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Article

Keywords: fermi surface, charge density wave, electronic raman spectroscopy, QEP

Posted Date: September 3rd, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-846651/v1>

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Fermi surface instabilities in electronic Raman scattering of the metallic kagome lattice CsV_3Sb_5

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(Dated: August 26, 2021)

Understanding the link between a charge density wave (CDW) instability and superconductivity is a central theme of the 2D metallic kagome compounds AV_3Sb_5 ($A=K, Rb$, and Cs). Using polarization-resolved electronic Raman spectroscopy, we shed light on Fermi surface fluctuations and electronic instabilities. We observe a quasielastic peak (QEP) whose spectral weight is progressively enhanced towards the superconducting transition. The QEP temperature-dependence reveals a steep increase in coherent in-plane charge correlations within the charge-density phase. In contrast, out-of-plane charge fluctuations remain strongly incoherent across the investigated temperature range. In-plane phonon anomalies appear at $T^* \sim 50$ K in addition to right below $T_{CDW} \sim 95$ K, while showing no apparent evidence of reduced symmetry at low temperatures. In conjunction with the consecutive phonon anomalies within the CDW state, our electronic Raman data unveil additional electronic instabilities that persist down to the superconducting phase, thereby offering a superconducting mechanism.

I. INTRODUCTION

Two-dimensional kagome metals are at the forefront of correlated topological physics as their electronic structure entails flat bands, van Hove singularities, and Dirac cones. The sought-after many-body states in kagome materials include topological superconductivity, unconventional charge density wave (CDW), fractional quantum Hall, and Majorana fermions^{1–9}.

The recently discovered kagome metals AV_3Sb_5 ($A=K, Rb$, and Cs ; $P6/mmm$ space group) have opened a new avenue toward establishing the relationship between CDW, anomalous Hall effect (AHE), and superconductivity (SC)¹⁰. In this V-based kagome family, an alkali metal layer and a V_3Sb_5 layer are alternately stacked, forming quasi-2D kagome layers of the V ions. CsV_3Sb_5 features $\mathbb{Z}_2$ band topology and a two-stage symmetry breaking: a CDW transition at $T_{CDW} \sim 95$ K with a subsequent superconducting transition at $T_C \sim 2.7$ K^{10–14}. The CDW order with inverse star-of-David pattern involves 2×2 and 1×4 charge orders^{15,16}. The nesting of the Fermi surface assisted by the M-point van Hove singularity may provide a primary cause of the CDW transition¹⁵. Strikingly, a large AHE

concurr with the CDW order despite the lack of long-range magnetic ordering^{10,12,15,17–19}.

To account for spontaneous time-reversal symmetry breaking, orbital currents with different closed-loop patterns have been invoked^{16,20–22}. Currently, there are opposing experimental reports on the existence of a chiral flux current phase and the field tunability of chiral charge order^{23–25}. At low pressures, double-dome SC behavior is observed within the CDW phase and a new SC dome emerges at higher pressures where the CDW transition is completely repressed^{26–30}. Aside from the rich pressure-induced SC phases, contradictory views exist on the SC pairing mechanism and symmetry. The residual linear term of thermal conductivity along with Majorana modes and pair-density wave observed by scanning tunneling microscopy (STM) favour unconventional nodal superconductivity^{31–33}. Conversely, the magnetic penetration depth and the Hebel-Slichter coherence peak of ⁵¹V $1/T_1T$ support an *s*-wave superconductor^{34,35} or multiband superconductivity with sign-preserving order parameter^{36–38}.

To pin down the nature of the SC phase, it is pivotal to elucidate Fermi surface instabilities below the CDW state and their connection to SC. In this work, we employ polarization-resolved Raman spectroscopy to probe low-energy electronic fluctuations and to evaluate their possible role in the superconducting pairing mechanism. We uncover an additional phonon anomaly and an electronic instability below T_{CDW} , which may provide a gluing channel to superconductivity. The angular-dependent Raman data show a dominant *s*-wave character of the electronic correlations down to $T = 5$ K, which constrains the possible pairing symmetry.

