Evidence for vertical line nodes in Sr₂RuO₄ from nonlocal electrodynamics

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(Received 22 December 2023; revised 10 May 2024; accepted 7 August 2024; published 17 September 2024)

By determining the superconducting lower and upper critical fields $H_{c1}(T)$ and $H_{c2}(T)$, respectively, in a highpurity spherical Sr_2RuO_4 sample via ac-susceptibility measurements, we obtain the temperature dependence of the coherence length ξ and the penetration depth λ down to $0.04T_c$. Given the high sample quality, the observed T^2 dependence of λ at low temperatures cannot be explained in terms of impurity effects. Instead, we argue that the weak type-II superconductor Sr_2RuO_4 has to be treated in the nonlocal limit. By comparing our data with existing theory in that limit, the penetration depth in Sr_2RuO_4 agrees with a gap structure having vertical line nodes, while horizontal line nodes cannot account for the observation. The work highlights the potential benefits of purifying other unconventional superconductors in order to access the fascinating nonlocal regime in more materials and to determine their Cooper pair wave functions.

DOI: 10.1103/PhysRevB.110.L100503

Understanding the superconductivity of Sr_2RuO_4 is a challenge that has now spanned nearly three decades [1]. The high purity of the crystals available for study, combined with the relatively simple and well-understood normal state [2], means that this should be a soluble problem, and it has become a milestone for the whole field of unconventional superconductivity [3–9]. Progress is hindered by the lack of a complete understanding of its superconducting order parameter. Recent studies of the spin susceptibility in the superconducting state have called into question the long-held paradigm of a spin-triplet, odd-parity order parameter, and provided strong evidence for a spin-singlet, even-parity state [10–12]. The question of whether the order parameter breaks time-reversal symmetry or not is also the subject of ongoing investigations [13–16].

To further inform the rejuvenated theoretical effort that these results have stimulated, it is important to find new ways to address a related issue about which there is apparently conflicting information: the nodal structure of the superconducting gap. It is widely agreed, on the basis of, for example, ultrasound [17], penetration depth [18], heat capacity [19–21], and thermal conductivity [22–24], that the gap in Sr₂RuO₄ has nodes, but different conclusions have been reached about whether these are horizontal (perpendicular to the *c* axis of the tetragonal crystal structure) [20,25] or vertical (parallel to the *c* axis) [19,22,24]. Any information on this issue is important, because horizontal line nodes would imply mechanisms incorporating interplane or interorbital pairing [26–29], while vertical line nodes would be consistent with pairing states formed from in-plane electronic states [29–32], the latter being natural for a quasi-2D material.

As well as giving a low-energy spectrum with well-defined power laws that can be studied by any probe sensitive to the density of states, nodal Bogoliubov quasiparticles profoundly affect the screening currents that are at the heart of the macroscopic coherence of a superconductor. The effective coherence length v_F/Δ_k diverges at T = 0 along the nodal directions, where the gap Δ_k vanishes, and is replaced by the thermal de Broglie wavelength $\xi_T \sim v_F/(k_BT)$ at finite T. As shown by Kosztin and Leggett [33], this leads to a length-scale-dependent ability of supercurrents to screen an external magnetic field and the London equation must be generalized to $\nabla \times \mathbf{j}(\mathbf{r}) = -\int d^3 \mathbf{r}' K_{\perp}(\mathbf{r} - \mathbf{r}') \mathbf{B}(\mathbf{r}')$ with a nonlocal transverse current response. The electromagnetic response for a wavelength of the magnetic field smaller than ξ_T is reduced due to thermally excited nodal quasiparticles

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and qualitatively changes the T dependence of the magnetic penetration depth [33]. In strongly type-II superconductors, the range of temperatures over which these nonlocal effects are relevant becomes vanishingly small. This is one reason why nonlocal effects have largely been disregarded in most of the previous interpretations of experimental penetration depth results on type-II superconductors although they have been discussed in [34,35]. However, Sr₂RuO₄, being not far from the type I-type II phase border, allows for a much larger range of temperatures for which the nonlocal physics becomes prominent. Nevertheless, even in Sr₂RuO₄, this has not been unambiguously identified [18] because impurities can cause the same T dependence of the penetration depth in nodal superconductors, when the impurity bandwidth is high [36]. This ambiguity also has so far prevented scientists from employing a particularly attractive feature of electrodynamic calculations in the nonlocal limit: Existing theory predicts that they distinguish the responses from vertical and horizontal line nodes [33,37].

