Broken time-reversal symmetry probed by muon spin relaxation in the caged type superconductor $Lu_5Rh_6Sn_{18}$

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The superconducting state of the caged type compound $Lu_5Rh_6Sn_{18}$ has been investigated by using magnetization, heat capacity, and muon spin relaxation or rotation (μSR) measurements, and the results interpreted on the basis of the group theoretical classifications of the possible pairing symmetries and a simple model of the resulting quasiparticle spectra. Our zero-field μSR measurements clearly reveal the spontaneous appearance of an internal magnetic field below the transition temperature, which indicates that the superconducting state in this material is characterized by broken time-reversal symmetry. Further, the analysis of the temperature dependence of the magnetic penetration depth measured using the transverse-field μSR measurements suggests an isotropic s-wave character for the superconducting gap. This is in agreement with the heat capacity behavior, and we show that it can be interpreted in terms of a nonunitary triplet state with point nodes and an open Fermi surface.

The understanding of the pairing mechanism in unconventional superconductors in strongly correlated electron systems is a major theoretical challenge [1,2]. In conventional "s-wave" superconductors, only the gauge symmetry is broken. If the pairing is not conventional, then some other symmetries of the Hamiltonian may be broken below the superconducting transition. Symmetries which might be broken include the lattice point and translation group operations and spin rotation symmetries, in addition to the global gauge symmetry that is responsible for the Meissner effect, flux quantization, and Josephson effects. The nature of the broken symmetry in the pairing state is reflected in the symmetry properties of the order parameter. Superconductors whose crystal structure features a center of inversion can be classified via the parity of the Cooper pair state: The spin-singlet pair state ($S = 0$) corresponds to an orbital pair wave function $\psi(k) \sim \psi(-k)$ with even parity [i.e., $\Delta(k) = \Delta(-k)$]; the spin-triplet state (total spin $S = 1$) has a superconducting order parameter with odd parity [$\psi(k) \sim -\psi(-k)$] [3]. A few compounds have been reported to be spin-triplet superconductors, for example, the $4d$-electron system Sr_2RuO_4 [4–6], and the $5f$-electron systems UPt_3 [7] and UNi_2Al_3 [8].

Broken symmetry can modify the physics of a system, which results in novel and uncommon behavior. Superconductivity is one of the finest illustrations of a symmetry breaking phenomenon. A particularly interesting case is time-reversal symmetry (TRS) breaking. This is rare and has only been observed directly in a few unconventional superconductors, e.g., Sr_2RuO_4 [4,9], UPt_3 [7], (U;Th)Be$_{13}$ [10], (Pr;La)(Os;Ru)$_4$ Sb$_{12}$ [11], PrPt$_4$ Ge$_{12}$ [12], LaNiC$_2$ [13], LaNiGa$_2$ [14], and Re$_6$Zr [15]. A direct manifestation of

broken TRS is the appearance of spontaneous weak magnetic fields, detected in these systems by zero-field muon spin relaxation (ZF-μSR). ZF-μSR is useful in the search for TRS breaking fields; The presence of such fields limits the possible superconducting states and the associated pairing symmetry. For example, TRS is a prerequisite for any state with a one-dimensional representation (singlet, triplet, or admixed), and its breaking is associated with special kinds of states which have a degenerate representation. The presence of two or more nearly degenerate superconducting phases naturally leads to a spatially inhomogeneous order parameter near the resulting domain walls; this creates spontaneous supercurrents and hence magnetic fields near those regions. Another possible origin of TRS breaking fields is from intrinsic magnetic moments due to spin polarization (for spin-triplet pairing) and the relative angular momentum of the Cooper pairs [2]. Specifically, one can prove, using group-theoretical arguments [14], that nonunitary triplet pairing (thought to occur in noncentrosymmetric LaNiC$_2$ [13] and centrosymmetric LaNiGa$_2$ [14]) leads to a small bulk magnetization M. The latter acts as a subdominant order parameter of the superconducting instability, i.e., it grows only linearly with decreasing temperature, $|M| \sim |T_c - T|$ [14]. Recently, the size of this magnetization has been obtained within a nonunitary triplet pairing model of Sr_2RuO_4 [16].

