Optical investigations of RbV₃Sb₅: Multiple density-wave gaps and phonon anomalies

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Temperature-dependent reflectivity studies on the non-magnetic kagome metal RbV₃Sb₅ in a broad energy range (50 cm⁻¹ – 20000 cm⁻¹; 6 meV – 2.5 eV) down to 10 K are reported. Below T_{CDW} = 102 K, the optical spectra demonstrate a prominent spectral-weight transfer from low to higher energies as the fingerprint of the charge-density wave (CDW) formation with the opening of a partial gap. A detailed analysis reveals two energy scales of, respectively, ~ 100 meV and 45 meV, the latter appearing only below 50 K. Additionally, two modes at, respectively, 160 cm⁻¹ and 430 cm⁻¹ can be traced over the whole measured temperature range. Strong anomalies are observed for both of these modes already above T_{CDW} with a further renormalization across T_{CDW}, suggesting the importance of the electron-phonon coupling in RbV₃Sb₅ in both normal and CDW states. While the 160 cm⁻¹ mode can be attributed to the E₁u phonon, the 430 cm⁻¹ mode could not be reproduced in our phonon calculations. The antiresonance nature of this mode gives hints for a nontrivial electron-phonon coupling in RbV₃Sb₅.

I. INTRODUCTION

Kagome metals became model compounds for studying effects of correlations along with topologically non-trivial electronic states [1]. Driven by the spatially separated metallic kagome planes, electronic structures of kagome metals feature flat bands and linearly dispersing topological Dirac bands, as has been shown in several magnetic systems of recent interest [2–5].

The non-magnetic AV₃Sb₅ (A = K, Rb, Cs) series opens up new opportunities to study electronic properties of the kagome metals [6]. These compounds crystallize in the P6/mmm space group with V atoms forming a kagome lattice and Sb1 atoms filling the centers of the hexagons. The kagome networks are stacked along the c-axis and separated by Sb2 honeycomb layers and A alkali atoms. This results in an overall quasi-2D structure with well-isolated kagome planes. All key features of a nearest-neighbor kagome metal – linear bands, saddle points, and flat bands – can be indeed distinguished in electronic structures of AV₃Sb₅ [7].

Transport and magnetic measurements on AV₃Sb₅ reveal a strong anomaly at T_{CDW} = 102 K for RbV₃Sb₅ [8] (78 K for KV₃Sb₅ [6] and 94 K for CsV₃Sb₅ [9]). Moreover, RbV₃Sb₅ becomes superconducting at T_c = 0.92 K [8] (0.93 K for KV₃Sb₅ [10] and 2.5 K for CsV₃Sb₅ [9]). While the exact nature of the high-temperature anomaly is still under debate, the robust nature of this transition under external magnetic fields suggests charge-density wave (CDW) as the plausible origin. Indeed, band saddle points (van Hove singularities) observed in AV₃Sb₅ in the vicinity of the Fermi level [11–13] should lead to a CDW instability of a kagome metal [14–16]. The resulting CDW state is quite unusual, as it features large anomalous Hall effect [17, 18], multiple energy gaps [11, 19, 20], and intrinsic chirality [18, 21, 22]. One general and hitherto unresolved issue is the clear distinction between the bulk and surface effects in the CDW state of AV₃Sb₅. For example, two different coexisting superstructures were observed in surface-sensitive mea-

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measurements of CsV$_3$Sb$_5$ [23], but only one of them is believed to occur in the bulk [24, 25]. A temperature-driven re-arrangement of the CDW order has been proposed for CsV$_3$Sb$_5$ as well [26].

Despite many similarities shared by all three compounds – KV$_3$Sb$_5$, RbV$_3$Sb$_5$, and CsV$_3$Sb$_5$ – they feature somewhat different energy scales and, possibly, different underlying interactions. Density-functional-theory (DFT) band-structure calculations could perfectly reproduce the optical response of CsV$_3$Sb$_5$ [27], but not of KV$_3$Sb$_5$ [28], suggesting that deviations from the band picture may be enhanced across the series as the size of the alkaline metal is reduced. This motivates a comparative optical study of RbV$_3$Sb$_5$ as the possible intermediate case.