In this Letter, we approach the problem of nodal excitation using a technique that has not so far been employed in the study of Sr₂RuO₄. We perform magnetic susceptibility measurements for $\mu_0 H \parallel c$ on a nearly spherical sample sculpted from an ultrahigh-purity single crystal with $T_{\rm c} =$ 1.5 K. The measurements enable a quantitative determination of the temperature dependence of both the lower and upper critical fields, H_{c1} and H_{c2} . From this information, we derive the temperature dependence of the in-plane penetration depth λ and superconducting coherence length ξ to temperatures below 50 mK. The extremely low impurity concentration in our samples leads to a very low crossover temperature of $0.05T_{\rm c}$, below which impurities should play a role for the temperature dependence of the penetration depth. Because this temperature is near our lowest measured temperature we are able to rule out impurities as an origin of the observed Tdependence. Instead, our results show clear evidence that nonlocal electrodynamics must be used to analyze the penetration depth in Sr₂RuO₄ with this level of purity. Importantly, our data are consistent with the prediction for vertical nodes.

Experimental results. To study the critical fields of Sr_2RuO_4 , we measured the magnetic field dependence of the ac susceptibility at different temperatures with the external magnetic field μ_0H applied parallel to the crystallographic *c* axis. We used a high-purity sample cut into a sphere with a diameter of 470 µm using focused ion beam (FIB) milling as shown in Fig. 1(a) (for details, see the Supplemental Material (SM) [38]). The spherical shape gives a well-defined demagnetization factor independent of the magnetic field direction and removes uncertainties from other shapes that can strongly influence the measured field values, particularly that of H_{c1} . The field orientation was ensured by a FIB mark on the sphere and the frequency of quantum oscillations occurring in higher fields [38].

The temperature dependence of the real part of the magnetic ac susceptibility $\chi'(T)$ at H = 0 is shown in Fig. 1(a). The superconducting critical temperature $T_c = 1.5$ K reveals a high sample purity, comparable to samples having the highest T_c values [39]. Figure 1(b) shows the field dependence of the ac susceptibility $\chi'(H)$ at 100 mK. These are the upsweep data that have been corrected for the remanent field of the



FIG. 1. (a) Temperature dependence of ac susceptibility $\chi'(T)$ of the spherical sample of Sr₂RuO₄. The T_c is defined at the onset of the superconducting transition, highlighted with an arrow. (b) Magnetic field dependence (upsweep) of $\chi'(H)$ at 100 mK. The upper critical field H_{c2} is defined at the onset of the transition, and the features related to the vortex physics, H_p and H_v , are defined at the peak and valley as indicated by arrows. The inset shows a zoom near H_{c1}^* , defined as the field where the susceptibility departs from the constant minimum value.

magnet [38]. We identify four features: the lower critical field H_{c1}^* , which is uncorrected for demagnetization, the upper critical field H_{c2} , a peak H_p , and a valley H_v , the latter two being likely associated with the superconducting vortex physics [40,41]. H_{c2} is defined as the onset of the normal state transition and H_{c1}^* as the first deviation of the susceptibility from the full screening in the Meissner state, as defined in the inset to Fig. 1(b) (see also the SM for more details [38]). To obtain the actual lower critical field $H_{c1} = H_{c1}^*(1 - N)^{-1}$, knowledge of the demagnetization factor N is required. Crucial for our analysis is that for a spherical sample N = 1/3 [42]. It is important to note that the feature in the ac susceptibility due



FIG. 2. (a) Magnetic field dependence (upsweep) of $\chi'(H)$ at different selected temperatures of the spherical sample with the field applied along the *c* axis. The H_{c1}^* , H_p , H_v , and H_{c2} are indicated with symbols. (b) Superconducting phase diagram extracted from (a).

to H_{c1}^* remains the same if we sweep the magnetic field up, starting in zero-field cooled or under field-cooled conditions (see the SM [38]). These results show that we can clearly detect a sharp signature at H_{c1}^* .

Figure 2(a) exhibits the magnetic field dependence of the susceptibility at different temperatures. The four features described above are again indicated with arrows on the susceptibility curves. From this study, we identified the signature of H_{c1}^* up to 1.25 K. The peak/valley (H_p , H_v) vortex features can be observed up to 1.1 K since they approach each other as we increase the temperature, making them indistinguishable and undetectable above that. From the data in Fig. 2(b), we extrapolated to T = 0 K, $H_{c1}^*(0) = 9.27$ mT, $H_p(0) = 49$ mT, $H_v(0) = 60$ mT, and $H_{c2}(0) = 67$ mT. Using the demagnetizing factor of a sphere, we obtain the lower critical field $H_{c1}(0) = 13.9$ mT.

