






Broken time-reversal symmetry in the cubic skutterudite-like superconductor $Y_3Ru_4Ge_{13}$ A. Kataria ¹, J. A. T. Verezhak,² O. Prakash,³ R. K. Kushwaha ¹, A. Thamizhavel,³ S. Ramakrishnan ⁴,
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The microscopic properties of the superconducting cubic skutterudite-like material $Y_3Ru_4Ge_{13}$ are investigated using muon spin relaxation and rotation (μ SR) measurements. Zero-field μ SR measurements reveal the presence of a spontaneous internal field with a magnitude of $\simeq 0.18$ mT below the superconducting transition temperature, indicating broken time-reversal symmetry in the ground state. In line with previous experiments, transverse-field μ SR measurements are consistent with a fully developed superconductivity gap in $Y_3Ru_4Ge_{13}$. Our observations point towards the relevance of electronic correlations beyond electron-phonon coupling as the superconducting origin and indicate that spin-orbit coupling is likely not the key driving force behind the spontaneous breaking of time-reversal symmetry in this system.

DOI: [10.1103/PhysRevB.108.214512](https://doi.org/10.1103/PhysRevB.108.214512)**I. INTRODUCTION**

Breaking additional symmetries, such as time-reversal or rotational symmetry, in a superconductor is a defining feature of unconventional superconductivity and lies outside the scope of BCS theory [1,2]. In the superconducting state, the breaking of the time-reversal symmetry (TRS) is manifested by the spontaneous presence of a magnetic field around inhomogeneities below the superconducting transition temperature. This additional symmetry breaking in superconductivity can contribute to novel properties and rich and exciting physics, which motivates a detailed investigation of the ground state and attracts tremendous research attention. Studying such broken symmetries in superconductors is also crucial for understanding the pairing mechanism and the structure of the order parameter.

The phenomenon of broken TRS has been extensively investigated in cage-type superconducting systems due to their intriguing properties and complex structure. In this family, $PrOs_4Sb_{12}$ is reported as the first strongly correlated heavy-fermion superconductor with broken TRS, a complex order parameter, and multigap nodal structure [3,4]. Further studies of sister compounds with doping suggest the importance of Pr single-ion excitations, Pr-Pr interactions, and spin-orbit coupling (SOC) for TRS breaking in the superconducting state [5–13]. In contrast to this, $PrRu_4Sb_{12}$ exhibits no signs of broken TRS [7,14]. Similarly, other cage compounds PrA_2M_{20} , (where $A = V, Ir, Rh$ and $M = Al, Zn$), do not show evidence of broken TRS, further complicating the understanding

of TRS breaking in cage compounds [15–17]. Meanwhile, tetragonal cage compounds of the form $R_5Rh_6Sn_{18}$ ($R = Sc, Y, Lu$) exhibit unconventional superconducting characteristics, including broken TRS, an anisotropic gap structure, and rattling vibrations [18–21]. Yet again, the ruthenate composition $R_5Ru_6Sn_{18}$ shows BCS superconductivity and preserved TRS [22]. Given this complex behavior and the limited number of studies on the cage structure family revealing different results, there is a huge gap that has to be overcome to be able to understand the superconducting order parameter, the mechanisms behind TRS breaking, and the pairing mechanism. The role of crystal structure, electron correlation, SOC, and rattling modes on the unconventional properties of cage-type superconductors remains largely unexplored, motivating a thorough examination of the ground-state properties.

In this context, the cubic skutterudite-like family $R_3M_4X_{13}$, where R represents a rare-earth metal, M is a transition metal, and X is a group-14 element, provides an exciting platform for investigating superconductivity with low rattling modes [23,24]. Within this family, the weakly correlated stannides exhibit unconventional properties with a BCS coupling mechanism and a strong interaction with crystal structure [25–28]. Furthermore, recent investigations on the low-carrier-density superconductor $Lu_3Os_4Ge_{13}$ have revealed a multigap superconducting state with broken TRS, indicating the possibility of an electron-electron interaction as a pairing mechanism [29,30]. Therefore, the sister compound $Y_3Ru_4Ge_{13}$, with a transition temperature T_c of 2.8 K, low carrier density, low SOC, and few or no rattling modes, offers a promising avenue to understand the effect of carrier concentration and SOC on the pairing mechanism of the cubic skutterudite family [31–33]. In this paper, we investigate the microscopic su-