The possibility of singlet-triplet pairing in noncentrosymmetric superconductors makes them prime candidates to exhibit TRS breaking. In spite of this, it is well established theoretically [17] and experimentally [18] that singlet-triplet mixing does not necessarily imply broken TRS. On the other hand, broken TRS has been observed in Re$_6$Zr [15], where we expect a strong singlet-triplet admixture. In contrast, for LaNiC$_2$, symmetry analysis implies that the superconducting instability is of a purely triplet type, with a spin-orbit coupling that is comparatively weak and with mixing of singlet and triplet pairing being forbidden by symmetry [17].

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Caged type structures have received considerable attention due to their fascinating properties [19]. Three cage-type compounds have been comprehensively studied over the past decade as "rattling-good" materials: Ge/Si clathrates, filled skutterudites (RT_4X_{12}), and β-pyrochlore oxides (AOs_2O_6) [19]. Typically, they possess three-dimensional skeletons surrounding large atomic cages, inside of which reasonably small atoms are situated and can "rattle" with large atomic excursions due to the virtual size inconsistency, weak structural coupling, and strong electron-phonon (rattler) coupling, leading to a considerable anharmonicity for the rattling vibration. For instance, rattling of the A atoms in the OsO_6 cages induces extremely strong-coupling superconductivity in AOs_2O_6 [20]. A strong interplay between the quadrupolar moment and superconductivity has been pointed out in RT_4X_{12} [21] and RT_2X_{20} [22]. $R_5Rh_6Sn_{18}$ (R = Sc, Y, Lu), which can also be categorized as cage-type compounds, exhibit superconductivity with a transition temperature $T_c = 5$ K (Sc), 3 K (Y), and 4 K (Lu) [23]. These compounds have a tetragonal structure with the space group $I4_1/acd$ and $Z = 8$, where R occupies two sites of different symmetry [24]. In this Rapid Communication, we report on ZF-μSR and transverse-field (TF) μSR measurements for $Lu_5Rh_6Sn_{18}$. The results unambiguously reveal the spontaneous appearance of an internal magnetic field in the superconducting (SC) state, providing clear evidence for broken time-reversal symmetry.

Single crystals of $Lu_5Rh_6Sn_{18}$ were grown by a conventional Sn-flux method in the ratio of Lu:Rh:Sn = 1:2:20. A detailed discussion on the crystal growth can be found in Ref. [23]. Well defined Laue diffraction spots indicated the good quality of the single crystals with a typical size $3 \times 3 \times 3$ mm^3. Powder x-ray diffraction patterns were indexed as the $Lu_5Rh_6Sn_{18}$ phase with the space group $I4_1/acd$ [23]. The magnetic measurements were performed using a Quantum Design magnetic property measurement system (MPMS). Specific heat measurements were performed down to 500 mK by a relaxation method calorimeter [Quantum Design physical property measurement system (PPMS) equipped with a ^{3}He refrigerator].

Muon spin relaxation (μSR) experiments were carried out on the MUSR spectrometer at the ISIS pulsed muon source of the Rutherford Appleton Laboratory, UK [25]. The μSR experiments were conducted in zero-field (ZF), longitudinal-field (LF), and transverse-field (TF) modes. A high quality single crystal of $Lu_5Rh_6Sn_{18}$ was mounted on a sample plate made of 99.995% silver, which was placed in a dilution refrigerator with a temperature range of 100 mK to 4.5 K. Using an active compensation system, the stray magnetic fields at the sample position were canceled to a level of 1 μT. TF-μSR experiments were performed in the superconducting mixed state in an applied field of 400 G, well above the $\mu_0H_{c1} = 20$ G of this material. Data were collected in the field-cooled mode, where the magnetic field was applied above the superconducting transition and the sample was then cooled down to a base temperature. Muon spin relaxation is a dynamic method to resolve the type of pairing symmetry in superconductors [26]. The mixed or vortex state in the case of type-II superconductors gives rise to a spatial distribution of local magnetic fields, which demonstrates itself in the μSR signal through a relaxation of the muon polarization.
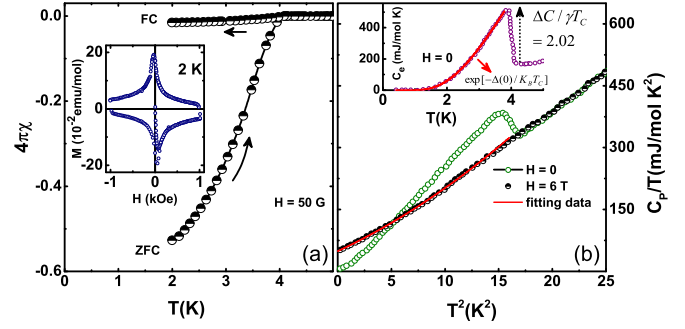

