

Unconventional Superconductivity in La_7Ir_3 Revealed by Muon Spin Relaxation: Introducing a New Family of Noncentrosymmetric Superconductor That Breaks Time-Reversal Symmetry

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(Received 14 August 2015; published 30 December 2015)

The superconductivity of the noncentrosymmetric compound La_7Ir_3 is investigated using muon spin rotation and relaxation. Zero-field measurements reveal the presence of spontaneous static or quasistatic magnetic fields below the superconducting transition temperature $T_c = 2.25$ K—a clear indication that the superconducting state breaks time-reversal symmetry. Furthermore, transverse-field rotation measurements suggest that the superconducting gap is isotropic and that the pairing symmetry of the superconducting electrons is predominantly s wave with an enhanced binding strength. The results indicate that the superconductivity in La_7Ir_3 may be unconventional and paves the way for further studies of this family of materials.

DOI: [10.1103/PhysRevLett.115.267001](https://doi.org/10.1103/PhysRevLett.115.267001)

PACS numbers: 74.20.Mn, 74.70.Dd, 76.75.+i

To this day, the microscopic theory of superconductivity presented by Bardeen *et al.* [1] forms the basis to the theoretical and experimental understanding of the phenomenon of superconductivity. The conventional superconducting state is formed of electrons bound in spin-singlet Cooper pairs, with the attractive force mediated by the electron-phonon interaction. The Pauli principle requires that the total Cooper pair wave function is antisymmetric; thus, a spin-singlet state has even parity, and a spin-triplet pair has odd parity [2]. In centrosymmetric materials, parity is a good quantum number, and no mixing of pair states is allowed. Systems lacking a center of inversion exhibit a nonuniform lattice potential, which gives rise to an antisymmetric spin-orbit coupling [3,4]. This leads to a splitting of the Fermi surface into spin-up and spin-down contributions [5]. Consequently, Cooper pairs may form where the composite electrons belong to different parts of this split Fermi surface—a completely different situation from the conventional case, which leads to rich and interesting new physics. This includes the potential for a ground state that is an admixture of spin-singlet and spin-triplet superconducting channels [6], upper critical fields exceeding the Pauli limit [7], and nontrivial line or point nodes in the order parameter [2].

One of the best methods of detecting an unconventional ground state is muon spin rotation and relaxation (μSR) [8–10]. The flux line lattice (FLL) that is established in the mixed state of a type-II superconductor leads to a distinctive field distribution in the sample. The positive muon can be employed as an extremely sensitive probe of local

magnetic environments and directly measures the distribution of fields associated with the FLL. In this way, μSR is used to calculate the temperature evolution of the magnetic penetration depth λ and thus can determine the presence of nodes in the superconducting order parameter. The technique is also sensitive to the very small magnetic moments associated with the formation of spin-triplet electron pairs, and measurements in zero field provide one of the most unambiguous methods of detecting this broken time-reversal symmetry [11]. Time-reversal symmetry breaking (TRSB) is an extremely rare phenomenon, which has only been reported for a handful of unconventional superconductors: the candidate chiral p -wave superconductor Sr_2RuO_4 [12,13], the heavy fermion superconductors UPt_3 and $(\text{U,Th})\text{Be}_{13}$ [14–17], the filled skutterudites $(\text{Pr,Lu})(\text{Ru,Os})_4\text{Sb}_{12}$ [18,19], $\text{PrPt}_4\text{Ge}_{12}$ [20] and centrosymmetric LaNiGa_2 [21], and recently the caged-type superconductor $\text{Lu}_5\text{Rh}_6\text{Sn}_{18}$ [22]. μSR studies have been carried out on many other noncentrosymmetric superconductors (NCSs), including $\text{Ca}(\text{Ir,Pt})\text{Si}_3$ [23], $\text{La}(\text{Rh,Pt,Pd,Ir})\text{Si}_3$ [24–26], $\text{Mg}_{10}\text{Ir}_{19}\text{B}_{16}$ [27], and Re_3W [28]. No spontaneous magnetization has been observed in these materials, implying that the superconductivity in these systems occurs predominantly in a spin-singlet channel.

