Order-disorder Peierls instability in the kagome metal (Cs,Rb)V$_3$Sb$_5$

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The nature of the charge density wave phases in the kagome metal compound AV$_3$Sb$_5$ has raised many questions and their origin is still under debate. Here, we combine thermal diffuse scattering and inelastic x-ray scattering to identify a 3-dimensional (3D) precursor of the charge order with propagation vector 0.5 0 0.5, which condenses into a 3D-CDW through a first order phase transition. The quasi-elastic critical scattering indicates that the dominant contribution to the diffuse precursor is the elastic central peak without phonon softening. However, the inelastic spectra show a small broadening of the Einstein-type phonon mode on approaching the phase transition. Our results point to the situation where the Fermi surface instability at the L point is of order-disorder type with a critical growth of quasi-static domains. The results go beyond the classical weak-coupling Peierls transition dynamics and are discussed within models including strong-electron phonon coupling and non-adiabaticity.

The intricate relationship between lattice geometry and topological electronic behavior determines the ground state properties of materials. The non-trivial band topology of the kagome lattice is being extensively explored as candidates to engineer Dirac fermions [1], topological flat bands [2], magnetic Weyl semimetals [3] or quantum spin liquid behavior [4, 5]. Besides, the combination between the non-trivial topology and strong electronic correlations, derived from the interplay of orbital, charge and spin degrees of freedom, introduces topological correlated physics [6] with more exotic phenomena, like topological Majorana modes.

Recently, a new family of non magnetic kagome metals AV$_3$Sb$_5$ (A=K, Rb, Cs) has emerged as a fertile playground to investigate correlation-driven topological phases [7, 8]. Derived from the kagome structure, this family of materials features a quasi 2D electronic band structure with van Hove singularities (vHs) and Dirac crossings close to the Fermi level [9, 10]. Superconductivity sets in at low temperature and forms an intertwined ground state with a charge density wave phase (CDW) [11]. Although the electronic structure derived from density functional theory (DFT) calculations is well established, controversy surrounds the origin and stabilization of the CDW, and the mechanism of superconductivity and its gap symmetry [12–14].

The rich phase diagram of the kagome lattice was investigated against Fermi surface instabilities within the Hubbard model, revealing spin and CDW orders and unconventional superconductivity emerging upon tuning the band filling to the vHs [15, 16]. Diffraction experiments and scanning tunneling microscopy (STM) unraveled multiple-q CDW modulations with an inverse Star of David structure [17, 18], following the phonon instabilities at the M and L points predicted by DFT [14]. The q vector at M connects neighbouring vHs in the vicinity of the Fermi surface of AV$_3$Sb$_5$, suggesting that the CDW is triggered by the Peierls mechanism [9, 10]. On the other hand, the CDW of CsV$_3$Sb$_5$ has been shown to break the time reversal symmetry [19], unveiling a superconductivity enhanced charge order chirality [17]. Nevertheless, the chiral CDW has raised many questions and their origin is still under debate. Here, we combine thermal diffuse scattering and inelastic x-ray scattering to identify a 3-dimensional (3D) precursor of the charge order with propagation vector 0.5 0 0.5, which condenses into a 3D-CDW through a first order phase transition. The quasi-elastic critical scattering indicates that the dominant contribution to the diffuse precursor is the elastic central peak without phonon softening. However, the inelastic spectra show a small broadening of the Einstein-type phonon mode on approaching the phase transition. Our results point to the situation where the Fermi surface instability at the L point is of order-disorder type with a critical growth of quasi-static domains. The results go beyond the classical weak-coupling Peierls transition dynamics and are discussed within models including strong-electron phonon coupling and non-adiabaticity.

