Testing electron–phonon coupling for the superconductivity in kagome metal CsV$_3$Sb$_5$

In crystalline materials, electron-phonon coupling (EPC) is a ubiquitous many-body interaction that drives conventional Bardeen-Cooper-Schrieffer superconductivity. Recently, in a new kagome metal CsV$_3$Sb$_5$, superconductivity that possibly intertwines with time-reversal and spatial symmetry-breaking orders is observed. Density functional theory calculations predicted weak EPC strength, $\lambda$, supporting an unconventional pairing mechanism in CsV$_3$Sb$_5$. However, experimental determination of $\lambda$ is still missing, hindering a microscopic understanding of the intertwined ground state of CsV$_3$Sb$_5$. Here, using 7-eV laser-based angle-resolved photoemission spectroscopy and Eliashberg function analysis, we determine an intermediate $\lambda=0.45–0.6$ at $T=6$ K for both Sb 5p and V 3d electronic bands, which can support a conventional superconducting transition temperature on the same magnitude of experimental value in CsV$_3$Sb$_5$. Remarkably, the EPC on the V 3d-band enhances to $\lambda\sim0.75$ as the superconducting transition temperature elevated to 4.4 K in Cs(V$_{0.93}$Nb$_{0.07}$)$_3$Sb$_5$. Our results provide an important clue to understand the pairing mechanism in the kagome superconductor CsV$_3$Sb$_5$.

The kagome lattice, made of corner-shared triangles, is an exciting platform for emergent quantum phenomena$^{1-3}$. Due to the wavefunction interference, the electronic structure of the kagome lattice features flat band, Dirac fermion, and van Hove singularities that result in a rich interplay between topology, geometry, and correlations$^{4,5}$. For kagome metals near the van Hove singularities, the high density of states combining with the frustrated lattice geometry are predicted to support novel electronic orders$^{6-8}$. Recently, in a topological kagome metal CsV$_3$Sb$_5$, superconductivity that possibly intertwines with charge density wave (CDW)$^{9-12}$ (Fig. 1a), nematicity$^{13-16}$ and loop current$^{17,18}$ is observed. To date, the origin of superconductivity and its interplay with the other symmetry-breaking orders remain rigorous debate. Angle-resolved photoemission spectroscopy (ARPES) studies$^{19,20}$ observed multiple van Hove singularities from V 3d-electrons near the Fermi level ($E_F$), highlighting electronic driven instabilities$^{6,8}$ (Fig. 1b). Furthermore, the density functional theory (DFT) calculated EPC strength$^{21}$, $\lambda\sim0.25$, in CsV$_3$Sb$_5$ fails to support the superconducting transition temperature$^9$, $T_c\sim2.6$ K, indicating unconventional pairing mechanism. However, a recent ARPES study of a cousin compound KV$_3$Sb$_5$ revealed a clear kink$^{22}$ in the electronic band structure near the van Hove singularity, suggesting a moderate EPC. Therefore, an

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enhanced by about 50% in the isovalent-substituted CsV3Sb5.

Experimental estimation of orbital- and momentum-dependent characters (three nesting wavevectors. The von Hove singularities has mainly V 3 singularities at the M point of the Brillouin zone boundary which are connected by pressure/doping phase diagram. The inset shows the crystal structure of CsV3Sb5.)

**Results**

Fig. 1a, c shows the crystal structure and Fermi surface (FS) topology of CsV3Sb5, respectively. In agreement with previous DFT and ARPES studies, the Sb 5p-band forms a circular FS, marked as α, at the BZ center and the V 3d bands yield hexagonal and triangle FSs, marked as β and γ in Fig. 1c, respectively. Figure 1d shows a typical ARPES intensity plot of the α band corresponding to the black cut shown in Fig. 1c. The coupling between electrons and bosonic modes is manifested by the intensity and dispersion anomalies, known as kink near a binding energy \( E_B \approx 32 \text{ meV} \). This many-body effect can be quantified by fitting the ARPES momentum distribution curves (MDCs) with a Lorentzian function:

\[
I(k, \omega) \propto A(k, \omega) = \frac{1}{\pi} \frac{\text{Im} \Sigma(\omega)}{\epsilon(k) - E_F + \text{Re} \Sigma(\omega) + i \text{Im} \Sigma(\omega)}.
\]