Below, we show that this compound is indeed intermediate in terms of its bands near the Fermi level and their saddle points. We further confirm that several pertinent features – hindered electron dynamics witnessed by the low-energy localization peak, and strong electron-phonon coupling revealed by the broadening of the phonon modes – are common across the AV$_3$Sb$_5$ series. Additionally, we are able to pinpoint two distinct CDW gaps in RbV$_3$Sb$_5$. These gaps are integral to the putative topological state but could be previously observed with surface-sensitive techniques only [12, 29]. Our optical study confirms their bulk nature, thus setting a benchmark for the CDW state of AV$_3$Sb$_5$.

II. METHODS

A. Experimental

High-quality single crystals were prepared as explained in Refs. [6, 10]. For the optical measurements, a sample with the dimensions of ~1.5 × 1.5 mm$^2$ surface area was used, while the thickness of the specimen was around 0.2 mm. The sample was freshly cleaved prior to the optical experiments. On the same crystal, four-point resistivity measurements were performed in order to determine the CDW transition temperature and confirm the stoichiometry. The kink at 102 K marking the CDW transition [Fig. 2(b), inset], as well as the overall behaviour agrees well with the previous reports [8].

Temperature-dependent reflectivity measurements in the ab-plane over a broad frequency range from 50 to 20000 cm$^{-1}$ (6 meV – 2.5 eV) were performed down to 10 K. While for the high-energy range ($\omega > 600$ cm$^{-1}$) a Bruker Vertex 80v spectrometer with an incorporated Hyperion IR microscope was used, the low-energy range was measured with a Bruker IFS113v spectrometer and a custom-built cryostat. Freshly evaporated gold mirrors served as reference in these measurements. The absolute value of the reflectivity was obtained by an in-situ gold-overcoating technique in the far-infrared range, as described in Ref. [30].

Below 50 cm$^{-1}$, we use standard Hagen-Rubens extrapolation, considering the metallic nature of our sample, while for the high-energy range we utilize x-ray scattering functions for extrapolating the data [31]. The optical conductivity is then calculated from the measured reflectivity via Kramers-Kronig analysis.

B. Computational

Density-functional-theory (DFT) calculations of the band structure and optical conductivity were performed in the Wien2K [32, 33] code using Perdew-Burke-Ernzerhof flavor of the exchange-correlation potential [34]. We used experimental structural parameters from Ref. [6] for the undistorted RbV$_3$Sb$_5$ structure, whereas possible CDW structures were obtained by a structural relaxation in VASP [35, 36] similar to Ref. [28]. Spin-orbit coupling was included for the calculations of band structure and optical conductivity.

Self-consistent calculations and structural relaxations were converged on the $24 \times 24 \times 12$ k-mesh for the undistorted RbV$_3$Sb$_5$ structure (normal state) and $12 \times 12 \times 12$ k-mesh for the distorted structures (CDW). Optical conductivity was calculated on the k-mesh with up to 100 × 100 × 50 points for the normal state and 36 × 36 × 36 points for the CDW state.

III. RESULTS AND DISCUSSION

A. Optical discussion

Fig. 2 (a) displays the temperature-dependent reflectivity of RbV$_3$Sb$_5$. The high reflectivity values at low frequencies, as well as the Drude-like increase in the optical conductivity [Fig. 2 (b)] demonstrate the metallic nature of the sample. Conductivity values in the $\omega \rightarrow 0$ limit are obtained from the Hagen-Rubens fit of the reflectivity and match well with the four-probe dc resistivity measurements performed on the same sample, as shown in the inset of Fig. 2 (b). Upon going through the CDW transition, significant changes occur in the optical properties. Around 0.15 eV, a dip develops in the reflectivity, echoed by a spectral weight transfer to an additional peak in the optical conductivity below $T_{\text{CDW}}$. The low-energy optical conductivity is highlighted in Fig. 3, where the spectral-weight transfer is shown by the green arrow. The solid circles in the figure represent the dc conductivity values, whereas the orange arrow highlight the Fano-resonance.