Central to the interplay between CDW and $Z_2$ band topology is the nature of the CDW phase. From the thermodynamic point of view, specific heat measurements [7, 8] show a small release of entropy through the CDW transition, suggesting a first order phase transition, and tight binding models describe the CDW as electronically mediated [23, 24]. Despite DFT predictions of soft modes at M and L points of the Brillouin zone [14], inelastic x-ray scattering (IXS) does not observe any anomaly of the low energy acoustic branches [25]. However, ARPES data have reported deviations from the linear electronic
were performed with a Dectris PILATUS3 1M X area detector at the ID28 beamline at the European Synchrotron Research Facility (ESRF) with an incoming energy $E_i=17.8$ keV. Orientation matrix refinement and reciprocal space reconstructions were done using the CrysAlis software package. Final TDS images were processed with an in-house code. Low energy phonons were measured by inelastic x-ray scattering (IXS) at the ID28 IXS station at ESRF ($E_i=17.8$ keV, $\Delta E=3$ meV) and at the HERIX beamline at the Argonne Photon Source (APS) ($E_i=23.72$ keV, $\Delta E=1.5$ meV). The components $(h k l)$ of the scattering vector are expressed in reciprocal lattice units (r.l.u.), $(h k l)=h \mathbf{a}^* + k \mathbf{b}^* + l \mathbf{c}^*$, where $\mathbf{a}^*$, $\mathbf{b}^*$, and $\mathbf{c}^*$ are the reciprocal lattice vectors.

Figure 1 (a) and (b) sketches the structure of CsV$_3$Sb$_5$ ($P6/mmm$ space group). The unit cell is described as individual sublattices of a kagome network of V coordinated by Sb1 atoms and a hexagonal net of Sb2 above and below each kagome layer [30]. The lattice parameters derived from the TDS at room temperature ($a=7.59\,\text{Å}, c=9.34\,\text{Å}$) are consistent with the reports in the literature. The temperature dependence of the CDW instability at the $L$ point is displayed in the figure 1 (d), showing a sharp onset at $\sim 95$ K, indicative of a first order-like phase transition. Bragg reflections with the same temperature dependence (not shown) were also measured with propagation vectors $(0.5\,0\,0)$ and $(0.5\,0\,0.25)$ r.l.u. [25].

Besides the sharp and intense CDW diffraction spots in x-ray diffraction, diffuse intensity is present at $T>T_{CDW}$ due to lattice vibrations and correlated disorder. Figure 2 summarizes the processed TDS maps for the CsV$_3$Sb$_5$ sample at 90 and 100 K for the $h k 0$ and $h 0 l$ planes. The image reconstruction of the $h k 0.5$ plane at 100 K (figure 2 (a) bottom) reveals the presence of DS corresponding to freezing of a transverse component of phonons (streaks highlighted in the yellow square), with most pronounced intensity around the $3\,0\,0.5$ reciprocal lattice vector. Other satellites are equivalent according to the symmetry operations of the $P6/mmm$ space group. The DS is visible below 150 K and condense into a 3-fold CDW below $T_{CDW}$ (figure 2 (a) top). The spatial correlation of the charge precursor ($\xi^{-1}$) along the short axis of the yellow ellipse drawn in figure 3 (b) remains nearly constant as we cool down to $T_{CDW}$ and extends to 4-5 unit cells, while along the large axis reduces to less than one unit cell. The fluctuating precursor observed in figure 2 (a) along the $<1\,0\,1>$ direction is in agreement with the phonon calculations that predict a lattice instability at the $L$ point. Remarkably, DS is absent for $T>T_{CDW}$ in the $h k 0$ plane (figure 2 (b) bottom), which probes the acoustic modes, and $h 0 l$ plane (figure 2 (c) bottom) that maps the non-transverse in-plane component of the $0.5\,0\,0.25$ and $0.5\,0\,0.5$ CDWs [17, 20, 25]. However, CDW reflections are again present at $T<T_{CDW}$, matching the hard x-ray diffraction data of Fig. 1 (d). The presence of diffuse intensity only as satellites of the $0.5\,0\,0.5$ reciprocal lattice vector indicates that the Fermi

dispersion [26], providing that the electron-phonon coupling gives sufficient strength for the CDW formation. Furthermore, optical spectroscopy ascribed the opening of the CDW gap to the suppression of density of states at the $M$ point [27] with strong phonon renormalizations [28] without electronic anomalies at $L$. Finally, in addition to the enigmatic driving force of the CDW, the multiple $q_{CDW}$ and the different energy scales [29] hinder a detailed study of the ground state of AV$_3$Sb$_5$, and the origin and stabilization of the CDW phase transition remains unclear.

