Anomalous amplitude modes and thinning-induced loss of crystallinity in the kagome metal CsV$_3$Sb$_5$

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We report Raman scattering spectroscopy of the kagome metal CsV$_3$Sb$_5$ to investigate its charge-density wave (CDW) phase. Temperature-dependent measurements reveal a multitude of CDW-induced modes, in agreement with density-functional theory calculations assuming the inverse Star of David lattice distortion. They are shown to be strongly hybridized amplitude modes and zone-folded modes, different from those in other known CDW materials. These CDW modes show negligible magnetic-field dependence and a signature of $c$-axis CDW modulation. Reducing the thickness to tens of nanometers dramatically enhances the change in the lattice phonon parameters across the CDW transition and strengthens its first-order character, indicating sizable electron-phonon coupling. Further thickness reduction leads to destruction of the crystal lattice. Our results provide essential structural information to unravel the CDW mechanism in CsV$_3$Sb$_5$.

I. INTRODUCTION

Materials with a kagome lattice can host rich phenomena encompassing quantum magnetism [1, 2], Dirac fermions [3, 4], nontrivial topology [5–7], density waves, and superconductivity [8–10]. The recently discovered kagome metals AV$_3$Sb$_5$ ($A = K$, Rb, or Cs) [11, 12] offer a new platform to study the interplay of these phenomena. These compounds have Fermi levels close to Dirac points or van Hove singularities [12–14], leading to a plethora of possible intriguing ground states. Indeed, charge-density waves and superconductivity have been discovered [12, 15, 16], with ample evidence showing that both types of orders are exotic. For example, the CDW transition is accompanied possibly by a large anomalous Hall effect [17, 18], and the superconductivity features a pair-density wave state [19]. The weakly bonded layers can be mechanically exfoliated [11], enabling the exploration of the effects of dimensionality on these phenomena and their carrier-density tuning [20, 21].

The nature of the CDW state and the mechanism for its formation have been under close scrutiny. In this work, we focus on the CsV$_3$Sb$_5$ compound, which shows a CDW transition temperature $T_{CDW} = 94$ K [12]. The transition is accompanied by appreciable partial gapping of the Fermi surface [22–26] and reportedly the lack of Kohn anomalies [27], although density functional theory (DFT) calculations found two phonon instabilities at the $M$ and $L$ points of the Brillouin zone [28–30]. Currently, there is still no consensus on the structural configuration of the CDW ground state. X-ray diffraction [12] and scanning tunneling microscopy (STM) found a 2×2 modulation of the crystal lattice [19, 31–33]. The CDW was shown to be three-dimensional, featuring a $c$-axis modulation that either doubles [33] or quadruples the original periodicity [34]. STM also found a 1×4 unidirectional modulation emerging only below 50 K [19, 31, 32], which was suggested to be of surface origin [32, 35]. It was further proposed that the CDW is chiral and exhibits time-reversal symmetry breaking [32, 36–38]. This was ascribed to a chiral flux phase, for which a later spin-polarized STM study indicated a negative result [39]. The roles of Fermi-surface nesting and electron-phonon coupling are also debated. Because the period of the 2×2 superlattice matches perfectly with the Fermiology of the van Hove singularity, it is natural to ascribe the CDW transition to Fermi surface nesting [28]. The renormalization of the Fermi surface observed by infrared and angle-resolve photoemission spectroscopy supports this scenario [22–26]. However, the calculated electronic susceptibility lacks the expected divergence [40, 41], and the effect of electron-phonon coupling may not be dismissed [40, 42].

Because the CDW phase features lattice distortions, studies of the lattice degree of freedom can offer insight into the CDW state. Raman scattering is a valuable tool in this respect. The collective excitation of the charge order manifests as Raman-active amplitude modes, providing a direct probe of the CDW order parameter [43, 44]. The temperature dependence of such modes as well as the zone-folded modes, which become Raman-active due to superlattice-induced zone folding, can both reflect the CDW transition [45–49]. The lattice distortion may affect the normal phonon modes of the pristine lattice and manifest as anomalous temperature dependence of the phonon parameters [50]. Combined with symmetry in-
formation from polarization-resolved measurements, constraints can be set on the possible CDW ground state.

