathrm{XXZ} = -\sum_{i,j}[X_{ij}(S_{ix}S_{jx} + S_{iy}S_{jy}) + Z_{ij}S_{iz}S_{jz}] + \sum_i \Delta_\mathrm{CF} S_{iz}^2$, where i (j) denotes a site of U ions. In this model, the order of the XX (xy) components of $\boldsymbol{S}$ indicates an octupole order, and a slight canting of $\boldsymbol{S}$ from the xy plane induces a tiny magnetic moment along the c-axis. The present model, therefore, possesses the potential to account for the close proximity of weak antiferromagnetism to the hidden order in URu_2Si_2.

We show here that an incommensurate octupole order based on the XXZ model accounts for the behaviours of the ^{29}Si NMR linewidth [5]. To describe an incommensurate order of the octupole moments, we transform the Hamiltonian $\mathcal{H}_\mathrm{XXZ}$ to the wavevector representation as $\mathcal{H}_\mathrm{XXZ} = -\sum_{\boldsymbol{q}}[X(\boldsymbol{q})\{S_x(\boldsymbol{q})S_x(-\boldsymbol{q}) + S_y(\boldsymbol{q})S_y(-\boldsymbol{q})\} + Z(\boldsymbol{q})S_z(\boldsymbol{q})S_z(-\boldsymbol{q})] + \sqrt{N}\Delta_\mathrm{CF}S_z^2(\boldsymbol{0})$ by the use of $X(\boldsymbol{q}) = \sum_j X_{ij}\mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{R}_j - \boldsymbol{R}_i)}$, $S_x(\boldsymbol{q}) = N^{-1/2}\sum_i S_{ix}\mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}_i}$ etc, where $\boldsymbol{R}_i$ denotes the position vector for the i site of U ions, and N the number of U sites. We apply the mean-field approximation (MFA) to this Hamiltonian, and seek a solution consistent with the ^{29}Si NMR linewidth data. The criterion of our calculation is that the ^{29}Si NMR linewidth λ for $H \to 0$ and $T \to 0$ tends to 10 G, regardless of the direction of H, namely $\lambda_\parallel(H \parallel c) = \lambda_\perp(H \perp c) = 10$ G. As such a solution, we obtain the XX (xy) order with the wavevector $\boldsymbol{Q} = (0.13, 0.12, 0)2\pi/a$ or $(0.12, 0.13, 0)2\pi/a$, which is stabilized if $X(\boldsymbol{Q})$ has the maximum value. For $X(\boldsymbol{Q}) = 34$ K, $\Delta_\mathrm{CF} = 63$ K, we have $T_0 = 17.5$ K as the octupole transition temperature.

Here we add notes to the MFA calculation and the determination of the ordering wavevector $\boldsymbol{Q}$. In the MFA, an order with $\boldsymbol{Q}$ corresponding to the maximum of $X(\boldsymbol{q})$ is realized for $T \leqslant T_0$. The second-order transition temperature T_0 is simply obtained from $k_\mathrm{B}T_0/X(\boldsymbol{Q}) = \delta/\ln[(1+\delta)/(1-\delta/2)]$, where $\delta \equiv \Delta_\mathrm{CF}/X(\boldsymbol{Q})$. Note that T_0 vanishes for $\delta \geqslant 2$. The weight (w_5) of the upper Γ_5 doublet in the ground state is given by $w_5 = (1 - \delta/2)/2$.

In the present case, $\delta = 1.84$ and $w_5 = 0.04$. The value of $\boldsymbol{Q} = (0.13, 0.12, 0)2\pi/a$ (or $(0.12, 0.13, 0)$) is obtained for a superfluous accuracy of $\lambda_\parallel = \lambda_\perp = 10.0$ G. If we take deviated values of $\boldsymbol{Q}$ as $(0.11, 0.11, 0)2\pi/a$ and $(0.14, 0.14, 0)2\pi/a$, we obtain $(\lambda_\parallel, \lambda_\perp) = (9.0, 10.2)$ and $(\lambda_\parallel, \lambda_\perp) = (11.0, 9.7)$ in G, respectively. Therefore, a rather wide range of $\boldsymbol{Q}$ may be compatible with the experimental data.