II. RESULTS

Electronic Raman scattering and phonons. To disentangle in-plane from out-of-plane dynamical electronic behavior, we performed temperature-dependent Raman measurements in two different scattering geometries, as sketched in Fig. 1a. In Fig. 1b, we show representative Raman spectra obtained at $T = 5$ K and 300 K in in-plane and out-of-plane polarizations. The Raman spectra comprise sharp, well-defined phonon peaks superimposed on a broad electronic background. We decompose the electronic Raman scattering into two contributions: a rather symmetric maximum centered around 17 meV (shaded pale green) and a quasielastic peak (QEP) (shaded pale red). The former is assigned to an interband

FIG. 1. **Polarization-resolved Raman spectra of CsV₃Sb₅.** **a** Crystal structure of CsV₃Sb₅ and a sketch of the experimental Raman scattering geometries on a kagome lattice comprised of V atoms. The green arrows depict the incident and scattered light directions for in-plane and out-of-plane measurements. The shaded sine curves mark the direction of the electric field vector. **b** Raman spectra measured in three distinct symmetry channels A_{1g}+E_{2g}, E_{2g}, and E_{1g} at two temperatures $T = 300$ K and 5 K. The symbols mark symmetry-allowed optical phonons and a broad electronic background (green pentagon).

transition and the latter QEP pertains to a long-wavelength dynamical electronic response. The in-plane QEP forms a maximum at 2.5 meV, while the out-of-plane QEP is centered around zero energy. The difference of the QEP maximum between the in-plane and out-of-plane channels signifies anisotropic electronic scattering processes.

Besides, we observe two Raman-active A_{1g} and E_{2g} phonons at 17.9 meV and 14.7 meV, respectively, for in-plane scattering geometries. In the out-of-plane configuration, a single E_{1g} mode appears at 19.2 meV, in accordance with the Raman tensors (see Methods). We further note that a weak remnant of the A_{1g} phonon can be seen in the E_{1g} channel (the triangle in brackets). Since a fresh cut along the *c* axis was performed at a random in-plane orientation, a finite leakage from the A_{1g} channel might be inevitable. This is contrasted by no leakage of the E_{1g} phonon in our in-plane measurements.

Thermal evolution of the Raman response and conductivity. In Fig. 2a, we plot the temperature dependence of the Bose-corrected Raman response $\chi''(\omega, T)$ (see Supplementary Fig. S1 for the as-measured Raman intensity). With decreasing temperature, the in-plane low-energy $\chi''_{\text{ip}}(\omega, T)$ displays a gradual buildup of its spectral weight below 22

FIG. 2. **Electronic Raman scattering in CsV₃Sb₅.** **a** Bose-corrected Raman spectra $\chi''(\omega)$ and **b** Raman conductivity $\chi''(\omega)/\omega$ measured between 5 K and 300 K in an in-plane scattering channel. **c** Color plot of in-plane $\chi''(\omega)/\omega$ vs temperature. The white arrows point towards CDW-induced phonons. **d** Modeling of the in-plan Raman conductivity in terms of the low-energy quasielastic peak (pale red) and an electronic continuum at finite energies (pale green) **e-h** Out-of-plane $\chi''(\omega)$, $\chi''(\omega)/\omega$, and a color contour map of $\chi''(\omega)/\omega$ as a function of temperature, together with fits of the electronic Raman response, respectively.

meV. The Raman conductivity $\chi''_{\text{ip}}(\omega, T)/\omega$, shown in Fig. 2b, highlights the thermal growth of low-energy electronic scattering. Evident from the color contour plot of $\chi''_{\text{ip}}(\omega, T)/\omega$ in Fig. 2c, the QEP is slightly enhanced for temperatures below $\approx T_{\text{CDW}}$. Besides, CDW-induced phonons appear at 5.5 meV and 12 meV (see Fig. 4d and below). We analyze the Raman conductivity by a sum of two individual electronic excitations as demonstrated for $T = 300$ K and 5 K in Fig. 2d. The low-energy QEP is described by a collision-dominated model applied to the Bose corrected Raman response $\chi''_{\text{QEP}}(\omega) \sim A\Gamma/(\omega^2 + \Gamma^2)$, where A is the scattering amplitude and $\Gamma(T)$ is the scattering rate. We model the higher-energy excitation by a Gaussian profile.