where \( \text{Re} \Sigma(\omega = E_F) \) and \( \text{Im} \Sigma(\omega = E_F) \) are the real and imaginary parts of the single-particle self-energy, \( \epsilon(k) \) is the non-interacting bare band that can be approximated as a linear dispersion crossing \( E_F \). Figure 1e demonstrates the extracted self-energy of the α band. We subtract a linear bare band from the experimentally extracted band to obtain \( \text{Re} \Sigma(\omega) \) (see supplementary note 1). To extract the electron-boson coupling induced \( \text{Im} \Sigma(\omega) \), the electron-electron and electron-impurity scatterings induced self-energy effects are removed, as suggested by previous practices (see supplementary note 2). At \( E_F \approx 32 \text{ meV} \), a peak near in \( \text{Re} \Sigma(\omega) \) and a step jump in \( \text{Im} \Sigma(\omega) \) prove strong many-body interactions. Since the self-energy anomalies persist above CDW (supplementary Fig. S7), we attribute the self-energy anomaly to EPC.

Figures 2a–c compares the EPC-induced kinks on the α and β bands at 6 K. The ARPES intensity plots of the α and β bands shown in Fig. 2b correspond to the black cuts in Fig. 2a. While the kink near \( E_F \approx 32 \text{ meV} \) is clear on both the α and β bands, an additional kink is observed at a lower \( E_F \approx 12 \text{ meV} \) on the β band (Fig. 2c). The 12-meV kink is also

**References**

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prominent in Reː(ω). As we show in Fig. 2d, Reː(ω) of the β band shows a peak near E_F-12 meV, proving strong d-electron–phonon coupling near 12 meV. In contrast, Reː(ω) of the α band only shows a broad shoulder.

The observation of clear EPC effects on both 5p and 3d bands points to a non-neglectable role of EPC for superconductivity in CsV₃Sb₅. To test the EPC-driven superconductivity, we extract the Eliashberg function, α²F(ω), and λ(ω) using the maximum entropy method. A fit of the Reː(ω) and the extracted α²F(ω) are shown in Fig. 2d, e, respectively. λ and the logarithmic mean phonon frequency are obtained via\textsuperscript{30,39}:

\[
\lambda = 2 \int_{0}^{\omega_{\text{max}}} \frac{\alpha^2 F(\omega)}{\omega} d\omega,
\]

\[
\ln \omega_{\text{log}} = 2\lambda \int_{0}^{\infty} \ln \left[ \frac{\alpha^2 F(\omega)}{\omega} \right] d\omega,
\]

where \(\omega_{\text{max}}\) is the maximum frequency of the phonon spectrum. As shown in Fig. 2e, the orbital dependence of the EPC is mirrored in the different shapes of α²F(ω), where phonon modes near 32 meV are accounted for 70% of the total EPC strength on the α band, λₐ, but less than 50% for the EPC strength on the β band, λₜ. Interestingly, due to the spectral weight redistribution in α²F(ω) (shaded area in Fig. 2e), the extracted λₜ and λₜ are similar with λₜ≈0.45±0.05. We also employed the MEM fits the extracted ImΣ(ω), which yields a λ consistent with the ReΣ(ω) fits (see supplementary note 4 and Fig. S2). Theoretically, λ can approximately be derived from a simpler approach\textsuperscript{39} following \(\lambda_{\text{dev}} = -\text{dReΣ(ω)}/\text{dω}|_{E_F} \approx \lambda\), when T is far lower than the Debye temperature. At T = 6 K, this method yields a λ_{dev}≈0.6±0.1, qualitatively consistent with Eq. (2) within the experimental uncertainty (Fig. 3f).

Generally, EPC can exhibit momentum dependence. Figure 3 summarizes the momentum-dependent kinks on the α and β bands. The ARPES intensity plots and extracted band dispersions along representative directions are shown in Fig. 3a, b and Fig. 3d, e, respectively (see supplementary Fig. S3 for complete dataset). The extracted λ_{dev}(k) for the α and β bands of two independent samples (supplementary Fig. S4) are summarized in Fig. 3f, which shows a nearly isotropic behavior within experimental uncertainties.