FIG. 1. (Color online) (a) The temperature dependence of the dc magnetic susceptibility of $Lu_5Rh_6Sn_{18}$. The inset in (a) shows the isothermal field dependence of magnetization at 2.0 K. (b) shows the C_P/T vs T^2 curve. The solid line shows the fit (see text). The inset in (b) shows the temperature dependence of electronic specific heat C_e under zero field after subtracting the lattice contribution for $Lu_5Rh_6Sn_{18}$.

Magnetization measurements indicate that $Lu_5Rh_6Sn_{18}$ is a bulk superconductor with a superconducting transition temperature $T_c = 4.0 \pm (0.1)$ K, as shown in Fig. 1(a). Below T_c, the low-field $\chi(T)$ shows a robust diamagnetic signal. The shielding volume fraction is $\sim$53% at 2 K. The inset of Fig. 1(a) shows the magnetization $M(H)$ curve at 2 K, which is typical for type-II superconductivity. Resistivity [$\rho(T)$, not shown here] exhibits a very unusual temperature variation [27]. $\rho(T)$ is nearly independent of T down to about 120 K, and shows an increase on further cooling [27]. Figure 1(b) shows the $C_P(T)$ at $H = 0$ and 6 T. At 4.0 K a sharp anomaly is observed, indicating the superconducting transition which matches well with the $\chi(T)$ data. Since the normal-state specific heat was found to be invariant under external magnetic fields, the normal-state electronic specific heat coefficient γ and the lattice specific heat coefficient β were deduced from the data in a field of 6 T by a least-squares fit of the C_P/T data to $C_P/T = \gamma + \beta T^2 + \delta T^4$. The least-squares analysis of the 6 T data provides a Sommerfeld constant $\gamma = 48.10 \pm (0.5)$ mJ/(mol K^2), $\delta = 0.32 \pm (0.03)$ mJ/(mol K^6), and the Debye temperature $\Theta_D = 157 \pm (2)$ K. We obtained the specific heat jump $\Delta C_P(T_c) = 397 \pm (3)$ mJ/(mol K) and $T_c = 4.0 \pm (0.2)$ K, which yields $\Delta C/\gamma T_c = 2.06 \pm (0.03)$. From the exponential dependence of C_e, as shown in the inset of Fig. 1(b), we obtained $2\Delta(0)/k_BT_c$ to be $4.26 \pm (0.04)$. Because this value is relatively larger than that of the theoretical BCS limit of a weak-coupling superconductor (3.54), this compound can be categorized as a strong-coupling superconductor [28].

Figures 2(a) and 2(b) show the TF-μSR precession signals above and below T_c with an applied field of 400 G (well above H_{c1}). Below T_c the signal decays with time due to an inhomogeneous field distribution of the flux-line lattice. The TF-μSR asymmetry spectra were fitted using an oscillatory decaying Gaussian function,

