Optical detection of charge-density-wave instability in the non-magnetic kagome metal KV₃Sb₅

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Optical spectroscopy and density-functional calculations reveal electronic properties of the non-magnetic kagome metal KV₃Sb₅. Temperature and frequency-dependent optical measurements down to 10 K and up to 2 eV energy range confirm bulk nature of the charge-density-wave (CDW) state below 78 K and gauge the charge gap of Δ_{CDW} ≈ 60 meV at 10 K. We further detect strong phonon anomalies and the prominent low-energy localization peak indicative of the unconventional charge transport caused by electron-phonon or electron-electron interactions. Possible CDW structures of KV₃Sb₅, the star and hexagon (inverse star), are strongly reminiscent of p-wave states expected in the Hubbard model on the kagome lattice at the filling level of the van Hove singularity. The proximity to this regime may have intriguing and far-reaching implications for the physics of KV₃Sb₅ and related materials.

I. INTRODUCTION

Kagome geometry of corner-sharing triangles plays special role in condensed-matter physics. In magnetic insulators, it may cause the highly entangled quantum spin liquid state with exotic fractionalized excitations [1]. Kagome metals exploit another aspect of this peculiar geometry, the simultaneous presence of linear and flat bands. The former cross at the Dirac point that gives rise to Weyl nodes when time-reversal symmetry is broken. These interesting features of the electronic structure were indeed detected in several magnetic kagome metals [2–6], where spin texture – the type of magnetic order and its individual spin directions – was shown to have large impact on the energy bands and transport [7–9].

The discovery of AV₃Sb₅ (A = K, Rb, Cs) as a new family of non-magnetic kagome metals [10] opens the way to studying physics of kagome framework in the absence of magnetism. Here, the topological nature of the kagome bands can be intricately intertwined with electronic instabilities, such as superconductivity and charge order. Under ambient conditions, the AV₃Sb₅ family crystallizes in the hexagonal P6/mmm space group. V-atoms create the kagome network, while Sb1 atoms fill the centers of the kagome triangles. These V–Sb1 layers are sandwiched between graphite-like Sb2 layers that are, in turn, separated by the alkali-metal ions [Fig. 1(a)], creating easily exfoliable quasi-2D structures.

The AV₃Sb₅ compounds show abrupt changes in the electrical resistivity around 78 K (A = K [10]), 102 K (A = Rb [11]), and 94 K (A = Cs [12]) due to a charge-density-wave (CDW) instability. They further become superconducting below 0.9 K (A = K [13] and Rb [11]) and 2.5 K (A = Cs [10]). Superconductivity is often found in the vicinity of CDW states [14, 15], but its coexistence with CDW is far from trivial and raises questions on the exact nature of the CDW state as well as its microscopic origin. Understanding the CDW state seems also crucial in the context of the anomalous Hall effect, which is strongly enhanced in KV₃Sb₅ [16] and CsV₃Sb₅ [17] below the CDW transition and has been ascribed to scattering from spin clusters. With no signs of local magnetism detected by muon spin spectroscopy in this temperature range [18], other forms of spin degrees of freedom embedded in a CDW must be considered.

Here, we use broadband optical spectroscopy to examine the CDW formation in KV₃Sb₅. In contrast to very recent scanning-tunneling-spectroscopy studies [19], optical spectra are sensitive to bulk features of the electronic structure and simultaneously probe its interplay with lattice degrees of freedom. We demonstrate the bulk nature of the CDW state in KV₃Sb₅, gauge the associated charge gap, and detect signatures of strong electron-phonon coupling as the likely driving force of the CDW transition. Aided by ab initio band-structure calculations, we propose possible structural models of the CDW state and identify their similarity to phases predicted for the nearest-neighbor Hubbard model on the kagome lattice [20]. The respective filling factor puts the Fermi level at the van Hove singularity, in strong resemblance to KV₃Sb₅ with its high density of states at the Fermi energy. Combined with the presence of a low-energy localization peak in optical absorption, this hints at phonons and electronic correlations as two important ingredients of the KV₃Sb₅ physics.