Here, we report systematic Raman scattering measurements on CsV$_3$Sb$_5$. We observe multiple CDW-induced modes, whose symmetries and frequencies are in good agreement with DFT calculations for a single layer CsV$_3$Sb$_5$ under inverse Star of David distortion. In contrast to mostly independent amplitude modes and zone-folded modes in well-known CDW materials [46–48], we show that they hybridize strongly in CsV$_3$Sb$_5$, causing spectral weight redistribution to the latter and rendering them amplitude-mode-like. These CDW-induced modes are insensitive to magnetic fields, contrary to previous STM results on KV$_3$Sb$_5$ [51] and RbV$_3$Sb$_5$ [52]. The main lattice phonon parameters show clear anomalies in their temperature dependence, suggesting that electron-phonon coupling is significant. We also discovered that thickness reduction decreases the crystallinity and suppresses the CDW order. The complete loss of the main lattice modes in the Raman spectra in samples thinner than seven layers indicates collapse of the crystal lattice, informing future studies on this family of materials at reduced dimensions.

II. RAMAN-ACTIVE MODES IN CsV$_3$Sb$_5$

CsV$_3$Sb$_5$ crystallizes in a hexagonal lattice with the $P6_3/mmm$ space group [11]. Fig. 1(a) shows the unit cell of its crystal structure. The V atoms form a kagome net interspersed by Sb atoms (labeled Sb1), all within the $ab$-plane. The V atoms are further bonded by Sb atoms above and below the kagome plane (labeled Sb2). These V$_3$Sb$_5$ slabs are separated by Cs layers, with weak coupling between them. Factor group analysis yields three Raman-active phonon modes, $\Gamma_{\text{Raman}} = A_{1g} + E_{2g} + E_{1g}$. The former two can be detected when the photons are polarized in the $ab$-plane, satisfied by the back-scattering geometry used in our experiment (see methods in Appendix A). These intense modes are marked by dashed lines in Figs. 1(b, c). They involve only the Sb2 atoms, with their atomic vibrations along the $c$-axis and within the $ab$-plane for the $A_{1g}$ and $E_{2g}$ modes, respectively; see Fig. 1(a). The $E_{2g}$ modes are a pair of degenerate vibrations with opposite circling directions, i.e., opposite chiralities (see Appendix B). These two types of symmetries can be distinguished by polarization-resolved measurements. Specifically, the $A_{1g}$ modes can be detected in the XX and LL polarization configurations, whereas the $E_{2g}$ modes appear in the XX, XY, and LR configurations. Here, XX and XY represent collinear and cross-linear polarization for the incident and scattered photons, and LL and LR involve circularly polarized light with left (L) and right (R) helicity. A comparison of data in all four configurations is included in Fig. 9.

Below $T_{\text{CDW}}$, multiple peaks emerge, highlighted by the dotted lines in Figs. 1(b, c). Their origin will be discussed in the next sections. These modes are rather weak compared to the main lattice phonons. Their disappearance at 100 K suggests a close correlation with CDW formation. In contrast, many weak peak-like structures below 100 cm$^{-1}$ lack temperature dependence, whose origin is unclear.

Fig. 1(d) compares the observed Raman mode frequencies with those from DFT calculations for a single layer of CsV$_3$Sb$_5$ [28], considering two possible forms of lattice distortion, the Star of David (SD) and inverse Star of David (ISD) lattice distortions. The thick lines denote the main lattice phonons. The dots indicate modes undetected in our experiment.
FIG. 2. (a, b) Temperature-dependent Raman intensity color plot for CsV$_3$Sb$_5$, measured in the (a) LL and (b) LR configurations. The normal phonon modes are labeled in black and the CDW-induced modes in white. The dashed lines mark $T_{\text{CDW}}$. (c, d) Temperature-dependent spectra for the (c) $A_2$ and (d) $E_3$ modes. (e–g) Frequency, linewidth, and amplitude for the $E_{2g}$ and $A_{1g}$ normal phonons. The frequency and amplitude are compared to the corresponding values at 200 K. (h) Temperature dependence of the Raman mode frequencies. (i) Temperature dependence of the linewidth of the $A_2$ and $E_3$ modes.

