Nodeless superconductivity in the kagome metal CsV$_3$Sb$_5$

Weiyan Duan$^{1,2}$, Zhiyong Nie$^{1,2}$, Shuaishuai Luo$^{1,2}$, Fanghang Yu$^3$, Brened R. Ortiz$^4$, Lichang Yin$^{1,2}$, Hang Su$^{1,2}$, Feng Du$^{1,2}$, An Wang$^{1,2}$, Ye Chen$^{1,2}$, Xin Lu$^{1,2}$, Jianjun Ying$^{3}$, Stephen D. Wilson$^4$, Xianhui Chen$^{3,5,6}$, Yu Song$^{1,2}$*, and Huiqiu Yuan$^{1,2,6,7}$*

$^1$Center for Correlated Matter and Department of Physics, Zhejiang University, Hangzhou 310058, China;
$^2$Zhejiang Province Key Laboratory of Quantum Technology and Device, Department of Physics, Zhejiang University, Hangzhou 310058, China;
$^3$Hefei National Laboratory for Physical Sciences at Microscale and Department of Physics, and CAS Key Laboratory of Strongly-coupled Quantum Matter Physics, University of Science and Technology of China, Hefei 230026, China;
$^4$Materials Department and California Nanosystems Institute, University of California Santa Barbara, Santa Barbara CA 93106, USA;
$^5$CAS Center for Excellence in Quantum Information and Quantum Physics, Hefei 230026, China;
$^6$Collaborative Innovation Center of Advanced Microstructures, Nanjing 210093, China;
$^7$State Key Laboratory of Silicon Materials, Zhejiang University, Hangzhou 310058, China

Received July 3, 2021; accepted July 8, 2021; published online July 27, 2021

The recently discovered kagome metal series AV$_3$Sb$_5$ (A=K, Rb, Cs) exhibits topologically nontrivial band structures, chiral charge order and superconductivity, presenting a unique platform for realizing exotic electronic states. The nature of the superconducting state and the corresponding pairing symmetry are key questions that demand experimental clarification. Here, using a technique based on the tunneling diode oscillator, the magnetic penetration depth $\Delta \lambda (T)$ of CsV$_3$Sb$_5$ was measured down to 0.07 K. A clear exponential behavior in $\Delta \lambda (T)$ with marked deviations from a $T$ or $T^2$ temperature dependence was observed at low temperatures, indicating an absence of nodal quasiparticles. Temperature dependence of the superfluid density and electronic specific heat can be described by two-gap $s$-wave superconductivity, consistent with the presence of multiple Fermi surfaces in CsV$_3$Sb$_5$. These results evidence nodeless superconductivity in CsV$_3$Sb$_5$ under ambient pressure, and constrain the allowed pairing symmetry.

kagome superconductor, order parameter, penetration depth

PACs number(s): 74.25.-q, 74.25.Bt, 74.25.Ha, 74.20.Rp

Citation: W. Duan, Z. Nie, S. Luo, F. Yu, B. R. Ortiz, L. Yin, H. Su, F. Du, A. Wang, Y. Chen, X. Lu, J. Ying, S. D. Wilson, X. Chen, Y. Song, and H. Yuan, Nodeless superconductivity in the kagome metal CsV$_3$Sb$_5$, Sci. China-Phys. Mech. Astron. 64, 107462 (2021), https://doi.org/10.1007/s11433-021-1747-7

1 Introduction

The unique geometry of the kagome lattice leads to magnetic frustration [1-3], topologically nontrivial electronic structures [4-6], and both electronic [6-8] and magnon [9-11] flat bands. Superconductivity with exotic pairing symmetries and properties were also predicted for the kagome lattice [12-16], although physical realizations of such exotic superconductors have been limited.

