Electronic Correlations and Evolution of the Charge-Density Wave in the Kagome Metals $AV_3Sb_5$ ($A = K, Rb, Cs$)

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The kagome metals $AV_3Sb_5$ ($A = K, Rb, Cs$) have attracted enormous interest as they exhibit intertwined charge-density wave (CDW) and superconductivity. The alkali-metal dependence of these characteristics contains pivotal information about the CDW and its interplay with superconductivity. Here, we report optical studies of $AV_3Sb_5$ across the whole family. With increasing alkali-metal atom radius from K to Cs, the CDW gap increases monotonically, whereas $T_{CDW}$ first rises and then drops, at variance with conventional CDW. While the Fermi surface gapped by the CDW grows, $T_c$ is elevated in CsV$_3$Sb$_5$, indicating that the interplay between the CDW and superconductivity is not simply a competition for the density of states near $E_F$. More importantly, we observe an enhancement of electronic correlations in CsV$_3$Sb$_5$, which suppresses the CDW but enhances superconductivity, thus accounting for the above peculiar observations. Our results suggest electronic correlations as an important factor in manipulating the CDW and its entanglement with superconductivity in $AV_3Sb_5$.

The kagome lattice, composed of hexagons and corner-sharing triangles, provides a fascinating playground for exploring exotic quantum phenomena. For instance, spins or magnetic moments on a kagome lattice are subject to high degree of geometric frustration that may lead to quantum spin liquids [12]: electrons in a kagome lattice form flat bands, Dirac points, and saddle points, which support intriguing quantum phenomena associated with nontrivial band topology [3, 7] and a wide variety of electronic instabilities [8–11]. Particularly at van Hove filling, as a function of the on-site repulsion $U$ and nearest-neighbor Coulomb interaction $V$, the kagome lattice exhibits a rich phase diagram consisting of various phases such as charge or spin bond order [11, 11], unconventional superconductivity [8, 10, 11], charge-density wave (CDW) [11], and spin-density wave (SDW) [8].

The recently discovered kagome metals $AV_3Sb_5$ ($A = K, Rb, Cs$) [12] with the Fermi level $E_F$ lying near the saddle points (van Hove filling) provide an excellent platform to realize the above exotic quantum states in real materials. Multiple topologically protected Dirac bands [13, 13] and superconductivity with a transition temperature $T_c$ of 0.92–2.5 K [13, 13] have been reported in these compounds. In addition, a CDW transition occurs at $T_{CDW} = 78, 103$, and 94 K for KV$_3$Sb$_5$, RbV$_3$Sb$_5$, and CsV$_3$Sb$_5$, respectively [12, 13], resulting in a three-dimensional (3D) $2 \times 2 \times 2$ superlattice [16, 17]. Upon entering the CDW state, a giant anomalous Hall effect [13, 13] and electronic nematicity [20, 23] also emerge. The application of pressure [24, 27], uniaxial strain [28], or chemical doping [29, 32] suppresses the CDW order, but enhances the superconductivity, signifying the competition between the CDW and superconductivity in $AV_3Sb_5$. While a variety of studies suggest that the saddle point or Fermi surface (FS) nesting plays an important role in driving the CDW instability [33–40], there is also evidence that the CDW phase is mainly driven by electron-phonon (e-ph) coupling [41, 41]. At the present time, the driving mechanism of the CDW and how it interacts with superconductivity in $AV_3Sb_5$ are subjects for intensive debate.

The evolution of the CDW and superconducting properties with alkali metal in $AV_3Sb_5$ may reveal key information about the factors controlling the CDW order and its interplay with superconductivity. In this Letter, we systematically study the optical properties of $AV_3Sb_5$ across the whole family. As the alkali-metal atom radius grows from K to Cs, the CDW gap $\Delta_{CDW}$ increases monotonically, whereas $T_{CDW}$ first rises but then drops, failing to exhibit a scaling relation with $\Delta_{CDW}$. This anomalous behavior is clearly at odds with the description of conventional CDW order. While the FS removed by $\Delta_{CDW}$ increases, $T_c$ is raised in CsV$_3$Sb$_5$, indicating that the interplay between the CDW and superconductivity is not simply a competition for the density of states (DOS) near $E_F$. Moreover, we observe an enhancement of electronic correlations in CsV$_3$Sb$_5$, which suppresses the CDW but promotes unconventional superconductivity, giving rise to the above peculiar behavior. Our results underline the importance of electronic correlations in manipulating the CDW and its entanglement with superconductivity in $AV_3Sb_5$. 
For all three compounds, above $T_c$, at several representative temperatures above and below $T_c$, a peak centered at zero frequency, can be clearly observed, respectively. The details of optical measurements were carried out to confirm the CDW transition at $T_{CDW}$ for KV$_3$Sb$_5$, RbV$_3$Sb$_5$, and CsV$_3$Sb$_5$, respectively. The inset displays $\Delta \sigma_1(\omega)$ at 5 K for each compound, where $\sigma_1(\omega)$ at $T$ just above $T_{CDW}$ is used as the base curve.