II. METHODS

High-quality single crystals were prepared as described elsewhere [10, 13]. Freshly cleaved samples with the di-
FIG. 1. (a) Crystal structure of $\text{KV}_3\text{Sb}_5$ consists of V-Sb kagome layers and graphite-like Sb layers separated by the potassium atoms. (b) Representative band structure and corresponding density-of-states (DOS) for the nearest-neighbor tight-binding model on the kagome lattice. Fermi energy is set to the van Hove singularity point at the filling factor of 5/12. (c) Calculated DOS of $\text{KV}_3\text{Sb}_5$ in its normal (undistorted) structure and in the CDW state (star structure, see Fig. 5) demonstrates a partial gap formation at the Fermi level ($E_F = 0$) across the transition. (d) dc resistivity of the sample used in the optical study is shown together with the resistivity values obtained by the Hagen-Rubens fits of the optical data. The inset display the CDW anomaly in the first derivative of the resistivity and in the magnetic susceptibility. (e) Calculated band structure of $\text{KV}_3\text{Sb}_5$ (normal state) demonstrates the linearly dispersing bands around the Fermi energy. (f) Fermi surface constructed from the calculated band structure and the cut along $k_z = 0$ for $\text{KV}_3\text{Sb}_5$. The high-symmetry points and the $k$-path are shown in orange.

mensions $\sim 2 \times 3 \times 0.2 \text{ mm}^3$ were used for optical measurements. In-plane component of the optical conductivity ($\sigma_{xx}$) was probed in all measurements. Additionally, dc resistivity [Fig. 1(d)] was measured with the standard four-point contact method on the exact piece used in optical experiments to quantify the amount of potassium in the sample. Magnetic anomaly has also been confirmed via magnetic susceptibility measurements. Here, a field of 1 T has been applied along $H \parallel c$ and the susceptibility has been measured in field-cooled (FC) configuration with a Quantum Design Magnetic Property Measurement System (MPMS). Comparison to the earlier literature suggest that our sample of $\text{KV}_3\text{Sb}_5$ is nearly stoichiometric, with the possible K deficiency level of well below 8% reported in Ref. [10] (see Appendix).

Broadband reflectivity measurements were performed with two Bruker Fourier Transform Infrared (FTIR) spectrometers. For the high-energy range (0.075−2.25 eV / 600 − 18000 cm$^{-1}$), an infrared microscope coupled to a VERTEX80v FTIR spectrometer is utilized, where the infrared light is focused to 200 µm$^2$. Freshly evaporated gold mirrors are used for the reference. For the low-energy measurements (0.01 − 0.1 eV / 70 − 700 cm$^{-1}$), an IFS 113v spectrometer coupled with a custom made cryostat is used. Absolute reflectivity of the sample has been obtained with the gold overcoating technique [21].

Optical conductivity is calculated via Kramers-Kronig (KK) analysis from the measured reflectivity. For the KK analysis, data are extrapolated using Hagen-Rubens relations to the low-energy range, while x-ray scattering functions have been utilized for the high-energy extrapolations [22].

Density-functional (DFT) band-structure calculations were performed in the VASP [23, 24] and Wien2K [25, 26] codes using Perdew-Burke-Ernzerhof flavor of the exchange-correlation potential [27]. Experimental structural parameters from Ref. [10] were chosen for the undistorted $\text{KV}_3\text{Sb}_5$ structure, with the Sb1 atom located at $(\frac{2}{3}, \frac{1}{3}, 0.7539)$ [28]. Crystal structures of possible CDW states were relaxed in VASP in the $2a \times 2a \times c$ supercell, with residual forces below 0.002 eV/Å. To facilitate comparison with the optical data and kagome Hubbard model, we focus on the charge order in the $ab$ plane and disregard possible modulation along the $c$-axis [19].