All the five predicted $A_{1g}$ modes and five out of the eight predicted $E_{2g}$ modes are observed. The missing $E_{2g}$ modes are due to their weak scattering cross section. The observed $A_{1g}$ mode below 50 cm$^{-1}$ is unaccounted for by our calculations. This mode was also observed by pump-probe time-resolved spectroscopy, which, when compared with calculations taking into account interlayer coupling, was assigned as a Cs-mode due to CDW modulation along the c-axis [29]. These results suggest that the CDW ground state consists of weakly coupled layers with ISD-type distortion, but CDW modulation along the c-axis is also indispensable.

### III. TEMPERATURE DEPENDENCE

Figs. 2(a, b) show the temperature dependent Raman intensity color plot for CsV$_3$Sb$_5$, obtained in the LL and LR configurations, respectively. The intense $A_{1g}$ and $E_{2g}$ main lattice modes are the most conspicuous features. Figs. 2(c–g) show the frequency (with the corresponding value at 200 K subtracted), linewidth (full width at half maximum), and normalized integrated area for both modes, extracted from Lorentzian fits of the peaks. The $A_{1g}$ frequency sharply increases below $T_{\text{CDW}}$, whereas the $E_{2g}$ frequency exhibits a subtle kink across the CDW transition. This is consistent with the planar ISD lattice distortion mainly involving V atoms, forcing the Sb$_2$ atoms to displace along the c-axis, hence affecting the out-of-plane vibration of the $A_{1g}$ mode more effectively. The calculated phonon vibration patterns and frequencies confirm this picture (see Fig. 10 and Tab. I). The CDW transition also causes a faster decrease in the linewidths below $T_{\text{CDW}}$. This can be understood as being due to the CDW-induced partial gapping of the Fermi surface [22–26], which reduces the electron-phonon interaction. The integrated peak intensity for both phonons

|   | $A_{1g}$ | $E_{2g}$ |
|---|---------|---------|
| Pristine DFT | 136.69  | 128.62  |
| $A_1$ | 108.08  | 131.52  |
| $A_2$ | 202.93  | 241.89  |
| $A_3$ | 141.45  |         |
| $A_4$ | 141.45  |         |
| $A_{1g}$ |         |         |
| Expt. | 44.64   | 104.04  |
| $E_1$ | 127.24  | 197.42  |
| $E_2$ | 241.51  | 136.72  |
| $E_{2g}$ |         |         |
| CDW DFT | 99.03   | 212.22  |
| $E_1$ | 221.00  | 233.50  |
| $E_2$ | 129.02  |         |
| Expt. | 101.45  | 205.93  |
| $E_3$ | 222.02  | 237.19  |
| $E_{2g}$ |         | 118.55  |
| DFT | 57.51   | 124.81  |
| $E_1$ | 175.23  |         |
increases upon warming, in line with increased thermal phonon populations. The rate of increase is faster when approaching \( T_{CDW} \) from below, and interestingly, the value saturates below approximately 50 K. The renormalization of the phonon parameters across the CDW transition evidences sizable electron-phonon coupling.

CDW-induced modes are labeled in Figs. 2(a, b). Except for the \( A_1 \) mode, there appears to be two types of modes, represented by \( A_2 \) and \( E_3 \). \( A_2 \) exhibits appreciable softening and broadening upon warming toward \( T_{CDW} \). It is overdamped before disappearing, visualized in the color plot in Fig. 2(a) as the streak of signal below 100 cm\(^{-1}\) between 60–90 K. These are signatures of a CDW amplitude mode [45–49], caused by the collapse of coherent CDW order near \( T_{CDW} \). \( E_3 \) shows a smaller change of frequency and much less broadening, more consistent with the characteristics of a zone-folded mode [46], as this type of mode arises from folding a zone-boundary phonon to the zone center, making its temperature dependence as weak as that of normal phonons. Figs. 2(c, d) compare the distinct temperature dependence of these two types of modes. While \( A_2 \) broadens significantly above 40 K, \( E_3 \) maintains its linewidth and suddenly vanishes above ~80 K. The dramatic difference in the linewidth broadening is quantified in Fig. 2(i). Fig. 2(h) shows the frequencies of all the observed Raman modes on the same scale. Upon warming, the CDW-induced modes (\( A_1 \) excluded) soften more dramatically than the main lattice modes. While it is tempting to assign most of them as amplitude modes because of the apparent softening behavior, we show below that they are in fact zone-folded modes, mixed with the amplitude modes to partially inherit their properties.