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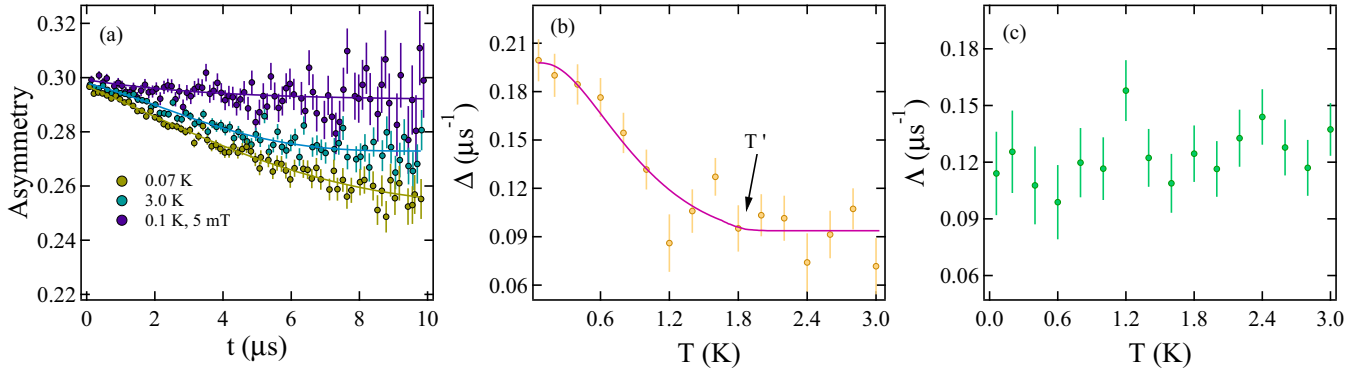


FIG. 1. (a) ZF asymmetry spectra at 0.07 K ($T < T_c$) and 3.0 K ($T > T_c$) with LF asymmetry spectra at 0.1 K and 5 mT, where solid lines represent the respective fit. (b) The relaxation rate Δ [see Eq. (2)] increases at a temperature T' below the superconducting T_c (see the text for details), while the rate Λ in Eq. (1) stays approximately constant (c).

perconducting properties of the low-carrier-density system $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ via muon spin rotation/relaxation (μSR) measurements. Zero-field (ZF) μSR measurements suggest broken TRS, as indicated by an increased relaxation rate below the superconducting transition temperature. As the associated magnetic field scale is even slightly larger than in the sister compound $\text{Lu}_3\text{Os}_4\text{Ge}_{13}$ [29] with stronger SOC, we conclude that SOC is not essential for stabilizing a TRS-breaking superconducting order parameter in these systems.

II. EXPERIMENTAL DETAILS

The single crystals of $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ used for μSR measurements have already been characterized and studied in previous work [32,33]. Phase purity and superconducting properties with $T_c = 2.8$ K were investigated using resistivity, magnetization, and specific-heat measurements. μSR measurements have been performed on single crystals with the incident muon beam oriented along the [110] axis at the ISIS Neutron and Muon Pulsed Source, Rutherford Appleton Laboratory, U.K. The methodology and instruments are detailed in Refs. [34,35]. In the longitudinal configuration, measurements were performed with both an applied field (LF) and zero field (ZF), to infer the presence of an internal field in the superconducting state, while in the transverse-field (TF) measurements, a magnetic field was applied perpendicular to the incident muon beam direction to investigate the gap structure in the vortex state of a superconductor.