Wien2K was employed for calculating all band dispersions and optical conductivities, whereas VASP was used for phonon calculations via the built-in procedure with frozen atomic displacements of 0.015 Å. All calculations were performed on full-relativistic level with effects of spin-orbit coupling (SOC) included. Whereas SOC has minor influence on lattice energies and phonons, it strongly affects band structure in the vicinity of the Fermi
III. RESULTS AND DISCUSSION

A. Normal state

In Fig. 2(a), frequency-dependent optical conductivity of KV₃Sb₅ is plotted as a function of temperature. In the high-frequency range, it shows rather frequency-independent behavior with several interband transitions, which get sharper upon cooling. This frequency-independent behavior is interrupted with a very pronounced absorption in the low-energy range, below 0.3 eV.

A closer look reveals the details. The low-energy part of the spectra can be decomposed into several contributions as demonstrated in Fig. 2(b). Narrow peaks that do not shift in frequency upon cooling represent two phonon modes. A broader peak around 0.12 eV does not move either and can be associated with the low-energy interband transition, discussed in detail in the following paragraph.

Finally, an even broader peak is observed around 0.06 eV at room temperature and shows a red shift upon cooling. We interpret this feature as a localization peak and defer its discussion until Sec. III D.

The interband transitions are readily identified using DFT calculations for the undistorted P6/mmm crystalline structure of KV₃Sb₅ [Fig. 3]. Linear bands appear in the vicinity of the Fermi level and arise from the underlying Kagome geometry of the V atoms [Fig. 1]. Band-resolved calculations show that in the 0.25 – 1.0 eV range optical conductivity is dominated by two nearly frequency-independent contributions that are indeed expected for transitions between linear bands in 2D Dirac systems [29, 30]. We assign these contributions to the transitions between linear parts of bands A-C and B-D, respectively.

The sharp absorption peak around 0.12 eV has a different origin and can be traced back to the transitions between bands B and C having parallel segments along the A – L reciprocal direction. We note in passing that the size of this peak is quite sensitive to the exact position of the Fermi level and to the addition of electronic correlations within DFT+U. Best agreement with the experimental spectrum was achieved upon shifting the Fermi level by +64 meV in DFT, or upon adding U = 2 eV.
These changes control the occupation of bands B and C around the M-point, see Appendix for details.

Notwithstanding the complex nature of the real KV$_3$Sb$_5$ band structure, several similarities to the idealized bands of a nearest-neighbor kagome metal can be identified [Fig. 1(b)]. First, crossing points of the linear bands are located below the Fermi level, suggesting the effective filling factor above $\frac{1}{2}$. Second, the Fermi level sits at the energy where bands A–C cross in the vicinity of the M-point. Their inflection points are remarkably similar to the band shape expected in the idealized kagome metal near its van Hove singularity at the filling factor of $5/12$. Indeed, in its normal state KV$_3$Sb$_5$ also shows a high density of states at the Fermi level [Fig. 1(c)]. This suggests that the CDW instability in KV$_3$Sb$_5$ may be associated with an instability of a correlated kagome metal at its van Hove singularity [20].

### B. CDW state

Having identified general trends in the optical conductivity, we inspect the low-temperature behavior in more detail. A transport and magnetic anomaly is observed at $T_{\text{CDW}} = 78$ K [Fig. 1(d)]. Its insensitivity to the magnetic field [10] suggests an electronic rather than magnetic nature of this instability. Indeed, in the optical spectra we observed clear signatures of this transition. The (partial) gap at the Fermi energy is witnessed by the reduction of the low-energy optical conductivity, which is transferred toward higher energies as dictated by the optical sum rules. These observations prove the formation of a bulk CDW state in KV$_3$Sb$_5$ [31]. The phonon modes persist below $T_{\text{CDW}}$, albeit with significant anomalies that will be discussed in Sec. III C below.

**FIG. 4.** (a) Difference optical conductivity in the CDW state. The spectral weight transfer from low to high energies is observed. Arrows indicate the zero crossing and the maximum of the transferred peak. (b) Temperature evolution of the spectral weight transfer, where the peak position is normalized to the 10 K point. A clear gap opening below 80 K is marked that also deviated from the mean-field behavior (solid line).