The CDW transition also affects the free carriers dynamics, as seen in the temperature dependence of the screened plasma frequency, $\omega_p^{\text{screened}}$, which can best be estimated from the maximum in the dielectric loss function, $-\text{Im}(1/\varepsilon)$ [37]. The real plasma frequency is masked by the interband transitions, but it can be calculated as $\omega_p = \omega_p^{\text{screened}} \sqrt{\varepsilon_\infty}$ with the high-energy contributions, $\varepsilon_\infty$, fixed around 6 at all temperatures.
Therefore, the abrupt increase in $\omega_p^{\text{screened}}$ around $T_{CDW}$, as shown in the inset of Fig. 2 (c), is expected in the genuine plasma frequency, too. As the plasma frequency can also be given as $\omega_p^2 \propto n/m^*$ (n is the carrier density and $m^*$ is the effective mass), the overall increase in the plasma frequency can be attributed to either increasing carrier density or decreasing effective mass. For RbV$_3$Sb$_5$, as well as for its sibling compounds, a non-monotonic change of the carrier concentration was found in recent Hall effect measurements [8, 17, 18], thus giving support to the former interpretation.

**B. Decomposition**

Different contributions to the optical spectra can be modeled with the Drude-Lorentz approach, with $\varepsilon_\infty$ being the high-energy contributions to the real part of the dielectric permittivity [$\varepsilon = \varepsilon_1 + i\varepsilon_2$].

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_p^2 \text{Drude}}{\omega^2 + i\omega/\tau \text{Drude}} + \sum_j \frac{\Omega_j^2}{\omega_{0,j}^2 - \omega^2 - i\omega\gamma_j}. \quad (1)$$

Here, $\omega_p \text{Drude}$ and $1/\tau \text{Drude}$ are the plasma frequency and the scattering rate of the itinerant carriers, respectively. $\omega_{0,j}$, $\Omega_j$, and $\gamma_j$ describe the resonance frequency, width, and the strength of the $j^{th}$ excitation, respectively. The complex optical conductivity $[\tilde{\sigma} = \sigma_1 + i\sigma_2]$ is then calculated as

$$\tilde{\sigma}(\omega) = -i\omega[\varepsilon - \varepsilon_\infty]/4\pi. \quad (2)$$

An example of the decomposed experimental spectrum is given in Fig. 4 (a) for the room-temperature data. Several interband transitions at high frequencies are modeled by multiple Lorentzians, as shown in orange.

In addition to the classical Lorentzian and Drude contributions, a Fano-like shaped peak around 400 cm$^{-1}$, as well as a strongly temperature-dependent absorption feature in the same energy range, are observed. In line with the previous optical studies of the kagome metals, this feature is assigned to a so-called localization peak [27, 28, 38] that signals hindered electron dynamics.

**C. Localization Peak**

Having described the general decomposition of the optical spectrum, we now turn to a more detailed analysis of the localization peak. Its temperature evolution is plotted in Fig. 4 (b), after subtracting the Drude, phonon, and interband contributions from the experimental data. The strong and linear red-shift of the peak position upon cooling, given in the inset of Fig. 4 (b), makes this feature clearly distinguishable from the interband transi-
tions. Furthermore, the interband transitions are well reproduced by DFT calculations, as discussed in Sec. III F, however DFT does not show any indications for this additional peak. This gives further evidence for interpreting this peak as arising from an intraband process.