The recent discovery of superconductivity in the two-dimensional kagome metal series AV$_3$Sb$_5$ (A = K, Rb, Cs) provides a much-desired platform to investigate potentially
exotic superconducting states on the kagome lattice [17-20]. In addition to superconductivity, these systems also exhibit a chiral charge order with unusual characteristics [21-25], topological band crossings [17, 18], and a giant anomalous Hall effect in the absence of magnetic local moments [18, 26-28]. The nature of the superconducting state in the presence of these electronic states, remains to be clarified. Nonetheless, tantalizing evidence for an unusual superconducting state has emerged from the observation of spin-triplet supercurrent in $K_1-xV_3Sb_5$ Josephson junctions [29], possible Majorana bound states inside the superconducting vortex cores of $CsV_3Sb_5$ [30], and superconducting domes around the suppression of charge order in the temperature-pressure phase diagrams of $AV_3Sb_5$ [31-34].

A fundamental question that needs to be addressed is the pairing symmetry of the $AV_3Sb_5$ series, and specifically whether it is unconventional with a sign-changing superconducting order parameter. While direct phase-sensitive evidence for such a sign-change is difficult to obtain, for a number of pairing symmetries the sign-change mandates nodes in the superconducting order parameter. Therefore, detecting the presence or absence of such nodal quasiparticles is critical for determining the pairing symmetry of newly discovered superconductors. In the case of $CsV_3Sb_5$, while thermal conductivity measurements down to 0.15 K indicated possible nodal quasiparticles [31], the tunneling spectrum measured by scanning tunneling microscopy could be described by an $s$-wave gap [30]. To convincingly address the existence of nodal quasiparticles and clarify the pairing symmetry of the $AV_3Sb_5$ series, experiments highly sensitive to low-energy electronic excitations are imperative.

In this article, the magnetic penetration depth and specific heat of the kagome metal $CsV_3Sb_5$ were measured to study its superconducting gap structure. The change of the magnetic penetration depth reveals a clear exponential behavior down to 0.07 K, and deviates significantly from a $T$ or $T^2$ temperature dependence. Such a behavior suggests an absence of nodal quasiparticles, and instead points to fully-gapped superconductivity in $CsV_3Sb_5$ under ambient pressure. By analyzing the derived superfluid density and electronic specific heat, a two-gap $s$-wave superconducting order parameter is found to capture the experimental data. These results provide evidence for nodeless superconductivity in $CsV_3Sb_5$, and rule out pairing with symmetry-enforced nodes.

2 Experimental details

Single crystals of $CsV_3Sb_5$ were synthesized using the self-flux method, at the University of Science and Technology of China (sample A) [28] and the University of California, Santa Barbara (sample B) [18]. Eight samples were studied with various approaches, and are labeled samples #A-1 to #A-5, and samples #B-1 to #B-3. The $ab$-plane electrical resistivity $\rho(T)$ was measured in a $^3$He cryostat, using a standard four-probe method. Specific heat was measured using a Quantum Design Physical Property Measurement System (PPMS) with a $^3$He insert, using a standard pulse relaxation method. The change in the London penetration depth $\Delta \lambda(T) = \lambda(T) - \lambda(0)$ was measured using a tunnel diode oscillator (TDO) [35-37], with an operating frequency of about 7 MHz. Here $\Delta \lambda(T)$ can be obtained from the TDO frequency shift $\Delta f(T)$ through $\Delta \lambda(T) = G \Delta f(T)$, where the $G$ factor is determined by the sample geometry [36]. The TDO method can precisely measure the temperature dependence of $\Delta \lambda(T)$, offering a powerful probe of low-energy excitations in the superconducting state. TDO measurements down to 0.35 and 0.07 K, with noise levels as low as 0.1 and 0.5 Hz, were carried out in $^3$He and dilution refrigerators, respectively. The ac field generated by the coil (20 mOe) is far below the lower critical field $H_{c1}$ of $CsV_3Sb_5$, ensuring that the sample remains in the full Meissner state once the temperature is slightly below $T_c$. In our TDO measurements, the samples are mounted on a sapphire stage connected to a copper stage, where the thermometer is mounted. Particular attention was paid to possible thermal gradients in our measurements. Dimensions of the four samples measured using the TDO are $\approx 870 \mu m \times 870 \mu m \times 110 \mu m$ (sample #A-3), $\approx 870 \mu m \times 870 \mu m \times 40 \mu m$ (sample #A-4), $\approx 640 \mu m \times 640 \mu m \times 50 \mu m$ (sample #A-5), and $\approx 830 \mu m \times 830 \mu m \times 50 \mu m$ (sample #B-3). Samples #A-3, #A-4 and #B-3 were studied with applied field along the $c$-axis, with respective estimated $G$ factors being 7.8, 9.1 and 9.8 $\mu$Hz. Sample #A-5 was studied with field perpendicular to the $c$-axis, with its $\Delta \lambda(T)$ scaled to that of sample #A-3, due to difficulties in accurately determining the $G$ factor for a thin sample.