The orbital- and momentum-dependent results demonstrate that the EPC strength λ in CsV₃Sb₅ falls in the intermediate range of 0.45–6.0, which is about 2 times larger than the previous DFT prediction λ_{DFT}=0.25 (ref. 21). Using McMillan’s formula\textsuperscript{30} and taking the lower and upper limits of the experimentally estimated λ and the logarithmic mean phonon frequency -17.1 meV obtained from Eq. (3), we derive Tｃ in a range from 0.8 K to 3 K (see supplementary note 6). The upper limit is comparable to the experimentally determined T_{c} in CsV₃Sb₅ (Fig. 4a). We shall note that the CDW gap near the M point\textsuperscript{29,31} (supplementary Fig. S6) flattens the 6 bands near E_{F}, hindering the precise estimation of EPC strength. However, strong self-energy anomalies are observed on the 6 bands and they have the same energy scales as the α and β bands (supplementary Fig. S6).

As shown in Figs. 4a and 1a, T_{c} of CsV₃Sb₅ is increased with chemical substitutions or external pressure\textsuperscript{29,33}. We thus continue to examine the EPC in a 7% Nb-doped Cs(V₀.93Nb₀.07)₃Sb₅ with T_{c}=4.4 K\textsuperscript{33}. Electronic Kinks are observed on both the α and β bands as shown in Fig. 4b for the ARPES intensity plots and Fig. 4c for the extracted band dispersions. Figure 4d shows the extracted ReΣ(ω) on the α and β bands of Cs(V₀.93Nb₀.07)₃Sb₅. The shaded area corresponds to the
and \( \alpha \) gap comparable to the kink energy. We also note that the \( \beta \) band is similar in Cs(V_{0.93}Nb_{0.07})_3Sb_5 and CsV_3Sb_5, on the \( \beta \) band, it shows a strong enhancement in the Nb-doped sample, especially near \( E_F \). Based on the extracted \( \alpha\Gamma(k) \), shown in Fig. 4(e), we find that \( \lambda_{\alpha.c}=0.75 \pm 0.05 \) is enhanced by about 50\% in Cs(V_{0.93}Nb_{0.07})_3Sb_5. Such giant enhancement is also manifested by the slope of \( \Re \Sigma(\omega) \) near \( E_F \) (Fig. 4d). Consequently, the enhanced \( \lambda_{\alpha.c} \) in Cs(V_{0.93}Nb_{0.07})_3Sb_5 is expected to elevate \( T_c \) up to 4.5 K (see supplementary note 6), which is comparable to the experimental value of 4.4 K (Fig. 4a). Such synchronous enhancements of \( \lambda_{\alpha.c} \) and \( T_c \) may indicate that the V 3d-electron–phonon couplings are the main driver of the superconductivity in CsV_3Sb_5.

Finally, we discuss the influences of CDW order on the quantitative extraction of \( \lambda \) at \( T<T_{CDW} \). The formation of a CDW gap will modify the bare band to deviate from a linear dispersion near \( E_F \). As we show in the supplementary Fig. S3, within the experimental resolution, we do not observe a CDW gap on the \( \alpha \) and \( \beta \) bands. Therefore, for the \( \alpha \) and \( \beta \) bands, the CDW modified bare band dispersion below \( T_{CDW} \) is \( \sqrt{e^2(k) + A_{CDW}^2} \approx e_0(k) \), where \( e_0(k) = v_0 h k \) is the linear bare band dispersion above \( T_{CDW} \). In this case, the linear bare band assumption used in our study is a good approximation. Indeed, the excellent agreement of \( \Re \Sigma(\omega) \) and \( \Im \Sigma(\omega) \) linked by Kramers-Kronig transformation\(^{2,26} \) validates the linear bare band assumption for the \( \alpha \) and \( \beta \) bands (supplementary Figs. S1c-d). The linear bare band assumption, however, does not apply to the \( \delta \) band that forms a CDW gap comparable to the kink energy.\(^{2,6} \) We also note that the formation of CDW will also modify the electronic self-energy. As we show in the supplementary Fig. S7e, \( \lambda_{dev} \) shows an inflection point at \( T_{CDW} \), which may suggest an enhanced EPC strength below \( T_{CDW} \). However, it can also be a consequence of the CDW-corrected electronic self-energy effect (see supplementary note 9).