In contrast to the in-plane behavior, the out-of-plane $\chi''_{\text{op}}(\omega, T)$, shown in Fig. 2e, gains progressively in low-energy spectral weight and shifts to zero frequency with decreasing temperature. $\chi''_{\text{op}}(\omega, T)/\omega$ and its color plot in Fig. 2f,g visualize the steep growth of QEP

FIG. 3. **Raman susceptibility, scattering rate and the symmetry of electronic scattering.**

a Raman susceptibility derived from in-plane and out-of-plane $\chi''(\omega)/\omega$. The blue and red lines are fits to a Curie-Weiss behavior. **b** Scattering rate extracted from fittings of the low-energy quasielastic peak to a collision-dominated model (see text) measured in in-plane (red squares) and out-of-plane (blue circles) channels. The dashed lines denote a linear temperature dependence. **c** The ratio of in-plane vs. out-of-plane scattering rate as a function of temperature. The dashed black line marks the close-to-linear temperature-dependence above T_{CDW} . **d** Angular dependence of the in-plane electronic Raman scattering intensity measured at $T = 5$ K (left) and $T = 80$ K (right). **e** Temperature-dependent energy, linewidth, and intensity of the electronic continuum centered around 17 meV.

as the temperature is lowered through T_{CDW} . Overall, the divergent or steady increase of $\chi''(\omega, T)$ below T_{CDW} , depending on the scattering channels, alludes to a secondary electronic instability of the pre-existing CDW ordered state and anisotropic scattering mechanism. Fig. 2h shows the decomposition of the out-of-plane electronic Raman conductivity into QEP and broad continuum.

Raman susceptibility and symmetry of electronic scattering. To shed light on the divergent QEP behavior and its link to an electronic instability, we analyze the dynamic Raman susceptibility χ^{dyn} at zero frequency using the Kramers–Kronig relation³⁹:

$$\chi^{\text{dyn}} = \lim_{\omega \rightarrow 0} \chi(k = 0, \omega) \sim \int_0^{\infty} \frac{\chi''(\omega)}{\omega} d\omega. \quad (1)$$

$\chi^{\text{dyn}}(T)$ allows us to access dynamic electronic fluctuations for a given symmetry channel, thereby we may quantify in-plane vs. out-of-plane electronic correlations. $\chi^{\text{dyn}}(T)$ is ob-

122 tained by integrating $\chi''(\omega)/\omega$ from 1 meV up to 50 meV and plotted in Fig. 3a for both
 123 scattering channels. Over the entire measured temperature range of $T = 5 - 300$ K, the
 124 temperature evolution of $\chi^{\text{dyn}}(T)$ is described by a Curie-Weiss law $\chi^{\text{dyn}}(T) \propto \frac{1}{T - T_{\text{CW}}}$ with
 125 the Curie-Weiss temperature T_{CW} . The lacking divergence of $\chi^{\text{dyn}}(T)$ at T_{CDW} alludes to
 126 an unconventional CDW order. Rather, the fact that the Curie-like increase of $\chi^{\text{dyn}}(T)$
 127 persists to the SC state supports the notion of an incipient electronic instability in addition
 128 to the CDW transition. From the in-plane $\chi^{\text{dyn}}(T)$, we extract $T_{\text{CW}}^{\text{ip}} \approx -22$ K, while the
 129 out-of-plane $\chi^{\text{dyn}}(T)$ yields $T_{\text{CW}}^{\text{op}} \approx -96$ K, i.e., very close to T_{CDW} (see Supplementary Fig.
 130 S2 for $1/\chi^{\text{dyn}}$ plots).