To date, the only noncentrosymmetric superconductors reported to break TRS are LaNiC_2 [29], Re_6Zr [30], and the locally noncentrosymmetric SrPtAs [31]. It is clearly important to search for new noncentrosymmetric structures that exhibit TRSB. Binary transition metal compounds with

the Th_7Fe_3 structure have been found to host a large number of superconducting combinations [32,33]. These materials crystallize in a hexagonal structure with space group $P6_3mc$. In this Letter, evidence for TRSB in a member of this family La_7Ir_3 is presented, a development that introduces a new system of potential materials in which to investigate the phenomenon of spin-triplet superconductivity.

A polycrystalline sample of La_7Ir_3 was prepared by arc melting stoichiometric quantities of La (99.9%, Alfa Aesar) and Ir (99.99%, Alfa Aesar) on a water-cooled copper hearth in a high-purity Ar atmosphere. Part of the sample was powdered and characterized on a PANalytical powder x-ray diffractometer. μSR measurements were carried out on the MuSR instrument at the ISIS pulsed muon and neutron spallation source. MuSR receives 40 pulses of 100% spin-polarized muons per second; these ensembles of muons are implanted into the sample and rapidly thermalize, sitting at interstitial positions in the crystal lattice. The muon spin precesses at the Larmor precession frequency before decaying with a lifetime of $2.2 \mu\text{s}$. The decay positron is emitted preferentially along the direction of the muon spin vector, and the emitted positrons are recorded by scintillation detectors positioned in circular arrays around the sample. The MuSR spectrometer can be rotated through 90° in order to change the geometry of the experiment; a full description of the two detector geometries is given in Ref. [10]. Stray fields at the sample position due to the Earth and neighboring instruments are canceled to within $1 \mu\text{T}$ using three sets of orthogonal coils and an active compensation system. The powdered La_7Ir_3 was mounted on a silver holder and placed in a dilution fridge, which operated in the temperature range $0.1 \leq T \leq 3.6 \text{ K}$. Silver is used as it gives a nondepolarizing background that can be easily accounted for during data analysis.

The powder x-ray diffraction (XRD) data showed that the sample had crystallized into the Th_7Fe_3 noncentrosymmetric structure with space group $P6_3mc$ and lattice parameters $a = 10.2376(3) \text{ \AA}$ and $c = 6.4692(3) \text{ \AA}$. No impurity phases were detected in the sample to within the sensitivity of the XRD technique. The sample was ground to a fine powder for the μSR experiments in a high-purity Ar atmosphere and transported in a sealed, evacuated quartz tube in order to reduce the effect of oxidization. The superconducting transition temperature T_c was determined to be 2.25 K by magnetization measurements in agreement with a previous report [32].

Transverse-field μSR (TF μSR) was performed in the field range $10 \leq \mu_0 H \leq 50 \text{ mT}$. The field was applied above T_c before cooling through the superconducting transition to a temperature of 100 mK in order to stabilize a well-ordered flux line lattice in the mixed state of the superconductor. Asymmetry signals collected above and below T_c are shown in Fig. 1. The time evolution of the

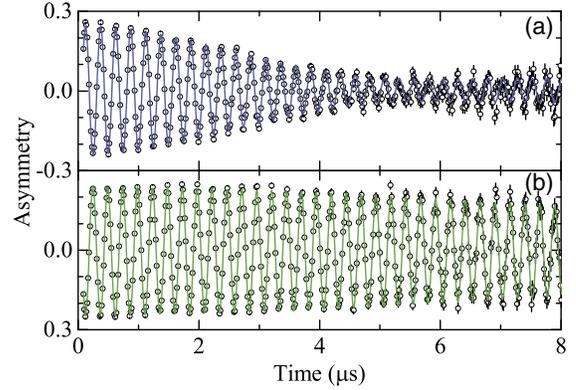


FIG. 1 (color online). Representative TF μSR signals collected at (a) 100 mK and (b) 3.0 K in an applied magnetic field of 30 mT. The solid lines are fits using Eq. (1). The effect of the flux line lattice can be seen in the top panel as the strong Gaussian decay envelope of the oscillatory function. Above T_c , the depolarization is reduced and is due to the randomly oriented array of nuclear magnetic moments.

asymmetry is described by a sinusoidal function damped with Gaussian relaxation plus a nondecaying oscillation that originates from muons stopping in the silver:

$$G_{\text{TF}}(t) = A_1 \exp\left(-\frac{\sigma^2 t^2}{2}\right) \cos(\gamma_\mu B_1 t + \phi) + A_2 \cos(\gamma_\mu B_2 t + \phi). \quad (1)$$