In summary, by investigating the electronic kinks, we determined an intermediate EPC that is twice larger than the DFT calculated value in the kagome superconductor CsV_3Sb_5 and Cs(V_{0.93}Nb_{0.07})_3Sb_5. Our results provide an important clue to understand the pairing mechanism in CsV_3Sb_5. The orbital, momentum of electronic kinks and their strengthening with the promoted \( T_c \) prove that the EPC in CsV_3Sb_5 is strong enough to support a \( T_c \) comparable to the experimental value and hence cannot be excluded as a possible pairing mechanism. While the exact microscopic pairing mechanism calls for further scrutiny, it is important to point out that the EPC-driven superconductivity is not incompatible with the recently observed pair-density wave (PDW) in CsV_3Sb_5.\(^{2,9,39} \) Indeed, PDW has been observed in another conventional superconductor NbSe_2, where the pair-density modulation is due to the real space charge density modulations.\(^{38} \) We also note that the EPC-driven superconductivity can coexist with the time-reversal symmetry-breaking (TRSB) orders or fluctuations,\(^{38,38} \) as proposed by theoretical studies.\(^{34} \) In those cases, the superconducting order parameter is expected to intertwine with the TRSB order parameter, which gives rise to an unconventional ground state.

**Methods**

**Growth and characterization of single crystals**

Single crystals of CsV_3Sb_5 were grown using CsSb_2 alloy and Sb as flux. Cs, V, Sb elements and CsSb_2 precursor were sealed in a Ta crucible in a
Temperature dependence of magnetic susceptibility for pristine CsV$_3$Sb$_5$ and Cs(V$_{0.93}$Nb$_{0.07}$)$_3$Sb$_5$. The samples were cleaved in situ and kept under a sample temperature was set to be 6 K if there is no special announcement. The samples were then separated from the quartz tube. The tube was heated up to 1273 K, maintained for 20 hours and then cooled down to 763 K slowly. Single crystals were provided by Jinggong New Materials (Yangzhong) Co., Ltd. The growth and characterizations of Cs(V$_{0.93}$Nb$_{0.07}$)$_3$Sb$_5$ were presented in ref.34. The magnetic susceptibility (Yangzhong) Co., Ltd. The growth and characterizations of Cs(V$_{0.93}$Nb$_{0.07}$)$_3$Sb$_5$. Both zero-field cooling (ZFC) and field cooling (FC) curves are presented. The Eliashberg function

\[ \alpha^2 F(\omega) = \int_{-\infty}^{\infty} d\omega f(\omega - \omega_0) f(\omega) \]

is related to the real part of the self-energy by the integration function

\[ \Re \Sigma(\omega) = \int_{-\infty}^{\infty} d\omega \alpha^2 F(\omega) \]

where \( K(y, y') = \int_{-\infty}^{\infty} dx f(x) \frac{f(x-y) f(y)}{y} \) and \( f(x) \) is the Fermi distribution function. It is an ill-posed problem to obtain the Eliashberg function from Eq. (4). In this work, we adopted the maximum entropy method (MEM)$^{25,26}$, which is frequently used to perform the analytic continuation$^3$. By considering the energy resolution of the laser-ARPES, we estimated that the error bar of the real part of the self-energy was 1 meV. MEM requires a model default function to define the entropic prior. Here, we adopted the following model:

\[ m(\omega) = m_0 \left( \frac{\omega}{\omega_0} \right)^2, \quad \begin{array}{ll} \omega & \omega_0 \\ \omega_0 \leq \omega \leq \omega_m, \end{array} \]

where \( m_0 = 15 \text{ meV} \), \( \omega_D = 10 \text{ meV} \), and \( \omega_m = 80 \text{ meV} \). This default model was also used in the previous study of the electron–phonon coupling on the Be surface$^26$.

**Data availability**

Data are available from the corresponding author upon reasonable request.