131 Several comments on the distinct T_{CW} values are in order. Given that $T_{\text{CW}}^{\text{ip}}, T_{\text{CW}}^{\text{op}} \leq T_{\text{CDW}}$,
 132 an instability of the Fermi surface may be inherited from the CDW reconstructed Fermi
 133 surface or a second CDW instability occurs in the primary CDW ordered state. On the
 134 other hand, $T_{\text{CW}}^{\text{ip}} \ll T_{\text{CW}}^{\text{op}} \approx T_{\text{CDW}}$ suggests that another in-plane electronic instability
 135 emerges well below T_{CDW} (see below) and may be inextricably tied to the SC formation.

136 Next, we discuss the QEP fitting parameters as a function of temperature. Detailed
 137 information on the individual fits is given in the Supplementary (Figs. S3 and S4). Since
 138 $A(T)$ and $\chi^{\text{dyn}}(T)$ convey essentially the same information, we focus on $\Gamma(T)$, plotted in Fig.
 139 3b. Both the in-plane and out-of-plane $\Gamma(T)$ exhibit a linear decrease at high temperatures.
 140 On approaching T_{CDW} , both quantities slightly increase with a clear peak at about T_{CDW} .
 141 The critical-like slowing down of CDW related excitations supports that the CDW order is
 142 mainly driven by electronic correlations. For lower temperatures, the out-of-plane $\Gamma^{\text{op}}(T)$
 143 tends to drop to zero as $T \rightarrow 0$ K. In stark contrast, the in-plane $\Gamma^{\text{ip}}(T)$ shows a strong
 144 upturn upon approaching T_c . This disparate behavior is highlighted in Fig. 3c, where the
 145 ratio of $\Gamma^{\text{ip}}/\Gamma^{\text{op}}$ evidences the increasingly anisotropic electronic correlations prior to the SC
 146 phase. In a weak coupling limit, the QEP can be understood as the Drude contribution to
 147 $\chi''(\omega)$ and $\Gamma(T)$ undergoes a renormalization by $\Gamma(T) \propto \Gamma_0(T - T_0)$ with the single-particle
 148 scattering rate Γ_0 ³⁹. We find $\Gamma_0^{\text{ip}}/\Gamma_0^{\text{op}} \approx 2$ above T_{CDW} . That is, the electronic fluctuations
 149 in the in-plane are twice as slow as in the out-of-plane channel prior to the CDW transition.
 150 The remarkable observation is the steep increase of $\Gamma_0^{\text{ip}}/\Gamma_0^{\text{op}}$ as $T \rightarrow T_c$, which goes hand
 151 in hand with the divergent $\chi^{\text{dyn}}(T)$.

152 To address the issue of in-plane anisotropies and possible symmetry lowering, we showcase
 153 angular plots of the in-plane electronic Raman scattering intensity measured at $T = 5$ K

FIG. 4. **Phonon dynamics in CsV_3Sb_5 .** Temperature-dependent phonon energy, linewidth, and normalized intensity of **a** the in-plane E_{2g} phonon, **b** the in-plane A_{1g} phonon, **c** the out-of-plane E_{1g} phonon, and **d** two CDW-induced modes. The solid red lines are fits to an anharmonic decay model (see Methods for details). The vertical dashed lines mark the CDW transition temperature and a second CDW mode onset temperature T^* .

(left panel Fig. 3d) and at $T = 80$ K (right panel Fig. 3d). Within our signal-noise ratio, we find no apparent angular dependence at both temperatures. This observation is at odds with a symmetry reduction of the electronic response through the CDW and approaching the SC state. Assuming that the secondary electronic instabilities below T_{CDW} give rise to SC, our angular variation data impose s -wave symmetry to the SC pairing. Nonetheless, we cannot exclude the possibility that a limited spectral resolution can obscure weakly broken rotational symmetry. Shown in Fig. 3e is the thermal evolution of the mid-energy electronic scattering signal centered around 17 meV. With decreasing temperature, all three quantities (energy, linewidth, and intensity) evidence a subtle discontinuous behavior around T_{CDW} . This may reflect the slight band modifications accompanying CDW.

