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Analysis of *f*-*p* model for octupole ordering in NpO_2

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In order to examine the origin of octupole ordering in NpO_2, we propose a microscopic model constituted of neptunium $5f$ and oxygen $2p$ orbitals. To study multipole ordering, we derive effective multipole interactions from the f-p model by using the fourth-order perturbation theory in terms of p-f hopping integrals. Analyzing the effective model numerically, we find a tendency toward Γ_{5u} antiferro-octupole ordering.

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In the research field of condensed matter physics, it is one of currently important issues to unveil the mechanism of multipole ordering phenomena frequently observed in f-electron systems. It has been widely recognized that quadrupole ordering realizes in several f-electron compounds, but recently, in addition to dipole and quadrupole ordering, a possibility of ordering of higher-order magnetic multipoles, i.e., octupoles, has been discussed intensively.

A typical candidate with octupole ordering is the low-temperature ordered phase of NpO_2,[1] since time reversal symmetry is broken in this phase,[2,3] but the detected internal field is too weak to be ascribed to dipole ordering.[2–4] Indeed, several experimental facts[5–8] can be consistently explained by assuming longitudinal triple-**q** Γ_{5u} octupole ordering.[9] In addition, recent experiments on the ^{17}O NMR also support the triple-**q** ordered state.[10,11]

In order to understand why such higher-order multipole order is realized in NpO_2, it has been highly required to proceed to the microscopic research. In general, it is difficult to develop a microscopic theory for complex multipole ordering in f-electron systems beyond the phenomenological level, but it has been recently proposed to construct a microscopic f-electron model on the basis of a j-j coupling scheme.[12] Following this proposal, we have studied the f-electron model on an fcc lattice composed of Np ions with hopping integrals via $(ff\sigma)$ bonding, and actually found the triple-**q** octupole ordering.[13] However, as shown in Fig. 1(a), oxygen anions exist between Np ions in the actual crystal structure. Thus, it is important to clarify how the octupole ordering in the f-electron model is affected by oxygen anions.

In this paper, we construct a more realistic model including also p orbitals of oxygen anions in addition to f orbitals of Np ions. We derive an effective multipole interaction model by evaluating the exchange of f electrons via p orbitals within the fourth-order perturbation with respect to p-f hopping integrals. By analyzing the effective model, we again find a tendency toward the triple-**q** octupole ordered phase, indicating that the f-electron model on the fcc lattice has grasped the essential point on the appearance of octupole ordering of NpO_2.

Let us discuss local f-electron states of actinide dioxides based on the j-j coupling scheme, in which we first include the spin-orbit interaction and consider only the states with total angular momentum $j=\frac{5}{2}$. From a quantitative viewpoint, this simplification may not be appropriate for actinide dioxides, since they are insulators, and the LS coupling scheme is expected to work well to describe f-electron states in these materials. However, both schemes are continuously connected to each other by changing the ratio of the strength of the spin-orbit interaction and the Coulomb interaction, as long as the symmetry of the ground state is not changed.[14] Thus, on the basis of a spirit of adiabatic continuation, we expect that qualitative properties at low temperatures can be grasped whether we choose the j-j coupling or the LS coupling schemes as a starting approximation. The $j=\frac{5}{2}$ states split into a Γ_7 doublet and a Γ_8 quartet under a cubic crystalline electric field (CEF). Since the Γ_7 wave function extends along the [111] direction and an oxygen anion is located in this direction, the Γ_7 level is expected to be higher than the Γ_8 level. If we assume that the level splitting Δ between Γ_8 and Γ_7 is large enough, CEF ground states for f^2, f^3, and f^4 are obtained by accommodating two, three, and four electrons into Γ_8 levels, leading to Γ_5, Γ_8, and Γ_1, respectively, consistent with experimental results for UO_2,[15] NpO_2,[16] and PuO_2,[17] respectively. Thus, we ignore the Γ_7 state to discuss the ground state of actinide dioxides in the j-j coupling scheme.