The following analysis is based on the model of a displaced Drude peak by Fratini et al. [39], where the classical Drude response is modified with the backscattering of the electrons, $1/\tau_b$, leading to a shift of the zero-frequency response to a finite value.

$$\sigma_1(\omega) = C \left(\frac{\tau_b}{\tau} - \frac{\tanh\left(\frac{\hbar \omega}{2k_B T}\right)}{\hbar \omega}\right) \cdot \text{Re}\left\{\frac{1}{1 - i\omega \tau} - \frac{1}{1 - i\omega \tau_b}\right\}.$$  

Here, $C$ is a constant, $\hbar$ is the reduced Planck constant, $k_B$ the Boltzmann constant, and $\tau$ is the elastic scattering time of the standard Drude model.

While its origin is still under debate, a temperature-dependent low-energy absorption feature is commonly found in strongly correlated electronic systems and is also observed in the sibling compounds, KV$_3$Sb$_5$ and CsV$_3$Sb$_5$ [27, 28, 40]. The formalism, as presented in Ref. [39], assumes possible localization effects, due to interactions of charge carriers with low-energy degrees of freedom, like phonons, electric or magnetic fluctuations, which lead to a backscattering of the electrons.

For a further insight, we plotted the elastic scattering and the backscattering of the localization peak in Fig. 5 (a). As noted by the arrow, the elastic scattering shows a slight change across the CDW transition. Considering the partial gapping of the Fermi surface [12, 29] and the slight change of the carrier density across $T_{CDW}$, this small change is expected. On the other hand, the backscattering mechanism seems to be unaffected by the CDW. In Fig. 5 (b), we have also shown the elastic scattering of the Drude component, which is overlayed with the dc resistivity. The remarkably similar temperature evolution suggests that the dc transport is mainly governed by the Drude component and not by the incoherent localization peak.

Regardless of the exact microscopic interpretation of this localization peak, a striking feature of its temperature evolution is the insensitivity to $T_{CDW}$. The effects that hinder electron dynamics seem to be purely thermal in nature and vanish in the $T \to 0$ limit. A similar behavior is observed in CsV$_3$Sb$_5$ [27], but not in KV$_3$Sb$_5$ where the localization peak also shifts toward low energies upon cooling, yet it saturates at around 300 cm$^{-1}$ and does not reach zero energy even at 10 K [28].

FIG. 3. Low-energy optical conductivity shown for selected temperatures. The solid lines are the fits to the experimental spectra as described in the text. While at 10 K the localization peak lies outside of the measurement range, its position can be estimated assuming linear temperature dependence and using the high-energy tail, which is still visible in our data. The solid circles are the fixed dc conductivity values obtained from the Hagen-Rubens fits. The orange arrow marks the Fano resonance, and the green arrow illustrates the spectral weight transfer to an additional peak that appears below $T_{CDW}$.

FIG. 4. (a) Decomposition of the optical conductivity at room temperature, consisting of a Drude peak (purple), a localization peak (green), a phonon (yellow), a Fano resonance (blue), and multiple interband transitions (orange). (b) Temperature dependence of the localization peak obtained after subtracting all other contributions from the experimental spectra. The inset shows the temperature evolution of the peak position.
D. Energy scale of the charge-density wave

One of the key features of the AV₃Sb₅ family is the formation of a density-wave state accompanied by a superstructure [9]. While there is a consensus on the 2 × 2 in-plane modulation [22, 41–45], both two-fold and four-fold modulations along the c axis have been proposed [23, 44, 46, 47]. This superstructure formation is concomitant with the reduction in the density of states at the Fermi level. Recent quantum oscillation [46] and ARPES [11, 12] studies suggested that vanadium bands around the M point become gapped, in line with theoretical proposals of band saddle points at M as the main origin of the density-wave instability [15, 48]. The presence of multiple d-bands in AV₃Sb₅ [7] implies that this instability can have more than one energy scale revealed by several gap features that were indeed detected spectroscopically, but only with surface-sensitive techniques, so far [11, 12, 20].