Accurate knowledge of H_{c1} is important for the rest of our analysis, so we have checked our results against others from the literature: $H_{c1}^* = 7$ mT was obtained in specific heat measurements at 60 mK in samples with a slab geometry [19]. A value of $H_{c1}^* = 7 \text{ mT}$ was determined using SQUID magnetometry at T = 20 mK [43], while thermal-conductivity measurements find $H_{c1}^* = 8 \text{ mT}$ at T = 320 mK [22] and 12 mT for a long plate-shaped sample parallel oriented to the field at T = 300 mK [23]. Those values reveal a strong influence of the geometry of the sample on the value of H_{c1}^* . For comparison, we use an estimated typical demagnetizing factor $N \approx 0$ for a plate-shaped sample oriented parallel to the field, while for a slab with proportions of $a \times b \times c =$ $0.5 \times 0.5 \times 0.33$, $N \approx 0.5$ (see the SM [38]). This leads to estimated H_{c1} values between 13 mT and 16 mT for these measurements. Our value of $H_{c1}(0) = 13.9 \text{ mT}$ is hence in very good agreement with previous measurements using different techniques.

We can perform a still more rigorous check of the accuracy of our critical field values by estimating some fundamental superconducting parameters, calculating the thermodynamic critical field, and comparing it to that deduced from specific heat data. To this end we use

$$H_{c2} = \frac{\Phi_0}{2\pi\xi^2}$$
 and $H_{c1} = \frac{\Phi_0}{4\pi\lambda^2}C(\kappa)$, (1)

which relate the two critical fields with the penetration depth λ and the coherence length ξ of a type-II superconductor. Here, Φ_0 is the flux quantum and $\kappa = \lambda/\xi > \kappa_c = \frac{1}{\sqrt{2}}$ the Ginzburg-Landau parameter. The function $C(\kappa)$ was determined numerically from the solution of the Ginzburg-Landau equations by Brandt [44], who also gave a simple analytic interpolation formula that is highly accurate ($<10^{-3}$) for all values $\kappa > \kappa_c$ and reproduces the limits $C(\kappa_c) = 1$ and $C(\kappa \gg 1) = \log_e \kappa + 0.49693$ [45] (see SM [38]).

Using Eq. (1) and our result for $H_{c2}(0)$ and $H_{c1}(0)$ yields $\xi_0 = 70$ nm and $\lambda_0 = 134$ nm for the zero-temperature values of coherence length and penetration depth, respectively. This is consistent with the measurement by muon spin rotation $(\mu SR) \lambda_0 = 126$ nm and the calculation of the contribution of each band to the magnetic penetration depth based on angleresolved photoelectron spectroscopy (ARPES) $\lambda_0 = 130 \text{ nm}$ [46]. (Note that μ SR uses data above H_{c1} and ARPES uses the normal state band structure to extract λ_0 . Therefore, these results are independent from the geometry of the sample.) From λ_0 and ξ_0 we obtain $\kappa_0 \equiv \kappa(T=0) = 1.92$; i.e., Sr₂RuO₄ is not a strong type-II superconductor as the value of the Ginzburg-Landau parameter is not far from the limit to type-I superconductivity ($\kappa_c \approx 0.71$). These results yield the thermodynamic critical field $H_c(0) = \frac{H_c(0)}{\sqrt{2\kappa_0}} = 24.7$ mT, in very good agreement with $H_c = (23 \pm 2)$ mT deduced from specific heat data [38]. In summary, our measurements give precise values of H_{c1} and superconducting parameters in full agreement with previous results.

Through Eq. (1), the temperature dependencies of $H_{c1}(T)$ and $H_{c2}(T)$ are directly related to the *T* variation of the in-plane penetration depth $\lambda(T)$ and coherence length $\xi(T)$, respectively. In the SM [38] we show both length scales as a function of temperature. Focusing on the temperature



FIG. 3. $\Delta\lambda/\lambda_0$ vs $(T/T_c)^2$ in Sr₂RuO₄. The gray and violet lines show the expected behavior of $\Delta\lambda/\lambda_0$ for vertical and horizontal line nodes in the nonlocal electrodynamic limit, respectively. The dotted lines are for the BCS value of the gap $2\Delta_{0,BCS}/(k_BT_c) = 3.53$ and the full lines for $2\Delta_{0,C}/(k_BT_c) = 3.16$ extracted from specific heat [38,47]. The inset shows the data in a larger temperature range with a straight line indicating the temperature range up to $\approx 0.5T_c$ in which a T^2 behavior is found.

variation of the penetration depth leads us to plot $\Delta\lambda(T)/\lambda_0$ in Fig. 3 showing a clear T^2 dependence below ≈ 0.6 K.