We have three comments on the CEF level scheme. (i) In our picture, the first excited state of PuO_2 should include three Γ_8 and one Γ_7 electrons, indicating that the excitation energy provides the lower limit for Δ. Since the CEF excitation energy of PuO_2 is 123 meV,[17] Δ should be larger than 1400 K, consistent with the initial assumption. (ii) The f^3 state in NpO_2 is regarded as the one hole state in Γ_8. In the following, we use a hole picture and the "electron" denotes such a hole. (iii) Among the f^2 states, the Γ_5 triplet is the

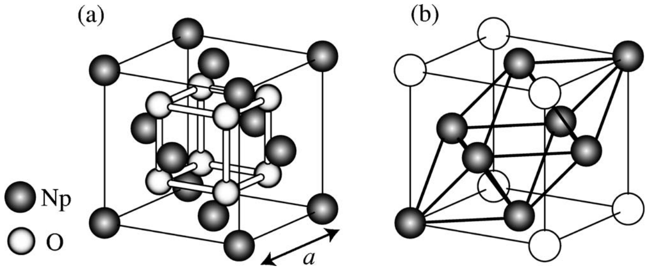

FIG. 1. (a) Crystal structure of NpO_2. a is the lattice constant. (b) eight-site cluster (gray spheres) on the fcc lattice taken in our numerical calculation.

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ground state as observed in UO_2. Since the CEF excitation energy in UO_2 is as large as 150 meV,[15] we consider only the Γ_5 triplet among f^2 intermediate states to study exchange processes of f^3 ions in NpO_2.

The Γ_8 quartet consists of two Kramers doublets, and it is convenient to introduce orbital index $\tau(=\alpha,\beta)$ to label the two Kramers doublets and spin index $\sigma(=\uparrow,\downarrow)$ to distinguish the two states in each Kramers doublet. In the second-quantized form, annihilation operators for Γ_8 electrons are defined by $f_{\mathbf{r}\alpha\uparrow}=\sqrt{5/6}a_{\mathbf{r}5/2}+\sqrt{1/6}a_{\mathbf{r}-3/2}$, $f_{\mathbf{r}\alpha\downarrow}=\sqrt{5/6}a_{\mathbf{r}-5/2}+\sqrt{1/6}a_{\mathbf{r}3/2}$, $f_{\mathbf{r}\beta\uparrow}=a_{\mathbf{r}1/2}$, and $f_{\mathbf{r}\beta\downarrow}=a_{\mathbf{r}-1/2}$, where $a_{\mathbf{r}j_z}$ is the annihilation operator for an f electron with the z component j_z of the total angular momentum $j=5/2$ at site $\mathbf{r}$. Multipole operators are usually expressed as $X_{\mathbf{r}}^{\Gamma\gamma}$, where Γ_γ denotes symmetry. The explicit forms of $X_{\mathbf{r}}^{\Gamma\gamma}$ in the Γ_8 subspace are found in Ref. 13.

Now we show the f-p model for NpO_2, given by

$$\mathcal{H}=\mathcal{H}_f+\mathcal{H}_p+\mathcal{H}_{\text{kin}}, \tag{1}$$

where $\mathcal{H}_f$ and $\mathcal{H}_p$ are the local f- and p-electron terms, respectively, and $\mathcal{H}_{\text{kin}}$ denotes the hybridization term between f and p electrons. Among them, the local f-electron term is explicitly given by

$$\begin{aligned}\mathcal{H}_f=\epsilon_f\sum_{\mathbf{r},\tau} n_{\mathbf{r}\tau}+U\sum_{\mathbf{r}\tau} n_{\mathbf{r}\tau\uparrow}n_{\mathbf{r}\tau\downarrow}+U'\sum_{\mathbf{r}} n_{\mathbf{r}\alpha}n_{\mathbf{r}\beta}\\ +J\sum_{\mathbf{r},\sigma,\sigma'} f^{\dagger}_{\mathbf{r}\alpha\sigma}f^{\dagger}_{\mathbf{r}\beta\sigma'}f_{\mathbf{r}\alpha\sigma'}f_{\mathbf{r}\beta\sigma}+J'\sum_{\mathbf{r},\tau\neq\tau'} f^{\dagger}_{\mathbf{r}\tau\uparrow}f^{\dagger}_{\mathbf{r}\tau\downarrow}f_{\mathbf{r}\tau'\downarrow}f_{\mathbf{r}\tau'\uparrow},\end{aligned} \tag{2}$$