CDW induced phonon anomaly. To elucidate phonon anomalies associated with the CDW transition, we present the phonon parameters (energy, linewidth, and intensity) as a function of temperature for all three phonons in Figs. 4a-c. The CDW transition involves no discernible effect on the out-of-plane E_{1g} phonon, while a minute softening can be seen in the energy of the in-plane E_{2g} phonon. In contrast, the in-plane A_{1g} phonon follows conventional anharmonic behavior only down to T_{CDW} . At lower temperatures, it pronouncedly hardens,

together with a discontinuous increase in intensity. In addition, two CDW modes of A_{1g} symmetry are activated by the charge-order-induced lattice modulations (as marked by the blue arrows in Fig. 1b and detailed in Fig. 4d). We recall that the CDW-induced modes are observed only in the in-plane polarization (see the vertical arrows in Fig. 1), indicative of the dominant two-dimensional nature of charge orders. Remarkably, the high-energy mode located around 12 meV, and thereby close to the A_{1g} phonon develops on the left shoulder of the electronic continuum below $T^* \sim 50$ K. It dramatically loses in intensity and broadens significantly well below T_{CDW} . Conversely, the lower energy CDW mode vanishes just below T_{CDW} , with its intensity following an order-parameter-like behavior. The rapid repression and damping of the 12 meV mode indicates its strong susceptibility to electronic fluctuations. The two characteristic temperatures T^* and T_{CDW} imply the presence of an extra electronic instability or a charge modulation deep inside the CDW state. Indeed, STM and ^{51}V NMR studies of CsV_3Sb_5 reveal another charge modulation at 40 K, which is interpreted in terms of a $4a_0$ charge modulation pattern or a roton pair density wave^{33,40}.

III. DISCUSSION

Using polarization-resolved electronic Raman scattering, we have detected a QEP that stems from long-wavelength electronic density fluctuations. The salient observations are that the QEP is continuously enhanced in χ'' upon approaching the SC state and that it features an anisotropic scattering rate between the intra- and inter-plane channels. The strong enhancement of the QEP for $T < T_{\text{CDW}}$ implies the presence of dynamical charge fluctuations in the background of the long-range charge orders in the CDW state and, thereby, raises the possibility of their link to SC.

The QEP associated susceptibilities show a Curie-like divergence as $T \rightarrow T_c$. The divergent behavior of $\chi^{\text{dyn}}(T)$ is reminiscent of a Pomeranchuk instability that is responsible for a nematic state of the Fe-based superconductors⁴¹. The presence of Fermi surface instabilities is further supported by the steeply increasing $\Gamma_0^{\text{ip}}/\Gamma_0^{\text{op}}$ at low temperatures. Recent theoretical studies have put forward Pomeranchuk fluctuations of a reconstructed Fermi surface or smectic bond-density-wave fluctuations as SC pairing mechanisms^{42,43}. Our angular-dependent Raman scattering study of the QEP closely resembles an isotropic s -wave character within the CDW phase (see Fig. 3d, and Supplementary Fig. S5), which seems to be

at odds with the original proposal that favors a d -wave Pomeranchuk channel. Nonetheless, both our phonon spectra and the ^{51}V NMR line independently demonstrate the occurrence of another charge modulation on top of the inverse star-of-David pattern⁴⁰. This hints towards a weak (possibly twofold) anisotropy, which could be a vestige of an emerging symmetry breaking within the superconducting phase. Putting aside the exact origin of the electronic instabilities, we expect dominant s -wave SC, if the observed electronic instabilities at $T > T_c$ are responsible for the gluing mechanism of Cooper pairs. Future Raman studies within the superconducting phase and under hydrostatic pressure are envisaged to identify the origin of the QEP unambiguously and its relation to SC as the CDW and SC display a distinct pressure evolution.

In summary, our Raman scattering study of CsV_3Sb_5 discloses secondary electronic instabilities in the CDW state, which arise from either Fermi surface fluctuations or another type of weak density wave or charge order. Their s -wave symmetry with tantalizing rotational symmetry breaking and two temperature scales will pose constraints on viable theories of establishing the relationship between CDW and SC.

