Fully-gapped superconductivity in single crystals of noncentrosymmetric Re$_6$Zr with broken time-reversal symmetry

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The noncentrosymmetric superconductor Re$_6$Zr has attracted much interest due to the observation of broken time-reversal symmetry in the superconducting state. Here we report an investigation of the superconducting gap structure of Re$_6$Zr single crystals by measuring the magnetic penetration depth shift $\Delta \lambda(T)$ and electronic specific heat $C_v(T)$. $\Delta \lambda(T)$ exhibits an exponential temperature dependence behavior for $T \ll T_c$, which indicates a fully-open superconducting gap. Our analysis shows that a single gap $s$-wave model is sufficient to describe both the superfluid density $\rho_s(T)$ and $C_v(T)$ results, with a fitted gap magnitude larger than the weak coupling BCS value, providing evidence for fully-gapped superconductivity in Re$_6$Zr with moderate coupling.

I. INTRODUCTION

Noncentrosymmetric superconductors (NCS) have attracted a great deal of attention due to the influence of antisymmetric spin-orbit coupling (ASOC) on the superconducting properties, which is induced by the antisymmetric potential gradient arising due to broken inversion symmetry$^{1,2}$. Strong ASOC may lift the spin degeneracy of the conduction bands, allowing for superconducting states which are an admixture of spin-singlet and spin-triplet pairing, giving rise to a variety of unique properties$^{1-4}$. Unconventional superconductivity was evidenced in the first heavy fermion NCS CePt$_3$Si, where a linear temperature dependence of the magnetic penetration depth at low temperatures and a constant Knight shift across $T_c$ were observed$^{5,6}$. While studies of heavy fermion NCS revealed a range of unusual findings, disentangling the role played by broken inversion symmetry from the effects of strong electronic correlations and magnetism is highly challenging, which spurred an interest in looking for weakly correlated NCS with singlet-triplet mixing. Evidence for such an admixture was demonstrated in Li$_2$(Pd,Pt)$_3$B, where Li$_2$Pd$_3$B has a fully open gap$^{7-9}$, but the gap of Li$_2$Pt$_3$B exhibits line nodes$^{9-11}$. This change from nodeless to nodal superconductivity upon switching Pd for Pt was explained using a model with a mixture of singlet and triplet states, where there is a relative increase in the size of the spin-triplet component of the order parameter, as the ASOC is increased$^9$. The Knight shift also shows a marked difference between the two compounds, corresponding to a decrease of the spin susceptibility below $T_c$ for Li$_2$Pd$_3$B, while for Li$_2$Pt$_3$B this remains constant, indicating greater influence of the ASOC on the superconductivity$^8,10$.

Subsequently, the order parameters of a wider range of NCS have been studied, where evidence for nodal superconductivity was also found in quasi-one-dimensional K$_2$Cr$_3$As$_3$ and the low carrier system YPtB$^{11}$. Meanwhile, although studies of Y$_2$C$_3$ at higher temperatures were accounted for by fully gapped superconductivity$^{14-16}$, very low temperature penetration depth measurements also indicate the presence of a nodal gap structure$^{17}$. However, most NCS have been found to be fully-gapped superconductors, such as BaPtSi$_3$,$^{18}$ BiPd$^{19,22}$, PbTaSe$_2$,$^{23,25}$ La$_7$Ir$_3$,$^{26}$ and Re$_6$Zr$^{27-30}$. In particular, many NCS have been found compatible with single-gap $s$-wave superconductivity, indicating that the size of any triplet component is very small, and the relationship between the ASOC strength and the degree of singlet-triplet pairing is not entirely understood. Moreover, it has been proposed that some NCS can exhibit topological superconductivity. Both theoretical calculations and experimental studies reveal topological surface states in the NCS PbTaSe$_2$ and BiPd$^{26,31-33}$, which may lead to possible Majorana fermions in the vortex cores when these states are sufficiently close to the Fermi level.

