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J. Phys.: Condens. Matter **17** (2005) L419–L424 [doi:10.1088/0953-8984/17/41/L04](http://dx.doi.org/10.1088/0953-8984/17/41/L04)

LETTER TO THE EDITOR

Heavy-fermion behaviours in URu₂Si₂

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Received 19 July 2005 Published 30 September 2005 Online at stacks.iop.org/JPhysCM/17/L419

Abstract

We analyse the magnetic susceptibility χ and the specific heat *C* of the heavyfermion material URu₂Si₂, assuming a variety of the crystal field level schemes. The heavy-fermion behaviours of χ and *C* above $T_N = 17.5$ K are shown to be reproduced fairly well in the singlet–singlet–singlet $(\Gamma_3-\Gamma_1-\Gamma_2)$ level scheme proposed by Santini and Amoretti, and also in a novel level scheme of Γ_1 singlet ground state with Γ_5 -doublet excited state. We discuss that the novel singlet–doublet level scheme is favourable for the close proximity of weak antiferromagnetism to the hidden order in $URu₂Si₂$, and the most probable candidate of the hidden order is a $J_x (J_y^2 - J_z^2)$ -type octupolar ordering.

The heavy-fermion superconductor URu_2Si_2 exhibits a second-order phase transition at $T_N = 17.5$ K to a mysterious *hidden* order, prior to the superconducting phase transition at $T_c = 1.2$ K [\[1\]](#page-6-0). Efforts in many experimental $[1–8]$ and theoretical [\[9–18\]](#page-6-1) investigations have been made to reveal the nature of the *hidden* order, but its order parameter is still uncertain. The identification of the order parameter is closely related to the electronic structure of 5f shell of U ions: practically, the crystal-field level structure of the groundstate multiplet ${}^{3}H_{4}$ of the main $(5f)^{2}$ configuration. In a tetragonal crystal field, the nine-fold degenerate ${}^{3}H_{4}$ multiplet splits into five singlets and two doublets. If we let $|M\rangle$ denote the wavefunctions $|JM\rangle$ of ³H_{J=4}, the crystal-field eigenstates necessary for the following the waverunctions $|JM\rangle$ or ${}^{\circ}H_{J=4}$, the crystal-field eigenstates necessary for the following
discussion are expressed as $|\Gamma_1\rangle = \alpha(|4\rangle + |-4\rangle) + \beta|0\rangle$ (singlet); $|\Gamma_2\rangle = (|4\rangle - |-4\rangle)/\sqrt{2}$ discussion are expressed as $|1_1\rangle = \alpha(|4\rangle + |-4\rangle) + \beta|0\rangle$ (singlet); $|1_2\rangle = (|4\rangle - |-4\rangle)/\sqrt{2}$
(singlet); $|\Gamma_3\rangle = (|2\rangle + |-2\rangle)/\sqrt{2}$ (singlet); and $|\Gamma_5\pm\rangle = \gamma|\pm 3\rangle + \delta|\mp 1\rangle$ (doublet), where $2\alpha^{2} + \beta^{2} = 1$ and $\gamma^{2} + \delta^{2} = 1$.

The properties of $URu₂Si₂$ interpreted as heavy-fermion behaviours are seen, for example, in the magnetic susceptibility χ_c for *H* \parallel *c* [\[1\]](#page-6-0), the resistivity ρ and the electronic part C_e of the specific heat [\[2\]](#page-6-2), which show large values above and near to T_N , accompanied by the peaks at \approx 50 K (χ_c), \approx 60 K (ρ) and \approx 30 K (C_e). Such behaviours are usually attributed to a kind of correlation effect in a heavy-fermion system. Many theories [\[15–17\]](#page-6-3) may support, explicitly or implicitly, the Γ_5 -doublet ground-state configuration, because its two-fold degeneracy is thought to be reconciled with the heavy-fermion behaviours above T_N . However, any quantitative calculation to analyse the heavy-fermion behaviours has not been reported so far.