Discussion. One possible explanation of the T^2 dependence, which is appropriate for systems such as the cuprates, is the effect of impurities in superconductors with line nodes. Hirschfeld and Goldenfeld showed that for an unconventional superconductor with vertical line nodes, the scattering due to impurities would lead to a change in the temperature dependence of $\lambda(T) \propto T$ to T^2 below a crossover temperature $T_{imp}^* \approx 0.83(\Gamma\Delta_0)^{1/2}$, where Γ is the scattering rate and Δ_0 is the magnitude of the superconducting gap [36]. In our extremely clean sample with $T_c \approx 1.5$ K, we obtain $T_{imp}^* \leqslant 0.05T_c$, such that the effect of impurities cannot explain the observed quadratic T dependence across the wide range of temperatures.

There have been several previous reports of T^2 behavior of $\Delta\lambda/\lambda_0$ in Sr₂RuO₄ within the Meissner state [18,48,49]. However, these were on crystals with lower T_c and higher T_{imp}^* . For the samples used for those measurements, we estimate $T_{\rm imp}^* \approx 0.2T_c$, using the relationship between the scattering rate Γ and T_c [50]. It was therefore uncertain whether the observed behavior should be attributed to impurity scattering or not. In our data, there is no such ambiguity: the data shown in Fig. 3 are in the clean limit. At first sight, this presents a puzzle. In the local electrodynamics, applicable to most unconventional superconductors, only point nodes can give a T^2 dependence of the penetration depth [51]. In contrast, point nodes are ruled out for Sr₂RuO₄ by most existing experimental results. Thermodynamic and transport properties in the low-temperature limit (as listed in the introduction) as well as the observation of an NMR spin-relaxation rate $1/T_1 \propto T^3$ in clean samples are fully consistent with line nodes but not with point nodes $(1/T_1 \propto T^5)$ [52]. Furthermore, Knight shift measurements in NMR and NQR give strong evidence for a singlet, even-parity order parameter. By symmetry, all the even-parity states under discussion as potential order parameters in this material either have line nodes or are fully gapped, giving only *T*-linear or exponential dependencies of $\Delta\lambda/\lambda_0$ at low temperatures.

The resolution to this apparent paradox lies in considering nonlocal electrodynamics. Nonlocal effects in the electromagnetic response below T_c go back to the analysis of Pippard for type-I superconductors [53], where spatial modes of the magnetic field with wavelengths smaller than the coherence length give rise to reduced screening currents that shield the external field, affecting the length scale up to which the field can penetrate. As shown in Ref. [33], a nonlocal response can also play a role in type-II superconductors if they possess nodes of the gap function. In the ground state, the entire Fermi surface contributes to the phase stiffness and the relative importance of the nodal points is negligible. However, the temperature dependence of the transverse current response $K_{\perp}(q)$ and hence of the penetration depth is dominated by thermal quasiparticle excitations near the nodes. With $\xi_T \sim v_F/(k_B T)$, the result by Kosztin and Leggett for a system with vertical line nodes can then be written in the form

$$\frac{\Delta\lambda(T)}{\lambda_0} \sim \left. \frac{\Delta\lambda(T)}{\lambda_0} \right|_{\rm loc} \frac{\lambda_0}{\xi_T},\tag{2}$$

where $\Delta\lambda(T)/\lambda_0|_{\text{loc}} = \log_e 2\frac{T}{\Delta_0}$ is the well-established result for the local electromagnetic response of a clean nodal superconductor. Hence, it follows that in the nonlocal limit $\Delta\lambda(T)/\lambda_0 \propto \kappa_0 T^2/\Delta_0^2$. Here Δ_0 is the gap amplitude that also enters in the low-temperature density of states $\rho(\omega) = \rho_F |\omega|/\Delta_0$ of the nodal superconductor. This nonlocality is tied to the condition $\lambda_0 \ll \xi_T$, which translates to $T \ll T_{\text{nl}}^* = \Delta_0/\kappa_0$. In the SM [38], we estimate T_{nl}^* to be as high as $0.8T_c$.