Here, we use a combination of thermal diffuse scattering (TDS) and inelastic x-ray scattering (IXS) to identify frozen phonon fluctuations of the $2 \times 2 \times 2$ ($L$ point) 3D-CDW in AV$_3$Sb$_5$ at $T>T_{CDW}$ that condense into a 3-fold CDW through a first order phase transition. The precursor of the charge order is absent for the $2 \times 2$ ($M$ point) and $2 \times 2 \times 2$ superstructures, indicating that the $2 \times 2 \times 2$ charge modulation is the leading Fermi surface instability. Furthermore, the intensity of the quasi-elastic central peak (CP) of IXS follows the DS intensity, indicating that the CDW is of an order-disorder transformation type. This is corroborated by the absence of a clear softening, but small broadening of the optical branch at $L$. Our results solve the controversy about the primary order parameter and highlight the critical role of the order-disorder phase transition to understand the controversial experimental reports.

Single crystals of AV$_3$Sb$_5$ (A=Cs, Rb) with $T_{CDW}=95$ and 104 K, were synthesized by the self flux growth method [30]. Thermal diffuse scattering measurements

Figure 1. (a) Unit cell of AV$_3$Sb$_5$. Orange, blue and red balls stand for A (Cs, Rb), V and Sb atoms. (b) Top view of the kagome structure, showing the V-kagome net interlaced with the Sb atoms. (c) Sketch of the Brillouin zone along the main symmetry directions. (d) Temperature dependence of the CDW reflection 2.5 0.5 0.5 r.l.u. in CsV$_3$Sb$_5$, showing a sharp drop of intensity at the $T_{CDW}=95$ K. (Inset, rocking scans of the 2.5 0.5 0.5 peak.)
surface instability at the $L$ position is the leading order parameter and drives the CDW formation in the kagome metal $AV_3Sb_5$.

The reconstruction of the $(h k 0.5)$ plane for $CsV_3Sb_5$ at different temperatures around $3 0 0.5$ cut is shown in figure 3 (a-c). Figure 3 (d) shows the profile of the diffuse precursor along the large axis of the yellow ellipse of the figure 3 (b). The critical scattering was modelled assuming a sum of pseudoVoigt and squared Lorentzian function, where the pseudoVoigt term arises from the instrumental resolution and the Lorentzian-squared from the diffuse scattering induced by random fields [31–33]. Although the data of figure 3 (d) could be well modelled, it does not prove the uniqueness of the pseudoVoigt and Lorentzian-square profile, but it demonstrates the presence of site-random potentials and disorder. The temperature dependence of the DS intensity is extracted by integrating the diffuse precursor and plotted in figure 3 (f). It starts at temperatures below 150 K and increases down to 100 K, below which it sharply diverges at the critical temperature, following the temperature dependence of the energy integrated experiment of figure 1 (d).

To disentangle static and dynamic contributions and to get further insights into the origin of the observed diffuse features, we have carried out IXS scanning the 2.5 0.5 0.5 satellite position. As shown in figure 3 (e), the IXS spectra consists on a (quasi)-elastic central peak (zero energy loss) evolving in intensity below 120 K and a low energy non dispersive phonon at 6 meV. The intensity of the CP sharply drops upon warming from 95 K and follows the intensity of the pretranslational fluctuations detected by DS. The CP of $AV_3Sb_5$ shows parallels to the critical scattering observed in SrTiO$_3$ [34, 35] and transition metal dichalcogenides [36] exhibiting a critical divergence on approaching $T_{CDW}$. On the other hand, directly visualized in the raw data of figure 3 (e), the intensity of the low energy mode increases smoothly upon cooling and drops below 100 K due to the modification of the dynamic structure factor on approaching the CDW phase, making the quasi-elastic contribution more important than the phonon intensity to the DS. Following the applicability of the Ornstein-Zernike correlation function [37], the generalized susceptibility, $\chi(q)$, proportional to the $q$ Fourier component of the displacement-displacement correlation function $\langle |u_q|^2 \rangle = k_B T \chi(q)$, displays a linear Curie-Weiss behavior for the quasi-elastic component of the CP (inset of figure 3(f)) and vanishes at the critical temperature, indicating a temperature dependence of the density of states. Nevertheless, $\chi(q)$ of the DS changes slope at $T^*$, coinciding with the drop of intensity of the low energy phonon at 100 K.