3 Results

The inset of Figure 1(a) shows the electrical resistivity $\rho(T)$ of $CsV_3Sb_5$ from 300 K down to 0.45 K. A clear kink can be observed around 94 K, due to the onset of charge order [17, 18]. The temperature evolution of $\rho(T)$ for samples #A-1 and #B-1 are highly similar, with residual resistivity ratios (RRR) of $\approx 57$ and $\approx 74$, respectively. Figure 1(a) zooms into $\rho(T)$ below 4.5 K, where a clear superconducting transition can be observed, which onsets around 3.5 K and zero resistance appears below $2.7 \mu$Ω cm in both samples. The residual resistivities just above the onset of superconductivity are comparable, close to 1 $\mu$Ω cm in both samples. Measurements of
the low temperature specific heat \(C(T)/T\) are shown in the inset of Figure 1(b) for samples #A-2 and #B-2, demonstrating the appearance of bulk superconductivity below \(T_c \approx 2.7\) K. In the normal state, the specific heat can be modeled using \(C(T)/T = \gamma + \beta T^2 + \delta T^4\), with \(\gamma = 20.03\) mJ mol\(^{-1}\) K\(^{-2}\), \(\beta = 3.306\) mJ mol\(^{-1}\) K\(^{-4}\), and \(\delta = 26.78\) \(\mu\)J mol\(^{-1}\) K\(^{-6}\) (solid line in the inset of Figure 1(b)). Here \(\gamma\) is the Sommerfeld coefficient, and the other two parameters characterize the contribution from phonons. After subtracting the phonon contribution, the electronic specific heat \(C_{el}(T)/\gamma T\) can be obtained, shown in Figure 1(b). Analysis of \(C_{el}(T)/\gamma T\) will be further discussed below. Combining the coherence length \(\xi \approx 26\) nm from \(H_c2 = 0.47\) T [18, 28]), residual resistivity \(\rho_0\), and the Sommerfeld coefficient \(\gamma\), mean free paths of 680 and 830 nm are estimated [38, 39] for samples #A-1 and #B-1, respectively. These values are much larger than the coherence length, indicating that the samples are in the clean limit. These characterizations indicate that both samples A and B are of high quality and behave similarly.

![Figure 1](image_url)

**Figure 1** (Color online) Temperature dependence of the low temperature (a) resistivity \(\rho(T)\) and (b) electronic specific heat \(C_{el}(T)/\gamma T\) for CsV\(_3\)Sb\(_5\). The inset in (a) shows \(\rho(T)\) from 0.45 K up to 300 K. The inset in (b) shows the specific heat \(C(T)/T\), with the blue solid line being a fit to the normal state specific heat, including contributions from electrons and phonons. The dashed and solid lines in (b) are fits to a single-gap and two-gap s-wave superconducting model, respectively.

Figure 2 shows the temperature dependence of the magnetic penetration depth for CsV\(_3\)Sb\(_5\). The inset in Figure 2(a) shows \(\Delta \lambda(T)\) for samples #A-3 and #A-5 from 4.5 K down to 0.07 K, exhibiting clear reductions upon cooling due to superconductivity, consistent with resistivity and specific heat measurements in Figure 1. Low-temperature \(\Delta \lambda(T)\) with applied field along and perpendicular to the \(c\)-axis are compared in Figure 2(a) for samples #A-3 and #A-5, revealing almost identical behaviors, suggesting that the superconducting state is unlikely to be strongly anisotropic. Figure 2(b) compares the low temperature penetration depth \(\Delta \lambda(T)\) for three CsV\(_3\)Sb\(_5\) samples with field applied along the \(c\)-axis. The data for different samples almost overlap, confirming the superconducting properties of samples A and B are similar, and demonstrate that our results are reproducible and reflect the intrinsic behavior of CsV\(_3\)Sb\(_5\).

