Local spectroscopic evidence for a nodeless magnetic kagome superconductor CeRu$_2$

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Abstract

We report muon spin rotation ($\mu$SR) experiments on the microscopic properties of superconductivity and magnetism in the kagome superconductor CeRu$_2$ with $T_c \approx 5$ K. From the measurements of the temperature-dependent magnetic penetration depth $\lambda$, the superconducting order parameter exhibits nodeless pairing, which fits best to an anisotropic $s$-wave gap symmetry. We further show that the $T_c/\lambda^{-2}$ ratio is comparable to that of unconventional superconductors. Furthermore, the powerful combination of zero-field (ZF)-$\mu$SR and high-field $\mu$SR has been used to uncover magnetic responses across three characteristic temperatures, identified as $T^*_1 \approx 110$ K, $T^*_2 \approx 65$ K, and $T^*_3 \approx 40$ K. Our experiments classify CeRu$_2$ as an exceedingly rare nodeless magnetic kagome superconductor.

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(Some figures may appear in colour only in the online journal)

1. Introduction

The unique kagome lattice, formed by an interwoven network of corner-sharing triangles, is well-known to host many fascinating physical phenomena [1–7]. Arising from the natural geometrical frustration, band structure calculations reveal several characteristic features in this atomic lattice, most frequently flat bands, van Hove singularities, and Dirac nodes, which can influence the electronic properties and give rise to topologically nontrivial phases when found near the Fermi energy. One of the most rare phenomena exhibited by kagome lattice materials is superconductivity, which often hosts competing magnetic [3] or otherwise unconventional [8] features. In our recent work on LaRu5Si2, which we identified as a robust s-wave kagome superconductor, we found that the critical temperature cannot be fully explained by electron-phonon coupling, but experiences additional enhancement from typical kagome band structure features found near the Fermi energy [8]. In further explorations of the recently discovered KV3Sb5 [9, 10] and sister compounds [3, 7, 11–13], we have identified time-reversal symmetry-breaking associated with the charge ordering transition at $T_{co} \simeq 80$ K, two orders of magnitude higher than the superconducting transition, $T_c \simeq 1.1$ K.

While the distorted Laves-phase superconductor CeRu2 takes a cubic structure (figure 1(a)) [14] with two different Ce sites, it reveals a pristine Ru kagome lattice (figure 1(b)) that contributes to the electronic properties. Indeed, the normal state band structure features a kagome flat band, Dirac points and van Hove singularities formed by the Ru-$dz^2$ orbitals near the Fermi level [15], which are predicted to support topologically nontrivial states [15]. Photoemission studies show the highly itinerant nature of the Ce electrons in CeRu2, identifying a strong hybridization effect in the itinerant 4f-bands [16, 17]. Additionally, much attention has been given to the unusual superconducting state in CeRu2, which shows two separate regions of magnetic hysteresis [18–22] and a rich $M - T$ phase diagram with multiple magnetic field-induced transitions [20]. Furthermore, nuclear magnetic resonance spectroscopy (NMR) nuclear quadrupole resonance spectroscopy (NQR), field-angle-resolved specific heat, and photoemission spectroscopy measurements [23–26] all suggest an anisotropic s-wave superconducting gap symmetry and show that quasiparticle excitations are gapped-out at a finite temperature. The importance of 4f-electrons in this material has been highlighted by de Haas–van Alphen (dHvA) measurements [27], finding no change in frequency with the onset of superconductivity, but a cyclotron mass dependence consistent only with f-electron superconductors. Previous muon spin relaxation ($\mu$SR) research has proposed the presence of weak magnetism below $T_M = 40$ K, a temperature much higher than the superconducting transition temperature ($T_c = 6$ K) [28]. Polarized neutron experiments [14] reported field-induced paramagnetic moments on the order of $4.4 \times 10^{-4} \mu_B T^{-1}$ per Ce atom and $4.5 \times 10^{-4} \mu_B T^{-1}$ per Ru atom. The $\mu$SR technique was also used to probe the magnetic penetration depth within the high-field anomalous ‘peak effect’ regime, suggesting the possibility of a Fulde–Ferrell–Larkin–Ovchinnikov state [29–31]. However, no microscopic study of the superconducting gap symmetry and its interplay with magnetism is reported so far.