III. RESULT

A. Zero-field μSR

In ZF- μSR measurement, the time-domain asymmetry spectra were measured in regular intervals from above the superconducting transition temperature to the lowest temperature. The measured ZF asymmetry spectra above and below T_c are shown in Fig. 1(a). The observed small relaxation and absence of a muon precession signal rule out the possibility of any long-range magnetically ordered in this sample. The asymmetry spectra are analyzed by considering relaxation due to static, randomly oriented nuclear moments, and fast fluctuation due to electronic moments. This can be expressed by the exponentially damped Gaussian Kubo-Toyabe (KT) function

with a flat background [36],

$$A(t) = A_0 G_z(t) \exp(-\Lambda t) + A_{\text{bg}}, \quad (1)$$

where

$$G_z(t) = \frac{1}{3} + \frac{2}{3}(1 - \Delta^2 t^2) \exp\left(-\frac{\Delta^2 t^2}{2}\right) \quad (2)$$

is the Gaussian Kubo-Toyabe function with Δ encoding the relaxation due to static or quasistatic local fields [37]; A_0 and A_{bg} are the initial and background asymmetries, respectively, and Λ represents the electronic relaxation rate. For the asymmetry spectra fitting, A_0 and A_{bg} are kept as temperature-independent global parameters, while the relaxation rates Δ and Λ serve as free parameters, demonstrating a temperature variation, as shown in Figs. 1(b) and 1(c). Λ remains temperature independent, while Fig. 1(b) shows the increase in Δ below the superconducting transition temperature corresponding to an increase in the typical local magnetic fields [31]. Further, an applied longitudinal field of 5 mT was sufficient to decouple the muons from the decaying channel [shown in Fig. 1(a)], indicative of the static or quasistatic nature of the field and broken TRS in the superconducting state of $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$. A similar increase in Δ , an indicative of broken TRS is also observed in $\text{PrOs}_4\text{Sb}_{12}$ [3], $\text{PrPt}_4\text{Ge}_{12}$ [5], LaNiGa_2 [38], and many Re-based superconductors [39–43]. The change in Δ [Fig. 1(b)] channel suggests a Gaussian-type field distribution at the muon-stopping site, which is different from other known TRS-breaking superconductors such as Sr_2RuO_4 where a change in the Λ channel indicating Lorentzian field distribution is observed [44].

The measured change in the relaxation rate Δ below T_c is $0.11 \mu\text{s}^{-1}$. The magnitude of the internal field associated with this change can be estimated by using the expression [45]

$$B_{\text{int}} = \sqrt{2} \frac{\Delta}{\gamma_\mu}, \quad (3)$$

where $\gamma_\mu = 2\pi \times 135.5$ MHz/T is the muon gyromagnetic ratio. The estimated value of $B_{\text{int}} \simeq 0.18(3)$ mT is comparable to reported values of 0.12 mT for filled skutterudite $\text{PrOs}_4\text{Sb}_{12}$ [3], 0.116 mT for Re_2Hf [43], and 0.11 mT for $\text{Lu}_3\text{Os}_4\text{Ge}_{13}$ [29]. However, this value is considerably larger than other known TRS-breaking superconductors.

