

## Why the Hidden Order in URu<sub>2</sub>Si<sub>2</sub> Is Still Hidden—One Simple Answer

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For more than two decades, the nonmagnetic anomaly observed around 17.5 K in URu<sub>2</sub>Si<sub>2</sub>, has been investigated intensively. However, any kind of fingerprint for the lattice anomaly has not been observed in the low-temperature ordered phase. Therefore, the order has been called “the hidden order”. One simple answer to why the hidden order is still hidden is presented from the space group analysis. The second-order phase transition from *I4/mmm* (No. 139) to *P4<sub>2</sub>/mmm* (No. 136) does not require any kind of lattice distortion in this system and allows the NQR frequency at a Ru site unchanged. It is compatible with *O<sub>xy</sub>*-type antiferro-quadrupole ordering with **Q** = (0, 0, 1). The characteristics of the hidden order are discussed based on the local *5f<sup>2</sup>* electron picture.

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A second-order phase transition in solid states causes a change in symmetry, such as gauge symmetry or time reversal symmetry. On crystal symmetry, it usually accompanies lattice distortion to reach a lower symmetry. In exchange for releasing entropy, it becomes a lower energy state at lower temperatures. The lattice distortion itself requires loss of energy; thus another mechanism is necessary to overcome the rising energy in a lattice system. The Jahn–Teller effect is known as an example. The degeneracy of electronic states is lifted, due to lattice distortion, to obtain a lower energy. Even when electronic states are not local, such an effect is widely discussed in terms of the Jahn–Teller effect.

The magnitude of such a distortion strongly depends on the mechanism to yield lower energy states. Lowering the symmetry changes the crystalline electric field (CEF) and the mixing strength, through the movements of atoms. The greater lattice distortion causes the lower energy of the electronic state. Therefore, the lattice is sometimes largely distorted, up to 10% of the lattice constant, as *1T-TaSe<sub>2</sub>*.<sup>1)</sup> In the ordered charge density wave phase, the electronic states produce a large gap between the bonding and antibonding states,<sup>2)</sup> where a strong Coulomb repulsion does not play an important role. In contrast, when the Coulomb interaction plays a crucial role, the distortion is kept very small, e.g., only about 0.05% in *PrRu<sub>4</sub>P<sub>12</sub>*,<sup>3)</sup> where the lattice distortion itself does not require to gain lower electronic states.<sup>4)</sup> Totally-symmetric antiferro-multipole ordering breaks the translational symmetry to open the gap in the conduction band, then the lattice is slightly and symmetrically distorted in order to relax multipoles developed on Pr sites.

URu<sub>2</sub>Si<sub>2</sub> shows a large peak in specific heat at 17.5 K (*T<sub>O</sub>*), indicating a second-order phase transition, then the ordered state coexists with superconductivity found below 1.5 K.<sup>5–7)</sup> Although intensive theoretical and experimental studies have been performed, the order parameter of the phase below *T<sub>O</sub>* is still unknown, and thus, it is called the hidden order (HO). Quite different theoretical proposals have been reported:<sup>8–16)</sup> some neglecting the itinerant character of *5f* electrons,

and some omitting the local specificity of U site; in the U<sup>4+</sup> valence state (*5f<sup>2</sup>* configuration), multipole ordering can be expected as recently discussed in the physics of Pr skutterudite systems.<sup>17)</sup>

On the experimental side, it seems now well established that the low-pressure HO phase is not antiferromagnetic, but that above *P<sub>x</sub>* ~ 0.5 GPa through the first-order transition long-range antiferromagnetism (AF) is established with the wave vector **Q<sub>AF</sub>** = (0, 0, 1) corresponding to a lattice doubling along the *c*-axis.<sup>18–20)</sup> Furthermore, the HO phase is characterized by a sharp resonance at the energy *E<sub>0</sub>* = 1.5 meV observed at the wave vector **Q<sub>O</sub>** = (1, 0, 0) [equivalent to (0, 0, 1) in a body-centered tetragonal lattice]. Switching to the AF phase at *P<sub>x</sub>* leads to the collapse of the resonance and to the observation of an elastic neutron magnetic signal at **Q<sub>O</sub>**.<sup>20)</sup> Taking into account that the same characteristic vector **Q<sub>O</sub>** emerges in the HO and AF phases, as well as the invariance of three Shubnikov de Haas frequencies through *P<sub>x</sub>*,<sup>21)</sup> the experimental data indicate the need to search for a model, where the transition from the PM phase to the HO or AF ground state corresponds to the same structural transition among the tetragonal classes, while the pressure switching from HO to AF induces a supplementary time-reversal breaking.