IV. METHODS

Sample preparation. Single crystals of CsV_3Sb_5 were grown from Cs ingot (purity 99.9 %), V powder (purity 99.9 %) and Sb grains (purity 99.999 %) using the self-flux method, similar to the growth of RbV_3Sb_5 ¹⁴. The eutectic mixture of CsSb and CsSb_2 is mixed with VSb_2 to form a composition with approximately 50 % Cs_xSb_y and 50 % VSb_2 . The mixture was placed into an alumina crucible and sealed in a quartz ampoule under a partial argon atmosphere. The sealed quartz ampoule was heated to 1273 K for 12 h and soaked there for 24 h. It was subsequently cooled down to 1173 K at 50 K/h and further to 923 K at a slower rate. Finally, the ampoule was removed from the furnace and decanted with a centrifuge to separate CsV_3Sb_5 single crystals from the flux. The obtained crystals have a typical size of $2 \times 2 \times 0.02 \text{ mm}^3$.

Raman spectroscopy. To avoid degradation of the sample surface, a freshly cleaved single crystal of $2 \text{ mm} \times 2 \text{ mm} \times 0.2 \text{ mm}$ was mounted onto the cold-finger of a helium-flow cryostat inside an Ar atmosphere glove box. Raman spectroscopic measurements were carried out using a $\lambda = 561 \text{ nm}$ laser (Oxxius LCX) and a Princeton Instruments TriVista

spectrometer equipped with a volume Bragg grating notch filter (Optigrate; low-energy cut-off: 6 cm⁻¹) and a liquid-Nitrogen cooled CCD (PyLoN eXcellon). The incident laser with a spot diameter of about 5 μm was kept at a power level of 0.3 mW to minimize heating effects.

Analysis of phonons. CsV₃Sb₅ belongs to the hexagonal space group $P6/mmm$, where the ions are located at the following Wyckoff positions: Cs – 1a, V – 3g, Sb1 – 1b, Sb2 – 4h¹⁰. This leads to a total of three Raman-active modes $A_{1g} + E_{1g} + E_{2g}$, with their corresponding Raman tensors

$$A_{1g} = \begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & b \end{pmatrix}, E_{1g} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & c \\ 0 & c & 0 \end{pmatrix}, E_{2g} = \begin{pmatrix} d & 0 & 0 \\ 0 & -d & 0 \\ 0 & 0 & 0 \end{pmatrix}, E_{2g} = \begin{pmatrix} 0 & -d & 0 \\ -d & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

The temperature dependence of the phonon energies has been approximated by a cubic anharmonic, second-order model⁴⁴:

$$\omega(T) = \omega_0 - A \cdot \left(1 + \frac{2}{\exp(\hbar\omega_0/2k_B T) - 1} \right), \quad (2)$$

where ω_0 is the bare phonon energy at $T = 0$, and A is a fitting constant.

In metals electronic Raman scattering probes the long wavelength dynamical charge response in the symmetry channel i : $S_i(\omega) = \langle \rho_i^\dagger(\omega) \rho_i(\omega) \rangle$, where ω is the Raman shift and ρ_i is the electronic charge density. To derive the associated susceptibility, we first obtain the imaginary Raman response in the dynamical limit $\chi''(\omega)$ from as-measured Raman intensity $I(\omega)$ via the fluctuation-dissipation theorem by $I(\omega) = [1 + n(\omega)]\chi''(\omega)$. Here, $[1 + n(\omega)] = 1/(1 - e^{-\hbar\omega/k_B T})$ is the Bose factor.

ACKNOWLEDGMENTS

We thank Inkyung Song and Tae-Yeong Park for important experimental support. This work was supported by the National Research Foundation (NRF) of Korea (Grants No. 2020R1A2C3012367, and No. 2020R1A5A1016518) and by the Institute for Basic Science (Grant No. IBS-R009-Y3). The work at SKKU was supported by the National Research Foundation (NRF) of Korea (Grants No. 2020R1A2C3012367, and No. 2020R1A5A1016518). H.C.L. was supported by National Natural Science Foundation of

China (Grant No. 11822412 and 11774423), Ministry of Science and Technology of China (Grant No. 2018YFE0202600 and 2016YFA0300504) and Beijing Natural Science Foundation (Grant No. Z2000005).