In any case, such an incommensurate wavevector $\boldsymbol{Q}$ near the centre of the Brillouin zone indicates a long-range nature of the interaction, which is considered to result from the RKKY interaction. Furthermore, it may be necessary to take into account the itinerant nature as a heavy-fermion system. If we consider a quasiparticle band Hamiltonian together with $\mathcal{H}_{\mathrm{XXZ}}$, an itinerant octupole order will be obtained as an orbital density wave. It will be necessary to assume an appropriate $\boldsymbol{q}$ dependence of $X(\boldsymbol{q})$, which may not necessarily have the maximum just at such a $\boldsymbol{Q}$. Even in such a case, a Fermi surface instability, which is discussed in the studies based on the itinerant-localized duality model [18, 19], may cause an itinerant octupole order with $\boldsymbol{Q}$. In fact, the nesting feature of the Fermi surface of URu_2Si_2 has been suggested by the band calculation [20, 21], as pointed out in [18]. We further note that the value of $\Delta_{\mathrm{CF}} = 63$ K is approximately half of that for the itinerant model of [7]. This is not a serious discrepancy. The effective crystal-field splitting in the itinerant model is approximately half of the parameter Δ_{CF} itself, since the Fermi energy lies midway between the singlet (Γ_1) and the doublet (Γ_5) in the itinerant model.

In the MFA solution that we have obtained, the ground-state wavefunction for the U site at $\boldsymbol{R}_i$ is given by $|\Psi_0\rangle_i = \sqrt{1-2\epsilon^2}|\Gamma_1\rangle + \epsilon(\mathrm{e}^{-\mathrm{i}\boldsymbol{Q}\cdot\boldsymbol{R}_i}|\Gamma_5+\rangle + \mathrm{e}^{\mathrm{i}\boldsymbol{Q}\cdot\boldsymbol{R}_i}|\Gamma_5-\rangle)$, where $\epsilon = 0.1414$. The weight of the Γ_5 doublet is given as $w_5 = 2\epsilon^2 = 0.04$, which means that $|\Psi_0\rangle_i$ consists of the main (96%) Γ_1 singlet and the small (4%) admixed Γ_5 doublet. The charge and spin density operators for the 5f electrons in the $(5\mathrm{f})^2$ configuration of a U site are given by $\rho(\boldsymbol{r}) = \sum_{k=1}^{2}\delta(\boldsymbol{r}-\boldsymbol{r}_k)$ and $\boldsymbol{\sigma}(\boldsymbol{r}) = \sum_{k=1}^{2}\boldsymbol{s}_k\delta(\boldsymbol{r}-\boldsymbol{r}_k)$. Here $\boldsymbol{r}$ denotes a spacial point and $\boldsymbol{r}_k$ the position vectors of the electrons, both of which are measured from the subject nucleus, and $\boldsymbol{r}$ is represented as $\boldsymbol{r} = (r\sin\theta\cos\varphi, r\sin\theta\sin\varphi, r\cos\theta)$ in the polar coordinates. The expectation values of $\rho(\boldsymbol{r})$ and $\boldsymbol{\sigma}(\boldsymbol{r}) = (\sigma_x(\boldsymbol{r}), \sigma_y(\boldsymbol{r}), \sigma_z(\boldsymbol{r}))$ with respect to $|\Psi_0\rangle_i$ are expressed as ${}_i\langle\Psi_0|\rho(\boldsymbol{r})|\Psi_0\rangle_i = R_{5\mathrm{f}}(r)^2C_i(\theta,\varphi)$ and ${}_i\langle\Psi_0|\sigma_\alpha(\boldsymbol{r})|\Psi_0\rangle_i = R_{5\mathrm{f}}(r)^2\Sigma_{i\alpha}(\theta,\varphi)$, where $R_{5\mathrm{f}}(r)$ is the radial part of the 5f wavefunction, and the angular parts of the densities $C_i(\theta,\varphi)$ and $\Sigma_{i\alpha}(\theta,\varphi)$ ($\alpha = x, y, z$) are expressed by the spherical harmonics $Y_l^m(\theta,\varphi)$. For a site with a phase of $\boldsymbol{Q}\cdot\boldsymbol{R}_i = 2\pi\times$ (integer), namely a pure S_x (T_x^β) ordered state, the angular dependence of the charge and spin densities, $C_i(\theta,\varphi)$ and $\Sigma_{i\alpha}(\theta,\varphi)$, are shown in figures 1(a)–(d), where the sign of spin density is denoted by + (up) and − (down). The scales of the axes are arbitrary in figures 1(a)–(d), where the scale for $C_i(\theta,\varphi)$ is one order of magnitude larger than those for $\Sigma_{i\alpha}(\theta,\varphi)$ s. Note that these figures do not show the densities themselves but only the angular dependence. The radial wavefunction $R_{5\mathrm{f}}(r)$ has one node to make a radial fall-off, which is not shown here. For the other site, the direction of the principal axis (x-axis for figures 1) rotates by an angle of $\boldsymbol{Q}\cdot\boldsymbol{R}_i$, for which the corresponding rotated spin densities are obtained.