**FIG. 5.** (a) Energy diagram of the possible CDW structures in KV$_3$Sb$_5$. The energies are given per formula unit, and the bonds show V–V distances shorter than 2.71 Å in the CDW structures. (b,c) p-wave states predicted for the kagome Hubbard model at the van Hove singularity. Charge bond order (b) features spatial modulation of charge, whereas spin bond order (c) features spatial modulation of spin current.

Difference spectra illustrate the spectral weight transfer [Fig. 4(a)]. The low-energy part of $\sigma_1(\omega)$ suppressed below ~0.1 eV is recovered at higher energies with a maximum around ~0.17 eV. The CDW gap $\Delta_{\text{CDW}}$ is gauged by the energy scale of the spectral weight redistribution. We employed the zero-crossing of the difference spectra to determine $2\Delta_{\text{CDW}}$. Its temperature dependence is given in Fig. 4(b). Alternatively, one can choose the maximum of the SW transfer as an analogy to ARPES measurements, resulting in different absolute values of $\Delta_{\text{CDW}}$ but essentially the same temperature dependence when scaled to the 10 K value. This temperature dependence deviates from the mean-field behavior, $\Delta_{\text{mf}} \approx \Delta(0) \sqrt{1 - \frac{T}{T_{\text{CDW}}}}$ (around $T_{\text{CDW}}$), where we assume $T_{\text{CDW}} = 78$ K and $\Delta(0) = 59$ meV. The obtained CDW energy gap is of the same order as ~0.03 eV determined recently by STM [19]. Note however that here we probe the bulk gap, while STM reports on the surface gap only.

To identify possible CDW states in KV$_3$Sb$_5$, we rely on the experimental constraint of the $2 \times 2$ in-plane superstructure detected by scanning tunneling microscopy [19, 32] and single-crystal x-ray diffraction on CsV$_3$Sb$_5$ [12]. Different low-symmetry configurations constructed within the $2 \times 2$ supercell were allowed to relax, and two stable solutions were found [Fig. 5]. One of them is star of David-type charge order reminiscent of 1T-TaS$_2$ [33–35] and denoted as star in the following. The other one involves two types of vanadium clusters,
ences are only marginally affected by the choice of the exchange-correlation functional or adding the spin-orbit coupling, and all three structures are non-magnetic in DFT. Further on, both hexagon and star types of charge order are compatible with transport properties of KV₃Sb₅. The metallic behavior observed experimentally even below $T_{\text{CDW}}$ suggests that gap opening is only partial. Indeed, difference density of states [Fig. 6(a)] reveals a gap of about 0.1 eV in good agreement with $\Delta_{\text{CDW}}$ determined from optics, but the overall spectrum remains metallic with a non-zero density of states at the Fermi level [see also Fig. 1(c)]. The absorption peak due to the interband transition shifts toward higher energies. Indeed, the calculated optical conductivity reveals a peak around 0.2 eV in excellent agreement with the experimental data as demonstrated in Fig. 6(b). Therefore, both hexagon and star structures are likely candidates for the CDW state of KV₃Sb₅.

An important observation at this juncture is that the hexagon and star structures are close analogues of density-wave states predicted theoretically for the Hubbard model at the filling level of the van Hove singularity [20]. These states are shown in Fig. 5 and classified as charge bond order and spin bond order, or, respectively, as singlet and triplet $p$-wave states according to the symmetry of their order parameter [36]. The star structure is immediately recognized as charge bond order, whereas the hexagon structure may be tentatively associated with spin bond order, albeit with one caveat. Spin bond order is not a conventional spin-density wave with the spatial modulation of spin density and local magnetic moments. Instead, it entails a spatial modulation of spin current that can not be captured by DFT. The hexagon structure itself, as obtained by the DFT atomic relaxation, is of charge bond order type, too, and can be understood as an “inverse star”, because the short and long V–V bonds are merely inverted compared to the star structure. However, the resemblance of this structure to the spin bond order is far from accidental. The hexagon structure does not support spin-density waves and appears to be immune to conventional magnetism, whereas the star structure readily turns into a spin-density wave when magnetism is introduced on the DFT+$U$ level (see Appendix for details).