$$G_{z1}(t) = A_1 \cos(2\pi\nu_1 t + \phi_1) \exp\left(\frac{-\sigma^2 t^2}{2}\right)$$
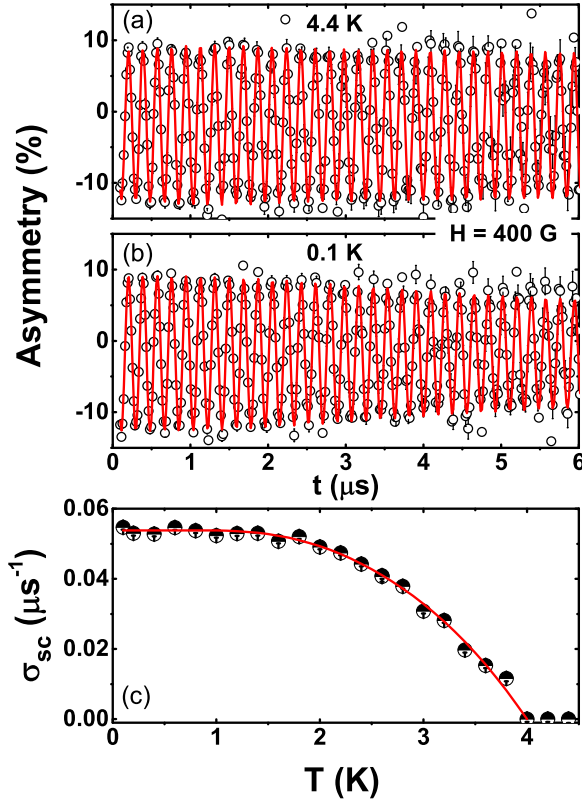$$+ A_2 \cos(2\pi\nu_2 t + \phi_2), \qquad (1)$$

FIG. 2. (Color online) The transverse-field muon time spectra (one component) for $Lu_5Rh_6Sn_{18}$ collected (a) at $T = 4.4$ K and (b) at $T = 0.1$ K in a magnetic field $H = 400$ G. (c) The temperature dependence of $\sigma_{sc}(T)$. The line is a fit to the data using an isotropic model [Eq. (2)].

where ν_1 and ν_2 are the frequencies of the muon precession signal and background signal, respectively, and ϕ_i ($i = 1,2$) are the initial phase offsets. The first term gives the total sample relaxation rate σ; there are contributions from both the vortex lattice (σ_{sc}) and nuclear dipole moments (σ_{nm}, which is assumed to be constant over the entire temperature range) below T_c [where $\sigma = \sqrt{(\sigma_{sc}^2 + \sigma_{nm}^2)}$]. The contribution from the
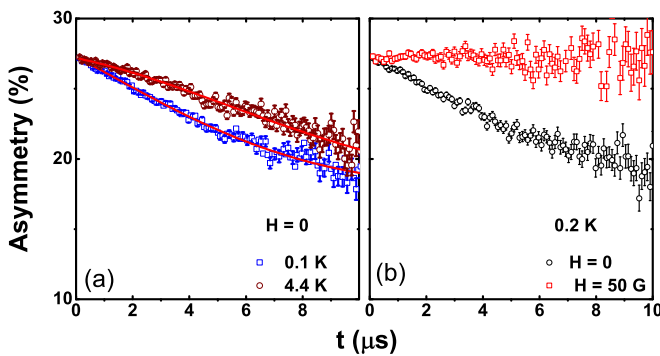
vortex lattice σ_{sc} was determined by quadratically subtracting the background nuclear dipolar relaxation rate obtained from spectra measured above T_c. As σ_{sc} is directly related to the magnetic penetration depth, the superconducting gap can be modeled by

$$\frac{\sigma_{sc}(T)}{\sigma_{sc}(0)} = \frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = 1 + 2\int_{\Delta(T)}^{\infty}\left(\frac{\delta f}{\delta E}\right)\frac{E dE d\phi}{\sqrt{E^2 - \Delta(T)^2}},$$

(2)

where $f = [1 + \exp(-E/K_B T)]^{-1}$ is the Fermi function [29]. The temperature dependence of the gap is approximated by the expression $\delta(T/T_c) = \tanh\{1.82[1.018(T_c/T - 1)]^{0.51}\}$ [30].