While we have no net dipole moment in the present case, an octupole moment can appear due to spatial distribution of magnetization, consisting of spin and orbital angular momentum densities, as shown in figures 1(b)–(d) for spin density. We note that, for a pure Γ_1 singlet, the charge density does not differ substantially from figure 1(a), whereas the net spin density vanishes everywhere. On the other hand, the present octupole order state $|\Psi_0\rangle_i$ has a small but finite spin density, which breaks time-reversal invariance and causes finite magnetic fields at ligand Si and Ru sites, as shown in the following.

We consider the origins of the NMR spectra of ligand ions surrounding a magnetic ion, which have been fully discussed by Abragam and Bleaney [22]. Following their notion, the

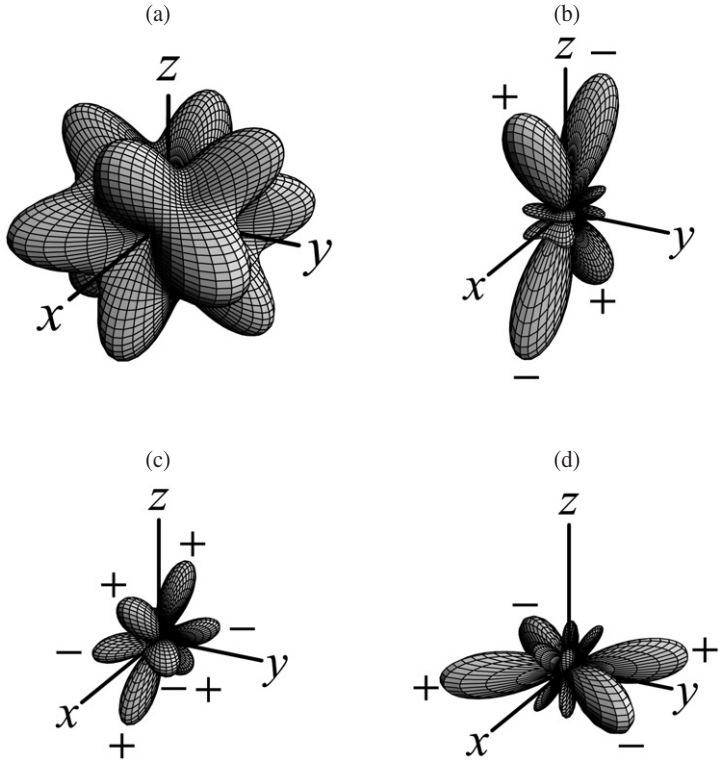

Figure 1. Angular dependence of (a) the charge density $C_i(\theta, \varphi)$ and the spin density: (b) $\Sigma_{iz}(\theta, \varphi)$; (c) $\Sigma_{ix}(\theta, \varphi)$; (d) $\Sigma_{iy}(\theta, \varphi)$, for an S_x (T_x^β) ordered state of $\sqrt{0.96}|\Gamma_1\rangle + \sqrt{0.02}(|\Gamma_5+\rangle + |\Gamma_5-\rangle)$, where + and − denote positive and negative spin density, respectively.

