Ultralow-Temperature Heat Transport Evidence for Residual Density of States in the Superconducting State of CsV_3Sb_5

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V-based kagome superconductors AV_3Sb_5 (A = K, Rb, and Cs) host a charge density wave (CDW) and a topological nontrivial band structure, thereby providing a great platform to study the interplay of superconductivity (SC), CDW, frustration, and topology. Here, we report ultralow-temperature thermal conductivity measurements of CsV_3Sb_5 and Ta-doped $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ and scanning tunneling microscopy (STM) measurements of CsV_3Sb_5. The finite residual linear term of thermal conductivity at zero magnetic field suggests the existence of a residual density of states (DOS) in the superconducting state of CsV_3Sb_5. This is supported by the observation of non-zero conductance at zero bias in STM spectrum at an electronic temperature of 90 mK. However, in $Cs(V_{0.86}Ta_{0.14})_3Sb_5$, which does not have CDW order, there is no evidence for the residual DOS. These results show the importance of CDW order for the residual DOS, and that a nodal s-wave gap or residual Fermi arc may be the origin of the residual DOS in such an unusual multiband kagome superconductor, CsV_3Sb_5.

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Superconductivity (SC) has been discovered in a family of vanadium-based kagome compounds, AV_3Sb_5 (A = K, Rb, and Cs).[1–4] The superconducting transition temperatures T_c are 0.93, 0.92, and 3 K for A = K, Rb, and Cs, respectively.[3–5] Although there are no local magnetic moments,[6] these V-based superconductors exhibit charge density wave (CDW) order at 78, 103, and 94 K.[1–4] Various measurements have revealed a 2×2 in-plane superlattice modulation associated with the CDW transition in AV_3Sb_5; however, it remains unclear whether the three-dimensional CDW is $2 \times 2 \times 2$ or $2 \times 2 \times 4$ along the c axis.[5,7–13] Within the CDW state, many exotic properties, such as giant anomalous Hall effect,[7,14–16] time-reversal symmetry breaking,[17–19] possible loop current ordering,[20,21] and electronic nematicity,[21–23] have been discovered.

When AV_3Sb_5 enters a superconducting state, its properties attract considerable attention. Two superconducting domes can be obtained with increasing pressure or doping,[24–29] and the CDW order was intertwined with the superconductivity in an unconventional manner.[30–32] The presence of an unusual pair density wave (PDW) makes the complexity of a superconducting state.[8,33–36] Regarding the superconducting gap structure, some early studies claimed that CsV_3Sb_5 is fully gapped,[37–41] whereas others have suggested nodal superconductivity in AV_3Sb_5.[24,42] The latest angle-resolved photoemission spectroscopy (ARPES) measurements of high-quality CsV_3Sb_5 single crystals at 2 K revealed that the SC gap in the β Fermi surface (FS) of CsV_3Sb_5 is highly anisotropic, and the minimum SC gap is considerably smaller than the experimental energy resolution, suggesting the possible presence of a node.[43] However, some scanning tunneling microscopy (STM) studies ob-

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served non-zero conductance at zero bias,[5,8,44] which may be related to the residual Fermi arc.[34] Such complexity of the superconducting state does not appear in Ta-doped CsV_3Sb_5 with a higher T_c of 5 K and without a CDW.[19,45,46] For example, ARPES measurements of $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ revealed that the superconducting gaps were isotropic and lacked nodes.[45] Similarly, a U-shaped superconducting gap characterized by a flat bottom and zero density of states (DOS) at zero bias was observed in the STM spectrum of $CsV_{2.6}Ta_{0.4}Sb_5$.[19,46]

In this study, we conducted ultralow-temperature thermal conductivity and STM measurements of CsV_3Sb_5 to investigate its superconducting gap structure. The thermal conductivity in zero magnetic field shows a clear residual linear term κ_0/T, indicating the existence of a residual DOS within the superconducting state of CsV_3Sb_5. This is supported by the STM result, which provides a significant non-zero DOS at zero bias with an electronic temperature of 90 mK and a lattice temperature of 30 mK. Moreover, we measured the ultralow-temperature thermal conductivity of its substituted counterpart, $Cs(V_{0.86}Ta_{0.14})_3Sb_5$, which manifests no κ_0/T in the zero field and exhibits a completely different field dependence of κ_0/T than that of CsV_3Sb_5. We discuss the origin of the residual DOS in CsV_3Sb_5 from the perspective of the nodal gap and the interplay between CDW and superconductivity.