Here, A_1 and A_2 are the sample and background asymmetries, B_1 and B_2 are the average fields in the superconductor and silver, ϕ is a shared phase offset, and $\gamma_\mu/2\pi = 135.5 \text{ MHz T}^{-1}$ is the muon gyromagnetic ratio. The depolarization rate σ is related to the variance of the magnetic-field distribution in the superconductor. The temperature dependence of the muon depolarization rate $\sigma(T)$ extracted from fits to Eq. (1) is displayed in Fig. 2(a). The field distribution of the flux line lattice is broadened by the presence of randomly oriented nuclear magnetic moments in the sample. The depolarization due to this nuclear dipolar field σ_N is assumed to be temperature independent and adds in quadrature to the contribution from the flux line lattice σ_{FLL} :

$$\sigma^2 = \sigma_{\text{FLL}}^2 + \sigma_N^2. \quad (2)$$

Isothermal cuts perpendicular to the T axis of the $\sigma(T)$ data sets were used to determine the field dependence of the depolarization rate $\sigma(H)$ displayed in Fig. 2(b). Brandt [34] has derived a useful relation describing this field dependence, which is valid over the field range examined in this experiment:

$$\sigma_{\text{FLL}}^2 = 7.5 \times 10^{-4} (1-h)^2 [1 + 3.9(1-h)^2] \Phi_0^2 \lambda^{-4}, \quad (3)$$

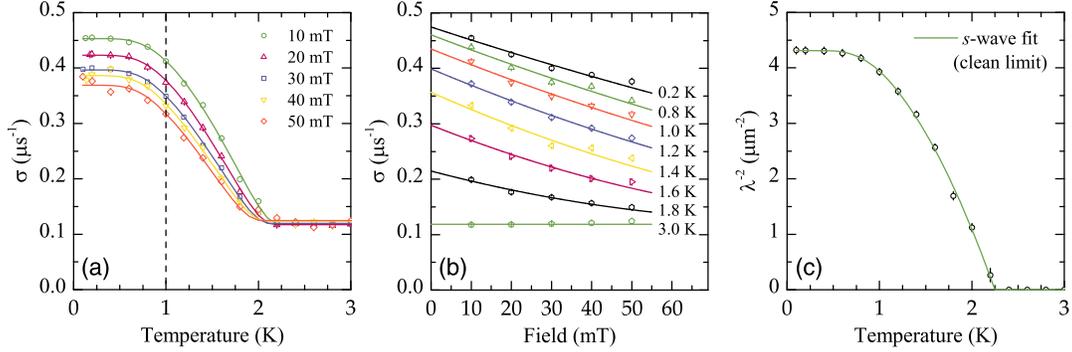


FIG. 2 (color online). (a) Temperature dependence of the TF μ SR spin-depolarization rate collected in a range of fields between 10 and 50 mT. The dashed line shows an example of the isothermal cuts used to find the field dependence of σ at a particular temperature. The solid lines are guides to the eye. (b) Field dependence of the muon spin-depolarization rate for a range of different temperatures. The solid lines are the results of fitting the data with Eq. (3) via Eq. (2). (c) Extracted temperature dependence of the inverse magnetic penetration depth squared. The solid line is the result of a fit using Eq. (4).

where $h = H/H_{C2}$ is the reduced field, and Φ_0 is the magnetic flux quantum. Inserting Eq. (3) into Eq. (2) yields a model that can be applied to the data in Fig. 2(b). A global fit was implemented in order to determine the temperature dependence of the inverse-squared penetration depth λ^{-2} with the background depolarization rate shared between all temperatures. The resulting fits to the $\sigma(B)$ data are displayed as solid lines in Fig. 2(b), with the fit at 3.0 K representative of the shared background depolarization rate, $\sigma_N = (0.116 \pm 0.003) \mu\text{s}^{-1}$. The temperature dependence of λ^{-2} is presented in Fig. 2(c), where λ^{-2} has been fixed to zero above T_c . Assuming London local electrodynamics, the temperature dependence of the superfluid density can be calculated for an isotropic s -wave superconductor in the clean limit using the following expression:

$$\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = 1 + 2 \int_{\Delta(T)}^{\infty} \left(\frac{\partial f}{\partial E} \right) \frac{E dE}{\sqrt{E^2 - \Delta^2(T)}}, \quad (4)$$