Figure 2(c) shows the T dependence of the σ_{sc}, which can be directly related to the superfluid density. From this, the nature of the superconducting gap can be determined. The data can be well modeled by a single isotropic gap of 0.75 ± 0.06 meV. This gives a gap of $2\Delta/k_B T_c = 4.4 \pm 0.02$, which is higher than the 3.53 expected for BCS superconductors. This is a further indication of the strong electron-phonon coupling in the superconducting state. $Lu_5Rh_6Sn_{18}$ is a type-II superconductor, assuming that roughly all the normal-state carriers (n_e) contribute to the superconductivity (i.e., $n_s \approx n_e$), and we have estimated the values of the effective mass of the quasiparticles $m^* \approx 1.32 m_e$ and the superconducting electron density $\approx 2.6 \times 10^{28}$ m^{-3}, respectively. More details on these calculations can be found in Refs. [31–33].

The time evolution of the ZF-μSR is shown in Fig. 3(a) for $T = 100$ mK and 4.4 K. In these relaxation experiments, any muons stopped on the silver sample holder give a time independent background. No signature of precession is visible, ruling out the presence of a sufficiently large internal magnetic field, as seen in magnetically ordered compounds. The only possibility is that the muon spin relaxation is due to static, randomly oriented local fields associated with the nuclear moments at the muon site. The ZF-μSR data are well described
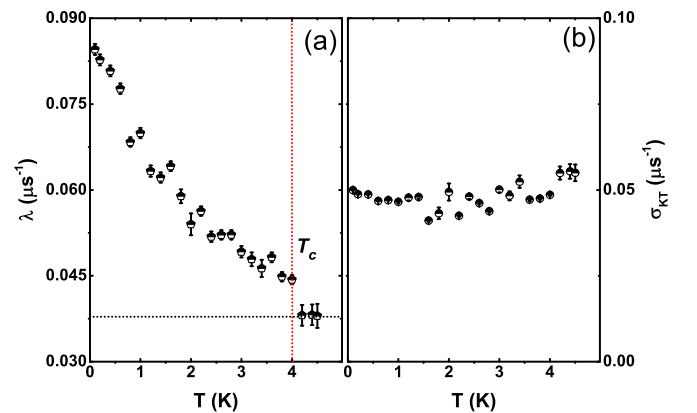

FIG. 3. (Color online) (a) Zero-field μSR time spectra for $Lu_5Rh_6Sn_{18}$ collected at 0.1 K (square) and 4.4 K (circle) are shown together with lines that are least-squares fits to the data using Eq. (3). These spectra, collected below and above T_c, are representative of the data collected over a range of T. (b) A LF-μSR time spectrum taken in an applied field of 5 mT at 0.2 K is also shown.

FIG. 4. (Color online) (a) The temperature dependence of the electronic relaxation rate measured in zero magnetic field of $Lu_5Rh_6Sn_{18}$ with $T_c = 4.0$ K is shown. The lines are guides to the eye. The extra relaxation below T_c indicates additional internal magnetic fields and, consequently, suggests the superconducting state has broken time-reversal symmetry. (b) The Kubo-Toyabe depolarization rate σ_{KT} vs temperature in zero field shows no temperature dependence.
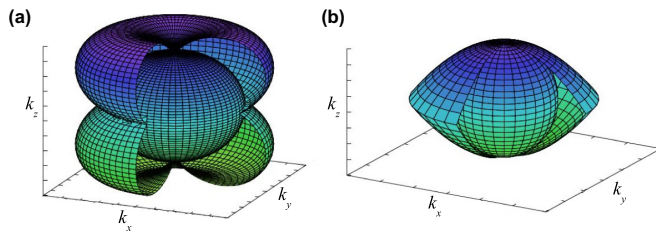
FIG. 5. (Color online) Nodal structure of the allowed (a) singlet and (b) triplet pairing states. See the Supplemental Material [37] for details.