The only quantitative analysis of χ_c , C_e , as well as the transition temperature T_N , has been carried out by Santini and Amoretti [\[10\]](#page-6-4). They have proposed a quadrupolar ordering model of localized 5f electrons for the *hidden* order, and performed a mean-field calculation to analyse the experimental results of χ_c and C_e above T_N as a crystal-field effect of the singlet– singlet–singlet $(\Gamma_3 - \Gamma_1 - \Gamma_2)$ level scheme. They have obtained a fairly good agreement with the experimental data. Their calculation however has not taken into account the itinerancy of 5f electrons responsible for heavy-fermion behaviours, and consequently other theoretical proposals, sometimes based on different crystal-field level schemes, have been presented until quite recently [\[14–18\]](#page-6-5).

In this study, we perform a practical calculation taking account of the heavy-fermion band formation to analyse χ_c and C_e assuming a variety of crystal-field level schemes, in order to clarify which crystal-field level structure of $\Gamma_3 - \Gamma_1 - \Gamma_2$ -singlets, Γ_5 -doublet or other scheme is realized in URu2Si2. We adapt the formula based on the *generalized* periodic Anderson model developed by one of the authors $[19, 20]$ $[19, 20]$ $[19, 20]$ to the present case of the non-Kramers ${}^{3}H_{4}$ multiplet.

The formula of [\[19,](#page-6-6) [20\]](#page-6-7) is briefly summarized as follows. In the *generalized* periodic Anderson model with plane-wave conduction states $|\vec{k}\sigma\rangle$, the mixing matrix element is given as $V_{\vec{k}\sigma J'M',JM}^{(n)} = \langle \vec{k}\sigma | \langle \hat{f}^{n-1} J'M' | \mathcal{H}' | f^n J M \rangle = V_{kM'M}^{(n)} Y_{l=3}^m (\theta_{\vec{k}}, \phi_{\vec{k}})$, where $V_{kM'M}^{(n)}$ is a constant calculated in terms of the Clebsch–Gordan coefficients, and Y_l^m a spherical harmonic with $m = M - M' - \sigma$ ($\sigma = \pm 1/2$). For Ce compounds, we have $V_{\vec{k}\sigma 005/2M}^{(1)} = V_{k0M}^{(1)} Y_3^{M-\sigma}$, giving rise to the *usual* periodic Anderson (PA) model; and similarly for Yb compounds. Even for compounds of the other rare-earth elements such as Sm comprised of plural 4f electrons, the PA model, the periodic Coqblin–Schrieffer (PCS) model or the periodic slave-boson (PSB) model is shown to be applicable, if it is sufficient to consider only a ground-state singlet in the associate 4f-shell state. In these cases, a state $|JM\rangle$ of the main 4f-shell state at site *i* is represented by f_{iM}^{\dagger} |0), where f_{iM}^{\dagger} is a pseudo fermion operator and |0) the vacuum state. It has been discussed that the anisotropy of the mixing matrix elements between the pseudo fermion states and the conduction-electron states plays a significant role for the pseudogap formation in these compounds. If we apply the mean-field approximation (MFA) to the PCS or the PSB model, we obtain the PA model possessing reduced parameters without Coulomb interactions. Here we should note that there appears a phase transition in the MFA, and the PA model is valid only in a lower temperature (*T*) region than the transition point $T_c^{(0)}$. However, it has been shown in [\[21\]](#page-6-8) that $T_c^{(0)}$ is lifted due to the lowest-order fluctuation effects around the MFA solution for the PCS model. This result suggests that the MFA phase transition may be converted into the crossover from the low-*T* heavy-fermion to high-*T* local-moment region. We also note that the PA model with constant parameters is shown to account for the crossover behaviours of the thermodynamic quantities in a wide range of *T* [\[20\]](#page-6-7). It follows that the simple PA model can be commonly used to describe the heavy-fermion behaviours.