\section*{IV. CDW-INDUCED MODES}

Our DFT results suggest that the formation of CDW in CsV\(_3\)Sb\(_5\) is similar to that in other well-known systems [45–49], in the sense that a soft acoustic phonon mode at the CDW wavevector condenses and gives rise to a distorted lattice [43]. The imaginary phonon modes of pristine CsV\(_3\)Sb\(_5\) at three \( M \) points (see Fig. 11) transform as irreducible representation \( M_1^+ (A_g) \) of the space group \( P6/mmm \) (little co-group \( D_2h \)). Fig. 3 shows that after the 2 \( \times \) 2 \( \times \) 1 CDW transition, they are folded to \( \Gamma \) and form triply degenerate modes. With lattice distortion, they decompose to a singlet \( A_{1g} \) mode and a doublet \( E_{2g} \) mode under the point group \( D_{6h} \):

\[ 3M_1^+ \rightarrow A_{1g} \oplus E_{2g}. \]  

As the lattice distorts from the pristine phase to the stable ISD pattern, the imaginary \( A_{1g} \) and \( E_{2g} \) modes turn real with positive frequencies, expected to be observable as two Raman-active amplitude modes. The phonon wavefunctions of the soft \( A_{1g} \) and \( E_{2g} \) modes are shown in Figs. 4(a, b), dominated by vibrations of V atoms.

FIG. 3. Phonon band structures in the process of CDW distortion. Here, 100% (0%) refers to the fully stable ISD (2 \( \times \) 2 pristine) structure. 10% refers to the intermediate structure with 10% distortion from the pristine to ISD phases. After 2 \( \times \) 2 \( \times \) 1 band folding with no distortion, the imaginary triply-degenerate modes form at \( \Gamma \). A weak ISD-type distortion lifts the degeneracy and leads to \( A_{1g} \) and \( E_{2g} \) modes. The ISD distortion gradually transforms imaginary modes to real. We highlight all \( A_{1g} \) and \( E_{2g} \) modes by orange and blue dots, respectively, at the \( \Gamma \) point.

The \( A_{1g} \) mode is fully symmetric, involving breathing-type motion for the V triangles, V hexagons, and Sb2 atoms. The \( E_{2g} \) mode involves circling motion for the atoms forming the V hexagons, while the amplitude for the Sb2 vibration is almost ten times smaller.

DFT calculations further reveal that the amplitude modes strongly hybridize with the other CDW-induced modes (\textit{i.e.} the zone-folded modes always at positive frequencies in Fig. 3), rendering them amplitude-mode-like, hence their apparent temperature-dependent frequencies. Figs. 4(c, d) show the real space wavefunctions of all the CDW-induced modes in the 2 \( \times \) 2 \( \times \) 1 ISD phase. \( A_{2-5} \) and \( E_{1-4} \) correspond to those in Fig. 2, and \( E'_{1-3} \) are undetected experimentally. Comparison with Figs. 4(a, b) shows that \( A_{2-5} (E_{1-4}) \) all resemble the \( A_{1g} \) (\( E_{2g} \)) soft mode. Such similarity is a result of hybridization of phonon wavefunctions.

