

Pressure-induced unconventional charge-density-wave states in kagome metal AV_3Sb_5 ($\text{A} = \text{K}, \text{Rb}, \text{Cs}$)

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Since the discovery of charge density wave (CDW) and superconductivity, kagome metal AV_3Sb_5 ($\text{A} = \text{K}, \text{Rb}, \text{Cs}$) provides a new platform for exploring novel many-body quantum phenomena. In CsV_3Sb_5 , a stripelike CDW with commensurate wave vector $q = 3/8$ was observed under moderate pressures, which leads to a peculiar superconducting double-dome behavior in pressure-dependent phase diagram. Previous density functional theory (DFT) calculations indicate that the pressure-induced stripelike CDW is beyond conventional phonon softening scenario, suggesting a nontrivial role of electronic correlations. However, an in-depth understanding for the pressure-induced unconventional CDW remains elusive. Here, we performed pressure-dependent ^{51}V nuclear magnetic resonance (NMR) measurements on KV_3Sb_5 and RbV_3Sb_5 . Although the superconducting double-dome behavior is absent in pressurized KV_3Sb_5 and RbV_3Sb_5 , a pressure-induced CDW phase, ascribed to a possible incommensurate triple- Q CDW, is identified by NMR spectra in both materials, indicating that the pressure-induced unconventional CDW beyond DFT calculations is a common feature for kagome metal AV_3Sb_5 . In contrast to the stripelike CDW, the pressure-induced incommensurate triple- Q CDW does not strongly suppress the superconducting temperature (T_c) but coincides with an almost plateau behavior at intermediate pressure regime in the pressure-dependent superconducting phase diagram. Furthermore, by systematically analyzing the Korringa relation between Knight shift and nuclear spin-lattice relaxation rate in AV_3Sb_5 , Van Hove singularity-driven electronic fluctuations are revealed as an effective knob for the pressure-induced unconventional CDW. Finally, our present findings underscore the pressure-induced unconventional CDW as a novel correlated quantum state in kagome metal AV_3Sb_5 .

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I. INTRODUCTION

Vanadium-based kagome metals AV_3Sb_5 ($\text{A} = \text{K}, \text{Rb}, \text{Cs}$) have recently emerged as a versatile platform for exploring novel many-body quantum phenomena within geometrically frustrated lattices [1–4]. In this family, a triple- Q charge-density-wave (CDW) order emerges below $T_{\text{CDW}} \approx 78\text{--}102\text{ K}$ [2–4]. Crucially, this CDW state is unconventional, exhibiting intrinsic symmetry-breaking characteristics such as time-reversal symmetry breaking [5–15] and rotational symmetry breaking [11,16–20]. Upon further cooling, the electronic system evolves into a superconducting ground state [2–4], which is strongly affected by the preexisting CDW

[8,9,12,14,18,21–33]. Despite extensive research, the microscopic origin of this complex CDW state and its interplay with the superconducting condensate remain subjects of intense debate [5,14,18,26,34–67].

The complexity of the electronic ground state is largely attributed to the unique band structure of the kagome lattice. Recent experiments have identified multiple saddle points near the Fermi level (E_F) in these kagome metals [36,37,45–47,68]. The inherent nature of these Van Hove singularities (VHS) induces hopping interference effects, which significantly enhance electronic correlations and drive the formation of diverse electronic orders [34,69]. On one hand, theoretical studies based on correlated electronic models suggest that Fermi surface nesting between VHS drives the CDW transition [36–40,46,49,58,70,71]. This nesting enhances nonlocal electronic correlations [72–74], which may subsequently lead to unconventional superconducting pairing [27,33,49,72,75]. On the other hand, density functional theory (DFT) calculations highlight the critical role of electron-phonon coupling (EPC), evidenced by imaginary phonon frequencies at the M and L points [38,48,52,71,76–78]. Indeed, a growing body of experimental evidence from

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spectroscopic studies [48,51,52,55,57,61,79,80] and first-principles calculations [55,62,65,81] has underscored the essential contributions of strong EPC and lattice anharmonicity to CDW formation [55,61,62,65,66,79–83]. Notably, x-ray scattering experiments have unambiguously revealed acoustic phonon softening in KV_3Sb_5 [83] and, remarkably, even in CsV_3Sb_5 , where such behavior had long remained elusive [66].