The analogy to the $p$-wave states of the kagome Hubbard model is reinforced by the similar filling factors. The $p$-wave instabilities appear in the model near its van Hove singularity [20], whereas real band structure of KV₃Sb₅ lies close to this regime and shows a peak in the density of states around the Fermi level. On the other hand, atomic reconstruction in the CDW state must involve phonons, in contrast to the Hubbard model where only electronic instabilities are at play. This dichotomy – competing roles of Fermi surface nesting and phonons – has sparked vivid debates for many of the experimentally observed CDW’s.

In the 1D case, electronic instabilities and phonons come hand in hand [37]. Extending this picture to higher dimensions is usually based on the assumption

triangles and hexagons, and is labeled simply as hexagon for brevity. In both cases, the V–V distances inside the clusters are below 2.71 Å to be compared with 2.74 Å in the undistorted structure, whereas V–V distances between the clusters exceed 2.80 Å.

The hexagon and star structures lie lower in energy than the parent, undistorted structure by 1.1 meV/f.u. and 0.8 meV/f.u., respectively. These energy differences are only marginally affected by the choice of

FIG. 6. (a) Difference DOS of the undistorted and distorted structures representing the change across $T_{\text{CDW}}$. A partial gap opening for the distorted structures is demonstrated with only 50% reduction at the Fermi energy, where one can also trace the redistribution to the higher energy range. The gap of $\sim$0.1 eV and the redistribution energy of $\sim$0.25 eV is also in line with the experimental observation. (b) Comparison of the experimental interband transitions with the DFT calculations in the normal and CDW states.

FIG. 7. The calculated IR-active modes are given for all the structures. Vertical lines represent the phonon modes relevant to the experimental observations. The black line shows the phonon mode, where the example fits in Fig. 8(b) and the parameters in Fig. 8(d–f) are also given. The red line indicates the phonon modes that can create the overtone/combination mode in the IR spectra.
that Fermi surface nesting occurs at quasi-1D portion of the Fermi surface, and the resulting electronic reconstruction is sufficient to drive a metal-insulator transition or cause drastic changes in carrier scattering. However, other effects may be significant too. For instance, strong electron-phonon coupling was discussed as the source of the CDW observed in metallic 2D dichalcogenides, 2H-NbSe$_2$, [38, 39], where strong momentum-dependent electron-phonon coupling can lead to a CDW transition even in the absence of nesting. The situation becomes even more complex in high-temperature cuprate superconductors, where neither weak Fermi surface nesting near the antinodal region [40] nor the strong electron-phonon coupling [41] were found sufficient to explain the CDW instability. Strong electronic correlations were then proposed as another crucial ingredient [42].

The aforementioned effects may also contribute to the CDW formation in KV$_3$Sb$_5$. Their relative importance depends on the balance between electronic correlations and electron-phonon coupling. In the following, we discuss possible experimental fingerprints of these two major ingredients using optical data for KV$_3$Sb$_5$.

### C. Phonon Anomalies

Experimental optical spectra reveal two phonon modes at 188 cm$^{-1}$ and 482 cm$^{-1}$. Calculated frequencies of Γ-point phonons allow the assignment of the lower mode to the IR-active $E_{1u}$ phonon (black line in Fig. 7). In contrast, the upper mode could not be reproduced, because the highest optical phonon is at 299 cm$^{-1}$. On the other hand, DFT calculations reveal two IR-active modes at 237 cm$^{-1}$ and 241 cm$^{-1}$, fairly close to each other [red line in Fig. 7 (b)]. This makes the high energy mode observable either as a combination of these modes or as an overtone of one, possibly the 241 cm$^{-1}$ mode, as the resonance frequency is exactly doubled. Furthermore, this mode displays a strong fano-like behavior, indicating that it unusually couples to the electronic background. This mode can be traced at high temperatures, but the CDW gap opening hinders its clear observation below $T_{CDW}$.