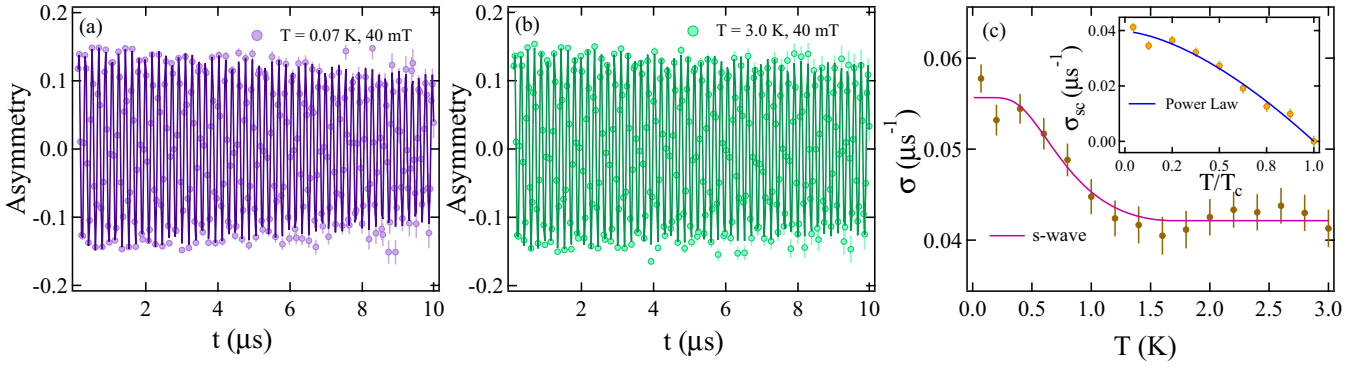


FIG. 2. TF- μ SR asymmetry spectra at 40 mT (a) in the superconducting state (at $T = 0.07$ K) and (b) in the normal state, $T = 3.0$ K, with the solid lines representing the fitting using Eq. (4). (c) The muon relaxation rate σ variation with temperature fitted with the s -wave model with the inset showing the fitting of σ_{sc} with temperature using the power law.

Furthermore, in Fig. 1(b), it is observed that the actual increase in Δ occurs at a temperature of $T' = 1.7$ K, which is below the resistive superconducting transition temperature of $T_c = 2.8$ K with a transition width of 0.4 K [33]. Measurement of the ac susceptibility of Ref. [31] indicated the onset of the superconducting transition at a temperature of 2.2 K in the same sample of $Y_3Ru_4Ge_{13}$. Both of these values are significantly different from the T' observed for our sample. The possible reason behind this discrepancy is discussed in Sec. IV.

B. Transverse-field μ SR

The superconducting gap structure of $Y_3Ru_4Ge_{13}$ can be evaluated from the TF- μ SR measurements. Asymmetry spectra were measured in a field-cooled protocol up to a temperature above T_c at fixed intervals. Figures 2(a) and 2(b) show the TF asymmetry spectra above and below T_c under an applied magnetic field of 40 mT. No significant changes can be found in the two asymmetry spectra. The time-domain TF asymmetry spectra were further analyzed by fitting the Gaussian-damped oscillatory relaxation function [46,47],

$$A(t) = A_1 \exp\left(-\frac{1}{2}\sigma^2 t^2\right) \cos(\gamma_\mu B_1 t + \phi) + A_2 \cos(\gamma_\mu B_2 t + \phi), \quad (4)$$

where A_1 and A_2 are the initial asymmetries of the sample and background, respectively. σ is the Gaussian relaxation rate, ϕ is the initial phase, and B_1 and B_2 are the local magnetic fields sensed by the muons in the sample and the sample holder (background), respectively. The solid line represents the fitting of the spectra in Figs. 2(a) and 2(b). The extracted variation of σ with temperature is shown in Fig. 2(c), revealing only a small change in σ below T_c within the experimental resolution of the time window of μ SR. The temperature dependence of σ has been modeled (solid red line) using a single nodeless s -wave model in the clean limit [48], providing a superconducting energy gap $\Delta(0) = 0.17(1)$ meV. The obtained value differs from that reported in other measurements, including the specific heat (0.21 meV) [32] and tunnel-diode oscillator measurements ($\simeq 0.40$ meV) [30].

The small change in σ vs T below T_c suggests that the vortex lattice has only a minor relaxation contribution, σ_{sc} . An upper bound can be estimated using $\sigma_{sc} = (\sigma^2 - \sigma_{dip}^2)^{1/2}$,

where σ_{dip} is the relaxation due to the nuclear moments measured in the normal state. The weak temperature dependence of the nuclear relaxation rate is also observed in $La_2(Cu_{1-x}Ni_x)_5As_3O_2$ similar to our case [49]. We fit the temperature variation of σ_{sc} with $T_{c,\mu SR} = 1.6$ K using the power law (see Refs. [50,51])

$$\sigma_{sc} = \sigma_{sc}(0) \left[1 - \left(\frac{T}{T_{c,\mu SR}} \right)^N \right] \quad (5)$$

for $T \leq T_{c,\mu SR}$. The fitting yields $N = 1.56(5)$, much less than the value 4 expected for BCS superconductors. In dirty d -wave superconducting systems, it is suggested that $N \sim 2$ [49,52,53]. The value for N we obtain points towards a nodal superconducting order parameter, which is surprising and in contrast with the microscopic model. Thus, these small temperature variations of σ and low superconducting gap values with relatively large uncertainties are inadequate to accurately determine the superconducting gap symmetry. We leave this for future work.