To provide a bulk local spectroscopic probe of the superconducting gap symmetry and of its interplay with weak magnetism in CeRu2, we have carried out a combination of zero-field (ZF)-$\mu$SR and high-field $\mu$SR experiments. Our studies were performed on two Czochralski-pulled single crystals, and the details of the sample preparation, additional characterization methods, and analysis techniques can be found in the supplemental information. The superconducting order parameter achieves best agreement with an anisotropic s-wave symmetry. In the superconducting state, the ratio $T_c/\lambda^{s2}$ (where $T_c$ is the superconducting transition temperature and $\lambda^{s2}$ is the superfluid density) is comparable to those of unconventional superconductors. The relatively high $T_c$ for the low carrier density may hint at an unconventional pairing mechanism in CeRu2. The measured superconducting (SC) gap value $\Delta_{max} = 0.76(5) \text{meV}$ yields a ratio $2\Delta/\kappa_B T_c \simeq 3.8$, suggesting that the superconductor CeRu2 is in the moderate coupling limit. Furthermore, we identified three magnetic anomalies at $T_1^{\parallel} \simeq 110$ K, $T_2^{\perp} \simeq 65$ K, and $T_3^{\perp} \simeq 40$ K. Importantly, these magnetic anomalies are strongly enhanced under a magnetic field of 8 T.

2. Results and discussion

For the case of a material known to exhibit weak magnetism, $\mu$SR studies provide the most powerful tool for investigation. During a $\mu$SR experiment, positive muons are implanted into the sample, where they thermalize at interstitial positions and precess in the local magnetic field. They decay radioactively after a mean lifetime of 2.2 $\mu$s and emit a positron $e^+$ preferentially along the spin direction [32]. The asymmetry of emitted positrons is detected and this time-dependent polarization $P(t)$ of the ensemble may be fitted with a number of different functions, elucidating the physics of the investigated material (see the supplemental material for details). The $\mu$SR technique provides a powerful tool to measure the magnetic penetration depth $\lambda$ in the vortex state (in the presence of a weak applied field $H_A \leq H_{gap} \leq H_{c2}$) of Type II superconductors in the bulk of the sample [33], in
Figure 1. (a) A three dimensional visualization of the atomic structure of CeRu$_2$. (b) When viewed along the [1 1 1] direction, a plane of Ru atoms constructs a pristine kagome lattice. (c) Resistivity measurements performed on a single crystal of CeRu$_2$ in different applied magnetic fields. The black arrow indicates the critical temperature, which was extracted at each field and used to construct panel (d), which shows the field dependence of the superconducting transition temperature. The data has been extrapolated with a straight line, giving a value of $H_{c2} \approx 5.6$ T at 1.5 K.

contrast to many techniques that probe $\lambda$ only near the surface. Additionally, ZF-$\mu$SR has the ability to detect internal magnetic fields as small as 0.1 G without applying external magnetic fields, making it a highly valuable tool for probing spontaneous magnetic fields due to time-reversal symmetry breaking.