Regarding superconductivity (SC), while the precise pairing mechanism remains controversial [18,27,33,55,56,63,64,84–87], recent studies have increasingly highlighted the significant role of EPC [48,56,62–65,81]. Consequently, a central challenge in kagome superconductors is to elucidate the cooperative effects of both electronic correlations and EPC in shaping the CDW and its interplay with superconductivity. Because these quantum phases are highly sensitive to the specific alkali metal ion [2–4], the AV_3Sb_5 family provides an ideal platform for systematic comparative studies. Theoretical calculations indicate that the VHS filling level in CsV_3Sb_5 is closest to E_F [82,88,89], suggesting that it should host the most prominent electronic correlations [88]. However, reported correlation strengths across the series vary significantly [82,90], likely stemming from sample-dependent variations in E_F . This intrinsic diversity underscores the necessity of systematic investigations to unravel the mechanisms governing the electronic states within the AV_3Sb_5 family.

Hydrostatic pressure serves as a powerful tuning parameter for investigating the interplay between SC and the CDW. Although pressure uniformly suppresses the primary CDW order across the AV_3Sb_5 family [8,21,22], the superconducting behaviors diverge significantly: CsV_3Sb_5 exhibits a double-dome superconducting phase diagram [8], whereas KV_3Sb_5 and RbV_3Sb_5 show a monotonic enhancement of the critical temperature (T_c) [21,22]. Recent studies have attributed this anomalous SC evolution in CsV_3Sb_5 to its competition with a pressure-induced CDW phase, identified as a stripelike CDW by nuclear magnetic resonance (NMR) [23] or a long-range $3/8$ charge-ordered phase by x-ray diffraction (XRD) [29]. Despite these advances, the pressure-dependent CDW evolution in KV_3Sb_5 and RbV_3Sb_5 remains largely unexplored. Previous transport measurements indicate that the maximum T_c is achieved at approximately 0.5 GPa in KV_3Sb_5 and 1.5 GPa in RbV_3Sb_5 [21,22]. Notably, while the CDW transition vanishes above 0.5 GPa in KV_3Sb_5 [21], it persists up to 2.4 GPa in RbV_3Sb_5 [22]. This discrepancy suggests a complex interplay between the CDW and SC in RbV_3Sb_5 , raising the possibility that a hidden CDW phase may emerge in the pressure regime between 1.5 and 2.4 GPa.

As a powerful local probe, NMR provides crucial microscopic insights into charge modulations. In this work, we systematically investigated the pressure-induced evolution of the CDW state in KV_3Sb_5 and RbV_3Sb_5 using ^{51}V NMR. Through an analysis of the spectral line shape evolution, we uncovered a pressure-induced incommensurate CDW phase in both compounds. Furthermore, once the incommensurate CDW phase is fully suppressed, the distinct deviations in the Knight shift and spin-lattice relaxation rate ($1/T_1T$) among AV_3Sb_5 highlight the pivotal role of VHS in shaping the

pressure-induced unconventional CDW phase and its interplay with superconductivity.

II. RESULTS

A. Pressure-dependent superconductivity and CDW state

At ambient pressure, the ^{51}V NMR spectrum of AV_3Sb_5 exhibits seven transition lines with the magnetic field applied along the c axis above T_{CDW} . Upon entering the CDW phase, the kagome lattice reconstructs into a staggered trihexagonal superlattice [Figs. 1(a) and 1(b)], generating two inequivalent vanadium sites within the hexagon cluster [V(I)] and the triangle cluster [V(II)]. Correspondingly, the ^{51}V NMR transition lines split into two sets of peaks with equal intensity [Fig. 1(c)] (see Fig. 8 for full spectra). As elucidated by our previous NMR experiments [91], the contribution of the second-order quadrupole effect to the resonance frequency of the central transition lines is negligible. Therefore, this equal splitting is predominantly driven by the magnetic Knight shift ($K = (f - \gamma H)/\gamma H$). In addition to the major splitting, both V(I) and V(II) sites develop a minor splitting with an intensity ratio of 2:1 [Fig. 1(c)], which is attributed to a bulk C_2 distortion arising from interlayer stacking [Fig. 1(b)]. For simplicity, we focus primarily on the central transition line to investigate the pressure-dependent spectral evolution.