The lower mode shows even more striking signatures of the electron-phonon coupling. The resonance frequency $(\omega_0)$, intensity $(\Delta \gamma)$, and damping (linewidth, $\gamma$) obtained from the Lorentzian fit of this mode are given in Fig. 8. Strong anomalies are observed across the CDW transition. Below $T_{CDW}$, a strong softening of the mode is accompanied with the significant increase in intensity. Furthermore, the linewidth is very high and by far exceeds the resolution of our measurement (1 cm$^{-1}$). While a combination of several weakly spaced modes could be responsible for this broadening in the CDW state where a much higher number of IR-active phonons modes is expected [Fig. 7], the broadening above $T_{CDW}$ must be caused by the electronic background. Moreover, the line gets broader upon cooling, while the sharpening is usually observed with decreasing thermal effects. Such unexpected broadening of the phonon modes upon cooling has been discussed in terms of electron-phonon coupling scenario [43, 44], for instance in the case of graphene. Here we used the same formalism and fitted temperature dependence of the linewidth [Fig. 8(e)] using

$$\gamma_{e-\text{ph}}(T) = \gamma_{e-\text{ph}}(0) \left[ f \left( \frac{\hbar \omega_0}{2k_B T} \right) - f \left( \frac{\hbar \omega_0}{2k_B T} \right) \right]$$

(1)

Here, $\hbar \omega_0 = 174$ cm$^{-1}$ is the calculated $E_{1u}$ phonon energy, $k_B$ is the Boltzmann constant, and $f(x) = 1/(e^x + 1)$. The intrinsic linewidth of $\gamma_{e-\text{ph}}(0) = 52$ cm$^{-1}$ signals the crucial role of electron-phonon coupling and makes it a plausible reason for the CDW instability in KV$_3$Sb$_5$ [41].

Microscopically, the aforementioned $E_{1u}$ mode involves out-of-plane motion of the vanadium atoms within the kagome plane [Fig. 8(a)]. Such deformations are likely...
to affect electronic structure and especially linear bands around the Fermi level, which originate from the flat kagome framework of the V atoms.

### D. Localization peak

Another distinct feature of KV$_3$Sb$_5$ is the presence of a localization peak in the low-energy part of the spectrum. This peak is identified by its red shift upon cooling and signals localization of charge carriers resulting in sub-diffusive transport and major deviations of the intraband optical conductivity from the Drude model [45–48]. Temperature evolution of this mode shown in Fig. 9 (a) reveals a redshift in frequency upon cooling, along with a sudden reduction in the spectral weight across $T_{\text{CDW}}$ [Fig. 9(b)], where $\sigma(\omega) = \int_{-\infty}^{\infty} \sigma_1(\omega) \, d\omega$. Here, $\omega_c$ is the cut-off frequency $\sim 2\,\text{eV}$, and $\sigma_1(\omega)$ is taken from Fig. 9(a) obtained through the fit of the whole spectra. While total spectral weight is conserved according to the sum rules, the weight of the localization peak drops at $T_{\text{CDW}}$, suggesting a redistribution of the intensity upon the CDW formation. This also leads us to assign the peak to intraband transitions, which are necessarily affected by the charge order.

Different microscopic scenarios of such a localization peak were discussed in the literature. Whereas mere disorder in the hopping paths [46] is excluded by the high quality of our crystals, both electron-phonon [49] and electron-electron [45] interactions are likely candidates. The former seems even more plausible in the light of the phonon anomalies discussed in Sec. III C. On the other hand, the concurrent observation [9] of a similar localization peak in the magnetic kagome metal Fe$_3$Sn$_2$, which is seemingly devoid of phonon anomalies, suggests that electronic correlations could also play a role.