Another notable feature of several NCS is the breaking of time-reversal symmetry (TRS). While broken TRS was previously discovered in the unconventional triplet superconductor Sr$_2$RuO$_4$,$^{14,25}$, evidence for this phenomenon has also been found in a number of NCS with fully gapped superconducting states, such as LaNiC$_2$,$^{36}$ La$_7$Ir$_3$,$^{26}$, and Re$_6$Zr$^{27}$. In the case of LaNiC$_2$, the symmetry analysis shows that for TRS to be broken at $T_c$, the effect of the ASOC on the superconductivity should be weak, indicating a lack of significant mixed parity pairing$^{37}$. On the other hand, Re$_6$Zr crystallizes in the cubic $\alpha$-Mn structure, where the presence of three dimensional irreducible representations of the point group potentially allows for TRS breaking with singlet-triplet mixing$^{27}$. It is therefore of particular importance to characterize the superconducting order parameter of Re$_6$Zr. Previous measurements of the gap structure of polycrystalline samples are accounted for by nodeless single gap $s$-wave superconductivity$^{27-30}$, but different conclusions are drawn from recent point contact spectroscopy results from single crystal measurements, which give evidence
for multiple gaps. As such, it is important to perform further measurements of Re$_6$Zr single crystals sensitive to low energy excitations to clarify this issue. In this paper, we study the superconducting order parameter of single crystalline Re$_6$Zr by measuring the magnetic penetration depth and specific heat of single crystals, which are both consistent with a single nodeless isotropic gap with a moderate coupling strength.

II. EXPERIMENTAL DETAILS

Single crystals of Re$_6$Zr were synthesized using the Czochralski method, as described in Ref. 38. Magnetization measurements were performed using a superconducting quantum interference device (SQUID) magnetometer with both field-cooling (FC) and zero-field-cooling (ZFC), with a small applied magnetic field of 10 Oe. The electrical resistivity $\rho(T)$ was measured by using a standard four-probe method from room temperature down to 0.3 K in a $^3$He refrigerator. The specific heat $C(T)$ was measured using a Physical Property Measurement System (PPMS) with a $^3$He insert down to 0.6 K. The temperature dependence of the magnetic penetration depth shift $\Delta \lambda(T) = \lambda(T) - \lambda(0)$ was measured using a tunnel-diode-oscillator (TDO) technique in a $^3$He cryostat down to 0.35 K. The operating frequency of the TDO setup is about 7 MHz, with a noise level as low as 0.1 Hz. For the TDO measurements, the sample was cut into a regular shape with dimensions of $700 \times 500 \times 200 \ \mu$m$^3$ and mounted onto a sapphire rod so that it may be placed into the coil without making contact. The sample experiences a very small ac field of about 20 mOe along the [100] direction induced by the coil, which is much smaller than the lower critical field $H_{\lambda 1}$, ensuring that the sample remains in the Meissner state, so that the change of the magnetic penetration depth $\Delta \lambda(T)$ is proportional to the frequency change $\Delta f(T) = f(T) - f(0)$. Here $\Delta \lambda(T) = G \Delta f(T)$, where the calibration constant $G$ is determined by the geometry of the sample and coil.

III. RESULTS AND DISCUSSION

Single crystals of Re$_6$Zr were characterized by the measurements of the electrical resistivity $\rho(T)$ and magnetic susceptibility $4\pi \chi(T)$, which are displayed in Fig. 1. $\rho(T)$ exhibits metallic behavior in the normal state with a residual resistivity of $\rho(7\,K) = 0.3 \ \text{m\,\Omega\,cm}$, just above the superconducting transition. This yields a mean free path of 2.1 nm, with a coherence length of $\xi = 5.5 \ \text{nm}$ deduced from the upper critical field, and a Sommerfeld coefficient of $\gamma_n = 27.4 \ \text{m\,J\,mol}^{-1}\text{K}^{-2}$ from our specific heat measurements described below. The calculated mean free path is less than half the coherence length, indicating that the sample is closer to the dirty limit. The inset of Fig. 1 (a) displays an enlargement of $\rho(T)$ at low temperatures, where a sharp superconducting transition occurs at $T_c = 6.8 \, \text{K}$, with a transition width less than 0.2 K. In addition, the temperature dependence of the magnetic susceptibility in Fig. 1(b) shows that the zero-field-cooled (ZFC) curve exhibits full diamagnetism, providing evidence for bulk superconductivity in the Re$_6$Zr single crystals.