where ϵ_f denotes the energy level of Γ_8, $n_{\mathbf{r}\tau\sigma}=f^{\dagger}_{\mathbf{r}\tau\sigma}f_{\mathbf{r}\tau\sigma}$, and $n_{\mathbf{r}\tau}=\Sigma_\sigma n_{\mathbf{r}\tau\sigma}$. The coupling constants U, U', J, and J' denote the intraorbital Coulomb, interorbital Coulomb, exchange, and pair-hopping interactions, respectively.

The local p-electron term is expressed as

$$\mathcal{H}_p=\epsilon_p\sum_{\mathbf{x}m,\sigma} p^{\dagger}_{\mathbf{x}m\sigma}p_{\mathbf{x}m\sigma}+(\text{on-site interactions for } p \text{ orbitals}), \tag{3}$$

where ϵ_p denotes the energy level of p orbitals of oxygen ions and $p_{\mathbf{x}m\sigma}$ is the annihilation operator for an electron with the z component m of orbital angular momentum and real spin σ at O site $\mathbf{x}$. Finally, $\mathcal{H}_{\text{kin}}$ is given by

$$\mathcal{H}_{\text{kin}}=\sum_{\mathbf{r},m,\sigma,s,\boldsymbol{\nu}}(t^{\boldsymbol{\nu}}_{m\sigma;s}p^{\dagger}_{\mathbf{r}+\boldsymbol{\nu}m\sigma}f_{\mathbf{r}s}+\text{H.c.}), \tag{4}$$

where $\boldsymbol{\nu}$ is a vector connecting nearest-neighbor Np and O sites and s symbolically denotes both the spin and orbital of an f electron. The hopping integrals $t^{\boldsymbol{\nu}}_{m\sigma;s}$ are represented in terms of $(pf\sigma)$ and $(pf\pi)$ by using the Slater-Koster table.[18]

From the f-p model (1), we derive an effective interaction by using the fourth-order perturbation theory with respect to the p-f hopping integrals. Here we note that the fourth-order processes are classified into two types: (a) Two electrons doubly occupy a Np ion in the intermediate states, as schematically shown in Fig. 2(a). (b)–(e) Two electrons occupy one or two oxygen sites simultaneously in the intermediate states, as shown in Figs. 2(b)–2(e). The effective Hamiltonian is arranged in the form of

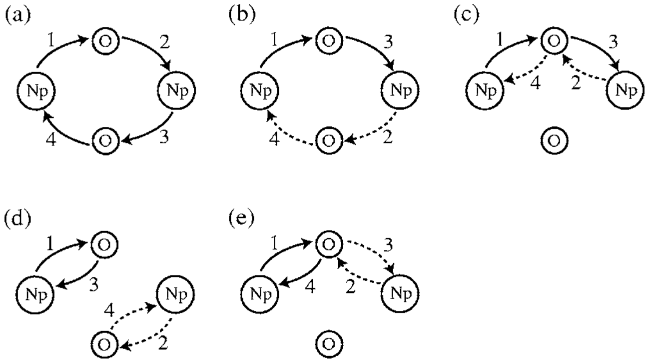

FIG. 2. Fourth-order processes with respect to p-f hoppings. (a) Electrons doubly occupy a Np site. Electrons occupy (b) two or (c) one O site simultaneously, and then exchange. Electrons occupy (d) two or (e) one O site simultaneously, and then return to the initial sites. The solid and dashed lines in (b)–(e) distinguish paths for different electrons.

$$\mathcal{H}_{\text{eff}}=\sum_{\langle\mathbf{r},\mathbf{r}'\rangle,s_1-s_4} I^{\mathbf{r}'-\mathbf{r}}_{s_3,s_4;s_1,s_2}f^{\dagger}_{\mathbf{r}s_3}f_{\mathbf{r}s_1}f^{\dagger}_{\mathbf{r}'s_4}f_{\mathbf{r}'s_2}, \tag{5}$$

where $\langle\mathbf{r},\mathbf{r}'\rangle$ denotes the pair of nearest-neighbor sites and the effective interaction I is given by

$$I^{\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2}=\sum_{\text{k=a–e}} I^{(\text{k})\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2}. \tag{6}$$

In the following, we will briefly explain each term included in I.