where $f = [1 + \exp(E/k_B T)]^{-1}$ is the Fermi function, and $\Delta(T) = \Delta(0) \tanh\{1.82[1.018(T_c/T - 1)]^{0.51}\}$ is the BCS approximation for the temperature dependence of the energy gap. The result of a fit to this model for the measured values of $\lambda^{-2}(T)$ is displayed as the solid line in Fig. 2(c). The fitted value for the energy gap $\Delta_0 = (0.369 \pm 0.007) \text{ meV}$ yields the BCS parameter $2\Delta_0/k_B T_c = 3.81 \pm 0.01$. This is larger than the value of 3.5 expected from the BCS theory in the weak coupling limit, implying that the strength of the superconducting pairing mechanism is enhanced in this system.

The magnetic penetration depth is directly related to (m^*/n_s) , where m^* is the effective mass of charge carrying electrons (in units of the electron rest mass m_e), and n_s the superconducting charge carrier density. Following the procedure described in Ref. [35], our experimentally determined value of $\lambda(0) = (482 \pm 2) \text{ nm}$ can be coupled with a heat capacity measurement of the Sommerfeld

constant $\gamma = (47 \pm 1) \text{ mJ mol}^{-1} \text{ K}^{-2}$ to yield $m^*/m_e = 13.9 \pm 0.2$ and $n_s = (0.169 \pm 0.003) \times 10^{28} \text{ m}^{-3}$. Consequently, an effective Fermi temperature $T_F = (432 \pm 8) \text{ K}$ is calculated. Uemura *et al.* [36–38] have described a method of classifying superconductors based on the ratio of the critical temperature to this effective Fermi temperature, which for this system is $T_c/T_F = 1/192$. This result implies that the superconductivity in La_7Ir_3 is unconventional and places this system in the vicinity of the heavy fermion superconductors under the Uemura classification scheme.

We now consider the results from the zero-field (ZF) and longitudinal-field (LF) experiments. Figure 3(a) shows the relaxation spectra collected above and below the superconducting transition temperature in ZF. There is a clear change in the relaxation behavior on either side of the transition. The increased relaxation below T_c has been verified with the μ SR instrument in both longitudinal and transverse geometries, which requires a physical rotation of the zero-field coils by 90° . This is significant, as any stray field that may erroneously be interpreted as a TRSB signal will be applied in orthogonal geometries and would appear differently in the relaxation spectra. There is no hint of an oscillatory component in the data, which would otherwise suggest the presence of an ordered magnetic structure. In the absence of atomic moments, the depolarization of the muon ensemble is due to the presence of static, randomly oriented nuclear moments. This behavior is modeled by the Gaussian Kubo-Toyabe equation [39]

$$G_{\text{KT}}(t) = \frac{1}{3} + \frac{2}{3}(1 - \sigma_{\text{ZF}}^2 t^2) \exp\left(-\frac{\sigma_{\text{ZF}}^2 t^2}{2}\right), \quad (5)$$

where σ_{ZF} measures the width of the nuclear dipolar field experienced by the muons. The spectra are well described by the function

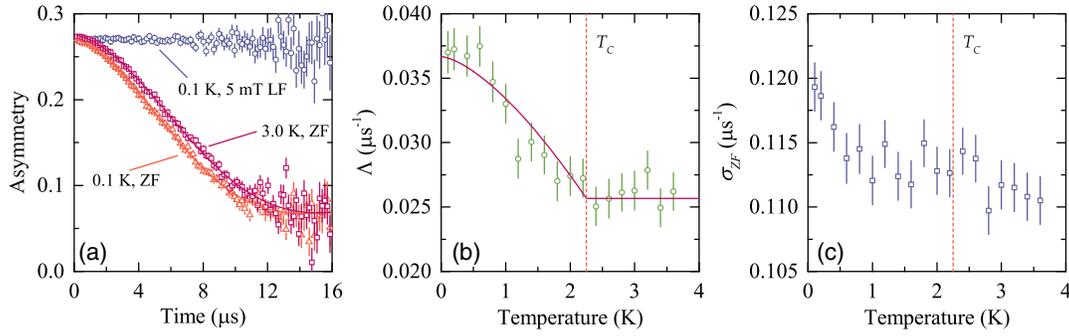


FIG. 3 (color online). (a) ZF and LF μ SR spectra collected above (squares) and below (triangles) T_c , with least-squares fits using the model of Eq. (6) (solid lines). In ZF, there is a clear difference between the spectra, indicating the presence of spontaneous fields in the superconducting state. The effect of applying a small LF field of 5 mT is also shown (circles). (b) Temperature dependence of the electronic relaxation rate Λ . A clear increase at T_c is observed. (c) Temperature dependence of the nuclear relaxation rate σ_{ZF} , which remains almost constant over the entire temperature range.

$$G(t) = A_0 G_{KT}(t) \exp(-\Lambda t) + A_{bg}, \quad (6)$$

where A_0 and A_{bg} are the sample and background asymmetries, respectively, and Λ measures the electronic relaxation rate.