Further evidence of the possible correlation effects in KV$_3$Sb$_5$ is gained from discrepancies in the description of its low-energy interband transitions by DFT. Experimental optical conductivity could be reproduced on the DFT level, but only upon shifting the Fermi level by +64 meV. This shift corresponds to doping the system with about 0.4e per formula unit, which is hard to reconcile with the chemistry of KV$_3$Sb$_5$, where hole doping is more likely because of the possible K deficiency. Our samples are close to stoichiometric, though, and the discrepancy with the DFT calculation for the exact KV$_3$Sb$_5$ stoichiometry can signal effects beyond DFT that influence band energies around the Fermi level. Indeed, DFT+$U$ largely improves the agreement with the experimental optical conductivity even without shifting the Fermi level (see Appendix for details).

We thus argue that band energies around the $M$-point should be renormalized by electronic correlations. Experimental study of the Fermi surface could be a stringent test for this conjecture. In the undoped KV$_3$Sb$_5$, the Fermi level around the $M$-point lies in between bands B and C and eliminates Fermi surface at the zone boundary. The presumed renormalization of band energies will create Fermi surface around $M$, in agreement with the ARPES results, where sizable intensity has been observed at the zone boundary [16].

### IV. CONCLUSIONS AND OUTLOOK

Our broadband optical spectroscopy experiments prove bulk nature of the CDW state in KV$_3$Sb$_5$. They further reveal two unusual features of this material, the phonon anomalies and the presence of a localization peak in low-energy optical absorption. Two scenarios can account for these observations. The first one relies on phonons as the key ingredient. The electron-phonon coupling drives the CDW formation and disturbs diffusive charge transport, resulting in the localization peak. The CDW state is then a mere non-magnetic phase with the star or hexagon (inverse star) type of charge order. Such a non-magnetic phase would hardly explain large extrin-
sianomalous Hall effect [16] in KV$_3$Sb$_5$, as well as chiral anisotropy detected in the recent tunneling spectroscopy experiment [19].

The second scenario stems from phonons and electronic correlations as two equally important aspects of the KV$_3$Sb$_5$ physics. Both electron-phonon and electron-electron interactions can be in fact responsible for the localization peak and CDW formation. Roles of these interactions may even be hard to separate. An intriguing observation in this context is that our search for the possible CDW structures of KV$_3$Sb$_5$ led to exact symmetries expected for density-wave states of the kagome Hubbard model. Electron-electron interactions central to this model will then lead to much richer physics than in the purely phonon case.

Theoretical results available for the kagome Hubbard model at the van Hove singularity [20, 50] and in other regimes [51–56] may offer several interesting insights into the KV$_3$Sb$_5$ physics. Spin bond order (triplet $p$-wave) embeds spin degrees of freedom in the form of spin currents that can not be detected via conventional probes, such as muon spectroscopy or neutron scattering [36], but may have influence on charge transport, including anomalous Hall effect. Our hexagon structure is the most stable type of CDW in KV$_3$Sb$_5$ and displays an intriguing similarity to this theoretically predicted spin bond order. On the other hand, doping the kagome Hubbard model away from the van Hove singularity leads to superconducting instabilities [50, 57, 58] that allow further interesting, yet to be explored connections to the recently established superconducting properties of KV$_3$Sb$_5$ and related materials [11–13, 32, 59–63].

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[FIG. 10. Dc resistivity comparison of different KV$_3$Sb$_5$ samples from the literature with the one used in the optical measurements.]

### Appendix A: Comparison of the K-content

KV$_3$Sb$_5$ samples are prone to be K-deficient, and intentional deintercalation of K atoms can even be used to tune the Fermi energy in these materials. We compared the sample used in the optical measurements with the ones from the literature, where the K-content was known. The comparison in Fig. 10 suggests that the sample used in the optical measurements is nearly stoichiometric.