Focusing on the inelastic part of the IXS spectra, figure 4 (a) shows the momentum dependence of the phonon dispersion along the $h h 0.5$ direction ($\Delta E=3$ meV). The spectra consist on 3 weakly dispersive branches with 2.5, 6 and 11 meV at $h=0$. The lowest energy mode merges the 6 meV branch at 0.1 r.l.u. and, at the $L$ point, only two optical-like modes are observed. These two modes are better visualized in figure 4 (b) and its energy dispersion, obtained from the fitting to damped harmonic oscillators (DHO) convoluted with the experimental resolution, is displayed in the figure 4 (c). They exhibit a weak dispersion in reciprocal space and can be described as a system of independent quantum oscillators or localized atomic vibrations (Einstein-type phonon modes). To isolate the contribution of each mode, we increased the energy resolution down to 1.5 meV and work at the 3.5 2.5 0.5 reciprocal lattice vector. Figure 4 (d) shows the temperature dependence of the 3 branches. From these data, the energy of the three phonons remains constant as the temperature approaches the CDW transition. This clearly demonstrates the absence of a Kohn anomaly and discards a soft phonon nature of the CDW phase transition; not phonon driven but of order-disorder type. This does not prevent the presence of a soft mode below $T_{CDW}$, which is, in fact, a fingerprint of both order-disorder and displacive phase transitions. We need to stress here that it is not the crystal quality, but the inherent dynamical disorder that stabilizes the CDW in $AV_3Sb_5$. On the other hand, a detailed analysis of the

Figure 2. Thermal diffuse scattering at 90 K and 100 K for $CsV_3Sb_5$ for the (a) $h k 0.5$ plane, showing the CDW Bragg reflections (weak spots highlighted within the yellow circle in the zoomed in area) and the precursor of the 3D CDW, marked with arrows. (b) and (c) $h k 0$ and $h 0 l$ planes, respectively (no precursor).
Figure 3. Temperature dependence of the DS maps of CsV$_3$Sb$_5$ at (a) 90 K, (b) 100 K and (c) 150 K around the h k 0.5 cut (d) Profile of the diffuse scattering and its fitting to pseudoVoigt and squared-Lorentzian profiles (see text). (e) Temperature dependence of the IXS spectra at the 2.5 0.5 0.5 reciprocal lattice vector, highlighting the enhancement of the elastic line and drop of intensity of the low energy phonon at 96 K. (f) Temperature dependence of the intensity of the DS signal, elastic central peak and optical phonon. Inset, temperature dependence of the generalized susceptibility.

Figure 4. (a) Color map of the low energy phonons of CsV$_3$Sb$_5$ along the h h 0.5 direction at 120 K. Three modes are identified at 2.5 and 6 and 11 meV and highlighted with symbols. (b) IXS spectra and (c) phonon dispersion along the h 1-h 0.5 direction ($\Delta E = 3$ meV). (d) Temperature dependence high resolution IXS ($\Delta E = 1.5$ meV) at the 3.5 2.5 0.5 reciprocal lattice vector. Inset, fitting details of the scan at 150 K. (e) Temperature dependence of the linewidth of $\omega_1$.