In order to derive multipole interaction $I^{(\text{a})}$ from the process (a), we simply replace f-electron hopping $t^{\boldsymbol{\mu}}_{s;s'}$ in the effective interaction I of the f-electron model[13] with the effective f-f hopping $T^{\boldsymbol{\mu}}_{s;s'}$ via p orbitals. For the purpose, we define f-electron hopping between sites $\mathbf{r}$ and $\mathbf{r}+\boldsymbol{\mu}$ via p orbitals of the oxygen ion at $\mathbf{r}+\boldsymbol{\nu}$, given by

$$T^{\boldsymbol{\mu},\boldsymbol{\nu}}_{s;s'}=(\epsilon_f-\epsilon_p)^{-1}\sum_{m\sigma}(t^{\boldsymbol{\nu}-\boldsymbol{\mu}}_{m\sigma;s})^{*}t^{\boldsymbol{\nu}}_{m\sigma;s'}. \tag{7}$$

Then, we obtain $T^{\boldsymbol{\mu}}_{s;s'}=\Sigma_{\boldsymbol{\nu}}T^{\boldsymbol{\mu},\boldsymbol{\nu}}_{s;s'}$, since the effective f-electron hopping consists of two paths through different oxygen sites. For instance, along the [110] direction, the effective f-f hopping integral is given by $T^{(a/2,a/2,0)}_{s;s'}=T^{(a/2,a/2,0),(a/4,a/4,a/4)}_{s;s'}+T^{(a/2,a/2,0),(a/4,a/4,-a/4)}_{s;s'}$, where a is the lattice constant. Using $T^{\boldsymbol{\mu}}_{s;s'}$, we express $I^{(\text{a})}$ as

$$\begin{aligned}I^{(\text{a})\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2}=-(U'-J)^{-1}\sum_{u,s,s'}[(T^{\mathbf{r}'-\mathbf{r}}_{s';s_4})^{*}P^{*}_{u;s_3,s'}P_{u;s_1,s}T^{\mathbf{r}'-\mathbf{r}}_{s;s_2}\\ +(T^{\mathbf{r}-\mathbf{r}'}_{s';s_3})^{*}P^{*}_{u;s_4,s'}P_{u;s_2,s}T^{\mathbf{r}-\mathbf{r}'}_{s;s_1}],\end{aligned} \tag{8}$$

where $P_{u;s,s'}$ denotes the inner product between one of the Γ_5 triplet states denoted by u and the f^2 state labeled by s and s'.

We note that the effective f-f hopping integrals have the same form as those via $(ff\sigma)$ bonding,[13] for instance,

$$T^{(a/2,a/2,0)}_{\tau\sigma;\tau'\sigma'} \propto [\delta_{\tau\tau'}\delta_{\sigma\sigma'} + c_1\sigma^z_{\tau\tau'}\delta_{\sigma\sigma'} + c_2\sigma^y_{\tau\tau'}\sigma^z_{\sigma\sigma'}], \quad (9)$$

where $\boldsymbol{\sigma}$ are Pauli matrices, and c_1 and c_2 are constants depending on $(pf\sigma)$ and $(pf\pi)$. This fact indicates that the form of the hopping integrals are restricted by f-electron symmetry, and the simple $(ff\sigma)$ model may grasp properties of actual materials with complex structures.