To quantify the mode mixing, we calculated the overlap between the soft modes and all the real modes of the stable ISD phase by projecting the phonon wavefunctions, \( P_f = |\langle u_f | u_{SM} \rangle|^2 \), where \( |u_{SM} \rangle \) refers to the wavefunction of the soft modes shown in Figs. 4(a, b) and \( |u_f \rangle \) refers to the wavefunction of the mode at frequency \( f \) in the ISD phase. The results in Fig. 5 show that as the soft \( A_{1g} \) and \( E_{2g} \) modes shift from negative to positive frequencies and turn into amplitude modes, they hybridize with most of the zone-folded modes belonging to the same irreducible representation. The amount of calculated projection correlates reasonably well with the observed mode intensity. \( A_2 \) and \( E_1 \) are residual ampli-
tude modes after mode mixing. $E'_{1-3}$ show minor projection from the $E_{2g}$ soft mode because of negligible wavefunction overlap, and accordingly their scattering cross section is weak. $E_{2-4}$ all involve V triangles, indicating that they have contributions unrelated with the $E_{2g}$ soft mode. Indeed, as discussed earlier, $E_3$ shows clear

experimental signatures of a zone-folded mode.

The anomalously large hybridization between the amplitude modes and zone-folded modes is unprecedented, because they are mostly decoupled in the canonical CDW materials, with the amplitude modes dominating the spectral intensity [45–49]. The fact that the amplitude modes in CsV$_3$Sb$_5$ couple strongly with multiple zone-folded modes spanning a wide frequency range suggests that the hybridization occurs indirectly, through interaction with the common electronic system. As the Fermi surface instability associated with the van Hove singularity is from the V bands [28], modes mainly involving Sb (including $E'_{1-2}$ and the $A_{1g}$ and $E_{2g}$ main lattice modes) naturally mix weakly with the amplitude modes. Although the CDW in CsV$_3$Sb$_5$ may be of electronic origin [28], these results offer unambiguous signatures of appreciable electron-phonon coupling.

The $A_2$ mode was also observed in another Raman study [53] and by time-resolved pump-probe spectroscopy [29, 54]. However, in these works, it was suggested to emerge below $\sim 60$ K [29, 53, 54], hence ascribed to another phase transition associated with the unidirectional $1 \times 4$ order [19, 31, 32]. According to our data, shown in Fig. 2(a), the $A_2$ mode survives above 60 K, and there is no clear evidence for two distinct phase transitions. Its vibration pattern shown in Fig. 4(c) confirms no relation with the unidirectional order. Our Raman data are quite different from those in Ref. [27], and the cause of the discrepancy is unclear.
V. MAGNETIC-FIELD DEPENDENCE

The pronounced temperature dependence of the CDW-induced modes discussed above demonstrates the high sensitivity of Raman scattering to the detailed characteristics of the CDW phase. We extend the technique to investigate the effects of a magnetic field on the CDW, motivated by the report of anomalous Hall effect concurrence with the CDW transition [18, 32], the chiral nature of the CDW and its magnetic-field tunability [32, 51, 52], and the possible link to a hidden flux state that spontaneously breaks the time-reversal symmetry [38, 55].

Fig. 6 shows the Raman spectra of CsV₃Sb₅ measured under perpendicular magnetic fields at 1.7 K. Magnetic fields up to ±8 T were applied, and the data at ±6 T are shown. Within experimental uncertainties, we did not observe clear field dependence of the frequency, linewidth, and intensity for any of the CDW-induced modes. Small intensity variation for the main lattice $E_{2g}$ (not shown in full scale) and $A_{1g}$ modes can be noted, but this was confirmed to be present also above $T_{CDW}$, therefore ascribed to slight misalignment of the polarization optics and the unavoidable Faraday effect from the magnet.

The negligible field dependence observed here suggests that the charge orders are robust and do not show evidence of coupling with the applied magnetic field, consistent with a spin-polarized STM study [39]. These results are at odds with the field tunability of the CDW observed by STM in the sister compounds KV₃Sb₅ [51] and RbV₃Sb₅ [52], but another STM study found the field dependence to be absent in KV₃Sb₅ [56]. More experiments are required to resolve this discrepancy.