V. AUTHOR CONTRIBUTIONS

K.-Y.C. conceived and designed the experiments. Q.Y., Z.T., C.G., and H.L. synthesized the single crystal. D.W., S.L., and Y.C. performed Raman spectroscopic experiments. K.-Y.C., D.W., S.L., and Y.C. analyzed the data. D.W., K.-Y.C, and S.L. wrote the manuscript with input from all authors. All authors discussed the results and commented on the manuscript.

Competing interests: The authors declare no competing interests. **Data and materials availability:** All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. All data are available from the corresponding authors upon reasonable request.

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VI. SUPPLEMENTARY

In Supplementary Fig. S1 we present the raw, as-measured Raman scattering intensity for the in-plane scattering configuration ($A_{1g}+E_{2g}$; panel a), as well as for the out-of-plane scattering configuration (E_{1g} , panel b), covering the temperature range 5 K – 300 K.

The detailed inverse dynamic Raman susceptibility $1/\chi^{\text{dyn}}$ is shown in Fig. S2 for both in-plane (red squares) and out-of-plane (blue circles) configurations. The Curie-Weiss behavior is indicated by the dashed lines, yielding the Curie-Weiss temperatures of $T_{\text{CW}}^{\text{op}} = -96$ K and $T_{\text{CW}}^{\text{ip}} = -22$ K for the out-of-plane and in-plane scattering geometries, respectively.

The full set of collision-dominated fits to the Bose corrected Raman intensity χ'' at all measured temperatures is plotted in Fig. S3 for the in-plane polarization, and in Fig. S4 for the out-of-plane polarization. For these fits, the data points in spectral ranges dominated by other excitations (phonons, as well as the electronic continuum centered around 17 meV; indicated by the red-colored symbols) were neglected.

In Fig. S5 we plot the angular dependence of the as-measured Raman intensity recorded

FIG. 5. **S1: Temperature dependence of as-measured Raman intensity.** **a** Raman scattering intensity measured in in-plane $A_{1g}+E_{2g}$ configuration. **b** Raman intensity measured in out-of-plane E_{1g} configuration. Spectra are stacked in intensity for clarity.

FIG. 6. **S2: Curie-Weiss fits to the dynamic Raman susceptibility.** The inverse dynamic Raman susceptibility $1/\chi^{\text{dyn}}$ measured in-plane (squares) and out-of-plane (circles). Dashed lines indicate the linear Curie-Weiss behavior.

FIG. 7. **S3: In-plane collision-dominated fits.** Bose-corrected Raman response χ'' (black and red symbols) measured in the in-plane $A_{1g}+E_{2g}$ channel over a temperature range from 5 K – 300 K. Green lines denote the collision-dominated fit, for which the black data symbols were considered. The red symbols denote the masked spectral range dominated by other excitations. The resulting fitting parameters Γ and A are indicated in each panel.

FIG. 8. **S4: Out-of-plane collision-dominated fits.** Bose-corrected Raman response χ'' (black and red symbols) measured in the out-of-plane E_{1g} channel over a temperature range from 5 K – 300 K. Green lines denote the collision-dominated fit, for which the black data symbols were considered. The red symbols denote the masked spectral range dominated by other excitations. The resulting fitting parameters Γ and A are indicated in each panel.

FIG. 9. **S5: Angular dependence at various temperatures.** Raman scattering intensity $I(\omega)$ obtained in in-plane configuration with the polarization of the light rotated in-plane over a range from 0° to 180° at fixed temperatures of 5 K, 80 K, and 300 K.

in the in-plane polarization over a range of $0^\circ - 180^\circ$ at $T = 5$ K (i.e., close to T_c), at $T = 80$ K (i.e., below T_{CDW}), and at $T = 300$ K ($T \gg T_{\text{CDW}}$). As seen from these three plots, we cannot resolve rotational symmetry breaking for phonons, charge-density-wave modes, as well as for electronic Raman scattering. Our angular variation data demonstrate that s -wave-like symmetry is largely retained for the electronic phases, while alluding to a subtle nature of a rotational symmetry broken state.