For a nodal superconductor in the clean limit, it is expected that the magnetic penetration depth exhibits a power-law behavior in the low temperature limit, i.e., \(\Delta \lambda \sim T^n\), with \(n = 1\) and 2 respectively corresponding to line nodes and point nodes being present in the gap structure. From Figure 2(b), it can be seen that the experimentally measured \(\Delta \lambda(T)\) obviously deviates from a \(T\) or \(T^2\) behavior, but is reasonably described by \(\Delta \lambda(T) \sim T^{2.9}\) with a slight deviation at low temperatures. To further analyze the power law dependence of \(\Delta \lambda(T)\), the experimental data is fit by \(\Delta \lambda(T) \sim T^n\) from 0.07 K to various temperatures, with the best fit \(n\) shown in the inset of Figure 2(b). It can be seen that the exponent \(n \approx 3\) appears for \(T > 0.2T_c\), and becomes significantly enhanced at lower temperatures. Such a power law behavior \((n \geq 3)\), in particular the progressive increase of \(n\) with decreasing temperature, is consistent with an exponential behavior, and suggests an absence of gap nodes in the superconducting state of CsV\(_3\)Sb\(_5\).

To further analyze the magnetic penetration depth, \(\Delta \lambda(T)\) is fit to an s-wave gap at low temperatures, with

\[
\Delta \lambda(T) \sim T^{-\frac{2}{3}} \exp\left(\frac{-\Delta(0)}{k_B T}\right),
\]

where \(\Delta(0)\) is the gap value at zero temperature. It can be seen that such an s-wave model fits the experimental data well at low temperatures, providing strong evidence for nodeless superconductivity in CsV\(_3\)Sb\(_5\). The derived small superconducting gap of \(\Delta(0) = 0.59k_B T_c\) indicates a saturation of \(\Delta \lambda(T)\) at very low temperatures, as seen in Figure 2(b), and the presence of multiple superconducting gaps.

To further extract information about the superconducting state, the normalized superfluid density is obtained through \(\rho_s(T) = [\rho(0)/\rho(T)]^2\). Here the value of the zero-temperature penetration depth \(\rho_s(0) = 387\) nm was estimated using \(\rho_s(0) = \sqrt{\phi_0 H_{c2}(0)} / \sqrt{24\pi M(0)}\) [40], where \(\phi_0\) is the magnetic-flux
quantum, the Sommerfeld coefficient $\gamma$ is obtained from specific heat measurements (inset of Figure 1(b)), $H_{c2}(0) = 0.47 \, T$ [18, 28] and the weak-coupling limit of BCS theory with $\Delta(0) = 1.76 \, k_B T_c$ is assumed. $\rho_s$ obtained this way is shown as a function of the reduced temperature $T/T_c$ for sample #A-3 in Figure 3(a), and is fit to several different models of the superconducting gap function $\Delta_k$, which is related to $\rho_s$ through [41]

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

where $f(E, T) = \left[ 1 + \exp\left( \frac{E}{k_B T} \right) \right]^{-1}$ is the Fermi-Dirac distribution, and $\langle \ldots \rangle_{FS}$ represents averaging over the Fermi surface. Temperature dependence of the superconducting order parameter is approximated using [42]

$$\Delta(T) = \Delta(0) \tanh \left[ 1.82 \, [1.018 \, (T_c/T - 1)]^{0.51} \right].$$