Shown in figure 2(a) are the Transverse-field (TF)-$\mu$SR time spectra recorded in the normal state (10 K) and in the superconducting vortex state (0.27 K), measured on a single crystal of CeRu$_2$ with a field of 30 mT applied perpendicular to the $a$-axis. The cylindrical Czochralski-pulled sample, with $\varnothing \approx 6$ mm and a length of $\approx 8$ mm, was placed directly on the sample fork in the muon beam. Any muons not stopped in the sample passed through the aluminated mylar tape and were stopped in the veto detectors behind the sample; in this way, the additional signal of muons not stopped in the sample is immediately removed from the spectrum. The spectrum in the normal state shows a weak depolarization due to random local fields from the nuclear moments, which can be fitted by a single Gaussian distribution, while in the superconducting state the relaxation rate is strongly enhanced due to the formation of the flux-line lattice. As one can see in the field distribution (figure 2(b)), the profile is asymmetric and we fitted it with a sum of two Gaussian distributions. A single central field can then be extracted from the two Gaussian distributions, as detailed in the supplementary information. The difference between the applied field (clearly visible as the center of the Gaussian field distribution in the sample in the normal state, see figure 2(b) at 10 K) and the central field in the superconducting state (see figure 2(b) at 0.27 K) constitutes the diamagnetic shift, plotted in figure 2(d). Furthermore, the second moment of the two Gaussian field distributions can also be extracted and combined to obtain the relaxation rate (see supplementary material) as displayed in figure 2(c). From the temperature dependence of the diamagnetic shift $\Delta B_{dia} = \mu_0 (H_{int,SC} - H_{int,NS})$ (i.e. the difference between the internal field $\mu_0 H_{int,SC}$ measured in the SC fraction and $\mu_0 H_{int,NS}$ measured in the normal state at $T = 10$ K), we can clearly see the large diamagnetic response of 0.7 mT associated with the superconducting transition at $T_c = 4.7$ K in figure 2(d). The temperature dependence of the superconducting muon spin depolarization rate, $\sigma_{sc}$, is shown in figure 2(c). In order to investigate the symmetry of the SC gap, we note that $\lambda(T)$ is related to the depolarization rate $\sigma_{sc}(T)$ in the presence
of a perfect triangular vortex lattice with \( H_{\text{app}} \ll H_c \) by the equation [34]:

\[
\frac{\sigma_{\mu}(T)}{\gamma_\mu} = 0.06091 \frac{\Phi_0}{\lambda^2(T)},
\]

where \( \gamma_\mu \) is the gyromagnetic ratio of the muon and \( \Phi_0 \) is the magnetic-flux quantum. The temperature dependence of the superfluid density \( \lambda^{-2}(T) \) was then fitted with nodeless \( (s\text{-} and \text{anisotropic } s\text{-wave}) \) and nodal \( (d\text{-wave}) \) models to determine the superconducting gap symmetry (see supplementary material). Considering the quality of fit and \( \chi^2 \) values (see figure 2(c) and table 1), it is clear that the anisotropic \( s\)-wave fits the data best, meaning that there is an angular dependence to the superconducting gap value (similar to \( d\)-wave superconductivity) but the minimum gap value is nonzero. The ratio of the minimum gap value to the maximum gap value we obtained was \( \Delta_{\text{min}}/\Delta_{\text{max}} = 0.47(1) \). This is in fairly good agreement with values near \( \Delta_{\text{min}}/\Delta_{\text{max}} = 0.33 \) obtained from NMR studies [25] and in excellent agreement with the value obtained by photoemission experiments \( \Delta_{\text{min}}/\Delta_{\text{max}} = 0.447 \) [24].

From the measured muon relaxation rate in the superconducting state, we can calculate the superfluid density using equation (1). The ratio of the superconducting gap to \( T_c \) was estimated to be \( 2\Delta_{\text{max}}/k_B T_c = 3.8 \), which is in excellent agreement with the predicted value [24].

### Table 1.

Summary of the parameters obtained for fits of the superconducting gap structure to the superfluid density measured in CeRu\(_2\) by means of \( \mu\)SR. The reduced \( \chi^2 \) values indicated in the rightmost column clearly show the best fit is obtained by the anisotropic \( s\)-wave gap structure. The fitting procedure and functions used can be found in supplementary material.

| Symmetry | \( \lambda_0 \) (nm) | \( T_c \) (K) | \( \Delta_{\text{max}} \) (meV) | \( \Delta_{\text{min}} \) (meV) | \( \chi^2 \) |
|----------|---------------------|-------------|-----------------|-----------------|--------|
| an. \( s\)-wave | 284(5) | 4.68(3) | 0.76(5) | 0.36(1) | 0.76 |
| s-wave | 287(4) | 4.72(3) | 0.73(4) | — | 1.05 |
| \( d\)-wave | 271(4) | 4.67(2) | 1.12(7) | — | 0.85 |
agreement with NMR, dHvA effect, photoemission, surface impedance, and tunneling results [22–24, 27, 35]. This ratio is consistent with the moderate coupling limit Bardeen-Cooper-Schrieffer (BCS) expectation [33]. However, a similar ratio can also be expected for the Bose Einstein condensate-like picture as pointed out in [36]. The Uemura ratio [37] between the critical temperature and the superfluid density extrapolated to \( T = 0 \) K is estimated to be \( T_c/\lambda^2 \approx 0.377 \), which is an order of magnitude smaller than for hole-doped cuprate superconductors, but still far away from conventional phonon-mediated BCS superconductors [38]. Interestingly, the ratio for CeRu\(_2\) is almost identical to that for LaRu\(_3\)Si\(_2\) [8], for charge density wave superconductors 2H-NbSe\(_2\) and 4H-NbSe\(_2\) as well as Weyl-supersymmetry \( T_d'\)-MoTe\(_2\) [33]. This finding hints at an unconventional pairing mechanism in CeRu\(_2\) with a low density of Cooper pairs and similar electron correlations as in LaRu\(_3\)Si\(_2\), 2H-NbSe\(_2\) and \( T_d'\)-MoTe\(_2\), but much weaker electron correlations than in cuprates and twisted bilayer graphene.