High-quality single crystals of AV$_3$Sb$_5$ ($A = K, Rb, Cs$) were synthesized using the self-flux method and characterized by x-ray diffraction. Transport measurements were carried out to confirm the CDW transition at $T_{CDW} = 78, 103$, and 92 K in KV$_3$Sb$_5$, RbV$_3$Sb$_5$, and CsV$_3$Sb$_5$, respectively. The details of optical measurements can be found in the Supplemental Material.

Figures 1(a)–1(c) show the measured reflectivity $R(\omega)$ up to 3000 cm$^{-1}$ at several representative temperatures for KV$_3$Sb$_5$, RbV$_3$Sb$_5$, and CsV$_3$Sb$_5$, respectively. Above $T_{CDW}$, $R(\omega)$ for all three compounds exhibits metallic behavior: a very high $R(\omega)$ in the far-infrared range that approaches unity in the zero-frequency limit and increases with decreasing $T$. Below $T_{CDW}$, a suppression of $R(\omega)$ in the frequency range of 1000–1500 cm$^{-1}$ occurs for all three materials, signaling the opening of a CDW gap. As the radius of the alkali-metal atom grows from K to Cs, the suppression in $R(\omega)$ shifts to higher frequency and deepens, suggesting that the CDW gap increases in energy and the gap-induced FS modification intensifies with growing alkali-metal atom radius.

Figures 2(a)–2(f) show the optical conductivity $\sigma_1(\omega)$ at several representative temperatures above and below $T_{CDW}$ for KV$_3$Sb$_5$, RbV$_3$Sb$_5$, and CsV$_3$Sb$_5$, respectively. For all three compounds, above $T_{CDW}$, a Drude response, i.e., a peak centered at zero frequency, can be clearly observed in the low-frequency $\sigma_1(\omega)$, in good agreement with the metallic nature of these materials; below $T_{CDW}$, a dramatic suppression of the low-frequency $\sigma_1(\omega)$ sets in, and the removed spectral weight [the area under $\sigma_1(\omega)$] is transferred to higher frequencies, which is the prototypical response of the CDW gap in $\sigma_1(\omega)$. The detailed evolution of $\sigma_1(\omega)$ with $T$ is traced out in the 2D temperature-frequency ($T$-$\omega$) maps in Figs. 2(a)–2(c) for all three materials. The horizontal dashed line in each panel denotes $T_{CDW}$ for KV$_3$Sb$_5$, RbV$_3$Sb$_5$, and CsV$_3$Sb$_5$, respectively. Below $T_{CDW}$, the opening of the CDW gap leads to the presence of a blue region in the low-frequency range [a suppression of the low-frequency $\sigma_1(\omega)$] and a shift of the green/cyan region to higher frequencies. A comparison of Figs. 2(a)–2(c) reveals that as the radius of the alkali-metal atom in AV$_3$Sb$_5$ increases, the low-frequency blue region moves to higher frequencies and grows in area. These observations indicate that not only does the CDW gap value increase, the removed spectral weight due to the opening of the CDW gap also grows with increasing alkali-metal atom radius.

The CDW gap $\Delta_{CDW}$ can be determined from the zero-crossing point in the difference optical conductivity,

$$\Delta \sigma_1(\omega) = \sigma_1^{T < T_{CDW}}(\omega) - \sigma_1^{N}(\omega),$$

where $\sigma_1^{T < T_{CDW}}(\omega)$ represents $\sigma_1(\omega)$ at $T < T_{CDW}$; $\sigma_1^{N}(\omega)$ refers to $\sigma_1(\omega)$ in the normal state, namely at
The values of $\Delta S$ at $T = 5$ K determined from Eq. (2) are 6.00, 6.49, and $6.74 \times 10^6 \Omega^{-1} \text{cm}^{-2}$ for KV$_3$Sb$_5$, RbV$_3$Sb$_5$, and CsV$_3$Sb$_5$, respectively. The increase of $\Delta S$ with increasing alkali-metal atom radius indicates that a larger portion of the FS is removed by $\Delta CDW$ in AV$_3$Sb$_5$ with a larger alkali-metal atom. We plot $\Delta S$ at $T = 5$ K as a function of alkali metal in Fig. 3(d) for further discussions.