The parameters A_0 and A_{bg} are found to be approximately temperature independent. The nuclear depolarization rate σ_{ZF} remains approximately flat, except as $T \rightarrow 0$ K where a slight increase is observed. The electronic relaxation rate Λ shows a systematic increase below the superconducting transition temperature [see Figs. 3(a) and 3(b)]. An exponential relaxation process is generally attributed to the field distribution arising from electronic spins fluctuating quickly enough to motionally narrow the effective depolarization of the muons. However, a weak magnetic field of only 5 mT is enough to fully decouple the muon from this exponential relaxation channel. This implies that the relaxation mechanism is actually static or quasistatic with respect to the muon lifetime. Furthermore, spin fluctuations associated with the proximity to a quantum critical point would be expected to exhibit a Curie-Weiss-like temperature dependence, as opposed to the onset at T_c observed [40].

Thus, it is likely that the source of the ZF signals observed below T_c is unique to the La_7Ir_3 and corresponds to the onset of a superconducting channel that breaks time-reversal symmetry. Aoki *et al.* have discussed the probable sources of the spontaneous field in superconductors with TRSB. In systems where the Cooper pairs have nonzero spin and orbital moments, regions in the sample where the order parameter becomes spatially inhomogeneous, such as grain boundaries, surfaces, and impurity sites, act as field sources due to the undamped supercurrents that arise there [41]. Alternatively, if the Cooper pairs have only nonzero spin moments, a hyperfine field may be generated at the interstitial μ^+ sites.

The TRSB signals are observed in the Λ relaxation channel akin to the NCS LaNiC_2 and Sr_2RuO_4 [12]. This

implies that the sources of field are dilute, producing a Lorentzian field distribution that is randomly sampled by the muons. If the field sources are caused by inhomogeneities in the order parameter, one would expect the Cooper pairs to possess a nonzero orbital momentum. However, the temperature dependence of the magnetic penetration depth is well described by an isotropic s -wave model. A further complication for NCS is that the ground state may be an admixture of spin-singlet and spin-triplet superconductivity. If the Cooper pairs associated with the spin-triplet channel do, indeed, possess an orbital moment, the greater relative strength of the singlet to triplet channels have made its detection difficult given the sensitivity of the current experiment.

In conclusion, TF and ZF μ SR measurements have been carried out on the noncentrosymmetric superconductor La_7Ir_3 . A spontaneous magnetization has been clearly observed at the superconducting transition temperature, confirming that time-reversal symmetry is broken in the superconducting state. This marks the discovery of the first hexagonal NCS to exhibit broken time-reversal symmetry. The superconducting order parameter has been described well by an isotropic gap with s -wave pairing symmetry and enhanced electron-phonon coupling. The results have implied that La_7Ir_3 has an unconventional superconducting ground state that features a dominant s -wave component, with the exact nature of the triplet component undetermined. In order to determine if the superconductivity is nonunitary, further experimental work on high-quality single crystals is vital, coupled with group theory calculations to determine the allowed pairing symmetries. This work paves the way for further studies of the large number of superconductors in the Th_7Fe_3 family in the hunt for unconventional behavior.

The authors would like to thank T.E. Orton for valuable technical support. J.A.T.B. acknowledges ISIS and the STFC for studentship funding through Grant

No. ST/K502418/1. R. P. S. acknowledges the Science and Engineering Research Board, Government of India for the Ramanujan Fellowship, through Grant No. SR/S2/RJN-83/2012. This work was funded by the EPSRC, U.K., through Grant No. EP/I007210/1. Some of the equipment used in this research was obtained through the Science City Advanced Materials project, Creating and Characterizing Next Generation Advanced Materials project, with support from Advantage West Midlands and partially funded by the European Regional Development Fund.

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