As illustrated in Figs. 1(d)–1(f), T_c is characterized by the temperature-dependent relative change in the resonant frequency ($-\Delta f/f$) of the NMR tank circuit at zero magnetic field (see Fig. 9 for details). With increasing pressure, T_c of both compounds is gradually enhanced, reaching a maximum of 3.15 K at 0.52 GPa for KV_3Sb_5 and 4.38 K at 1.49 GPa for RbV_3Sb_5 . Notably, above these critical pressures (P_{c1}), T_c exhibits only a slight decrease in both compounds. This behavior stands in stark contrast to the rapid suppression of T_c observed in CsV_3Sb_5 [Fig. 1(f)] [23]. These results are consistent with previous reports [21,22], confirming a single-dome evolution of pressure-dependent superconductivity in KV_3Sb_5 and RbV_3Sb_5 . In CsV_3Sb_5 , the emergence of a stripelike CDW shapes a double-dome superconducting phase diagram. Therefore, elucidating the microscopic nature of the CDW and its evolution under pressure is essential for understanding the superconducting landscape in KV_3Sb_5 and RbV_3Sb_5 .

Previous transport measurements indicate that while the CDW transition persists up to 2.4 GPa in RbV_3Sb_5 , it vanishes above 0.5 GPa in KV_3Sb_5 [21,22]. As shown in Figs. 2(a) and 2(b), the characteristic two-peak structure of the ^{51}V NMR spectra is preserved in both KV_3Sb_5 and RbV_3Sb_5 below P_{c1} . However, distinct anomalies emerge at P_{c1} : In KV_3Sb_5 , both peaks broaden toward the central frequency, whereas in RbV_3Sb_5 , a “third peak” abruptly appears between the original two peaks. Above P_{c1} , the spectra in both compounds evolve continuously, forming broad “bell-like” line shapes. Eventually, the spectra restore the single-peak structure characteristic of a perfect kagome lattice above $P_{c2} = 1.28$ GPa for KV_3Sb_5 and $P_{c2} = 2.86$ GPa for RbV_3Sb_5 , signaling the complete suppression of the CDW order. These phenomena bear a strong resemblance to our previous work on pressurized CsV_3Sb_5 [Fig. 2(c)] [23], where a new spectral line