A previous band structure calculation demonstrates that AF opens a large gap at the Fermi level; thus, it is suggested that the occurrence of large magnetic fluctuations preserves a Fermi surface construction at *T<sub>O</sub>* in the HO phase similar to that proposed for the transition from PM to AF above *P<sub>x</sub>*.<sup>16)</sup> Recently, by using an LDA+DMFT method, an idea similar to our proposal has been developed by Haule and Kotliar,<sup>22)</sup> but the selected order parameter of HO is a hexadecapole.

In this letter, we propose that the space group in the HO phase is No. 136 and that the transition at 17.5 K does not require any kind of lattice distortion and maintains a Ru site with 4-fold symmetry. Therefore, most of the experimental techniques are unable to detect the characteristic charge distribution in the HO phase. This is one simple answer to why the HO is still hidden.

After a second-order phase transition, the lower symmetry must belong to one of the subgroups of the mother

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Table I. Multiplicity, Wyckoff letter, and site symmetry of each atom in URu<sub>2</sub>Si<sub>2</sub>. First row: the case of No. 139, the mother space group; below the second row: the maximal non-isomorphic  $k$  subgroups of No. 139. All the subgroups belong to  $D_{4h}$ . The site coordinates are shown, when the notation changes. In No. 131, the origin of the lattice changes, but the lattice does not need to be distorted.

Space group	U site	Ru site	Si site
No. 139 $I4/mmm$	$2a$ $4/mmm$ (0,0,0)	$4d$ $\bar{4}m2$ (0, 1/2, 1/4), (1/2, 0, 1/4)	$4e$ $4mm$ (0, 0, $z$ ), (0, 0, $\bar{z}$ )
No. 123 $P4/mmm$	$1a$ $4/mmm \oplus 1d$ $4/mmm$	$4i$ $2mm$ . (0, 1/2, $z$ ), ...	$2g$ $4mm$ (0, 0, $z$ ), ... $\oplus$ $2h$ $4mm$ (1/2, 1/2, $z'$ ), ...
No. 126 $P4/nnc$	$2a$ $422$	$4d$ $\bar{4}..$	$4e$ $4..$
No. 128 $P4/mnc$	$2a$ $4/m$	$4d$ $2.22$	$4e$ $4..$
No. 129 $P4/nmm$	$2c$ $4mm$ (0, 1/2, $z$ ), ...	$2a$ $\bar{4}m2$ (0, 0, 0), ... $\oplus$ $2b$ $\bar{4}m2$ (0, 0, 1/2), ...	$2c$ $4mm$ (0, 1/2, $z$ ), ... $\oplus$ $2c$ $4mm$ (0, 1/2, $z'$ ), ...
No. 131 $P4_2/mmc$	$2c$ $mmm$ . (0, 1/2, 0), ...	$2e$ $\bar{4}m2$ (0, 0, 1/4), ... $\oplus$ $2f$ $\bar{4}m2$ (1/2, 1/2, 1/4), ...	$4i$ $2mm$ . (0, 1/2, $z$ ), ...
No. 134 $P4_2/nmm$	$2a$ $\bar{4}2m$	$4d$ $2.22$	$4g$ $2.mm$
No. 136 $P4_2/mnm$	$2a$ $m.mm$	$4d$ $\bar{4}..$	$4e$ $2.mm$
No. 137 $P4_2/nmc$	$2a$ $\bar{4}m2$	$4d$ $2mm$ . (0, 1/2, $z$ ), ...	$4c$ $2mm$ .