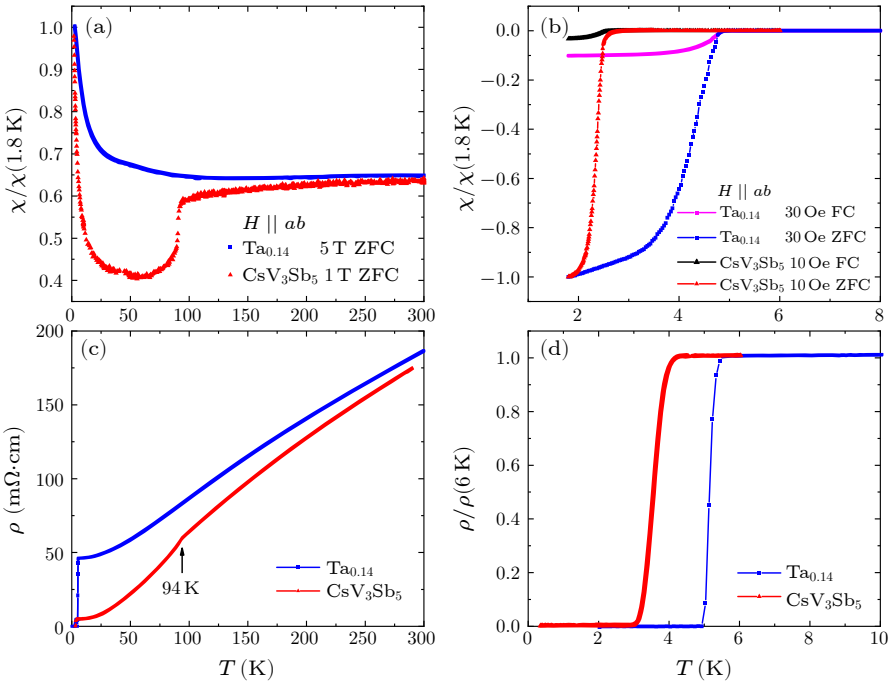

Fig. 1. (a) Temperature dependence of the normalized dc magnetization for CsV_3Sb_5 and $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ single crystals. (b) Normalized dc magnetization for CsV_3Sb_5 and $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ at low temperatures, with zero-field and field cooling modes. (c) Temperature dependence of in-plane resistivity for CsV_3Sb_5 and $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ single crystals. The arrow denotes the charge density wave order at 94 K for CsV_3Sb_5. (d) Normalized resistivity curves at low temperatures showing clear superconducting transitions.

Figure 1(a) shows the normalized DC magnetization of the CsV_3Sb_5 and $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ single crystals measured at 1 T and 5 T in zero-field cooling modes, respectively. The sharp drop at 94 K in CsV_3Sb_5 corresponds to a previously reported CDW transition,[2] whereas the CDW was suppressed by Ta doping in $Cs(V_{0.86}Ta_{0.14})_3Sb_5$.[19] In Fig. 1(c), the CDW transition manifests as a resistivity kink at 94 K for CsV_3Sb_5, and no anomaly can be observed in the resistivity of $Cs(V_{0.86}Ta_{0.14})_3Sb_5$. For the low-temperature magnetization results shown in Fig. 1(b), without considering the demagnetization factor, the calculated superconducting volume fractions are 128% and 106% for CsV_3Sb_5 and $Cs(V_{0.86}Ta_{0.14})_3Sb_5$, respectively. These values demonstrate the bulk superconductivity of the samples. At low temperature, the resistivity of both CsV_3Sb_5 and $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ show a superconducting transition at 3.0 K and 4.9 K (defined at zero resistivity), respectively, as shown in Fig. 1(d). The T_c values obtained from the resistivity are slightly higher than those obtained from the magnetization in Fig. 1(b). Further details regarding the sample growth, low-temperature resistivity in the field, upper critical field, and measurement procedure can be found in the Supplemental Material.[47]