We adapt the formula of $[19, 20]$ $[19, 20]$ $[19, 20]$ to the case of heavy-fermion U compounds whose main 4f-shell multiplet is ³H₄ of (5f)². We assume that the associate multiplet is ²F_{5/2} of (5f)¹, for which the mixing matrix elements are given by $V_{\vec{k}\sigma 5/2M/4M}^{(2)} = V_{kM'M}^{(2)} Y_3^{M-M'-\sigma}$. In contrast to the cases discussed in [\[19,](#page-6-6) [20\]](#page-6-7), the associate state is not a singlet but now a sextet ${}^{2}F_{5/2}$, for which the PCS model is suitable to describe heavy-fermion behaviours. To a good approximation, the sextet is treated as a fully (six-fold) degenerate state as an intermediate state of the PCS model. Furthermore, if the MFA is applied, we have a PA model with reduced parameters, in which the mixing matrix elements are given by $\tilde{V}_{\vec{k}\sigma M} \equiv \sum_{M'=-5/2}^{5/2} V_{kM'M}^{(2)} Y_{3}^{M-M'-\sigma}$. For the mixing matrix elements comprised of several Y_l^m , the mixing matrix elements must be finite for all \vec{k} , and hence can be assumed to be constants to a good approximation. We finally obtain a simple PA model with reduced constant parameters to analyse the properties of heavy-fermion U compounds. Ohkawa and Shimizu [\[16\]](#page-6-9) have argued that the periodic s–d model is mapped to the extended PA model, which might be essentially the same reasoning as ours.

For the present non-Kramers system, we must use the fundamental formula of [\[22\]](#page-6-10): the hybridized band energy is determined by the equation that $A_{\vec{k}\sigma}A_{\vec{k}\bar{\sigma}} - B_{\vec{k}\sigma}B_{\vec{k}\bar{\sigma}} = 0$, where $A_{\vec{k}\sigma} = \omega - \epsilon_k - \sum_M |\tilde{V}_{\vec{k}\sigma M}|^2 / (\omega - E_M)$, $B_{\vec{k}\sigma} = \sum_M \tilde{V}_{\vec{k}\sigma M}^* \tilde{V}_{\vec{k}\sigma M} / (\omega - E_M)$, ϵ_k and E_M are the conduction and the f electron energies, respectively. We simply assume that the density of conduction-band states per spin is a constant *N*/2*D* of width 2*D*, where *N* is the number of U sites. The number of pseudo fermions representing crystal-field eigenstates per U site ($n_f \equiv$ N_f/N must be fixed to unity, whereas the number of conduction electron per U site (n_c) may be taken as a fitting parameter. We should note that the condition $n_f = 1$ corresponds to the number of *real* 5f electrons per U site being 2, since *n*^f is the number of *pseudo* fermions representing $(5f)^2$ per U. Therefore, the description of quasiparticle band structure in the present formulation is unconventional; its correspondence with the conventional Fermi liquid band description is as follows. We consider first a typical case of a well-separated singlet ground-state system, for which the band constructed from the singlet state is fully occupied for $n_f = 1$. This situation corresponds to that of a single band fully occupied by 2*N* real 5f electrons in the conventional Fermi liquid description, for which the system is inactive for elementary excitations. If a crystal-field excited state comes down closer to the ground singlet state,both of the states spread out in energy (become dispersive) due to the hybridization with the conduction electrons. In this case, the condition $n_f = 1$ makes the Fermi energy ϵ_F pinned in the intermediate region between both of the states. The density of states at ϵ_F is always finite and considerably high, accompanied by an appreciable amount of the excited state, giving rise to a heavy-fermion metal responsible for elementary excitations. We will see the specific cases in the following.