Here we show for the first time that these multiple gaps can be also observed in the bulk optical probe. Fig. 6(a) displays the conventional spectral weight transfer from low to higher energies, as expected in the CDW formation. This spectral weight transfer from the peak at 70 meV is observed down to 50 K. A very different behavior is seen at lower temperatures where the low-energy spectral weight starts growing again [Fig. 6(b)]. A convenient way to trace these spectral weight transfers is by using the spectral weight ratio below and above T_{CDW}, similar to, e.g., iron pnictides [49].

The spectral weight (SW) is obtained as \( \frac{120}{\pi} \int_0^\infty \sigma_1(\omega)d\omega \). The cutoff value of \( \omega \) is chosen through the entire measured range taking into account the interband transitions only as shown in Fig. 6. The SW ratio, namely, SW(T)/SW(110 K), gives the characteristics of the SW transfers (transfer direction, energy scales, etc.) in the CDW state. If there is a SW transfer from high to low energies (e.g., Drude narrowing), the ratio is above “1” at low energies and then approaches unity. On the other hand, when there is a SW transfer from low to high energies, the ratio is below “1” until the energy transfer is completed. Here, we analyze only the spectral weight related to the interband transitions and thus directly probe the energy scales associated with the density wave.

In Fig. 7(a), the SW ratio for several temperatures below T_{CDW} is plotted. The SW transfer happens in the energy range below 0.35 eV, as the ratio levels off to unity at higher energies. The inflection point reflects the appearance of the new absorption channel that can be associated with the energy scale of the density-wave gap, \( \Delta \). It is better represented by the second derivative, as presented in Fig. 7(b). While a clear peak at around 90 meV marks the energy scale of the gap, an additional peak becomes clearly visible below 50 K. We suggest that this second energy scale is also related to the density-wave formation and serves as the bulk probe for the multigap scenario inferred from the ARPES measurements that reported the gaps of 130 meV and 80 meV, both of them highly anisotropic across the Brillouin zone [12]. Since this anisotropy is averaged out in the optical measurement, the exact gap values may differ from those in ARPES, but the qualitative behavior is remarkably similar.

Furthermore, larger energy gap shows a rather abrupt increase right below T_{CDW}, where the behavior is in line with the other AV₃Sb₅ systems [27, 28]. On the other hand, the smaller energy gap seems to develop gradually with a mean-field like behavior. However, we should point out that the exact onset temperature for this second gap cannot be determined with the current measurements.

The unconventional appearance of the CDW in dc resistivity also support the multiband scenario. Usually, opening of a density wave gap accompanies by the metal-insulator transition or jumps in dc resistivity due to the
reduction of the density of states [50]. However, here we only observe a kink in dc resistivity suggest that the carriers contributing to the dc conduction are not significantly affected by the density wave formation. Here possibly the carriers giving rise to the localization peak are the ones that are gapped out, which would also explain the non-responsive dc resistivity that is mainly governed by the Drude component [Fig. 5(b)].

E. Phonon modes

Besides these electronic features, two modes reminiscent of phonon excitations are visible at, respectively, 160 cm\(^{-1}\) and 430 cm\(^{-1}\) in the room-temperature spectrum [Fig. 8(b)]. While the low-energy mode is readily assigned to the IR-active E\(_{1u}\) phonon according to our DFT calculations [Fig. 8(a)], the high-energy mode cannot be interpreted in the same manner, because no corresponding IR-active phonons are found above 250 cm\(^{-1}\) in DFT [Fig. 8(a)]. Experimentally, both modes show strong anomalies across \(T_{CDW}\), as well as indications for a strong coupling to the electronic background.