Ultralow-temperature heat transport measurement is a well-established bulk technique for probing superconducting gap structures.[48] Figure 2(a) shows the in-plane thermal conductivities of five CsV_3Sb_5 single crystals in zero field. At ultralow temperatures, the thermal conductivity can be fitted to $\kappa/T = a + bT^{\alpha-1}$,[49,50] where the terms aT and bT^{α} represent contributions from electrons and phonons, respectively. Power α is typically between 2 and 3 because of the specular reflections of phonons at the boundary.[49,50] For samples

#1–#5 below 0.5 K, the fitting provides a finite residual linear term $(\kappa_0/T)_1 = 0.14 \pm 0.002\,\mathrm{mW{\cdot}K^{-2}{\cdot}cm^{-1}}$, $(\kappa_0/T)_2 = 0.15 \pm 0.004\,\mathrm{mW{\cdot}K^{-2}{\cdot}cm^{-1}}$, $(\kappa_0/T)_3 = 0.22 \pm 0.038\,\mathrm{mW{\cdot}K^{-2}{\cdot}cm^{-1}}$, $(\kappa_0/T)_4 = 0.35 \pm 0.013\,\mathrm{mW{\cdot}K^{-2}{\cdot}cm^{-1}}$, and $(\kappa_0/T)_5 = 0.42 \pm 0.057\,\mathrm{mW{\cdot}K^{-2}{\cdot}cm^{-1}}$, respectively. Furthermore, it can be observed that most of the obtained power α values are larger than three, with $\alpha_1 = 3.87 \pm 0.03$, $\alpha_2 = 3.85 \pm 0.05$, $\alpha_3 = 2.70 \pm 0.12$, $\alpha_4 = 3.28 \pm 0.05$, and $\alpha_5 = 3.19 \pm 0.08$. This anomalously large α suggests the existence of thermally excited electrons with increasing temperature, even at low temperatures. Considering that the T_c of CsV_3Sb_5 is quite low and the observation of a highly anisotropic superconducting gap in one of the complex FSs, a considerably small superconducting gap is gradually closed by the temperature. Although the second term $bT^{\alpha-1}$ may contain the contribution from thermally excited electrons, the finite residual linear term extrapolated from the ultralow-temperature thermal conductivity provides evidence for the existence of a residual DOS in the superconducting state of CsV_3Sb_5.

For s-wave nodeless superconductors, there are no fermionic quasiparticles to conduct heat as $T \to 0$ because the FS is entirely gapped.[49,50] Therefore, there is no residual linear term, κ_0/T, as observed for InBi and $NbSe_2$.[51,52] However, nodal superconductors exhibit a significant κ_0/T at zero field owing to nodal quasiparticles.[48] For example, κ_0/T of the overdoped ($T_c = 15$ K) d-wave cuprate superconductor $Tl_2Ba_2CuO_{6+\delta}$ (Tl-2201) is $1.41\,\mathrm{mW{\cdot}K^{-2}{\cdot}cm^{-1}}$, which is approximately 36% of the normal-state value κ_{N0}/T.[53] For the clean Fe-based superconductor KFe_2As_2, $\kappa_0/T = 3.8\,\mathrm{mW{\cdot}K^{-2}{\cdot}cm^{-1}}$ is approximately 3.2% of the normal-state value κ_{N0}/T.[54] For ultraclean $YBa_2Cu_3O_7$ (i.e., considerably low ρ_0 and significantly high κ_0/T in the normal state), a universal κ_0/T value of $0.16\,\mathrm{mW{\cdot}K^{-2}{\cdot}cm^{-1}}$ was observed for d-wave superconducting gap.[55] Therefore, one possible origin of the residual DOS, indicated by the finite κ_0/T of CsV_3Sb_5 in the zero field, may be the nodal quasiparticles. This issue will be discussed later.