We have considered a variety of the crystal-field level structures such as $\Gamma_3 - \Gamma_1 - \Gamma_2$ (singlet–singlet–singlet) [\[10\]](#page-6-4); $\Gamma_1 - \Gamma_2$ (singlet–singlet) [\[14\]](#page-6-5); Γ_5 (doublet) [\[16\]](#page-6-9); and a novel scheme of $\Gamma_1-\Gamma_5$ (singlet–doublet), and then analysed the experimental data of χ_c [\[1\]](#page-6-0) and C_e/T , where $C_e = C(URu_2Si_2) - C(ThRu_2Si_2)$ [\[2\]](#page-6-2), shown by the open circles in figures [1\(](#page-4-0)a) and (b), respectively. We have obtained fairly good agreement for $\Gamma_3 - \Gamma_1 - \Gamma_2$ and $\Gamma_1 - \Gamma_5$, but poor agreement for the other schemes, the reason for which is clarified in the following discussion. Here we show the results of the former successful cases.

from $\Gamma_1 - \Gamma_5$ (singlet–doublet), we may simply assume that $V_0 \equiv \sqrt{2} \tilde{V}_{\vec{k}\sigma M} = \sqrt{2} \tilde{V}_{\vec{k}\bar{\sigma}M}$, $E_0 \equiv E_M$ for $M = \Gamma_1$; and $V_1 \equiv \tilde{V}_{\vec{k}\sigma M} = \tilde{V}_{\vec{k}\sigma \bar{M}}$, $E_1 \equiv E_M = E_{\bar{M}}$, $\tilde{V}_{\vec{k}\sigma M} = \tilde{V}_{\vec{k}\sigma \bar{M}} = 0$ for $M = \Gamma_5 +$ and $\overline{M} = \Gamma_5 -$. Then, we can determine the eigenvalues $\omega = \omega_k^*$ by solving the equation $\{\omega - \epsilon_k - V_1^2/(\omega - E_1)\}\{\omega - \epsilon_k - V_0^2/(\omega - E_0) - V_1^2/(\omega - E_1)\} = 0$. We have five solutions for each \vec{k} , which construct rather complicated hybridized (quasi-particle) band structure. Because of $n_f = 1$, the Fermi energy ϵ_F (chemical potential μ) is pinned in the finite density-of-state region of $E_0 < \omega_k^* < E_1$, regardless of the value of n_c . Consequently, we always obtain a heavy-fermion metal. Noting that the eigenvalues of the Zeeman energy for the field *H* applied along the *c*(*z*)-axis is given as $\langle \Gamma_5 \pm | \mathcal{H}_Z | \Gamma_5 \pm \rangle = \pm g_J \mu_B (3\gamma^2 - \delta^2) H$ with $g_J = 4/5$, the magnetic susceptibility χ_c is calculated as shown in figure [1\(](#page-4-0)a), and also the electronic part of specific heat divided by temperature C_e/T in figure [1\(](#page-4-0)b), by the use of the formula described in [\[20,](#page-6-7) [22\]](#page-6-10). Here we have taken $\Delta = V^2/D = 24$ K with $V = V_0 = V_1$, Δ_{CF} = $E_1 - E_0$ = 143 K, γ^2 = 0.794, n_c = 1, D/Δ = 100, for which $V = 238$ K and $2D = 4760$ K. The behaviours of χ_c and C_e/T are governed mainly by the two significant parameters Δ_{CF} (crystal-field splitting) and $\Delta (=V^2/D)$ which characterizes the order of the heavy-fermion band width [\[20\]](#page-6-7). We note that, if we dare to take a somewhat artificial value of the ratio as $D/\Delta = 10$, we obtain the results shown by the dotted lines in figure [1,](#page-4-0) where $\Delta = 49$ K, $\Delta_{CF} = 147$ K, $\gamma^2 = 0.835$, $n_c = 1$, in better agreement with the experimental data.