As can be seen, the data are clearly incompatible with a single-gap $s$-wave model ($\Delta_s = \Delta_0$), a $p$-wave model with point nodes ($\Delta_k = \Delta_0 \sin \theta$), and a $d$-wave model with line nodes ($\Delta_k = \Delta_0 \cos 2\phi$). Here, $\theta$ is the polar angle and $\phi$ is the azimuthal angle. These models exhibit significant deviations from the data, especially at low temperatures (inset of Figure 3(a)). On the other hand, a two-gap $s$-wave model describes the data well over the entire temperature range (solid line in Figure 3(a)). The fit gap values are $\Delta_1(0) = 1.42 \, k_B T_c$ and $\Delta_2(0) = 0.58 \, k_B T_c$, with weights of the two gaps being 87% and 13%, respectively. The deduced small gap of $\Delta_2 = 0.58 \, k_B T_c$ is consistent with $\Delta = 0.59 \, k_B T_c$ derived from fitting $\Delta(T)$ at low-temperature (Figure 2(b)). Considering possible uncertainties in the estimate of $\Delta(0)$ and to test the robustness of our conclusions, $\Delta(0)$ is varied by $\pm 20\%$ from 387 nm and the analyses are repeated, with results shown in Figure 3(b). It is found that the two-gap $s$-wave model describes $\rho_s$ well in all cases (solid lines), while other models in Figure 3(a) exhibit clear deviations. For $\Delta(0) = 300 \, nm$, $\Delta_1(0) = 1.23 \, k_B T_c$ and $\Delta_2(0) = 0.50 \, k_B T_c$ are obtained, while $\Delta_1(0) = 1.63 \, k_B T_c$ and $\Delta_2(0) = 0.66 \, k_B T_c$ are obtained for $\Delta(0) = 470 \, nm$. Our analyses of the superfluid density therefore support the idea that CsV$_3$Sb$_5$ exhibits

![Figure 2](image1.png)

![Figure 3](image2.png)
multiband nodeless superconductivity.

To confirm the above conclusion, the low temperature electronic specific heat \( C_\text{el}(T)/\gamma T \) of CsV\(_3\)Sb\(_5\) is analyzed, shown in Figure 1(b). Similar to the superfluid density, it is found that a single s-wave gap does not capture the behavior of \( C_\text{el}(T)/\gamma T \) (dashed line in Figure 1(b)), while a two-gap s-wave model gives an excellent description of the experimental data over the full temperature range (solid line in Figure 1(b)). The fit gap values are \( \Delta_1(0) = 1.62 \, k_B T_c \) and \( \Delta_2(0) = 0.63 \, k_B T_c \), with weights of 79\% and 21\%, respectively. These values are in reasonable agreement with analysis of the superfluid density, and supports the notion that CsV\(_3\)Sb\(_5\) exhibits fully-gapped multiband superconductivity. It is noted that values of the larger gap \( \Delta_1(0) \) from our analysis of the superfluid density and electronic specific heat are close to but still slightly smaller than the BCS value of 1.76 \( k_B T_c \), the origin of which is not clear. It is possible that the contribution of an additional band or the effect of an anisotropic gap may lead to such a weak deviation.

### 4 Discussion and conclusion

The low temperature exponential behavior of \( \Delta_1(T) \) provides evidence for nodeless superconductivity, while the extracted small gap and analysis of both the superfluid density and the electronic specific heat evidence multiband superconductivity. This is consistent with the presence of multiple Fermi surfaces revealed by angle-resolved photoemission spectroscopy measurements and electronic structure calculations [18]. Such a picture is supported by the consideration that the pocket around \( \Gamma \) and the Fermi surfaces around \( M \) are mainly associated with Sb \( p_z \) and V \( d \) orbitals, respectively [25], and are likely associated with superconducting gaps of different sizes. It should be noted that our analyses do not exclude an anisotropic s-wave superconducting gap. However, considering that this is a multiband system, and the penetration depth along different directions are similar (Figure 2(a)), multi-gap superconductivity appears more likely.

Since a circular Fermi surface around \( \Gamma \) is present in CsV\(_3\)Sb\(_5\) [18], all even-parity basis gap functions associated with the \( D_{5h} \) point group would lead to symmetry-enforced nodes, except that of the \( A_{1g} \) channel [43]. Therefore, the finding of nodeless superconductivity indicates the pairing symmetry of CsV\(_3\)Sb\(_5\) belongs to the even-parity \( A_{1g} \) representation, or odd-parity nodeless representations. While our results can be described by a superconducting state with two s-wave gaps, and is supported by the presence of multiple Fermis surfaces in CsV\(_3\)Sb\(_5\), our measurements do not rule out some exotic unconventional superconducting states, for example, spin-triplet nodeless superconductivity [44], \( s^\pm \)-pairing as in the iron-based superconductors [45], and pairing with band-mixing as suggested for CeCu\(_2\)Si\(_2\) [46]. Further studies are needed to fully elaborate the pairing state of the AV\(_2\)Sb\(_5\) series.