Furthermore, considering the vortex lattice system of a type-II superconductor, an estimation of London penetration depth $\lambda(0)$ can be extracted by using the expression [54]

$$\frac{\sigma_{sc}(T)}{\gamma_\mu} = 0.0609 \frac{\Phi_0}{\lambda^2(T)}. \quad (6)$$

The calculated lower bound is $\lambda(0) \geq 1.6$ μ m. Moreover, using London's equation, the lower limit of the superconducting carrier density n can be extracted via the relation $\lambda^2 = m^*/\mu_0 n e^2$, where m^* is the effective mass (taken from Ref. [32]); the resulting bound $n \leq 1.7 \times 10^{25}$ carriers/ m^3 justifies the low-carrier-density limit of the system.

According to Uemura's classification system, which is based on the ratio of the Fermi temperature to the superconducting transition temperature T_F/T_c , $Y_3Ru_4Ge_{13}$ is categorized with other unconventional superconductors. To obtain an estimate, we assume a three-dimensional (3D) spherical Fermi surface and evaluate the Fermi temperature T_F from [50]

$$k_B T_F = \frac{\hbar^2}{2} (3\pi^2)^{2/3} \frac{n^{2/3}}{m^*}, \quad (7)$$

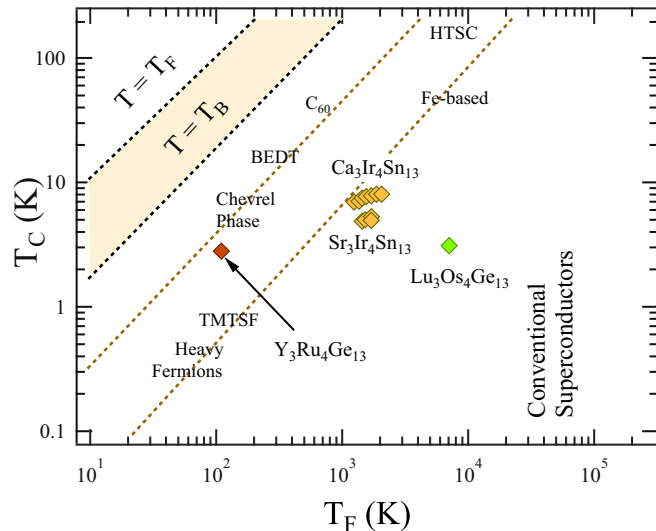


FIG. 3. Uemura plot of unconventional superconductors including $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ and other stannides of the family such as $\text{Ca}_3\text{Ir}_4\text{Sn}_{13}$ and $\text{Sr}_3\text{Ir}_4\text{Sn}_{13}$ at different pressure [25,55,56], where T_B is the Bose-Einstein condensation temperature.

where n is the carrier density, and m^* is the effective mass and resultant $T_F = 173(25)$ K. Figure 3 shows that $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ (red diamond) is positioned well inside the unconventional superconductor band. Further, the yellow diamonds represent $(\text{Ca}/\text{Sr})_3\text{Ir}_4\text{Sn}_{13}$ at different pressures, and the green diamond represents $\text{Lu}_3\text{Os}_4\text{Ge}_{13}$.

A small change in σ below T_c or no significant muon relaxation in the superconducting state has previously been observed in the Cu- or Sr-doped topological insulator Bi_2Se_3 [57,58], $\text{Ru}_{0.75}\text{Rh}_{0.25}\text{As}$ [59], noncentrosymmetric YPtBi [60], and most recently in CaSn_3 [61]. The common features among the materials mentioned above and $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ are low carrier density and nontrivial band topology. In the Heusler alloy YPtBi and A15 compound CaSn_3 , nontrivial topological band structures have recently been recognized [62]. Moreover, theoretical band-structure calculations from the topological material database also suggest $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ as a symmetry-enforced semimetal [63–66]. Additionally, a peak effect and anomalous susceptibility behavior with the magnetic field are observed for $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ [30]. These observations suggest weak vortex pinning at a low magnetic field and hint toward the possibility of a complex vortex lattice and field-induced disordering of the vortex lattice [30]. However, the precise mechanism governing the unconventional vortex state in low-carrier topological materials remains elusive. Further investigation in this direction is imperative to understand the intricacies of the vortex-state phenomenon.