VI. THICKNESS DEPENDENCE

We exploit the layered structure of CsV₃Sb₅ to investigate the effect of thickness reduction on the lattice dynamics and CDW properties. The material can be exfoliated down to atomically thin layers, but the obtained flakes degrade quickly under ambient conditions. We therefore prepared samples in an inert atmosphere and encapsulated them using a thin insulating layer of h-BN. A typical sample image is shown in the inset of Fig. 7.

Thickness reduction dramatically alters the properties of the material. The 68.2 nm region exhibits both the main lattice phonons and the CDW-induced modes, showing that it is still bulk-like. In the 33.0 nm region, all these spectral features are present, but their intensities are moderately suppressed. The intensity suppression progresses when the thickness is further reduced to 6.6–8.3 nm, which amounts to 7–9 layers (one unit cell of CsV₃Sb₅ is 0.93 nm thick [11]). Moreover, a broad hump centered at 150 cm⁻¹ emerges. In the 5.5 nm (6-layer) region, this hump becomes the most prominent feature, whereas the main lattice modes are barely noticeable. These results clearly show the loss of crystallinity in atomically thin CsV₃Sb₅. Sample preparation either in high-purity nitrogen or argon gas and double-side encapsulation using h-BN to completely seal thin flakes yielded consistent results. In view of the precautions taken to avoid sample degradation, it is more likely that the CsV₃Sb₅ lattice is intrinsically unstable in its atomically thin form.

The mechanism for the collapse of the crystal lattice in atomically thin CsV₃Sb₅ could be related with the high chemical activity of the alkaline metal Cs. STM studies found nonuniform cleaved surfaces with clustered Cs atoms [31]. Refinement of X-ray diffraction data revealed high atomic displacement parameters for the Cs atoms, indicating their rattling-like motion owing to their weakly bonded nature [57]. Since the Cs layers are essential for bonding the V₃Sb₅ slabs together, their instability induced by thickness reduction may lead to the
disintegration of the entire crystal lattice. Recent studies of mechanically exfoliated CsV$_3$Sb$_5$ found competing CDW and superconducting orders [20, 21]. We point out that thinning-induced suppression of crystallinity should be kept in mind to interpret these results.

The reduced crystallinity in thin CsV$_3$Sb$_5$ flakes strongly influences the CDW properties. Figs. 8(a,b) compare the temperature-dependent Raman intensity color plots for the 33.0 nm and 8.3 nm regions. They differ greatly in the broad hump centered at 150 cm$^{-1}$. In the thinner region, the temperature-dependent data confirm the absence of CDW modes, and a quasi-elastic peak emerges at high temperature. Figs. 8(c–h) compare the mode parameters for the main lattice phonons. The absence of anomalies for the 8.3 nm region suggests that the CDW order is extinguished. For the 33.0 nm region, we observe anomalous temperature dependence for all the phonon parameters, even more dramatic than that for the bulk sample shown in Figs. 2(e–g). These results indicate a transition temperature of approximately 70 K. This slightly lower value than in the bulk is probably due to the decreased crystallinity, which is detrimental for CDW as a coherent order. Nonetheless, the CDW appears to couple more strongly to the lattice, such that changes in the phonon parameters become more apparent, suggesting the first-order nature of the phase transition. This is consistent with evidence from nuclear magnetic resonance and nuclear quadrupole resonance measurements in the bulk limit [58–60]. These results imply that interlayer interaction tends to smear the CDW lattice distortion. Indeed, under a moderate pressure of $\sim$2 GPa, which enhances the interlayer interaction, CDW in CsV$_3$Sb$_5$ was found to be completely quenched [61, 62].