Further information on the superconducting gap structure can be obtained by investigating the field-dependent behavior of κ_0/T.[48] Figure 2(b) depicts the in-plane thermal conductivity of CsV_3Sb_5 sample #1 under magnetic fields of up to 0.6 T. The data were fitted using $\kappa/T = a + bT^{\alpha-1}$ to determine κ_0/T for each magnetic field. The fitting of the data under $\mu_0 H = 0.4$ T yields $\kappa_0/T = 4.50 \pm 0.02\,\mathrm{mW{\cdot}K^{-2}{\cdot}cm^{-1}}$. A further increase field to 0.6 T does not increase the thermal conductivity; therefore, we assume 0.4 T as the bulk $H_{c2}(0)$. This value is lower than that obtained from the resistivity measurements. Furthermore, the value of κ_0/T agrees well with the normal-state Wiedemann-Franz law expectation $L_0/\rho_0(0.4\,\mathrm{T}) = 4.54\,\mathrm{mW{\cdot}K^{-2}{\cdot}cm^{-1}}$, with the Lorenz number $L_0 = 2.45\times10^{-8}\,\mathrm{W{\cdot}\Omega{\cdot}K^{-2}}$ and $\rho_0(0.4\,\mathrm{T}) = 5.40\,\mu\Omega{\cdot}$cm (see the Supplemental Material[47] for more details). The validation of the Wiedemann-Franz law in the normal state confirmed the reliability of our thermal conductivity measurements.

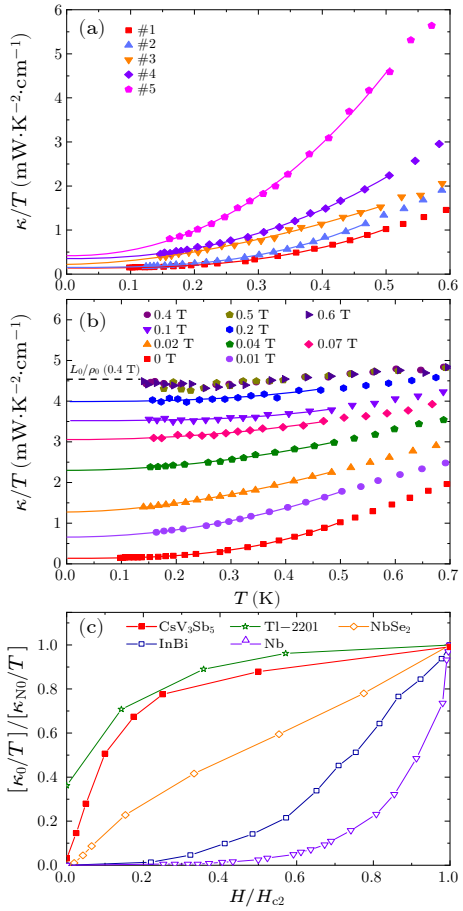

Fig. 2. (a) Temperature dependence of the in-plane thermal conductivity for five CsV_3Sb_5 single crystals in zero field. The solid line represents a fit to $\kappa/T = a + bT^{\alpha-1}$ below 0.5 K. (b) Thermal conductivity of CsV_3Sb_5 sample #1 in magnetic fields up to 0.6 T. The dashed line is the normal-state Wiedemann-Franz law expectation $L_0/\rho_0(0.4\,\mathrm{T})$, with the Lorenz number $L_0 = 2.45 \times 10^{-8}\,\mathrm{W{\cdot}\Omega{\cdot}K^{-2}}$ and $\rho_0(0.4\,\mathrm{T}) = 5.40\,\mu\Omega{\cdot}$cm. (c) Normalized residual linear term κ_0/T of CsV_3Sb_5 as a function of H/H_{c2}, with bulk $\mu_0 H_{c2} = 0.4$ T. Similar data of the clean s-wave superconductor Nb,[56] the dirty s-wave superconducting alloy InBi,[51] the multiband s-wave superconductor $NbSe_2$,[52] and an overdoped d-wave cuprate superconductor Tl-2201[53] are shown for comparison.