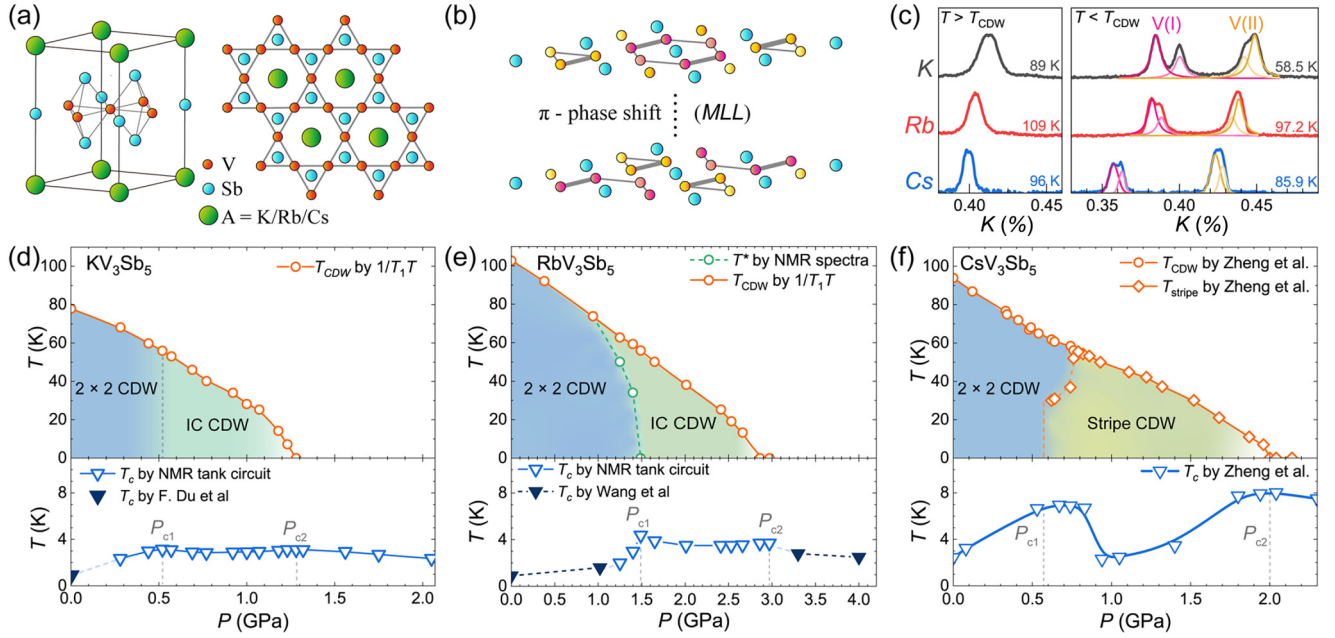


FIG. 1. Crystal structures and pressure-temperature (P - T) phase diagrams of AV_3Sb_5 ($A = K, Rb, \text{ and } Cs$). (a) The kagome lattice structure of the AV_3Sb_5 family. (b) Charge ordering pattern of the vanadium layers in the staggered trihexagonal structure. (c) ^{51}V NMR central transition lines of AV_3Sb_5 ($A = K, Rb, \text{ and } Cs$) measured above and below T_{CDW} . The pink and orange curves correspond to the vanadium sites in the hexagon cluster [V(I)] and the triangle cluster [V(II)], as depicted in panel (b). Each spectrum is fitted with four components, where the intensity ratio of the major splitting follows $V(I):V(II) = 1:1$, and the minor splitting on both V(I) and V(II) follows a 1:2 ratio. P - T phase diagrams for (d) KV_3Sb_5 , (e) RbV_3Sb_5 , and (f) CsV_3Sb_5 , summarizing the evolution of CDW states and superconductivity discussed in the text. The characteristic temperatures are defined in the preceding figures. Data in panel (f) are adopted from our previous work on CsV_3Sb_5 [23].

shape replaced the two-peak structure above P_{c1} , signaling the emergence of a new CDW phase (identified as a stripelike order with unidirectional $8/3a_0$ modulation) [Fig. 1(f)]. Furthermore, these spectral anomalies align with recent studies suggesting the emergence of distinct pressure-induced phases

in KV_3Sb_5 and RbV_3Sb_5 [92,93]. Consequently, the remarkable changes in spectral line shape observed near P_{c1} imply that the maximum T_c coincides with the boundary between two CDW phases, suggesting a complex interplay between the emergent CDW and superconductivity.