Figure 1. Temperature dependence of (a) the magnetic susceptibilities $\chi_{c,a}$ for *H* \parallel *c*, *a* and (b) the electronic part of the specific heat divided by temperature C_e/T for the $\Gamma_1-\Gamma_5$ scheme with $Δ = 24$ K, $Δ_{CF} = 143$ K, $γ^2 = 0.794$, $n_c = 1$, $D/Δ = 100$ (solid line); and for the $Γ_3 - Γ_1 - Γ_2$ scheme with $Δ = 33$ K, $Δ_{CF}^{(1)} = 133$ K, $Δ_{CF}^{(2)} = 266$ K, $α^2 = 0.432$, $n_c = 1$, $D/Δ = 100$ (dashed line), in comparison with the experimental data of χ_c (open circles) and χ_a (open triangles) after Palstra *et al* [\[1\]](#page-6-0) and C_e/T , where $C_e = C(\text{URu}_2\text{Si}_2) - C(\text{ThRu}_2\text{Si}_2)$ after Schlabitz *et al* [\[2\]](#page-6-2). The dash–dotted line in (a) shows that a calculated result failed to fit χ_a for Γ_1 – Γ_5 , and the dotted lines show the results for $\Gamma_1 - \Gamma_5$ with $D/\Delta = 10$ (see the values of parameters in the text).

For $\Gamma_3 - \Gamma_1 - \Gamma_2$ (singlet–singlet–singlet), we may simply assume that $V_0 = \sqrt{2} \tilde{V}_{\vec{k}\sigma M} = \sqrt{2} \tilde{V}_{\vec{k}\sigma M}$ For $13-1-12$ (singlet–singlet), we may simply assume that $v_0 = \sqrt{2}v_{k\sigma M} - \sqrt{2}v_{k\sigma M}$, $E_0 = E_M$ for $M = \Gamma_3$; $V_1 = \sqrt{2}v_{k\sigma M} = \sqrt{2}v_{k\sigma M}$, $E_1 = E_M$ for $M = \Gamma_1$;
and $V_2 = \sqrt{2}v_{k\sigma M} = \sqrt{2}v_{k\sigma M}$, $E_2 = E_M$ for $(\omega - \epsilon_k)(\omega - \epsilon_k - V_0^2/(\omega - E_0) - V_1^2/(\omega - E_1) - V_2^2/(\omega - E_2)) = 0$. There appear five solutions for each \vec{k} : four of these construct four hybridized bands separated by gaps; and the residual one corresponds to the unhybridized conduction-electron states, giving rise to a half of the original density of conduction states. We always have a heavy-fermion metal as well, for which we obtain the calculated results of χ_c and C_e/T shown by the dashed lines in figures [1\(](#page-4-0)a) and (b), where $\Delta = V^2/D = 33$ K with $V = V_0 = V_1 = V_2 = 332$ K, $\Delta_{\text{CF}}^{(1)} \equiv E_1 - E_0 = 133 \text{ K}, \, \Delta_{\text{CF}}^{(2)} \equiv E_2 - E_0 = 2 \Delta_{\text{CF}}^{(1)}, \, \alpha^2 = 0.432, n_c = 1, \text{ and } D/\Delta = 100$ $(2D = 6640 \text{ K}).$

In both cases, there exist finite matrix elements of J_z for χ_c only in the excited states in both cases, there exist finite matrix elements or J_z for χ_c only in the excited states
as $\langle \Gamma_5 \pm |J_z| \Gamma_5 \pm \rangle = \pm (3\gamma^2 - \delta^2)$ and $\langle \Gamma_1 | J_z| \Gamma_2 \rangle = 4\sqrt{2}\alpha$. This feature causes the peak structure of χ_c , or the decrease in low temperatures with decreasing *T*, as already pointed out for localized 5f electron models by Nieuwenhuys (singlet–singlet–singlet–doublet) [\[9\]](#page-6-1) and Santini–Amoretti (singlet–singlet–singlet) [\[10\]](#page-6-4). The results of the present calculation suggest that this feature is crucial to account for the *T* dependence of χ_c even for the case of itinerant character of 5f electrons included. For the cases without this feature such as of $\Gamma_1 - \Gamma_2$ (singlet–singlet) and Γ_5 (doublet), we have not obtained a clear peak structure of χ_c . At this stage, however, we cannot conclude which level scheme of $\Gamma_1 - \Gamma_5$ and $\Gamma_3 - \Gamma_1 - \Gamma_2$ is relevant to $URu₂Si₂$. For further discussion, it is undoubtedly significant to consider the nature of the *hidden* order, including its close proximity to *weak* antiferromagnetism.