symmetry. The space group in the high-temperature phase of URu<sub>2</sub>Si<sub>2</sub> (ThCr<sub>2</sub>Si<sub>2</sub>-type) is No. 139 ( $I4/mmm$ ;  $D_{4h}^{17}$ ), which has seven maximal non-isomorphic  $t$  subgroups (*translationengleiche*), eight maximal non-isomorphic  $k$  subgroups (*klassengleiche*), and two maximal isomorphic subgroups of lowest index.<sup>23</sup>  $t$  subgroups (No. 69 ( $Fmmm$ ;  $D_{2h}^{23}$ ), No. 71 ( $Immm$ ;  $D_{2h}^{25}$ ), No. 87 ( $I4/m$ ;  $C_{4h}^5$ ), No. 97 ( $I422$ ;  $D_4^9$ ), No. 107 ( $I4mm$ ;  $C_{4v}^9$ ), No. 119 ( $I4m2$ ;  $D_{2d}^9$ ), and No. 121 ( $I42m$ ;  $D_{2d}^{11}$ )) change the point group, and the isomorphic subgroup requires a superstructure [( $c' = 3c$ ) or ( $a' = 3a$ ,  $b' = 3b$ )]; thus, they should be easily observed by experiment. As the crystal class is unchanged, the point group of 8 maximal non-isomorphic  $k$  subgroups must be considered.

The multiplicity, Wyckoff letter, and site symmetry of each atom in URu<sub>2</sub>Si<sub>2</sub> are listed in Table I, with all the  $k$  subgroups. All the space groups have the point symmetry  $D_{4h}$  [from No. 123 ( $D_{4h}^1$ ) to No. 142 ( $D_{4h}^{20}$ )]. Let us now investigate the correspondence of atom positions between the mother group and the  $k$  subgroups. No. 139 ( $I4/mmm$ ;  $D_{4h}^{17}$ ) belongs to the body centered tetragonal lattice; thus, there are 16-point symmetry operations and (1/2, 1/2, 1/2) translation in the conventional simple lattice. 32 symmetry operations transfer an atom to a crystallographically equivalent position or rotate/reflect an atom at the same position. For example, U at (0,0,0) in No. 139 is transferred by (1/2, 1/2, 1/2), but not by the 16 point symmetry operations, resulting in the order of the site symmetry ( $4/mmm$ ;  $D_{4h}$ ) of 16. The multiplicity of the site (2) times the order of the site symmetry (16) is 32. In the cases of Ru and Si, the multiplicity of the site is 4, and the order of the site symmetry is 8 [ $\bar{4}m2$ ;  $D_{2d}$ ] or ( $4mm$ ;  $C_{4v}$ ), as listed in Table I.

In the maximal subgroups, 16 symmetry operations remain. The lost operation induces the splitting of the site into two inequivalent sites, or the lowering of the site symmetry. When the site is split as U and Si in No. 123, Ru and Si in No. 129, or Ru in No. 131, conventional experimental techniques can detect it. Even when the site is not split, the internal parameter arises in  $4i$  of No. 123,  $2c$  of No. 129, and  $4d$  of No. 137, resulting in the movements of atoms. However, any kind of lattice distortion is not expected when the lower space group is No. 126, No. 128, No. 134, or No. 136.

An NQR measurement is often considered a powerful tool for probing the phase transition.<sup>24,25</sup> Ru site NQR measurements have already been performed;<sup>26</sup> however they never

show any anomaly around  $T_0$ . If the Ru site loses the 4-fold symmetry axis owing to the phase transition, the asymmetric parameter  $\eta$  becomes nonzero and affects the NQR frequency. The unchanged NQR frequency indicates that the 4-fold axis of the Ru site survives even at lower temperatures. Therefore, the possibility of obtaining No. 128 or No. 134 is eliminated.

In No. 126, the local inversion symmetry at a U site is lost; thus, the U site belongs to  $422$  ( $D_4$ ). On the other hand, in No. 136, the local  $C_4$  ( $\pi/2$  rotation) at a U site is lost; thus, the U site belongs to  $mmm$  ( $D_{2h}$ ). In both cases, not the lattice distortion but the charge distortion at the U site, caused by the large Coulomb interaction between  $5f$  electrons, must be the origin of the symmetry lowering. The charge distortion without inversion symmetry could not occur without the movement of atoms. However, the charge distortion without  $C_4$  could be possible; it is ascribed to quadrupole ordering, i.e., a second-order phase transition from No. 139 to No. 136 could be compatible with  $O_{xy}$ -type antiferro-quadrupole ordering with  $\mathbf{Q} = (0, 0, 1)$ , which does not couple with any lattice distortion. The schematic charge density map of the HO phase is shown in Fig. 1.  $\mathbf{Q} = (0, 0, 1)$  is equivalent to  $\mathbf{Q} = (1, 0, 0)$  or  $(0, 1, 0)$  in the left panel of Fig. 1.