The lower 160 cm\(^{-1}\) mode can be represented with a single Lorentzian,

\[
\sigma_1(\omega) = \frac{\Delta \varepsilon \omega_0^2 \gamma}{4\pi[(\omega^2 - \omega_0^2)^2 + \gamma^2\omega^2]} \quad (3)
\]

Here, \(\omega_0, \Delta \varepsilon,\) and \(\gamma\) stand for the resonance frequency, intensity, and linewidth of the phonon mode, respectively. The obtained parameters in Fig. 8(c-e) reveal a strong increase in the intensity, \(\Delta \varepsilon\), along with a pronounced red shift upon cooling. At low temperatures, the mode is masked by the localization peak that has strong influence on the obtained parameters. The increased broadening upon cooling is a signature of strong electron-phonon coupling above \(T_{CDW}\). Below \(T_{CDW}\), this coupling probably persists, but also the splitting of the mode into multiple phonons within the CDW state may contribute to the broadening [28].

The higher 430 cm\(^{-1}\) mode is better reproduced by a Fano-like response,

\[
\sigma_1(\omega) = \frac{\Delta \varepsilon \omega_0 \gamma[\gamma(\omega^2 - 1) + 2\omega(\omega^2 - \omega_0^2)]}{4\pi[(\omega^2 - \omega_0^2)^2 + \gamma^2\omega^2]} \quad (4)
\]

The additional parameter, \(q\), is the dimensionless coupling constant that describes the asymmetry of the mode and also gauges the scale of the coupling to the electronic background. We note that \(q\) also takes negative values, as the high-energy mode is a strong antiresonance. The obtained parameters for the antiresonance are given in Fig. 8(f-i). A similar red-shift of the resonance frequency, \(\omega_0\), is accompanied by an increase in the intensity, \(\sigma_0\). In contrast to the low-energy mode, this feature sharpens upon cooling, which is the expected behavior with decreasing thermal effects. The coupling parameter, \(q \approx -3\), remains similar across the whole temperature range and suggests that the coupling persists even below \(T_{CDW}\). While the anomalies at \(T_{CDW}\) are clearly visible, a closer look at the intensity and the coupling constant reveals also the secondary anomalies around 50 K. The sudden increase in the intensity is accompanied by the singularity in \(q^2\), suggesting that the antiresonance responds to changes in the electronic structure when the second CDW gap appears.

The fact that the high-energy mode cannot be assigned to a Γ-point phonon goes hand in hand with the unusual antiresonance nature of this mode. A similar antiresonance has been observed in KV\(_3\)Sb\(_3\) around 480 cm\(^{-1}\), but in that case it could be regarded as an overtone of a Γ-point phonon [28]. Such an interpretation is clearly excluded in RbV\(_3\)Sb\(_3\) where no IR-active phonon appears in the 200 – 220 cm\(^{-1}\) range. On the other hand, such a mode could arise from a non-Γ-phonon as a result of a strong interaction with electronic degree of freedom. Similar antiresonances have been discussed in functionalyzed graphene and modeled with the involvement of the...
We find a good agreement between the Fermi level is shifted upwards by 41 meV. A similar calculation, and phonon modes have been subtracted to form the curves in (b) are given in (c–e) for the phonon mode and (f–i) for the Fano antiresonance. The blue and green lines correspond to $T_{CDW}$ and the onset temperature of the lower CDW gap, respectively.

$K$-point phonons. Another interesting observation is the prominent red shift that mirrors the red shift of the localization peak (Sec. III C), although the former appears only below $T_{CDW}$, while the latter is observed over the entire temperature range.

**F. Interband transitions**

The interband transitions are elucidated by DFT calculations of the optical conductivity. As seen in Fig. 9 (b), the low-energy contributions to the in-plane optical conductivity, $\sigma_{xx}$, are restricted to the transitions between bands $B$ and $C$, which occur in the vicinity of the $L$ and $M$ points of the Brillouin zone (see Fig. 9 (a)). In the $0.2−1.0$ eV range, the optical conductivity is dominated by three contributions that arise from the transitions $A \rightarrow C$, $B \rightarrow D$, and $B \rightarrow E$.