The quadrupolar ordering model for the *hidden* order proposed by Santini and Amoretti [\[10\]](#page-6-4) is based on the $\Gamma_3 - \Gamma_1 - \Gamma_2$ scheme, in which the matrix element of $\langle \Gamma_3 | O_2^2 | \Gamma_1 \rangle$ $(O_2^2 \propto J_x^2 - J_y^2)$ is responsible for the order. The ground-state wavefunction of this order must mix further with the second excited state $|\Gamma_2\rangle$ in order to possess the magnetic moment along the *c*(*z*)-axis, since only $\langle \Gamma_1 | J_z | \Gamma_2 \rangle$ is finite. Consequently, it may be hard to use this scheme to explain the proximity of the *hidden* order to the antiferromagnetism in URu₂Si₂, as already pointed out by Ohkawa and Shimizu [\[16\]](#page-6-9). Instead of this scheme, Ohkawa and Shimizu have proposed the Γ_5 -doublet scheme and the other quadrupolar ordering model of O_2^2 or $O_{xy}(\propto J_x J_y)$. This model may account for the close proximity of the antiferromagnetism, but cannot explain appropriately the heavy-fermion behaviours above T_N . Furthermore, the quadrupolar order parameter is invariant under time-reversal symmetry, while Si NMR measurements have strongly suggested that the *hidden* order parameter breaks time-reversal invariance [\[6,](#page-6-11) [17\]](#page-6-12). An octupole moment consistent with this observation becomes the most probable candidate for the *hidden* order, if we based on the novel level scheme of $\Gamma_1 - \Gamma_5$ (singlet–doublet), as shown in the following.

If we work within the subspace spanned by $|\Gamma_1\rangle$ and $|\Gamma_5\pm\rangle$, we should use $9(=3 \times 3)$ independent matrices, which represent three dipole and five quadrupole moments, as well as the unit matrix. We make use of the vector $\{|\Gamma_5+\rangle, |\Gamma_1\rangle, |\Gamma_5-\rangle\}$ as the base to represent the matrices, for which four matrices indispensable for the following discussion are defined as

$$
\tau_1 \equiv \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \qquad \tau_2 \equiv \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & -i & 0 \\ i & 0 & -i \\ 0 & i & 0 \end{pmatrix}, \qquad \tau_{3,4} \equiv \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \mp 1 \end{pmatrix}
$$

where the (3,3) component of τ_3 and τ_4 are −1 and 1, respectively. By the use of these matrices, the dipole moments are represented as $J_x = c_x \tau_1$, $J_y = c_y \tau_2$, $J_z = c_z \tau_3$, where $c_x = 2\alpha\gamma + \sqrt{10\beta\delta}$, $c_y = -c_x$, $c_z = 3\gamma^2 - \delta^2$; and the crystal-field Hamiltonian as $\mathcal{H}_{CF} = \Delta_{CF} \tau_4.$

From the analysis of χ_c and C_e , we have already determined the values of γ^2 and Δ_{CF} . If we attempt to fit χ_a^{exp} shown by the open triangles in figure [1\(](#page-4-0)a) within this subspace, we may take $\gamma = 0.891$ ($\gamma^2 = 0.794$), $\delta = -0.454$, $\alpha = 0.702$, $\beta = 0.117$ to obtain the calculated results as shown by the dash–dotted line in figure [1\(](#page-4-0)a). The calculated χ_a decreases with increasing *T* and deviates considerably from χ_a^{exp} , which is almost independent of *T*. It follows that the moments of J_x and J_y are almost inactive in this subspace, that is $c_x = 2\alpha\gamma + \sqrt{10\beta\delta} \approx 0$, and χ_a is a Van Vleck susceptibility induced by the off-diagonal matrix elements with the upper doublet, $\delta|\pm 3\rangle - \gamma|\mp 1\rangle$, separated by about 1500 K.