Concerning processes (b)–(e) in Fig. 2, we consider the effect of the Coulomb interaction at oxygen sites, symbolically expressed as "U_p". In this paper, we study two limiting cases, $U_p=0$ and $U_p=\infty$. For $U_p=0$, multipole interaction contains all the processes (b)–(e) in Fig. 2, given by

$$I^{(b)\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2} = 2(\epsilon_p-\epsilon_f)^{-1}\sum_{\boldsymbol{\nu}} T^{\boldsymbol{\mu},\boldsymbol{\nu}}_{s_4;s_1} T^{-\boldsymbol{\mu},-\boldsymbol{\nu}}_{s_3;s_2}, \quad (10)$$

$$I^{(c)\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2} = 2(\epsilon_p-\epsilon_f)^{-1}\sum_{\boldsymbol{\nu}} T^{\boldsymbol{\mu},\boldsymbol{\nu}}_{s_4;s_1} T^{-\boldsymbol{\mu},\boldsymbol{\nu}-\boldsymbol{\mu}}_{s_3;s_2}, \quad (11)$$

$$I^{(d)\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2} = 2(\epsilon_p-\epsilon_f)^{-1}\sum_{\boldsymbol{\nu}} S^{\boldsymbol{\nu}}_{s_4;s_1} S^{-\boldsymbol{\mu}+\boldsymbol{\nu}}_{s_3;s_2}, \quad (12)$$

and

$$I^{(e)\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2} = -I^{(d)\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2}, \quad (13)$$

respectively, where

$$S^{\boldsymbol{\nu}}_{s;s'} = (\epsilon_f-\epsilon_p)^{-1}\sum_{m\sigma} (t^{\boldsymbol{\nu}}_{m\sigma;s})^* t^{\boldsymbol{\nu}}_{m\sigma;s'}. \quad (14)$$

Note that the sum of processes (d) and (e) merely becomes an energy shift, since $I^{(e)\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2} + I^{(d)\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2} \propto \delta_{s_1s_3}\delta_{s_2s_4}$, and such terms can be eliminated in the present discussion. After all, the multipole interaction for $U_p=0$ is given by

$$I^{\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2} = I^{(a)\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2} + 2(\epsilon_p-\epsilon_f)^{-1} T^{\boldsymbol{\mu}}_{s_4;s_1} T^{-\boldsymbol{\mu}}_{s_3;s_2}. \quad (15)$$

Note that this term is expressed by only the effective f-f hopping integral.

Now we move on to another limiting case $U_p=\infty$, in which the effect of U_p is included by prohibiting processes (c) and (e). Thus, the effective interaction for $U_p=\infty$ is given by

$$I^{\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2} = I^{(a)\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2} + I^{(b)\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2} + I^{(d)\boldsymbol{\mu}}_{s_3,s_4;s_1,s_2}. \quad (16)$$

It should be noted that in the process (b), a couple of electrons exchange their sites by avoiding the effect of Coulomb interactions. Such a term is characteristic to the crystal structure of $\mathrm{NpO_2}$, and it has a tendency to stabilize the octupole ordering, as shown later.

Using the effective Hamiltonian, we evaluate numerically the multipole correlation function $\chi^{\Gamma\gamma}_{\mathbf{q}}=(1/N)\Sigma_{\mathbf{r},\mathbf{r}'}e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')} \times\langle X^{\Gamma\gamma}_{\mathbf{r}} X^{\Gamma\gamma}_{\mathbf{r}'}\rangle$, where $\langle\ \rangle$ denotes the expectation value using the ground-state wave function. Here we take $N=8$, as shown in Fig. 1(b). Figures 3(a) and 3(c) show phase diagrams, presenting the multipole moment which has the largest value in the correlation function at each parameter set, for $U_p=0$ and $U_p=\infty$, respectively. For $U_p=0$, as shown in Fig. 3(b) in a magnified scale, there is a very small, but finite region of the Γ_{5u} antiferro-octupole [$\mathbf{q}=(0,0,1)$ in units of $2\pi/a$, 5uAF] phase. For $U_p=\infty$, on the other hand, the region of the 5uAF phase becomes much larger than that for $U_p=0$. The 5uAF phase locates in the parameter region with $\epsilon_p-\epsilon_f \ll U'-J$, in which processes (b) and (d) work effectively. Since process (d) provides only a quadrupole interaction, we conclude that the stabilization of the 5uAF phase originates from the process (b).