### Appendix B: Additional computational results

#### 1. Optical conductivity

Fig. 11 shows optical conductivity calculated for different doping levels within DFT [panel (a)] and for the stoichiometric KV$_3$Sb$_5$ within DFT+$U$ [panel (b)]. Low-energy part of the spectrum is dominated by two peaks around 0.1 eV and 0.25 eV due to the B–C and B–D transitions, respectively. Relative intensities of these peaks strongly depend on the doping level and on adding correlations within DFT+$U$. Best agreement with the experimental optical conductivity is obtained upon shifting the Fermi level by +64 meV. This chemical potential corresponds to the doping level of 0.4e/f.u. A similar suppression of the 0.25 eV peak and enhancement of the 0.1 eV peak are observed in DFT+$U$ with the on-site Coulomb repulsion $U = 2$ eV and Hund’s exchange $J = 0$ eV that stabilize weak magnetic moment of 0.07 $\mu_B$ on vanadium atoms. Both methods seem to yield approximations to the real KV$_3$Sb$_5$ band structure, where band energies around the $M$-point should be renormalized compared to the DFT prediction.

We also note that low-energy optical conductivity of the CDW states, both star and hexagon structures, does
not show any significant dependence on the doping level and $U$ value. In this case, the absorption peak around 0.2 eV is observed systematically, in agreement with the experiment [Fig. 6(b)].

2. Possible spin states of CDW structures

Calculations on the DFT level (without adding $U$) suggest that both hexagon and star structures [Fig. 5] should be non-magnetic. Local magnetic moments can be nevertheless introduced in DFT+$U$ in the form of a conventional spin-density wave (SDW) where spin density is modulated throughout the crystal. Although different from spin bond order of the kagome Hubbard model, this is the best approximation that density-functional methods can offer. Here, we discuss such SDW’s that may appear in the charge-ordered hexagon and star structures. The values of energy and magnetic moment are quoted for $U = 2$ eV and $J = 0$ eV in DFT+$U$.

Two types of SDW, A and B, are possible in the star structure. Type-A SDW preserves six-fold symmetry and entails parallel spins on the inner hexagon of the star along with opposite spins on the outer sites of the star [Fig. 12(a)]. Slightly different magnetic moments of $0.85 \mu_B$ and $-1.0 \mu_B$, respectively, result in a net ferrimagnetic state. It is less stable than the antiferromagnetic state, type-B SDW with antiparallel spins on the inner hexagon [Fig. 12(b)]. It features higher magnetic moments of 1.3−1.4 $\mu_B$ and lies 50 meV/f.u. lower in energy compared to type-A SDW, which is in turn 35 meV/f.u. lower in energy than the non-magnetic state of the star structure. The total stabilization energy of the SDW is thus 85 meV/f.u. for the given choice of the DFT+$U$ parameters. Therefore, electronic correlations are likely to introduce local magnetism into the star structure and give rise to an antiferromagnetic type-B SDW that breaks six-fold symmetry of charge bond order.

Hexagon structure is very different. It does not support type-B SDW at all, whereas type-A SDW involves only tiny magnetic moments of 0.05 $\mu_B$ and shows no significant energy gain compared to the non-magnetic state (the energy gain due to the SDW is 0.5 meV/f.u. only). This result indicates that hexagon structure is not prone to the formation of conventional SDW’s, which then allows spin degrees of freedom to be embedded in more exotic forms, such as spatial modulation of spin currents in the spin bond order (tripllet $p$-wave) state of the kagome Hubbard model.

FIG. 11. Comparison of the experimental interband contribution to the optical conductivity at 90 K (above the CDW transition) with the DFT results: (a) for different hole and electron dopings expressed by the chemical potential $\mu$, and (b) for DFT+$U$ calculations at $\mu = 0$. Electron doping leads to a suppression of the peak around 0.25 eV and enhances the peak around 0.1 eV, resulting in the best match to the experiment. Similar effect can be reproduced with including $U$ on the DFT+$U$ level.

FIG. 12. SDW states of the star structure: (a) type-A SDW with parallel spins on the inner hexagon of the star; (b) type-B SDW with antiparallel spins on the inner hexagon. The type-B SDW breaks 6-fold symmetry of the charge bond order, increases local moment, and lowers total energy of the system. The high stability of this state indicates the proclivity of the star structure to the formation of a conventional spin-density wave.
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