While the unconventional nature itself of this superconductor makes it an interesting subject, a previous \( \mu \)SR study [28] found extremely weak magnetism in this material at a temperature much higher than the superconducting transition, \( T_M = 40 \) K. Motivated by the apparent similarity to recently-discovered KV\(_2\)Sb\(_3\), in which the onset of a charge density wave phase is accompanied by electron dynamics that break time-reversal symmetry [3], we similarly performed ZF-\( \mu \)SR measurements over a broad temperature range on CeRu\(_2\). We observe a clear increase in the relaxation rate, evidenced by the comparison of the \( \mu \)SR spectra observed at 5 K and 40 K, in figure 3(a). The \( \mu \)SR spectra were fitted with a Gaussian Kubo–Toyabe function. It has been previously shown that the muon spin relaxation rate originates from a static internal field distribution, as a longitudinal field of 1 mT is sufficient to fully decouple the depolarization [28]. We notice an upturn and a broad downturn with the onsets of \( T^*_\parallel \sim 110 \) K and \( T^*_\perp \sim 65 \) K. Consistent with the earlier report [28], we also notice a small increase of 0.03 \( \mu \)s\(^{-1}\) in \( \sigma_{mun} \) around 40 K, which we have denoted as \( T^*_\sigma \) in figure 3(b). With the application of 3 mT, we can more clearly identify the enhancement below 40 K, as seen in figure 3(c). It is interesting to note the reduction of ZF rate \( \sigma_{mun} \) below the superconducting transition temperature \( T_c \) (see figure 3(b)). This indicates a clear effect of superconductivity on the weak internal fields and supports the magnetic origin of the increased depolarization rate. More importantly, this behavior indicates an interplay between magnetism and superconductivity in CeRu\(_2\) involving competition for the same electrons. The strong suppression of the magnetism below the onset of superconductivity was also observed in the nodeless Fe-based high-temperature superconductors: NaFe\(_{1−x}\)Ni\(_x\)As [39], BaFe\(_{2−x}\)Co\(_x\)As\(_2\) [40, 41], BaFe\(_{2−x}\)Ni\(_x\)As\(_2\) [42], Ba\(_{1−x}\)K\(_x\)Fe\(_2\)As\(_2\) [43] and FeSe [44]. It was discussed that itinerant antiferromagnetic (AFM) and SC orders are generally expected to compete strongly for the same electronic states, which was captured within a simple Ginzburg–Landau free energy for the AFM and SC order parameters [39, 45].