author has analysed the ^{11}B NMR data of CeB_6 in detail [23], the situation of which is very similar to that of the ^{29}Si or ^{99}Ru NMR in URu_2Si_2. The hyperfine coupling of the electronic moments of the magnetic ions (U) with the nuclear moments of the ligands (Si, Ru) have two origins: the effect of covalent bonding (transferred hyperfine interaction) and the dipole–dipole interaction. We will show below that the ^{29}Si NMR can be accounted for only by the dipole–dipole interaction, while the ^{99}Ru NMR also needs the transferred hyperfine interaction. Here we show how to calculate the contribution from the dipole–dipole interaction.

The Hamiltonian of the dipole–dipole interaction between the nuclear moment $\boldsymbol{\mu}_n$ $(=\gamma_n\hbar\boldsymbol{I})$ and the electronic moment $\boldsymbol{\mu}_e$ separated by the vector $\boldsymbol{r}_{en}$ is given as $\mathcal{H}_{dd} = \boldsymbol{\mu}_n \cdot [\boldsymbol{\mu}_e/r_{en}^3 - 3\boldsymbol{r}_{en}(\boldsymbol{\mu}_e \cdot \boldsymbol{r}_{en})/r_{en}^5]$, where $\boldsymbol{\mu}_e = -\mu_B(\boldsymbol{l} + 2\boldsymbol{s})$. The position vector of the electron measured from the nucleus of the subject magnetic ion is defined by $\boldsymbol{r} \equiv \boldsymbol{r}_{en} - \boldsymbol{R}$, with $\boldsymbol{R}$ denoting the vector connecting the magnetic and the ligand sites. Now we use the Cartesian coordinates $\boldsymbol{r} = (x, y, z) \equiv (x_1, x_2, x_3)$ and define the direction cosines of $\boldsymbol{R}$ as $\boldsymbol{n} \equiv \boldsymbol{R}/R = (n_1, n_2, n_3)$. For $r \ll R$, we expand r_{en}^{-3} and r_{en}^{-5} in $\mathcal{H}_{dd}$ with respect to r/R to obtain

$$\frac{\delta_{\alpha\beta}}{r_{en}^3} - 3\frac{x_{en\alpha}x_{en\beta}}{r_{en}^5} = \frac{\delta_{\alpha\beta} - 3n_\alpha n_\beta}{R^3} + \frac{f_{\alpha\beta}}{R^5} + \cdots, \tag{1}$$

where $f_{\alpha\beta} = f_{\alpha\beta}(\boldsymbol{n}, \boldsymbol{r}) = (3/2)(5n_\alpha n_\beta - \delta_{\alpha\beta})r^2 - 3x_\alpha x_\beta + 15(n_\alpha x_\beta + n_\beta x_\alpha)(\boldsymbol{n} \cdot \boldsymbol{r}) - (15/2)(7n_\alpha n_\beta - \delta_{\alpha\beta})(\boldsymbol{n} \cdot \boldsymbol{r})^2$. The first term of equation (1) provides the classical dipole field