IV. DISCUSSION AND CONCLUSION

Finally, we discuss the implications of our findings for the microscopic pairing state and mechanism in $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$. While it has the same symmetries as $\text{Lu}_3\text{Os}_4\text{Ge}_{13}$ —both exhibit the space group No. 223 ($Pm\bar{3}n$) with the cubic point group O_h —the expected strength of SOC ($\propto Z^4$) is much weaker in $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$, given the significantly lower

atomic numbers of the involved atoms; more precisely, first-principles calculations indicate that the states around the Fermi level in $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ derive mostly from Ge ($Z = 32$) [67] and those in $\text{Lu}_3\text{Os}_4\text{Ge}_{13}$ also exhibit significant contributions from Os ($Z = 76$) [68]. Yet, the key observation of TRS breaking setting in at a temperature below the resistive superconducting transition is common in the present and our previous [29] μSR study of $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ and $\text{Lu}_3\text{Os}_4\text{Ge}_{13}$, respectively. In combination with the similar strength of the internal field B_{int} in these compounds, this strongly indicates that SOC is not the key driving force behind the TRS breaking.

By construction, the symmetry-based discussion of the possible pairing states for $\text{Lu}_3\text{Os}_4\text{Ge}_{13}$ presented in Ref. [29] applies here as well: If the superconducting state is reached by a single phase transition, the order parameter must transform under a two- or three-dimensional irreducible representation of O_h to allow for broken TRS. As a result of the complex structure of the group O_h , there are ten distinct TRS-breaking superconducting candidate phases; generically, all of them are expected to exhibit nodes. As pointed out above, our TF- μSR data are consistent with a full gap, but the small variation of σ might not be enough to rule out the presence of nodes. Since, however, more sensitive tunnel-diode oscillator measurements also indicate a fully gapped superconducting phase [30], alternative scenarios leading to a superconducting order parameter with broken TRS and a full gap should be discussed. Given that the signs of broken TRS in both $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ and $\text{Lu}_3\text{Os}_4\text{Ge}_{13}$ emerge at a temperature T' that is noticeably smaller than the resistive T_c , the most plausible scenario is that there are two superconducting transitions—at the first one, a (nodal or fully gapped) state sets in while only at a lower temperature, $T' < T_c$, a secondary order parameter appears, which, e.g., due to a nontrivial relative complex phase, breaks TRS; at least below T' , the superconducting order parameter has a full gap. Such a TRS-breaking complex phase can arise due to “frustrated” interactions (see Refs. [69–71] for example). Irrespective of which of these scenarios are realized, we expect electronic interactions beyond electron-phonon coupling to be present in order to stabilize the TRS-breaking superconducting phase below T' , making $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ a promising candidate for unconventional superconductivity.

In summary, the microscopic properties of the superconducting cubic skutterudite-like material $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$ were investigated using muon spin rotation and relaxation measurements. Zero-field μSR results suggest TRS breaking in the superconducting ground state, similar to its Lu sister compound and many other skutterudite compounds. A significant local internal magnetic field of magnitude $\simeq 0.18(3)$ mT is observed in the superconducting state. TF- μSR measurements are consistent with a fully gapped superconducting order parameter. Our findings indicate that SOC is likely not the key driving force for TRS-breaking superconductivity and that pairing interactions beyond the conventional electron-phonon coupling are at play. Nonetheless, there are still many open questions that deserve further investigations, such as determining the form of the superconducting order parameter and understanding the nature of the vortex state in $\text{Y}_3\text{Ru}_4\text{Ge}_{13}$. More generally, our work emphasizes that low-carrier-concentration systems and cage compounds are an intriguing playground for exotic superconducting physics.

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