It is a naive consequence that the space group in the HO phase is No. 136. To conclude this, it is not necessary to use the  $5f^2$  local picture for URu<sub>2</sub>Si<sub>2</sub>. However, we use the local picture in the following discussion to easily discuss the origin of the transition.

As mentioned in the introduction, the low-temperature phase should have a lower energy. Now, we propose the phase transition that does not require any movement of atoms. Therefore, the mechanism to reach the lower energy must be based on large Coulomb interactions. However, the antiferro-quadrupole ordering itself does not lower the system energy but raises it owing to the local electronic charge distortion. We need a strong coupling constant for the antiferro-quadrupole interaction.

One may expect a nesting character on the Fermi surface of the non- $f$  reference compound ThRu<sub>2</sub>Si<sub>2</sub>, as in LaRu<sub>4</sub>P<sub>12</sub> for PrRu<sub>4</sub>P<sub>12</sub>. In the local  $4f^2$  picture of PrRu<sub>4</sub>P<sub>12</sub>, corresponding to LaRu<sub>4</sub>P<sub>12</sub>, the conduction band with a strong nesting character provides a large coupling constant for a high multipole interaction.<sup>4</sup> However, no clear indication of such a nesting character was observed in the band structure of ThRu<sub>2</sub>Si<sub>2</sub>, as shown later.

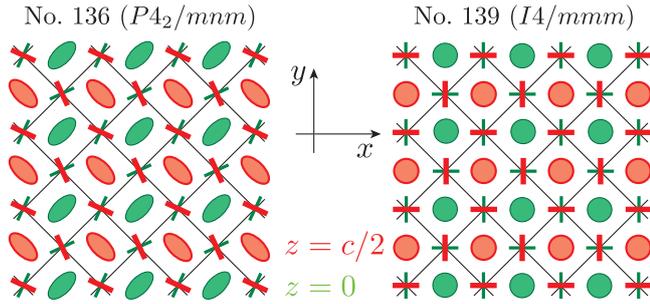


Fig. 1. Left: Schematic charge density map of the HO phase. Green and red ovals represent charge densities of U sites. Green (red) sites situate on the  $z=0$  ( $z=c/2$ ) plane. This is the  $O_{xy}$  type antiferro-quadrupole ordering. The crosses of green and red bars represent the schematic charge distribution at Ru sites on the  $z=c/4$  plane, which maintains the  $S_4$  symmetry even in the ordered phase. The charge distribution of Si (not shown) also lost the 4-fold symmetry, similar to that for U sites. Right: Schematic charge density map of the disordered phase. From right to left, any kind of lattice distortion is not allowed.

In this letter, we study which kind of orbitals can gain energy by introducing antiferro-quadrupole ordering. For simplicity, we consider a one-dimensional chain along the  $z$ -direction with  $p_x$  and  $p_y$  orbitals at each site. The lattice constant is  $a$ , and each lattice point has two sites with a distance  $a/2$ . Therefore, the energy dispersion is expressed by  $E = \varepsilon + 2t \cos(ka/2)$  for  $|k| \leq 2\pi/a$  for each of the  $p_x$  and  $p_y$  orbitals, where  $\varepsilon$  is the origin of the  $p_x$  and  $p_y$  orbitals, that is,  $\varepsilon_x$  and  $\varepsilon_y$ .  $t$  ( $< 0$ ) is the transfer or two-center integral  $[(pp\pi)]$  between the  $p_x$  (or the  $p_y$ ) orbitals along the  $z$ -direction. Then, let us now consider a unit cell with the lattice constant  $a$ , (then  $|k| \leq \pi/a$ ) which contains two sites, namely  $A$  and  $B$  with  $p_x$  and  $p_y$  orbitals each. The energy eigenvalues are expressed by the matrix

$$\begin{pmatrix} \varepsilon_x^A & 2t \cos(ka/2) & 0 & 0 \\ 2t \cos(ka/2) & \varepsilon_x^B & 0 & 0 \\ 0 & 0 & \varepsilon_y^A & 2t \cos(ka/2) \\ 0 & 0 & 2t \cos(ka/2) & \varepsilon_y^B \end{pmatrix}, \quad (1)$$

where  $\varepsilon_x^A$  is the origin of the  $p_x$  orbital at an  $A$ -sublattice site. Obviously,  $\varepsilon_x^A = \varepsilon_x^B = \varepsilon_y^A = \varepsilon_y^B$ ; thus, the 4 states are degenerated at the zone boundary  $|k| = \pi/a$ .