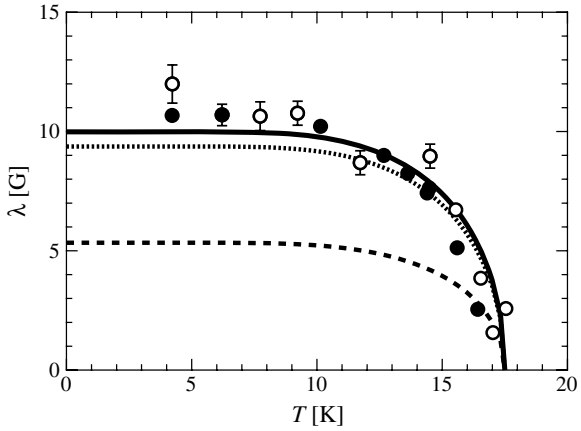

Figure 2. Temperature dependence of the ligand NMR linewidth λ. The solid line shows $\lambda_\parallel$ ($H \parallel c$) = $\lambda_\perp$ ($H \perp c$) for ^{29}Si NMR, in comparison with the experimental data of $\lambda_\parallel$ (open circles) and $\lambda_\perp$ (filled circles) from Bernal *et al* [5]. The broken line shows $\lambda_\parallel$ and the dotted line shows $\lambda_\perp$ for ^{99}Ru NMR, to be compared with the data from Bernal *et al* [8].

corresponding to the case where the electronic moment is regarded as a point dipole, which vanishes in the present model for no applied magnetic field. The second term of equation (1) is the correction arising from the octupole moment of the asymmetric spatial distribution of magnetic moment, which now provides the leading contribution to the ligand NMR. Consequently, we have the α component of the hyperfine field $\boldsymbol{h}$ ($\mathcal{H}_{\rm dd} = -\boldsymbol{\mu}_{\rm n} \cdot \boldsymbol{h}$) as $h_\alpha = (\mu_{\rm B}/R^5) \sum_{\beta=1}^{3} (l_\beta + 2s_\beta) f_{\alpha\beta}(\boldsymbol{n}, \boldsymbol{r})$. The magnetic field for the nuclear moments at the origin from the octupole correction of the 5f electrons at $\boldsymbol{R}$ is obtained from $H_\alpha^{(i)} \equiv {}_i\langle\Psi_0|h_\alpha|\Psi_0\rangle_i = c_i \mu_{\rm B} \langle r^2\rangle_{\rm 5f}/R^5$, where $\langle r^2\rangle_{\rm 5f} \equiv \int_0^\infty r^2 R_{\rm 5f}(r)^2 r^2 \, {\rm d}r$, and c_i is a factor to be calculated. We should note that, after taking the summation over β in the expression of h_α, the contribution from the orbital angular momentum $\boldsymbol{l}$ to $H_\alpha^{(i)}$ vanishes, while that from the spin $\boldsymbol{s}$ remains finite. This is the important feature for the case with no applied magnetic field, for which the direction of the electronic moment $\boldsymbol{\mu}_{\rm e}$ is not fixed.

For a Si site, there are four nearest- and one next-nearest-neighbour U sites separated by $R_0 = 0.3152$ nm and $R_1 = 0.3593$ nm, respectively [1]. Since $\langle r^2\rangle_{\rm 5f} \approx 2.0$ au $= 5.60 \times 10^{-3}$ nm^2 [24, 25], we have $\mu_{\rm B}\langle r^2\rangle_{\rm 5f}/R_0^5 \approx 16.7$ G. It follows that the hyperfine field of the order of 10 G observed in the ^{29}Si NMR experiment [5] is expected to be accounted for by this mechanism. Furthermore, the contributions from more distant U sites than R_1 may be negligible due to the dependence of R^{-5} and the sign alternation in $H_\alpha^{(i)}$, and hence we take into account the contributions from the five neighbouring U sites to the hyperfine field on ^{29}Si. According to our supposed incommensurate order, the environments of ^{29}Si nuclei vary from site to site, giving rise to a random distribution of the hyperfine fields, which may be observed as the linewidth of the NMR signal, as pointed out by Chandra *et al* [6]. We have taken the values of the parameters as well as the ordering vector so that the hyperfine fields are isotropic and distributed from -10 to 10 G for $T \to 0$. The results calculated for the NMR linewidth λ in G are shown in figure 2, where the solid line indicates λ for ^{29}Si NMR in comparison with the experimental results found by Bernal *et al* for $H \parallel c$ (open circles) and $H \perp c$ (filled circles) [5]. Note that the magnitude of λ depends on the coefficients of the wavefunctions as follows: the smaller α or the larger ϵ in the range $0 < \epsilon \leqslant 0.5$, the larger the value of λ that is obtained.