Figure 2 displays the temperature dependence of the penetration depth shift $\Delta \lambda(T)$ at low temperatures from TDO-based measurements. The frequency change $\Delta f(T)$...
displays the temperature dependence of the superconducting gap $\Delta(0)$, which is displayed as a function of the reduced temperature $T/T_c$ in Fig. 3. For a clean superconductor, $\rho_s(T)$ can be calculated using

$$\rho_s(T) = 1 + 2 \left( \int_{-\Delta}^{\infty} \frac{E dE}{\sqrt{E^2 - \Delta_k^2}} \frac{\partial f}{\partial E} \right)_{FS},$$

where $f(E, T) = [1 + \exp(E/k_BT)]^{-1}$ is the Fermi-Dirac distribution function and the superconducting gap function is defined as $\Delta_k(T) = \Delta(T) g_k$. This contains an angular dependent part $g_k$ and a temperature dependent component $\Delta(T)$, which can be approximated as $\Delta(T) = \Delta(0) \tanh \left\{ 1.82 \cdot [0.108 (T_c/T - 1)]^{0.51} \right\}^{32}$. Here the zero temperature gap magnitude $\Delta(0)$ is the only fitted parameter. As shown by the green dashed-dotted line in Fig. 3, a single gap $s$-wave model with $g_k = 1$ can well reproduce the experimental data with $\Delta(0) = 2.23 k_B T_c$. This gap magnitude is larger than the value of 1.76 $k_B T_c$ for weakly-coupled BCS superconductors, suggesting moderately strong coupling in Re$_6$Zr. On the other hand, since as discussed above the samples are near to the dirty limit, a dirty $s$-wave model was also applied, where $\rho_s(T) = \frac{\Delta(T)}{\Delta(0)} \tanh \left( \frac{\Delta(T)}{\Delta(0)} \right)$.$^{43}$ The results are shown by the solid red line, which also well accounts for the experimental data with $\Delta(0) = 2.1 k_B T_c$. In order to compare with nodal superconducting scenarios, both a $p$-wave model with $g_k = \sin^2 \theta$ and a $d$-wave model $g_k = \cos^2 \phi$ are also displayed, where $\theta$ is the polar angle and $\phi$ is the azimuthal angle. It is obvious that the superfluid density of $p$- and $d$-wave superconductors change significantly with temperature, even at very low temperatures, which is in contrast with the data showing near temperature independence below around 0.25 $T_c$. Furthermore, we also compare our results with measurements of polycrystalline samples using transverse-field muon-spin rotation ($\mu$SR) measurements from Ref. 27. These are displayed in Fig. 3 and show that comparable superconducting gap structures are inferred from measurements of single crystal and polycrystalline samples.