**Code availability**

Codes are available from the corresponding author upon reasonable request.
References

1. Syôzi, I. Statistics of kagomé lattice. Prog. Theor. Phys. 6, 306–308 (1951).
2. Zhou, Y., Kanoda, K. & Ng, T.-K. Quantum spin liquid states. Rev. Mod. Phys. 89, 025003 (2017).
3. Neupert, T., Denner, M. M., Yin, J.-X., Thomale, R. & Hasan, M. Z. Charge order and superconductivity in kagome materials. Nat. Phys. 18, 137–143 (2022).
4. Guo, H.-M. & Franz, M. Topological insulator on the kagome lattice. Phys. Rev. B 80, 113102 (2009).
5. Tang, E., Mei, J.-W. & Wen, X.-G. High-temperature fractional quantum Hall states. Phys. Rev. Lett. 106, 236802 (2011).
6. Kiesel, M. L., Platt, C. & Thomale, R. Unconventional fermi surface instabilities in the kagome Hubbard model. Phys. Rev. Lett. 110, 126405 (2013).
7. Wang, W.-S., Li, Z.-Z., Xiang, Y.-Y. & Wang, Q.-H. Competing electronic orders on kagome lattices at van Hove filling. Phys. Rev. B 87, 115135 (2013).
8. Park, T., Ye, M. & Balents, L. Electronic instabilities of kagome metals: saddle points and Landau theory. Phys. Rev. B 104, 035142 (2021).
9. Ortiz, B. R. et al. New kagome prototype materials: discovery of KV3Sb5, RbV3Sb5, and CsV3Sb5. Phys. Rev. Mater. 3, 094407 (2019).
10. Liang, Z. et al. Three-dimensional charge density wave and surface-dependent vortex-core states in a kagome superconductor CsV3Sb5. Phys. Rev. X 11, 031026 (2021).
11. Li, H. et al. Observation of unconventional charge density wave without acoustic phonon anomaly in kagome superconductors AV3Sb5 (A = Rb, Cs). Phys. Rev. X 11, 031050 (2021).
12. Ortiz, B. R. et al. CsV3Sb5: A Z2 topological kagome metal with a superconducting ground state. Phys. Rev. Lett. 125, 247002 (2020).
13. Jiang, Y.-X. et al. Unconventional chiral charge order in kagome superconductor KV3Sb5. Nat. Mater. 20, 1353–1357 (2021).
14. Xiang, Y. et al. Twofold symmetry of c-axis resistivity in topological kagome superconductor CsV3Sb5 with in-plane rotating magnetic field. Nat. Commun. 12, 6727 (2021).
15. Miao, H. et al. Geometry of the charge density wave in the kagome metal AV3Sb5. Phys. Rev. B 104, 195132 (2021).
16. Li, H. et al. Rotation symmetry breaking in the normal state of a kagome superconductor KV3Sb5. Nat. Phys. 18, 265–270 (2022).
17. Chen, H. et al. Roton pair density wave in a strong-coupling kagome superconductor. Nature 599, 222–228 (2021).
18. Miłek, C. et al. Time-reversal symmetry-breaking charge order in a kagome superconductor. Nature 602, 245–250 (2022).
19. Hu, Y. et al. Rich nature of Van Hove singularities in Kagome superconductor CsV3Sb5. Nat. Commun. 13, 2220 (2022).
20. Kang, M. et al. Twofold van Hove singularity and origin of charge order in topological kagome superconductor CsV3Sb5. Nat. Phys. 18, 301–308 (2022).
21. Tan, H., Liu, Y., Wang, Z. & Yan, B. Charge density waves and electronic properties of superconducting kagome metals. Phys. Rev. Lett. 127, 046401 (2021).
22. Luo, H. et al. Electronic nature of charge density wave and electron-phonon coupling in kagome superconductor KV3Sb5. Nat. Commun. 13, 273 (2022).
23. Valla, T., Fedorov, A. V., Johnson, P. D. & Hulbert, S. L. Many-body effects in angle-resolved photoemission: quasiparticle energy and lifetime of a Mo(110) surface state. Phys. Rev. Lett. 83, 2085–2088 (1999).
24. Lanzara, A. et al. Evidence for ubiquitous strong electron–phonon coupling in high-temperature superconductors. Nature 412, 510–514 (2001).
25. Sobotka, J. A., He, Y. & Shen, Z.-X. Angle-resolved photoemission studies of quantum materials. Rev. Mod. Phys. 93, 025006 (2021).