The ratio of the experimental kinetic energy $K_{exp}$ to the kinetic energy from band theory $K_{band}$ provides crucial information about the electronic correlations in AV$_3$Sb$_5$ [51, 52]. The electron’s kinetic energy can be conveniently derived from the area under the Drude feature in $\sigma_1(\omega)$ [51, 52],

$$K = \frac{2\hbar^2 c_0}{e^2} \int_0^{\omega_c} \sigma_1(\omega) \, d\omega,$$  

where $c_0$ is the distance between the V kagome layers, and $\omega_c$ is a cutoff frequency covering the entire Drude component in $\sigma_1(\omega)$. In order to determine $K_{band}$, we calculated the $ab$-plane $\sigma_1(\omega)$ for all three materials [44]. As depicted in Figs. 3(a), 3(c), the calculated $\sigma_1(\omega)$ spectra (blue curves) qualitatively agree with the measured ones (red curves). Using $\omega_c = 5000$ cm$^{-1}$ for both the measured and the calculated $\sigma_1(\omega)$, the values of $K_{exp}/K_{band}$ are obtained for all three compounds.
and Raman [44] measurements on CsV3Sb5, an increase of $\Delta_{CDW}$ coincides with a decrease of $T_{CDW}$, implying that besides e-ph coupling and FS nesting which should enhance both $\Delta_{CDW}$ and $T_{CDW}$, a competing factor that suppresses $T_{CDW}$ also exerts considerable influence on the CDW order. Previous studies on transition metal dichalcogenides have documented that while e-ph coupling, FS nesting, and a high DOS are beneficial to the formation of CDW order, electronic correlations act as a competing factor which tends to localize the carriers and prevents the formation of CDW order [50, 60]. Our optical results have attested to the decrease of $K_{exp}/K_{band}$ [Fig. 4(f)], i.e., the enhancement of electronic correlations in CsV3Sb5. These facts bring us to the possibility that the suppression of $T_{CDW}$ and the larger $2\Delta_{CDW}/k_B T_{CDW}$ in CsV3Sb5 may be related to the enhancement of electronic correlations.

Other interesting observations emerge from the comparison of $T_c$, $T_{CDW}$, $\Delta_S$, and $2\Delta_{CDW}/k_B T_{CDW}$. As shown in Figs. 4(b) and 4(e), the suppression of $T_{CDW}$ in CsV3Sb5 is accompanied by an enhancement of $T_c$, in accord with the competition relation between the CDW and superconductivity reported by previous work [24-30]. The competition between the CDW and superconductivity is not surprising, because the opening of the CDW gap depletes the DOS near $E_F$, resulting in a suppression of superconductivity. However, the comparison of Figs. 4(d) and 4(c) reveals that the rise of $T_c$ in CsV3Sb5 coincides with an increase of $\Delta S$. This observation implies that the CDW instability and superconductivity in AV3Sb5 do not share the same DOS, so that the competition between the CDW and superconductivity is not simply through competing for effective DOS near $E_F$, but controlled by another factor. Furthermore, $2\Delta_{CDW}/k_B T_{CDW}$ [Fig. 4(c)] and $T_c$ [Fig. 4(e)] exhibit identical alkali-metal dependence, hinting that the suppression of $T_{CDW}$ and the enhancement of $T_c$ in CsV3Sb5 are most likely induced by the same factor, namely electronic correlations. A recent theoretical study has shown that electronic correlations suppress the charge susceptibility but significantly enhance the spin susceptibility or spin fluctuations [59], which are believed to mediate unconventional superconductivity [61]. Moreover, extensive studies on cuprates and iron pnictides have underlined the importance of electronic correlations in generating unconventional superconductivity [57, 62]. Combining these studies with our observations, we suggest that electronic correlations should be taken into account when constructing a theory to describe the CDW phase and its entanglement with superconductivity in AV3Sb5.

To summarize, we performed a systematic investigation into the optical properties of AV3Sb5 ($A = K, Rb, Cs$) across the whole family. We found that as the alkali-metal atom radius grows from K to Cs, (i) while $\Delta_{CDW}$ increases monotonically, $T_{CDW}$ rises in RbV3Sb5 but then
drops in CsV₃Sb₅, at odds with conventional CDW; (ii) the FS removed by ΔCDW increases, whereas Tc is enhanced in CsV₃Sb₅, suggesting that the interplay between the CDW and superconductivity is not simply a competition for effective DOS near E_F; (iii) K_exp/K_band is reduced, indicating an enhancement of electronic correlations. An analysis considering all the above observations and previous work suggests that the enhancement of electronic correlations may be a decisive factor that controls the formation of the CDW order and its entanglement with superconductivity in AV₃Sb₅.

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