In figure 2, we also show the calculated results of the ^{99}Ru NMR linewidth for the same parameters as the analysis of the ^{29}Si NMR, where the broken line is for $\lambda_{\parallel}(H \parallel c)$ and the dotted line is for $\lambda_{\perp}(H \perp c)$. The anisotropy of $\lambda_{\perp}$ and $\lambda_{\parallel}$, including the ratio for ^{99}Ru NMR, is consistent with the experimental results of Bernal *et al* [8], but the magnitudes are approximately one third of the data. These results suggest that there exists a substantial contribution from the transferred hyperfine interaction for ^{99}Ru NMR, in contrast with the ^{29}Si NMR. According to band calculations [20, 21], the valence electrons of Ru contribute to the Fermi surface substantially, while those of Si make little contribution. To analyse the ^{99}Ru NMR data, therefore, we must take into account the transferred hyperfine interaction, as was done for the ^{11}B NMR in CeB_6 by Hanzawa [23].

Now we discuss the consistency of the present octupole order model with the other properties of URu_2Si_2. The calculated results of susceptibility χ and specific heat C agree with the experimental results qualitatively. The entropy released up to $T_0 = 17.5$ K is estimated to be $0.24R \approx 0.35R \ln 2$, which is somewhat larger than the experimental results [1]. The present calculation may be partly improved by taking account of fluctuation effects beyond the mean-field approximation. Furthermore, itineracy is certain to be included simultaneously to obtain better agreement with the experimental results, as shown in [7] for χ and C above T_0. Nevertheless, even within the present local-'spin' interaction model, we will be able to make a qualitative analysis of the phase diagrams of URu_2Si_2 [2, 26]. If the value of Δ_{CF} in $\mathcal{H}_{XXZ}$ is decreased, which may be realized by pressure p, the XX octupole order is converted to the Z dipole order in the first-order phase transition. Therefore, it is probable that the phase diagram of URu_2Si_2 on the T–p plane [2], as well as that on the H–T [26] plane, is explained by means of the present XXZ model qualitatively.

In conclusion, we have analysed the ligand NMR linewidth data of URu_2Si_2 in terms of an incommensurate octupole order model, and obtained good agreement with the data. The nature of an octupole order state has been shown to be nothing but the spatial distribution of magnetization, namely the spin and orbital angular momentum densities, as shown in figures 1(b)–(d) (see another example for CeB_6 in [23]). From this point of view, it is not essential to take the concept of microscopic eddy current in wavefunctions discussed in [17]. The possibility of an octupole order has also been discussed in other systems, such as NpO_2 [27] and $Ce_xLa_{1-x}B_6$ [28], but its existence is still under debate. To detect its order, a sophisticated technique is necessary, such as resonant x-ray scattering experiments performed for NpO_2 [29] and $Ce_{0.7}La_{0.3}B_6$ [30], as well as ligand NMR for URu_2Si_2 [5, 8]. Finally, we comment on the recent report by Takagi *et al* [31] that the ^{29}Si NMR linewidth in single crystals is suggested to be one order of magnitude smaller ($\approx$1 G) and not strictly isotropic, which is somewhat different from the results for c-axis oriented powder samples of Bernal *et al* [5]. Even if this is the case, the essence of the present discussion will not be altered, although the values of the parameters should be re-determined. A smaller value of λ will be obtained for a smaller ϵ, as described above. For example, if we assume $\delta = 1.99$ for the same values of α, β, γ and δ, we have $\lambda = 2$ G for $T \to 0$ along with $X(Q) = 56$ K and $\Delta_{CF} = 112$ K. The NMR linewidth is still isotropic for the same value of $\boldsymbol{Q}$, which will be redetermined so as to fit the experimental data if it is not isotropic.

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