The experimental interband conductivity is given in Fig. 10 (a) below and above $T_{CDW}$, after the Drude, localization, and phonon modes have been subtracted to allow for a direct comparison with the DFT results presented in Fig. 10 (b). We find a good agreement between experiment and calculations in the normal state when the Fermi level is shifted upwards by 41 meV. A similar upward shift by 64 meV was required in the case of KV$_3$Sb$_5$ [28], indicating that both compounds deviate from the band picture, and a slight renormalization of band energies is required for a proper description of their electronic structure.

For the density-wave state, two different distorted structures were considered according to the star-of-David and tri-hexagonal in-plane distortions (demonstrated in Fig. 1), as discussed in [28, 46, 51, 52]. The position of the additional peak in the spectrum below $T_{CDW}$ is best reproduced by the star-of-David CDW. On the other hand, this model also predicts a broad low-energy absorption feature, which we do not see in the experiment. Moreover, the peak around 70 meV in the experimental spectrum is not seen in the calculated star-of-David spectrum, however, it is reproduced by the tri-hexagonal CDW. We also note that the tri-hexagonal CDW has a higher stabilization energy of 5 meV/f.u. (relative to the normal state) compared to 1.5 meV/f.u. for the star-of-David CDW.

The direct comparison of the optical spectra of all three compounds of the AV$_3$Sb$_5$ series reveals important changes in the band structure and electron dynamics that appear prominently in the low-energy optical response. The interband transitions and the red-shifting localization peak are common across the whole series. Moreover, the sharp Drude peak mirrors the highly metallic nature of these systems. On the other hand, several differences need to be pointed out. (i) The Drude scattering is strongest in RbV$_3$Sb$_5$, causing a very broad Drude peak and a nearly flat optical conductivity in the low-energy range at high temperatures, as it overlaps with the localization peak. (ii) The low-energy interband spectrum of RbV$_3$Sb$_5$ strongly resembles the one of KV$_3$Sb$_5$ with one intense absorption peak below 1000 cm$^{-1}$, in contrast to the three weaker peaks in CsV$_3$Sb$_5$ (see Fig. 11 (a)). This change in the interband absorption reflects the
toward the inverted case of CsV$_3$. Two saddle points become closer in energy, thus evolving band picture across the $A$ version is clearly visible as the change in the interband electronic correlations in a series of materials, including cuprates, iron pnictides, and topologically nontrivial Dirac systems [53, 54]. In this comparison, the scaling parameter, $SW_{\text{experiment}}/SW_{\text{band}}$, is close to 1 for uncorrelated materials like simple metals and zero for the most correlated class of Mott insulators. Other classes of materials fall in between these two limits, with the ratios below 0.5 taken as indications for a highly correlated nature of the system.

In Fig. 12, we compare the scaling factors for the AV$_3$Sb$_5$ series. Experimental plasma frequencies for KV$_3$Sb$_5$ and CsV$_3$Sb$_5$ are taken from previous studies [27, 28]. While KV$_3$Sb$_5$ is the most correlated member of the AV$_3$Sb$_5$ family, correlations seem to play only a minor role in CsV$_3$Sb$_5$. This result corroborates our earlier observation that a renormalization of band energies should take place in KV$_3$Sb$_5$ but not in CsV$_3$Sb$_5$. Finally, RbV$_3$Sb$_5$ takes an intermediate position, but lies notably closer to KV$_3$Sb$_5$ and deviates from the band picture too. The similarities in the phonon anomalies and Fano antiresonance are also common to KV$_3$Sb$_5$ and RbV$_3$Sb$_5$. They may be associated with the moderately correlated nature of these compounds.

FIG. 9. (a) Calculated band structure of RbV$_3$Sb$_5$. (b) Calculated $\sigma_{xx}$ component of the optical conductivity and its band-resolved contributions. Fermi level is shifted upwards by 41 meV.