by the damped Gaussian Kubo-Toyabe (KT) function,

$$G_{z2}(t) = A_1 G_{KT}(t) e^{-\lambda t} + A_{bg}, \qquad (3)$$

where $G_{KT}(t) = [\frac{1}{3} + \frac{2}{3}(1 - \sigma_{KT}^2 t^2) e^{\frac{-\sigma_{KT}^2 t^2}{2}}]$, λ is the electronic relaxation rate, A_1 is the initial asymmetry, and A_{bg} is the background. The parameters σ_{KT} [Fig. 4(b)], A_1, and A_{bg} are found to be temperature independent. It is remarkable that λ shows a significant increase [Fig. 4(a)] with an onset temperature of 4.0 ± 0.1 K, indicating the appearance of a spontaneous internal field correlated with the superconductivity. This observation provides unambiguous evidence that TRS is broken in the SC state of $Lu_5Rh_6Sn_{18}$. Such a change in λ has only been observed in superconducting Sr_2RuO_4 [4], $LaNiC_2$ [13], and $SrPtAs$ [34]. This increase in λ can be explained in terms of a signature of a coherent internal field with a very low frequency, as discussed by Luke *et al.* [4] for Sr_2RuO_4. This suggests that the field distribution is Lorentzian in nature, similar to Sr_2RuO_4. Considering a similar temperature dependence of λ in Sr_2RuO_4, $LaNiC_2$, $SrPtAs$, and $Lu_5Rh_6Sn_{18}$, we attribute this behavior of λ to the TRS breaking below T_c in $Lu_5Rh_6Sn_{18}$. A longitudinal magnetic field of just 50 G [Fig. 3(b)] removes any relaxation due to the spontaneous fields and is sufficient to fully decouple the muons from this relaxation channel. This in turn shows that the associated magnetic fields are in fact static or quasistatic on the time scale of the muon precession. These observations further support the broken TRS in the superconducting state of $Lu_5Rh_6Sn_{18}$. The increase in the exponential relaxation below T_c is 0.045 μs^{-1}, which corresponds to a characteristic field strength $\lambda/\gamma_\mu = 0.5$ G. This is about the same as we observed in the B phase of UPt_3 and Sr_2RuO_4 [7]. Theoretical estimates of the characteristic field strength in $Lu_5Rh_6Sn_{18}$ are still lacking, however, we expect them to be comparable to those in Sr_2RuO_4 and UPt_3 as the fields should arise from a similar mechanism.

Our main observation, namely, the breaking of TRS on entering the superconducting state, has important implications for the symmetry of pairing and for the quasiparticle spectrum. In short, a standard symmetry analysis [2,35] carried out under the assumption of strong spin-orbit coupling yields two possible pairing states, one with a $d + id$ character (singlet) and another one nonunitary (triplet). As shown in Fig. 5, both states are nodal: The singlet has a line node and two point nodes, and the triplet has two point nodes. At temperatures $T \ll T_c$, the thermodynamics of the singlet state would be dominated by the line node, yielding, for example, $C \sim T^2$ for the specific heat. Similarly, the triplet state would be dominated by the point nodes, which happen to be shallow (a result protected by symmetry) and therefore also lead to $C \sim T^2$ [36]. However, because of the location of the nodes in the triplet case, fully gapped behavior may be recovered depending on the topology of the Fermi surface. Moreover, some limiting cases of the triplet state correspond to regular, i.e., linear point nodes ($C \sim T^3$), as well as to a more exotic state with a nodal surface (gapless superconductivity, $C \sim T$). The allowed pairing states and their quasiparticle spectra are discussed in detail in the Supplemental Material [37]. We note that the theoretical analysis presented there is valid for any superconductor with D_{4h} point group symmetry, strong spin-orbit coupling, and broken time-reversal symmetry, and may therefore be applied, for example, to Sr_2RuO_4 [38] as well as $Lu_5Rh_6Sn_{18}$.

In conclusion, we have used both ZF-μSR and TF-μSR to investigate the superconductivity of the caged type tetragonal system $Lu_5Rh_6Sn_{18}$. The ZF-μSR measurements show a spontaneous field appearing at the superconducting transition temperature. The presence of spontaneous internal magnetic fields in our measurements suggests that a time-reversal symmetry breaking mixed symmetry pairing state does occur below T_c. TF-μSR measurements yield a magnetic penetration depth that is exponentially flat at low temperatures, and so our data can be fit to a single-gap BCS model. Symmetry analysis suggests either a singlet $d + id$ state with a line node or, alternatively, nonunitary triplet pairing with point nodes, which may be linear or shallow and can become fully gapped depending on the Fermi surface topology.

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