The specific heat measurements of Re$_6$Zr single crystals were also analyzed, to further characterize the superconducting order parameter. In the inset of Fig. 4, the total specific heat as $C/T$ is displayed, which contains both electronic and phonon contributions. In the normal state, the data were fitted using $C(T) = \gamma_n T + \beta T^3 + \delta T^5$, where $C_e = \gamma_n T$ and $C_{ph} = \beta T^3 + \delta T^5$ represent the electron and phonon contributions respectively, and $\gamma_n$ is the Sommerfeld coefficient. The fitting results are shown by the dashed line with fitted parameters of $\beta = 0.39 \text{ mJ mol}^{-1} \text{K}^{-4}$, $\delta = 1 \mu \text{J mol}^{-1} \text{K}^{-6}$, and $\gamma_n = 27.4 \mu \text{J mol}^{-1} \text{K}^{-2}$, which are consistent with measurements of polycrystalline samples.$^{27}$

The main panel of Fig. 4 displays the temperature dependence of the electronic specific heat as $C_e/T$ in the superconducting state after subtracting the phonon contribution. A sizeable jump is observed around $T_c$ of
$\Delta C/\gamma_n T_c=1.76$, which is larger than the value of 1.43 for a weakly-coupled BCS superconductor, again indicating an enhancement of the coupling strength in Re$_6$Zr.

In the superconducting state, the entropy $S$ can be expressed as $^{44}$:

$$ S = -\frac{3\gamma_n}{\pi^3} \int_0^{2\pi} \int_0^{\infty} [f \ln f + (1 - f) \ln(1 - f)] dz d\phi. \tag{3} $$

and therefore the electronic specific heat in the superconducting state can be calculated as $C_e = T dS/dT$. Due to $C_e(T)/T$ becoming flat with decreasing temperature for $T \ll T_c$, the data were fitted using an isotropic s-wave model, and the results are shown in Fig. 4 by the solid red line. It can be clearly seen that this model well describes the experimental data, with a fitted parameter of $\Delta(0) = 1.95 k_B T_c$. This is consistent with the penetration depth and superfluid density results and also indicates a moderately enhanced superconducting gap magnitude.

Since Re$_6$Zr is a NCS, the ASOC will lift the spin degeneracy of the electron bands, which can potentially give rise to a mixture of spin singlet and spin triplet pairing states, leading to a two-gap structure $\Delta_{s} = \psi \pm |d|$, where $\psi$ and $d$ represent the singlet and triplet components$^1$. Therefore the experimental results being consistent with a single nodeless gap implies that the spin triplet component is very small relative to the spin singlet. The origin of TRS breaking in Re$_6$Zr still remains difficult to account for. In LaNiC$_2$, because of the low symmetry of the orthorhombic crystal structure, the breaking of TRS at $T_c$ implies that singlet-triplet mixing is weak, and moreover that the pairing corresponds to a non-unitary triplet state$^{36,37}$. Here the lack of a role played by ASOC was corroborated by the breaking of TRS in the similar but centrosymmetric LaNiGa$_2$$^{45}$, and the observation of two-gap superconductivity in both LaNiC$_2$ and LaNiGa$_2$ allowed for the proposal of a nodeless even-parity, non-unitary triplet state, where the overall pairing wave function is antisymmetric upon particle exchange due to an antisymmetric orbital index$^{46}$. Such a scenario is not readily applied to Re$_6$Zr, since the higher symmetry of the cubic $\alpha$-Mn crystal structure lifts the constraint that TRS breaking at $T_c$ must correspond to non-unitary pairing, and moreover most studies only show evidence for single-gap behavior. As the symmetry analysis of Re$_6$Zr also allows for mixed singlet-triplet pairing with broken TRS$^{27}$, further studies are highly desirable to understand the origin of TRS breaking and the role of the ASOC in the superconducting state of Re$_6$Zr.

**IV. CONCLUSIONS**

To summarize, we have measured the magnetic penetration depth and electronic specific heat of single crystals of the noncentrosymmetric superconductor Re$_6$Zr. Exponential behavior of $\Delta\lambda(T)$ was observed at $T \ll T_c$, which indicates the absence of low energy excitations, and a fully gapped superconducting state. Both the superfluid density and specific heat can be well accounted for by a single gap s-wave model across the whole temperature range with a gap magnitude larger than the weak coupling BCS value, providing strong evidence for fully-gapped superconductivity with moderately strong coupling in Re$_6$Zr.

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