Indeed, recent reflectivity measurements have reported a much larger CDW gap than the weak-coupling BCS value [27, 38]. As reported initially by Ortiz et al. [7], the A-cation disorder between kagome layers may result in local atomic displacements that contributes here to the DS. The effect of the dynamical disorder was theoretically modelled by Yu and Anderson [39] for the strong interaction of electrons and single dispersionless (Einstein-type) optical modes and further extended by Gor’kov [40] for any arbitrary ionic displacements. Within this scenario, the CDW in AV$_3$Sb$_5$ realizes in two stages. First, an ionic displacement in the kagome plane, presumably induced by the A-site disorder, introduces a strong EPI that traps the electronic cloud near an ion, changing the local electronic environment and breaking the adiabatic approximation. This gives rise to a quasi-elastic response above the Peierls transition with a Lorentzian-squared relaxation, an associated order-disorder dynamics and the absence of a Kohn anomaly. In an order-disorder type transformation, the harmonic potential well deforms into a double well and the phase transition is achieved by the critical growth and percolation of quasi-static domains on approaching the critical temperature [41]. Therefore, the electronic ordering happens due to domain interactions. In each domain, the potential within the atom moves is quasi-harmonic and phonon anomalies are expected. This naturally explains the change of slope in the diffuse susceptibility of figure 3(f) and the small broadening of the low energy mode in figure 4 (d) and (e). In
fact, the one dimensional Peierls instability of BaVS$_3$ has been observed to be of order-disorder type with a critical slowing down of cluster fluctuations [42, 43]. Strong EPI mechanisms have also been considered to explain the CDW formation in transition metal dichalcogenides [44], H-bond ferroelectrics [45] and the controversial charge order in high T$_c$ cuprates [46]. On the other hand, in the non-adiabatic dynamics, $\omega_{2k_F}$ fluctuates so rapidly that cannot couple with the electron–hole condensate during the electron–hole pair lifetime. Although the non-adiabatic dynamics has received less theoretical support, the rapidly fluctuating phonon reduces the screening between atoms and prevents the development of the Kohn anomaly as the Peierls transition is approached. Following this argument, the suppression of the Peierls transition of BaVS$_3$ under pressure was interpreted as a reduction of the $\tau_A$ [47]. This is a particularly appealing scenario since the $T_{CDW}$ of CsV$_3$Sb$_5$ is completely suppressed at low pressures and evolves towards a double superconducting dome [48–51].

We would like to finish our discussion by placing our results within the physics of the kagome metals Av$_3$Sb$_5$. An order-disorder transition is expected to be of first order type, thus the small release of entropy at $T_{CDW}$ [7, 8] fits within this scenario. Moreover, the data presented here does not conflict with the possible chiral charge density wave, but put constrains on the nematic order reported [20, 21]. Broken rotational symmetries can be affected by local dynamic disorder, fluctuating domains and defects that introduce pinning potentials. This pinning centers and local distortions introduce variations of the amplitudes of the local atomic displacements, which can be detected by pair distribution function (PDF) analysis [52].

In conclusion, we have presented thermal diffuse scattering and inelastic x-ray scattering data to demonstrate that the CDW in the kagome metal Av$_3$Sb$_5$ is of order-disorder type without phonon softening. A precursor of the CDW is observed in thermal diffuse scattering with propagation vector 0.5 0.5 0.5, indicating that the Peierls instability at the $L$ point is the leading Fermi surface instability (primary order parameter). Furthermore, we demonstrate that the diffuse signal contains intensity of the quasi-elastic central peak, likely due to the growth of fluctuating domains, thus favouring strong coupling theories to describe the CDW ground state of Av$_3$Sb$_5$. We acknowledge E. Efremov, A. Schnyder and J. Diego for fruitful discussions and critical reading of the manuscript. S.B.-C, D.S and L.S thank the MINECO of Spain through the project PGC2018-101334-A-C22. S.D.W and B.R.O gratefully acknowledge support via UC Santa Barbara NSF Quantum Foundry funded via the Q-AMASE-i program under award DMR-1906325. This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science user facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357.

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