The normalized values of $[\kappa_0/T]/[\kappa_{N0}/T]$ as a function of H/H_{c2} for CsV_3Sb_5 is plotted in Fig. 2(c), with $\kappa_{N0}/T = 4.54\,\mathrm{mW{\cdot}K^{-2}{\cdot}cm^{-1}}$ and $\mu_0 H_{c2} = 0.4$ T. For comparison, similar data of the clean s-wave superconductor Nb,[56] the dirty s-wave superconducting alloy InBi,[51] the multiband s-wave superconductor $NbSe_2$,[52] and an overdoped d-wave cuprate superconductor Tl-2201,[53] are also plotted. Note that the field dependence of κ_0/T for CsV_3Sb_5 exhibits a rapid increase at low fields. This is an indication of a nodal gap, as in Tl-2201, or of considerable small gaps in a multigap superconductor that can be suppressed by low fields.

To confirm the existence of residual DOS, we conducted ultralow-temperature STM measurements to investigate the SC state of CsV_3Sb_5. Figure 3(a) presents a typical STM image of the CsV_3Sb_5 surface, and Fig. 3(b) provides an enlarged view of the black box in Fig. 3(a). The

atomically resolved lattice topographies shown in Fig. 3(b) further confirm the kagome lattice structure. At an electronic temperature of 90 mK, the coexistence of 1×4 and 2×2 supercells in the superconducting state was observed. However, for $Cs(V_{0.86}Ta_{0.14})_3Sb_5$, the CDW is suppressed.[19,46] Furthermore, the superconducting gap of CsV_3Sb_5 exhibits a non-zero conductance at zero bias, as shown in Fig. 3(c). This appears to have a residual zero-energy DOS, which is consistent with our thermal conductivity measurements.

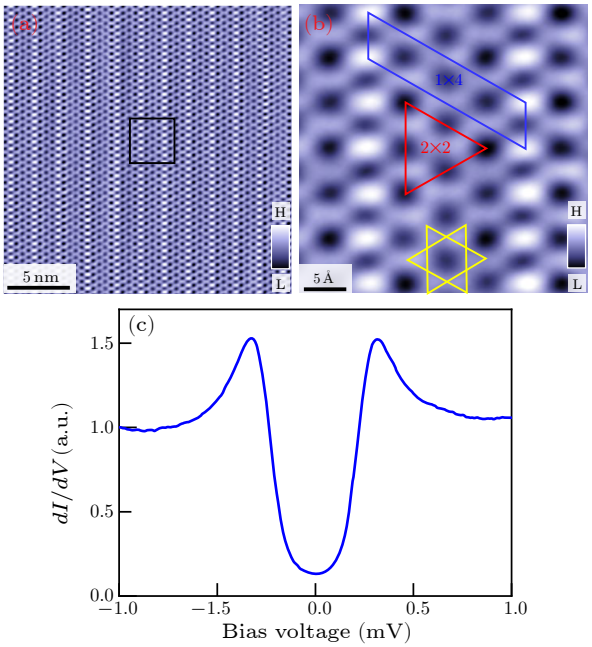

Fig. 3. (a) Typical STM images of the CsV_3Sb_5 surface. The scale bar is 5 nm. (b) Atomically resolved STM image of the sample surface taken in the dark box in (a). The blue quadrilateral and the red triangle represent the 1×4 and 2×2 supercells pattern, respectively. The yellow lines denote the underlying kagome lattice. (c) Spatially averaged dI/dV spectrum of CsV_3Sb_5 measured at an electronic temperature of 90 mK.