Once we introduce an antiferro-quadrupole ordering from an  $A$  sublattice site to a  $B$  sublattice site,  $\varepsilon_x^A = \varepsilon_y^B \neq \varepsilon_x^B = \varepsilon_y^A$ , resulting in a gap opening at the zone boundary  $|k| = \pi/a$ . A similar scenario has been discussed for filled skutterudites  $\text{PrRu}_4\text{P}_{12}$ , where a totally-symmetric antiferro-multipole ordering is introduced.<sup>4)</sup> Here, the gap ( $|\varepsilon_x^A - \varepsilon_x^B|$ ) originates in the  $O_{xy}$ -type  $5f^2$  charge distortion. This situation in band electrons with a gap at the zone boundary is very easily understood, when one imagines an antiferro spin ordering, then replaces the up and down spins with  $p_x$  and  $p_y$  orbitals. Obviously, the energy gain becomes maximum when the Fermi level is lying in the gap. However, the energy is more or less lowered when the bands are not fully occupied.

In a real three-dimensional system, flat band dispersion on the zone boundary plane  $|k_z| = \pi/c$  is necessary to open a clear gap. However, such flat dispersion could not be observed in the band structure of  $\text{ThRu}_2\text{Si}_2$ , which is shown

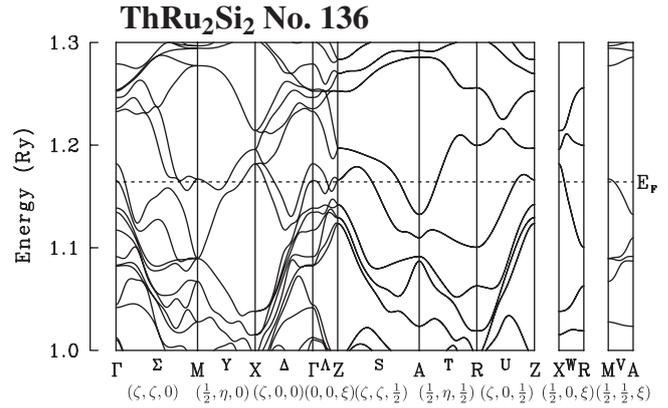


Fig. 2. Electronic band structure of  $\text{ThRu}_2\text{Si}_2$  within the space group No. 136. A self-consistent calculation results in an invisible antiferro  $O_{xy}$ -type potential; thus, this is the same as the band structure calculated within the space group No. 139 (body-centered tetragonal), and then folded into the 1st Brillouin zone of No. 136 (simple tetragonal). Note that two bands are degenerated on the boundary of the Brillouin zone (Y-, S-, T-, U-, W-, and V-axes). The degeneracies along the Y- and V-axes are due to the symmetry under No. 136, while those along the S-, T-, U-, and W-axes are accidental; thus, they will split once an antiferro  $O_{xy}$ -type potential is introduced. No flat band dispersion appears near the Fermi level  $E_F$  along the zone boundary axes, namely the S-, T-, and U-axes, on the  $|k_z| = (1/2)(2\pi/c)$  plane.

in Fig. 2. Even though it is not a clear gap opened in the vicinity of the Fermi level, introducing an antiferro  $O_{xy}$ -type potential can lower the energy of the itinerant electrons with an  $\{x, y\}$ - or  $\{xz, yz\}$ -type component.