In order to confirm the magnetic origin of the low-\( T \) relaxation rate, we performed high-field \( \mu \)SR experiments with the HAL-9500 instrument in 8 T applied along the cylindrical cut of the sample. The magnetic contribution should experience field-induced enhancement, but the nuclear contribution should remain unchanged [3]. Since the upper critical field in this superconductor is \( H_c^\parallel \approx 5.6 \) T [19, 21, 27], we were able to perform the \( \mu \)SR experiments purely in the normal state down to the base temperature of 3 K with complete suppression of the superconducting state. As illustrated in figure 4(a), the high-field \( \mu \)SR spectra are best described by a two-component Gaussian fit. We observed that a single-component fit was not sufficient to describe the field distribution in CeRu\(_2\) under applied field, and that a second Gaussian component was needed to fit the \( \mu \)SR spectra, as illustrated in figure 4(a). The contribution seen in figure 4(b) from the \( \sigma_2 \) component (i.e. the blue circles) accounts only for 30% of the total signal. These two components and their relative fractions may be related to the presence of both Ce\(^{3+}\) and Ce\(^{4+}\) states as evidenced by photoemission experiments [16] and crystallographic muon sites with different relative distances to the corresponding ions. Another possibility for the presence of two components is a phase separation into two spatially separated volumes with different magnetic properties. In figure 4(b) is displayed the temperature dependence of both components, which both show clear anomalies.
Namely, the relaxation rate anomalies are strongly enhanced under applied magnetic field. By ZF, the rate decreases. Both relaxation rates begin sharply increasing with the onset of the increase observed with ZF-magnetic susceptibility, measured in 0.005 T and 7 T. This clearly supports the electronic/magnetic origin of the anomalies seen under ZF, as the temperature dependence of the nuclear contribution to the relaxation cannot be significantly changed by an external field. It is also noteworthy to mention that the previously reported high-field Hall resistivity (being highly sensitive to magnetic contributions) exhibits very similar temperature dependence as our high-field μSR relaxation rate (see figure 4(c)).

The combination of ZF-μSR and high-field μSR results on CeRu₂ provides evidence of distinct magnetic responses with three characteristic temperatures $T_1^* \simeq 110$ K, $T_2^* \simeq 65$ K, and $T_3^* \simeq 40$ K. This may originate from the complex interaction between Ru-$d$-electrons and Ce-$4f$ electrons [16, 17]. We can not comment on the precise origin of magnetism in CeRu₂. However, since macroscopic susceptibility does not show any clear magnetic transitions (see figure 4(c)), magnetism is likely itinerant and antiferromagnetic. This calls for additional detailed experiments.

The presence of weak magnetism in CeRu₂ is reminiscent of kagome superconductors KV,Sb₂ and RbV,Sb₂ [3, 11], where μSR shows the emergence of a time-reversal symmetry-breaking state below 75 K and 120 K, respectively. However, in the 135 kagome superconductors, the weak magnetic signal occurs contemporaneously with topological charge ordering, which competes with superconductivity [11], occurring at much lower temperatures $T_c \simeq 1$ K. The $T_c$ of (K,Rb)V,Sb₂ are enhanced to $\simeq 4$ K under pressure, only after suppressing the charge order. Furthermore, the superconducting pairing symmetry is nodal for both (K,Rb)V,Sb₂ at low pressure when the system also exhibits charge order [11]. Upon applying pressure, the charge order is suppressed and the superconducting state progressively evolves from nodal to nodeless [11]. Thus, the high-pressure SC state in (K,Rb)V,Sb₂ without charge order is nodeless. No charge ordering has been reported for CeRu₂ even at ambient pressure and it exhibits a nodeless superconducting state with a relatively high critical temperature $T_c \simeq 5$ K, similar to kagome superconductor LaRu₄Si₁₂. All these observations strongly suggest that the presence of charge order in kagome superconductors can strongly influence the superconducting gap structure.

3. Conclusion

The distorted Laves-phase $f$-electron superconductor CeRu₂ exhibits a pristine Ru kagome network, which has been shown to host correlated electronic states. Using the bulk-sensitive magnetic microprobe μSR, we have spectroscopically identified CeRu₂ as a nodeless superconductor, with a temperature dependence of the superconducting order parameter which is best fitted by an anisotropic $s$-wave gap symmetry. The unconventional nature of superconductivity is additionally evidenced by the observed dilute superfluid density. Furthermore, the combination of highly-sensitive ZF-μSR and high-field μSR shows that this material exhibits a magnetic response with three characteristic temperatures, which we...
have identified as $T^*_1 = 110 \text{ K}$, $T^*_2 = 65 \text{ K}$ and $T_\gamma = 40 \text{ K}$. We furthermore show that the magnetic response is strongly enhanced by magnetic field. Our bulk spectroscopic characterization of the nodeless kagome superconductivity and magnetic order underline the competition between these two orders in CeRu$_2$.

**Data availability statement**

The data that support the findings of this study are available upon reasonable request from the authors.

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