To determine whether the CDW order is essential for the residual DOS in CsV_3Sb_5, we measured the thermal conductivity of $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ in which the CDW was suppressed by Ta doping. The in-plane thermal conductivity of the $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ single crystal in zero field is shown in Fig. 4(a). The fitting of the data below 0.5 K with $\kappa/T = a + bT^{\alpha-1}$ yields $\kappa_0/T = -0.4 \pm 4\,\mu W{\cdot}K^{-2}{\cdot}cm^{-1}$ and $\alpha = 2.28 \pm 0.035$. Considering our experimental error bar of $\pm 5\,\mu W{\cdot}K^{-2}{\cdot}cm^{-1}$, κ_0/T of $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ in the zero field is essentially zero. Power α also shows a normal value between two and three for phonons. Figure 4(b) shows the thermal conductivity of $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ under magnetic fields up to 2 T. Data below 0.5 K were fitted with $\kappa/T = a + bT^{\alpha-1}$ to determine κ_0/T for each magnetic field. The fitting of the data under $\mu_0 H = 1.8$ T yields $\kappa_0/T = 0.51 \pm 0.002\,mW{\cdot}K^{-2}{\cdot}cm^{-1}$. Further increasing the field to 2 T did not increase the thermal conductivity; therefore, we considered 1.8 T to be bulk $H_{c2}(0)$. This value of κ_0/T agrees well with the expectation from the normal-state Wiedemann-Franz law $L_0/\rho_0(1.8\,T) = 0.53\,mW{\cdot}K^{-2}{\cdot}cm^{-1}$, with the Lorenz number $L_0 = 2.45{\times}10^{-8}\,W{\cdot}\Omega{\cdot}K^{-2}$ and $\rho_0(1.8\,T) = 46.0\,\mu\Omega{\cdot}cm$ (see Supplemental Material[47] for more details). Figure 4(c) shows the normalized values of $[\kappa_0/T]/[\kappa_{N0}/T]$ as functions of H/H_{c2} for $Cs(V_{0.86}Ta_{0.14})_3Sb_5$. Here, we used $\kappa_{N0}/T = 0.53\,mW{\cdot}K^{-2}{\cdot}cm^{-1}$ and $\mu_0 H_{c2} = 1.8$ T. The absence of κ_0/T in zero field and the slow field dependence of κ_0/T suggests nodeless superconductivity in $Cs(V_{0.86}Ta_{0.14})_3Sb_5$, with no residual DOS. This is consistent with the STM results, which show a flat bottom with zero DOS.[19,46] Similar STM results were obtained for $CsV_{2.73}Ti_{0.27}Sb_5$ ($T_c = 3.7$ K),[28] showing that the effect of Ta doping is not unique in suppressing the CDW order and residual DOS. Consequently, the CDW order in pristine CsV_3Sb_5 seems to be essential for the existence of a residual DOS in the zero field.