If we take $c_x = 0$, we have $\gamma = 0.891$, $\delta = -0.454$, $\alpha = 0.531$, $\beta = 0.660$, for which the degrees of freedom corresponding to J_x and J_y should be carried by the octupole moments defined as $T_x^{\beta} \equiv 0$ √ $\sqrt{15/6}$ $(\overline{J_x J_y^2} - \overline{J_x J_z^2}) = -d\tau_1, T_y^{\beta} \equiv 0$ √ $15/6$ $(J_y J_z^2 - J_y J_x^2) =$ $-d\tau_2$ [\[23,](#page-6-13) [24\]](#page-6-14), where $d = (3$ $\sqrt{6}/4$ [5β($\sqrt{7}\gamma - 3δ$) + $\sqrt{10}\alpha(7\gamma + \sqrt{7δ})$] ≈ 38. Note that the other octupole moments are expressed by $T_x^{\alpha} = -e\tau_1$, $T_y^{\alpha} = e\tau_2$, $T_z^{\alpha} = f\tau_3$, $T_z^{\beta} = T_{xyz} = 0$ [\[23,](#page-6-13) [24\]](#page-6-14), where $e \approx 7.7 \ll d$ and $f \approx 5.2 \ll d$. Then, we have eight active moments of $\{J_z, O_2^0, O_2^2, O_{yz}, O_{zx}, O_{xy}, T_x^{\beta}, T_y^{\beta}\}$, among which $T_{x,y}^{\beta}$ have the largest components. We can therefore expect that the octupole moments $T_{x,y}^{\beta}$ are the primary order parameter of the *hidden* order. We should note that Kiss and Fazekas [\[18\]](#page-6-15) have also proposed the other type of octupolar ordering model based on a rather complicated crystal field level scheme of $\Gamma_1 - \Gamma_4 - \Gamma_5 - \Gamma_2$.

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Our octupolar ordering model may be consistent with the Si NMR measurements by Bernal *et al* [\[6\]](#page-6-11). The Si NMR in URu₂Si₂ is very similar to the B NMR in CeB₆, for which the hyperfine coupling between the nuclear spin of B and the octupole moments of Ce is derived by Sakai *et al* [\[25\]](#page-6-16). Based on their derivation, we obtain the hyperfine interaction for the nuclear spin \vec{l} of Si from the Fourier components of the moments of the 5f electrons at U as $h_{hf} = aI_zJ_z(\vec{Q}) + bI_xT_x^{\beta}(\vec{Q}) - bI_yT_y^{\beta}(\vec{Q})$ for $\vec{Q} = (0, 0, \pi/c)$, or a more complicated form for an arbitrary \vec{Q} . Such an interaction may account for the *T* and *H* dependence of the Si linewidth Γ , including the finite and nearly isotropic nature for $H \to 0$ for $T < T_N$. We should note that the microscopic origin of this interaction is considered to be an octupole correction of the dipole–dipole interaction [\[24\]](#page-6-14). An investigation on this subject is now in progress.

Finally, we point out that the close proximity of weak antiferromagnetism may be also accounted for by the present model, similarly to the doublet model [\[16\]](#page-6-9). The minimal interactions to discuss the *hidden* order may be those of $T_x^{\beta}(\tau_1)$, $T_y^{\beta}(\tau_2)$, $J_z(\tau_3)$, and $\mathcal{H}_{CF}(\tau_4)$, which present a spin-1 XXZ model with a uniaxial anisotropy on the body-centred hexagonal lattice $(XX: T_{x,y}^{\beta})$ interaction; Z: *J_z* one; and the anisotropy: \mathcal{H}_{CF}). An ordering of the XX (xy) components of *S* indicates an octupolar ordering, and a slight canting of *S* from the *xy* plane induces a tiny magnetic moment along the *c* axis. Such a model of local-spin interactions will lose the quantitative agreement with experiments obtained here, but may be useful to examine the phase diagrams of URu₂S₁² on the *H*–*T* [\[7\]](#page-6-17) and *T*–*p* [\[5,](#page-6-18) [8\]](#page-6-19) planes qualitatively.

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