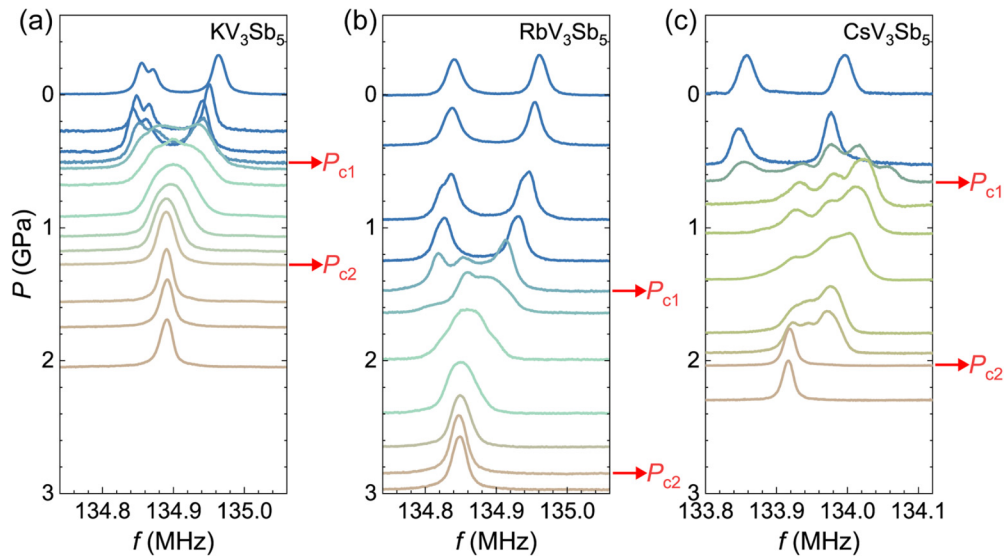


FIG. 2. Pressure-dependent evolution of the CDW states in AV_3Sb_5 ($A = K, Rb, \text{ and } Cs$). (a) KV_3Sb_5 , (b) RbV_3Sb_5 , and (c) CsV_3Sb_5 . All ^{51}V central transition lines are measured at 2 K with a magnetic field of 12 T applied along the crystallographic c axis. The red arrows indicate the critical pressures for the CDW transitions and suppression. Data in panel (c) are adopted from our previous work on CsV_3Sb_5 [23].