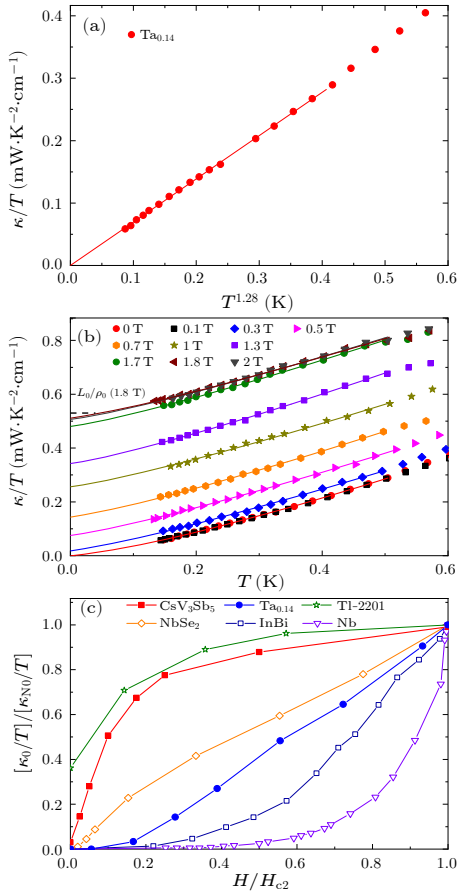

Fig. 4. (a) Temperature dependence of the in-plane thermal conductivity for $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ single crystal in zero field. The solid line represents a fit to $\kappa/T = a + bT^{\alpha-1}$ below 0.5 K, which yields $\kappa_0/T = -0.4 \pm 4\,\mu W{\cdot}K^{-2}{\cdot}cm^{-1}$ and $\alpha = 2.28 \pm 0.035$. (b) The thermal conductivity of $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ in magnetic fields up to 2 T. The dashed line is the normal-state Wiedemann-Franz law expectation $L_0/\rho_0(1.8\,T)$, with the Lorenz number $L_0 = 2.45 \times 10^{-8}\,W{\cdot}\Omega{\cdot}K^{-2}$ and $\rho_0(1.8\,T) = 46.0\,\mu\Omega{\cdot}cm$. (c) Normalized κ_0/T of $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ as a function of H/H_{c2}, with bulk $H_{c2} = 1.8$ T. Similar data of the pristine CsV_3Sb_5, the clean s-wave superconductor Nb,[56] the dirty s-wave superconducting alloy InBi,[51] the multiband s-wave superconductor $NbSe_2$,[52] and an overdoped d-wave cuprate superconductor Tl-2201[53] are shown for comparison.

Subsequently, we discuss the origin of the residual DOS in CsV_3Sb_5. As aforementioned, such a residual DOS usually originates from nodal quasiparticles. However, several phase-sensitive experiments[37,38,40,44] indicated the absence of sign reversal of the SC order parameter, and low-temperature STM with high resolution suggested a flat residual in-gap state instead of a V-shape or full gap.[34,35] These observations caused an outstanding paradox that has not been encountered in other superconducting systems. The underlying reason for this may be that CsV_3Sb_5 is an unusually multiband superconductor. Recently, ARPES measurements on high-quality CsV_3Sb_5 single crystals at 2 K showed that the SC gaps in the α FS (from the Sb $5p$ orbital) and δ FS (from the native V $3d$ orbital) are isotropic, whereas the SC gap in the β FS (from the induced V $3d$ orbital) is highly anisotropic.[43] It was interpreted that a nodal s-wave pairing state, which shows a node along ΓK direction and a maximum along the V-V bond direction (ΓM), is highly consistent with the ARPES results.[43] Such a nodal s-wave gap, without sign reversal of the SC order parameter, is consistent with our thermal conductivity results.

Another possible scenario is the strong anisotropic Cooper pairing of the tantalizing PDW order induced by the intertwining between a conventional wave SC and the CDW. The CDW occurs in the V d-orbital,[57] whereas the SC develops in the Sb p-orbital.[34,35,43] The PDW state is a finite momentum-paired state with the momentum of the CDW or related vectors, and only when the electrons on the FS are connected by the PDW vector is the gap opened at the Fermi level. Electrons not connected by the PDW vector are left as Bogoliubov Fermi states, such as the recently reported residual Fermi arcs.[34–36] Therefore, these residual Fermi arcs may be the origin of the residual DOS in CsV_3Sb_5 and contribute to the finite κ_0/T in the zero field.

In summary, we explored the superconducting gap structure of the V-based kagome superconductor CsV_3Sb_5 and its Ta-doped counterpart $Cs(V_{0.86}Ta_{0.14})_3Sb_5$ through ultralow-temperature thermal conductivity measurements, together with the ultralow-temperature STM measurements of CsV_3Sb_5. The presence of a finite κ_0/T in a zero magnetic field and non-zero conductance at zero bias observed in STM demonstrates the existence of a residual zero-energy DOS in CsV_3Sb_5. However, no residual DOS was observed for $Cs(V_{0.86}Ta_{0.14})_3Sb_5$, indicating the importance of CDW order for the residual DOS. For this unusual multiband superconductor, a nodal s-wave gap or residual Fermi arc may be the origin of the residual DOS in CsV_3Sb_5.

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