Intriguingly, the nonlocal regime offers a qualitative distinction between the effects of vertical and horizontal line nodes on $\Delta\lambda/\lambda_0$, because horizontal nodes lie in the plane of the screening supercurrents. This regime was analyzed by Kusunose and Sigrist in Ref. [37] and their result for horizontal line nodes can be formulated as

$$\frac{\Delta\lambda(T)}{\lambda_0} \sim \left. \frac{\Delta\lambda(T)}{\lambda_0} \right|_{\rm loc} \frac{\lambda_0}{\xi_T} \log_e \frac{\xi_T}{\lambda_0}. \tag{3}$$

Hence, the suppression of the electromagnetic response by quasiparticles with horizontal nodes is less strong and, with $\Delta\lambda(T)/\lambda_0 \propto T^2 \log_e(T_{nl}^*/T)$, in principle distinguishable from those of vertical line nodes where $\Delta\lambda(T)/\lambda_0 \propto T^2$. While these qualitative arguments are limited to the regime of lowest temperatures, in the SM [38], we demonstrate the full analysis of the electromagnetic response for horizontal and vertical line nodes. The theoretical curves, shown in Fig. 3 alongside the experimental data, depend on the two dimensionless numbers κ_0 , which we determined earlier, and $2\Delta_0/(k_BT_c)$. For the theoretical curves shown in Fig. 3, we used $\kappa_0 = 1.92$ and two values for $2\Delta_0$: the BCS value $2\Delta_{0,\text{BCS}}/(k_BT_c) = 3.53$ and the value extracted from specific heat $2\Delta_{0,C}/(k_BT_c) = 3.16$ [47]. The predictions for vertical line nodes are a systematically better match to the data than those for horizontal line nodes. This remains true if we allow

 $2\Delta_0/(k_BT_c)$ as an open fit parameter. Our conclusions are robust as long as there are no strong variations of the gap amplitude among the various Fermi surface sheets. Then, the data are compatible with the existence of purely vertical line nodes but not compatible with order parameters containing solely horizontal nodes. For mixed order parameters with both types of node, explicit calculations of $\Delta\lambda/\lambda_0$ would be required to determine whether or not the predictions are compatible within experimental error with our data.

In conclusion, we have used measurements of H_{c1} and H_{c2} on an extremely high purity single crystal of Sr₂RuO₄ to show that its in-plane low-temperature coherence length $\xi_0 = 70$ nm and penetration depth $\lambda_0 = 134$ nm, and that $\Delta\lambda/\lambda$ varies as T^2 in the clean limit. Analysis of our results using nonlocal electrodynamics confirms that the observations are compatible with vertical line nodes in its superconducting order parameter. Our measurements and analysis are of relevance to the ongoing quest to understand the order parameter symmetry of Sr₂RuO₄, and invite careful measurement and analysis of $\Delta\lambda/\lambda$ in other unconventional superconductors in which the nonlocal regime is experimentally accessible.

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Note added. We note that Ref. [54] reports measurements of $\Delta\lambda$ as a function of uniaxial pressure Sr₂RuO₄ that also highlight the importance of nonlocal effects. Furthermore, Ref. [55] shows calculations of penetration depth in the nonlocal limit comparing different superconducting order parameters in B_{1g} symmetry with purely vertical line nodes.

Acknowledgements. We thank D. Bonn, J.-P. Brison, S. Brown, P. Hirschfeld, G. Palle, R. Prozorov, A. Ramires, and H. Suderow for stimulating exchange and helpful discussions. E.H. acknowledges funding from Deutsche Forschungsgemeinschaft (DFG) via the CRC 1143-247310070 (project C10) and the Würzburg-Dresden cluster of excellence EXC 2147 ct.qmat Complexity and Topology in Quantum Matter, Project No. 390858490. J.S. and A.P.M. were supported by the DFG through CRC 288-422213477 Elasto-Q-Mat (projects A07 and A10, respectively). This work is supported by JSPS KAKENHI (Grants No. JP18K04715, No. JP21H01033, No. JP22H01168, and No. JP22K19093). J.S. also acknowledges the hospitality of KITP, where part of the work was done. KITP is supported in part by the National Science Foundation under Grants No. NSF PHY1748958 and No. NSF PHY-2309135.

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