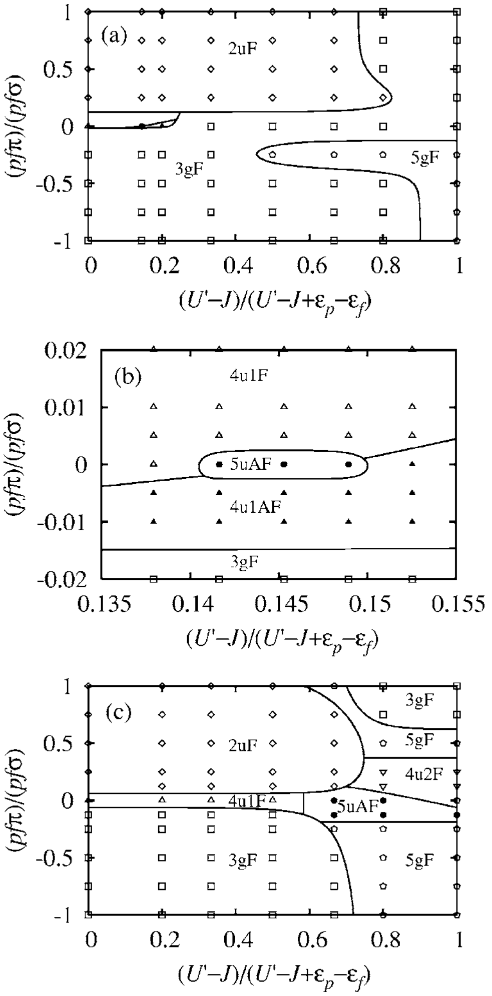

FIG. 3. Phase diagrams obtained from the multipole correlation functions. Here 3gF denotes the Γ_{3g} moment with $\mathbf{q}=(0,0,0)$, and so on. The antiferro (AF) ordering vector is $\mathbf{q}=(0,0,1)$ in units of $2\pi/a$ for all the AF phases. (a) Phase diagram for $U_p=0$. (b) Phase diagram for $U_p=0$ in a magnified scale around $(U'-J)/(\epsilon_p-\epsilon_f+U'-J)=0.145$ and $(pf\pi)=0$. (c) Phase diagram for $U_p=\infty$.

Note that the Coulomb energy in the f^2 intermediate state $U'-J$ is expected to be in the order of 1 eV, but we cannot estimate the difference of the energy levels of p and f electrons $\epsilon_p-\epsilon_f$ within the present theory. We also note that in the phase diagrams, the 5uAF phase appears for $(pf\pi)\simeq 0$. Since $(pf\sigma)$ and $(pf\pi)$ are treated as parameters in this paper, we have no clear answer as to why the absolute value of $(pf\pi)$ should be so small for the appearance of the 5uAF phase. In order to confirm the reality of the parameter region for the octupole phase obtained in this study is realistic or not, it is highly requested to perform the band-structure calculations for $\mathrm{NpO_2}$. This is one of future problems.

In summary, on the basis of the f-p model for $\mathrm{NpO_2}$, we have found a finite region of the 5uAF phase for both cases of $U_p=0$ and $U_p=\infty$. Thus, we expect that this property is retained in the actual situation for $\mathrm{NpO_2}$ with finite U_p. While the ordered state cannot be entirely determined within the present small-cluster calculation, it is confirmed that among structures with $\mathbf{q}=(0,0,1)$ and equivalent ones, the triple-$\mathbf{q}$ structure is energetically favorable,[13] since the Γ_{5u} moment in the Γ_8 subspace has the easy axis along the $[111]$ direction. We emphasize that the Γ_{5u} antiferro-octupole phase is also realized in the simple $(ff\sigma)$ hopping model on an fcc lattice even without oxygen p orbitals.[13] These findings indicate that the tendency toward Γ_{5u} antiferro-octupole ordering is common to Γ_8 models on fcc lattices.

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