VII. CONCLUSIONS

Our comprehensive Raman results offer informative insights into the CDW phase in CsV$_3$Sb$_5$. The observed multiple CDW-induced modes are in good agreement with DFT calculations for a single-layer model with the inverse Star-of-David superlattice, suggesting that only this type of in-plane distortion is present. The anomalously large hybridization between the amplitude modes and well separated zone-folded modes indicates indirect interaction between them through the common electronic system. The lack of magnetic-field dependence for the CDW-induced modes is inconsistent with the magnetic tunability observed in KV$_3$Sb$_5$ and RbV$_3$Sb$_5$, prompting further studies to substantiate the proposal of a chiral flux phase associated with CDW formation. Thickness reduction was found to decrease the crystallinity, which suppresses the CDW. However, in a thickness range where the CDW-induced modes are still preserved, the dramatic changes in the lattice phonon parameters across $T_{CDW}$ reveal a clear enhancement of the first-order character of the transition, indicating substantial electron-phonon coupling. These results form the structural basis for understanding the CDW in CsV$_3$Sb$_5$. 

FIG. 8. (a, b) Temperature-dependent Raman intensity color plot for CsV$_3$Sb$_5$ measured in the XX configuration for the (a) 33.0 nm and (b) 8.3 nm regions in Fig. 7. (c–e) Frequency, linewidth, and amplitude for the $E_{2g}$ and $A_{1g}$ main lattice phonons for the 33.0 nm region. The frequency and amplitude are compared to the corresponding values at 200 K. (f–h) The corresponding data for the 8.3 nm region.
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Appendix A: Methods

CsV$_3$Sb$_5$ single crystals were synthesized using the flux method [11]. The freshly cleaved surface of the samples was used in the study of bulk crystals. For the thickness-dependent study, the single crystals were mechanically exfoliated on silicone elastomer polydimethylsiloxane stamps and transferred on sapphire substrates. To avoid sample degradation, the process was completed in a glove box filled with nitrogen or argon gas, followed by encapsulation with thin h-BN. The sample thickness was determined by atomic force microscopy.

Raman scattering spectroscopy was performed using home-built confocal microscopy setups in the backscattering geometry with 532 nm laser excitation. The normally incident light was focused on the sample to a micron-sized spot, and the scattered light was directed through Bragg notch filters to access the low-wavenumber region. The Raman signal was collected using a grating spectrograph and a liquid-nitrogen-cooled charge-coupled device. Temperature- and magnetic-field-dependent data were acquired using two separate systems equipped with a Montana Instrument Cryostation and an attoDry superconducting magnet, respectively.

The DFT calculation results by Tan et al. are used to compare with the experiment. Details of the calculations can be found therein. In addition, we calculated the force constants by Vienna ab-initio Simulation Package (VASP) [63] and computed the phonon dispersion relation by Phonopy [64]. For the DFT calculation, a $5 \times 5 \times 5$ $k$ mesh and an energy cutoff of 400 eV were used.

Appendix B: Chiral character of $E_{2g}$ modes

For $E_{2g}$, the doubly degenerate modes correspond to states with opposite angular momentum ($l = \pm 1$), i.e., they are chiral. Although the wavefunctions are usually exported as real vectors by suppressing $l$, we can reconstruct the chiral phonons by a projection operation to chiral states. For $C_3$, the projection operator is defined as:

$$P_l = \sum_{k=0}^{2} (q^*_l C_3)^k.$$

$q_l = e^{i\frac{2\pi}{3}l}$ is the eigenvalue of $C_3$ with pseudoangular momentum $l = -1, 0, 1$. For an arbitrary phonon wavefunction $u$, the projected wavefunction $P_l u$ must be chiral, because

$$C_3 P_l = C_3 \sum_k (q^*_l C_3)^k = \sum_k (q^*_l)^k C_3^{k+1} = \sum_k (q^*_l)^{k-1} C_3^k = (q^*_l)^{-1} \sum_k (q^*_l C_3)^k = q_l P_l,$$

i.e. $C_3(P_l u) = q_l(P_l u)$.

Appendix C: Supplemental Data

FIG. 9. Raman spectra of CsV$_3$Sb$_5$ in the XX, XY, LL and LR configurations, measured at 4 K. The dashed and dotted lines mark modes with $A_{1g}$ and $E_{2g}$ symmetries, respectively.

FIG. 10. Wavefunctions of the main lattice phonons in the $2 \times 2 \times 1$ ISD phase. (a) The $A_{1g}$ mode. (b) The $E_{2g}$ mode. The Cs atoms are omitted in the crystal structure.

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