Upon pressure-tuning, superconductivity in CsV\(_3\)Sb\(_5\) exhibits significant modulations and exhibit two superconducting domes [31-33], suggesting the presence of competing superconducting ground states. As our results evidence a nodeless superconducting state under ambient pressure, whether the competing superconducting state stabilized under pressure remains nodeless or exhibits symmetry-enforced nodes, becomes an important question to be addressed in future works.

In conclusion, the superconducting pairing symmetry of CsV\(_3\)Sb\(_5\) single crystals is probed through magnetic penetration depth measurements down to 0.07 K. A clear exponential behavior at low temperatures provides evidence for nodeless superconductivity in CsV\(_3\)Sb\(_5\) under ambient pressure. Temperature dependence of the superfluid density and electronic specific heat can be described by two-gap s-wave superconductivity, consistent with the presence of multiple Fermi surfaces in CsV\(_3\)Sb\(_5\). Our results are inconsistent with pairing with symmetry-enforced nodes in CsV\(_3\)Sb\(_5\), but do not rule out fully-gapped unconventional superconductivity.

This work was supported by the National Key R\&D Program of China (Grant Nos. 2017YFA0303100, and 2016YFA0300202), Key R\&D Program of Zhejiang Province, China (Grant No. 2021C01002), and National Natural Science Foundation of China (Grant Nos. 11974306, and 12034017). S. D. Wilson and B. R. Ortiz gratefully acknowledge support via the UC Santa Barbara NSF Quantum Foundry funded via the Q-AMASE-I Program under award DMR-1906325. B. R. Ortiz also acknowledges support from the California NanoSystems Institute through the Elings Fellowship Program.

---

1. I. Syozi, Prog. Theor. Phys. 6, 306 (1951).
2. T. H. Han, J. S. Helton, S. Chu, D. G. Nocera, J. A. Rodriguez-Rivera, C. Broholm, and Y. S. Lee, Nature 492, 406 (2012), arXiv: 1307.5047.
3. C. Broholm, R. J. Cava, S. A. Kivelson, D. G. Nocera, M. R. Norman, and T. Senthil, Science 367, eaay0688 (2020).
4. L. Ye, M. Kang, J. Liu, F. von Cube, C. R. Wicker, T. Suzuki, C. Joziwiaik, A. Bostwick, E. Rotenberg, D. C. Bell, L. Fu, R. Comin, and J. G. Checkelsky, Nature 555, 638 (2018), arXiv: 1709.10007.
5. E. Liu, Y. Sun, N. Kumar, L. Muechler, A. Sun, L. Jiao, S. Y. Yang, D. Liu, A. Liang, Q. Xu, J. Kroder, V. Sujb, H. Bormann, C. Shekhar, Z. Wang, C. Xi, W. Wang, W. Schnelle, S. Wirth, Y. Chen, S. T. B. Goennenwein, and C. Felsers, Nat. Phys. 14, 1125 (2018), arXiv: 1712.06722.
6. M. Kang, L. Ye, S. Fang, J. S. You, A. Levitan, M. Han, J. I. Facio, C. Joziwiaik, A. Bostwick, E. Rotenberg, M. K. Chan, R. D. McDonald, D. Graf, K. Kazatschew, E. Vescovo, D. C. Bell, E. Kaxaras, J. van den Brink, M. Richter, M. Prasad Gumire, J. G. Checkelsky, and R. Comin, Nat. Mater. 19, 163 (2020), arXiv: 1906.02167.
7. Z. Lin, J. H. Choi, Q. Zhang, W. Qin, S. Yi, P. Wang, L. Li, Y. Wang, H. Zhang, Z. Sun, L. Wei, S. Zhang, T. Guo, Q. Lu, J. H. Cho, C. Zeng, and Z. Zhang, Phys. Rev. Lett. 121, 096401 (2018).
8. J. X. Yin, S. S. Zhang, G. Chang, Q. Wang, S. S. Tsirkin, Z. Guguchia, B. Lian, H. Zhou, K. Jiang, I. Belopolski, N. Shumiya, D. Mulher, M.