With the notation used in Ref. [15], the saddle points are identified as $\Gamma_1^+$ at $-105$ meV and $\Gamma_3^+$ at $-70$ meV in RbV$_3$Sb$_5$, $-120$ meV and $-60$ meV in KV$_3$Sb$_5$, and $-30$ meV and $-95$ meV in CsV$_3$Sb$_5$, respectively. The two saddle points are inverted in CsV$_3$Sb$_5$ with respect to the KV$_3$Sb$_5$ and RbV$_3$Sb$_5$. Experimentally, this inversion is clearly visible as the change in the interband absorption. From electronic structure point of view, RbV$_3$Sb$_5$ appears to be more similar to KV$_3$Sb$_5$, yet its two saddle points become closer in energy, thus evolving toward the inverted case of CsV$_3$Sb$_5$.

Another interesting point is the deviation from the band picture across the AV$_3$Sb$_5$ series. It has been detected in KV$_3$Sb$_5$ and RbV$_3$Sb$_5$, where an upward shift of the Fermi level was required in order to reproduce interband absorption with DFT. On the other hand, no such shift was needed in the case of CsV$_3$Sb$_5$ [27]. This difference can also be seen from the perspective of plasma frequencies that are compared between the experiment and DFT calculations. Previously, such a comparison of the experimental Drude spectral weight (plasma frequency squared) with the band theory was used as a gauge of electronic correlations in a series of materials, including cuprates, iron pnictides, and topologically nontrivial Dirac systems [53, 54]. In this comparison, the scaling parameter, $SW_{\text{experiment}}/SW_{\text{band}}$, is close to 1 for uncorrelated materials like simple metals and zero for the most correlated class of Mott insulators. Other classes of materials fall in between these two limits, with the ratios below 0.5 taken as indications for a highly correlated nature of the system.

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IV. CONCLUSIONS

A detailed temperature-dependent optical study is presented for RbV₃Sb₅ in the energy range of 10 meV–2 eV and at temperatures down to 10 K. Our data witness the highly metallic nature of the compound and the CDW transition at T_{CDW}. The evolution of the interband absorption across the AV₃Sb₅ series mirrors changes in the band structure and especially the re-arrangement of the band saddle points (van Hove singularities) around M. The detailed comparison suggest a close similarity between RbV₃Sb₅ and KV₃Sb₅.

An unexpected feature of the low-energy optical response is the prominent localization peak that signals hindered electron dynamics. In both RbV₃Sb₅ and CsV₃Sb₅, this peak shifts to low frequencies upon cooling and merges with the Drude peak at T → 0. Therefore, the localization effects in these compounds should be mostly temperature-driven and possibly related to phonons. On the other hand, the localization peak in KV₃Sb₅ remains at finite frequencies at T → 0. This difference is in line with the enhanced correlation effects observed in KV₃Sb₅ (Fig. 12).

Below T_{CDW}, RbV₃Sb₅ shows clear signatures of two energy scales associated with the CDW. These energy scales are in a good agreement with ARPES and confirm the bulk nature of both energy gaps that could be previously detected by surface-sensitive techniques only. We note that the agreement between the experimental and calculated optical conductivities is clearly less favorable in the CDW state than in the normal state. Each of the CDW models – tri-hexagonal and star-of-David – reproduces certain features of the experimental data but fails to reproduce the entire spectrum. Together with the presence of two distinct energy gaps, this may indicate the combination of multiple order parameters and the need to combine different types of distortions for a realistic modeling of the CDW in RbV₃Sb₅ and in the whole AV₃Sb₅ series.

Last but not least, the prominent modes at 160 cm⁻¹ and 430 cm⁻¹ both reveal a strong coupling to the electronic background and put forward electron-phonon coupling as an important ingredients of the AV₃Sb₅ physics. Intriguingly, the 430 cm⁻¹ mode shows strong antiresonance and can be assigned neither to an IR-active phonon, nor to an overtone. The intriguing electron-phonon coupling in the AV₃Sb₅ series certainly calls for a further dedicated investigation.

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