26. Yu, T. et al. Strong band renormalization and emergent ferromagnetism induced by electron-antiferromagnetic-magnon coupling. Nat. Commun. 13, 6560 (2022).
27. Jarrell, M. & Gubernatis, J. E. Bayesian inference and the analytic continuation of imaginary-time quantum Monte Carlo data. Phys. Rep. 269, 133–195 (1996).
28. Shi, J. et al. Direct extraction of the eliashberg function for electron-phonon coupling: a case study of Be(1010). Phys. Rev. Lett. 92, 186401 (2004).
29. Hofmann, P., Skylnadneva, I. Y., Rienks, E. & Chulkov, E. V. Electron–phonon coupling at surfaces and interfaces. N. J. Phys. 11, 125005 (2009).
30. McMillan, W. Transition temperature of strong-coupled superconductors. Phys. Rev. 167, 331 (1968).
31. Nakayama, K. et al. Multiple energy scales and anisotropic energy gap in the charge-density-wave phase of the kagome superconductor CsV3Sb5. Phys. Rev. B 104, L161112 (2021).
32. Yu, F. et al. Unusual competition of superconductivity and charge-density-wave state in a compressed topological kagome metal. Nat. Commun. 12, 3645 (2021).
33. Chen, K. et al. Double superconducting dome and triple enhancement of Tc in the kagome superconductor CsV3Sb5 under high pressure. Phys. Rev. Lett. 126, 247001 (2021).
34. Li, Y. et al. Tuning the competition between superconductivity and charge order in the kagome superconductor Cs(V1−xNbx)3Sb5. Phys. Rev. B 105, L180507 (2022).
35. Oey, Y. M. et al. Fermi level tuning and double-dome superconductivity in the kagome metal CsV3Sb5Sn. Phys. Rev. Mater. 6, L041801 (2022).
36. Liu, X., Chong, Y. X., Sharma, R. & Davis, J. S. Discovery of a Cooper-pair density wave state in a transition-metal dichalcogenide. Science 372, 1447–1452 (2021).
37. Xu, Y. et al. Three-state nematicity and magneto-optical Kerr effect in the charge density waves in kagome superconductors. Nat. Phys. 18, 1470–1475 (2022).
38. Khasanov, R. et al. Time-reversal symmetry broken by charge order in CsV3Sb5Sn. Phys. Rev. Res. 4, 023244 (2022).
39. Li, S., Hu, L.-H., Zhang, R.-X. & Okamoto, S. Topological superconductivity from forward phonon scatterings. Preprint at https://arxiv.org/abs/2207.09443 (2022).
40. Tazari, R., Yamakawa, Y. & Kontani, H. Charge-loop current order and Z3 nematicity mediated by bond-order fluctuations in kagome metal AV3Sb5 (A = Cs, Rub, K). Preprint at https://arxiv.org/abs/2207.08068 (2022).
41. Wu, X., Chakraborty, D., Schnyder, A. P. & Greco, A. Crossover between electron-electron and electron-phonon mediated pairing on the Kagome lattice. Preprint at https://arxiv.org/abs/2209.02351 (2022).
42. Kiss, T. et al. A versatile system for ultrahigh resolution, low temperature, and polarization dependent Laser-angle-resolved photoemission spectroscopy. Rev. Sci. Instrum. 79, 023106 (2008).
43. Li, S., Nocera, A., Kumar, U. & Johnston, S. Particle-hole asymmetry in the dynamical spin and charge responses of corner-shared 1D cuprates. Commun. Phys. 4, 217 (2021).

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Author contributions
Y.Z. and H.M. conceived the project. Y.Z. performed the ARPES experiment with the assistance from Y.D., K.A., and Y.A. and the guidance from T.K. and K.O.; T.K., K.O., and S.S. constructed the 7-eV laser-based ARPES system. Y.Z. S.L., Z.W., W.Z. and H.M. performed the theoretical analysis and simulations. H.X.L. and Y.G.S. grew the samples. H.X.L., H.N.L., and H.M. performed structural characterizations. Y.Z., H.M., T.K., and K.O. prepared the manuscript with inputs from all authors.

Competing interests
The authors declare no competing interests.

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