The band structure of  $\text{ThRu}_2\text{Si}_2$  in Fig. 2 is calculated by assuming the No. 136 space group, but using the same lattice as that for the No. 139 space group. The spin-orbit interaction is included; thus no degenerate bands appear even at the  $\Gamma$  point ( $E_g: \{xz, yz\}$  and  $E_u: \{x, y\}$  are the degenerated states, when the spin-orbit interaction is not included). The bands in the vicinity of the Fermi level consist of Ru- $d$  and Th- $d$  components. The antiferro  $O_{xy}$ -type potential yields the matrix element between the  $\{x, y\}$  (or  $\{xz, yz\}$ )-type orbitals of the bands reducing the system energy. This scenario is just a speculation so far. However, once it is ordered, the conduction bands are strongly modified, then the carrier number would be reduced, as observed in the HO phase.<sup>27,28)</sup> A realistic band structure calculation of the HO phase is now in progress by using an LDA+ $U$  method.

The Coulomb interaction between the local  $5f$  quadrupole moment pair ( $O_{\pm xy}^A - O_{\mp xy}^B$ ) and the conduction band pair  $[(x \pm y)^A - (x \mp y)^B]$  can be written as  $-I(S \cdot s)$ , where  $S$  ( $s$ ) is an Ising-type spin representing  $+$  or  $-$  for quadrupole states (orbital states). If it is written as  $(S \cdot s)$ , the Kondo behavior could be expected. It is emphasized that the coupling  $I$  is independent of lattice distortions in this system. This coupling survives even at high temperatures, escaping from the disturbance of the lattice vibrations.

Now, the CEF splitting of  $5f^2$  electrons in the local picture is worth mentioning. It is not necessary to consider a degenerated ground state. Due to the change in local symmetry from  $D_{4h}$  to  $D_{2h}$ , the off-diagonal term emerges to lower the ground state, even if it is a singlet. Then, the magnetic susceptibility shows a cusp at  $T_0$ , as observed.<sup>29)</sup>

Antiferro-quadrupole ordering was discussed previously, by using three singlet states<sup>8)</sup> or a doublet ground state.<sup>11)</sup> Although they did not distinguish  $O_{xy}$ -type quadrupole from  $O_{x^2-y^2}$ -type quadrupole, taking  $O_{xy}$ -type quadrupole and forming antiferro coupling are especially important. One simple way for realizing such a quadrupole ordering may be to use  $\Gamma_5^\pm$  [ $E_g, xz(+), yz(-)$ ] doubly degenerate states in a form or another. Detailed discussions will be given in a separate paper.

Quite recently, two characteristic phenomena which seem to have a potentiality to unveil the form of HO were reported. One is resistivity measurements at low temperature region: the resistivity along the  $a$ -axis,  $\rho_a$ , exhibits a non-Fermi liquid behavior,  $\rho_a = \rho_{0a} + A_a T$ , and that along the  $c$ -axis shows the usual Fermi liquid behavior,  $\rho_c = \rho_{0c} + A_c T^2$ .<sup>30)</sup> The other phenomenon is neutron scattering measurement: it was observed in a neutron scattering experiment at  $P = 0.67$  GPa that an inelastic mode of magnetic fluctuations  $m_z$  at the wave vector at  $\mathbf{q} = (1, 0, 0)$  exists only in the HO phase, not in the normal nor the large moment antiferromagnetic phases<sup>19)</sup>. The former unusual phenomenon can be explained, if the Nambu–Goldstone mode in the HO state is a linear combination of quadrupolar fluctuations of  $Q_{xz}$  and  $Q_{yz}$  representing  $\Gamma_5^\pm$  state in tetragonal symmetry.<sup>31)</sup>

The key point is still how to observe the symmetry lowering in HO. As many unsuccessful attempts have been tried, improvements in the sensitivity of the method are obviously necessary. For example, careful check of the anisotropy of the magnetization in the basal plane above  $T_0$  will be worthwhile as short correlations of the quadrupole pair ( $O_{xy} - O_{-xy}$ ) must enhance a 4-fold component. Refinement in resonant X-ray measurements (well-known tool in the proof of quadrupole ordering) is required as the last result fails to reveal any quadrupole order.<sup>32)</sup> Further experimental efforts are greatly encouraged.

In conclusion, the second-order transition to the HO phase in  $\text{URu}_2\text{Si}_2$  does not require any type of lattice distortions.  $\text{URu}_2\text{Si}_2$  is never a unique material without structural distortions, but it is the heart of materials in strongly correlated electron systems, where only electrons themselves bring about spatial symmetry lowering.

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