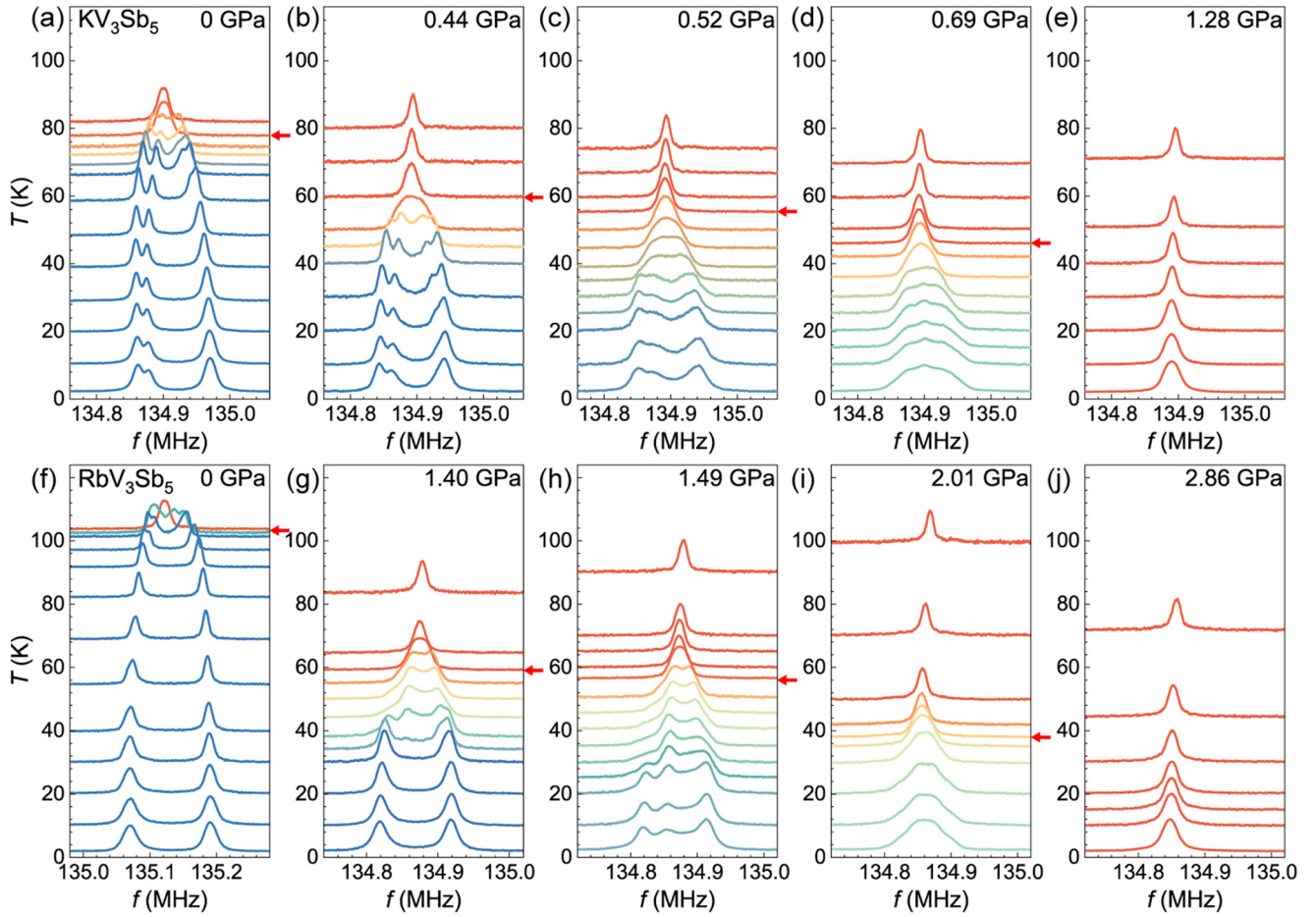


FIG. 3. Pressure-dependent multiple CDW transitions revealed by ^{51}V NMR spectra. (a)–(e) Temperature-dependent ^{51}V NMR spectra of KV_3Sb_5 measured under pressures of 0, 0.44, 0.52, 0.69, and 1.28 GPa. (f)–(j) Temperature-dependent ^{51}V NMR spectra of RbV_3Sb_5 measured under pressures of 0, 1.4, 1.49, 2.01, and 2.86 GPa. The red, blue, and green lines correspond to the kagome phase, the triple- Q CDW phase, and the incommensurate CDW phases, respectively. The red arrows indicate the CDW transition temperatures, as determined by $1/T_1T$ (see details in Fig. 4 and Sec. III of the Supplemental Material [94]).

B. Temperature-dependent evolution of CDW state under different pressures

Figure 3 presents the temperature-dependent NMR spectra for KV_3Sb_5 and RbV_3Sb_5 under various pressures. Below P_{c1} , the spectra consistently split into two peaks at low temperatures [Figs. 3(b) and 3(g)], corresponding to the preservation of the triple- Q CDW state. Notably, a two-step CDW transition is observed near P_{c1} in RbV_3Sb_5 . In Fig. 3(g), the spectra at 1.4 GPa first broaden below ~ 60 K, forming a distinct line shape. Upon further cooling, the two peaks characteristic of the triple- Q CDW emerge below 40 K, eventually restoring the standard two-peak structure at 30 K. At P_{c1} , while the splitting persists in KV_3Sb_5 , both peaks broaden asymmetrically toward the central frequency [Fig. 3(c)]. In RbV_3Sb_5 , the new spectral profile coexists with the characteristic two-peak structure down to 2 K [Fig. 3(h)], indicating the coexistence of the triple- Q CDW and the new CDW phase.

As pressure increases further above P_{c1} , the characteristic two-peak structure eventually disappears. The typical spectral line shapes for the second CDW phase in KV_3Sb_5 and RbV_3Sb_5 are shown in Figs. 3(d) and 3(i). Here, the single peak gradually broadens upon cooling, establishing

a “bell-like” line shape at 2 K, which confirms the establishment of a new emergent CDW phase. At higher pressures, the CDW transition becomes difficult to discern due to the diminished spectral linewidth at low temperatures (see Sec. II of the Supplemental Material [94]). To further probe the electronic state, we measured the temperature-dependent nuclear spin-lattice relaxation rate ($1/T_1$) of the ^{51}V nuclei (Fig. 4). Generally, the relaxation rate is described by $1/T_1T = (1/T_1T)^{\text{QP}} + (1/T_1T)^{\text{SF}}$, where the first term arises from quasiparticle contributions and the second from spin fluctuations. As shown in Figs. 4(a) and 4(f), at ambient pressure, $1/T_1T$ decreases rapidly below the CDW transition temperature due to the opening of a momentum-dependent CDW gap, as observed in previous angle-resolved photoemission spectroscopy (ARPES) studies [19,30,36]. Consistently, we define the onset temperature of this drop in $1/T_1T$ as T_{CDW} (Fig. 4), which gradually decreases with increasing pressure in both compounds (see Sec. III of the Supplemental Material for full characterization [94]). As shown in Figs. 4(e) and 4(j), the gap behavior vanishes completely at $P_{c2} = 1.28$ GPa for KV_3Sb_5 and $P_{c2} = 2.86$ GPa for RbV_3Sb_5 , confirming the full suppression of the CDW order.

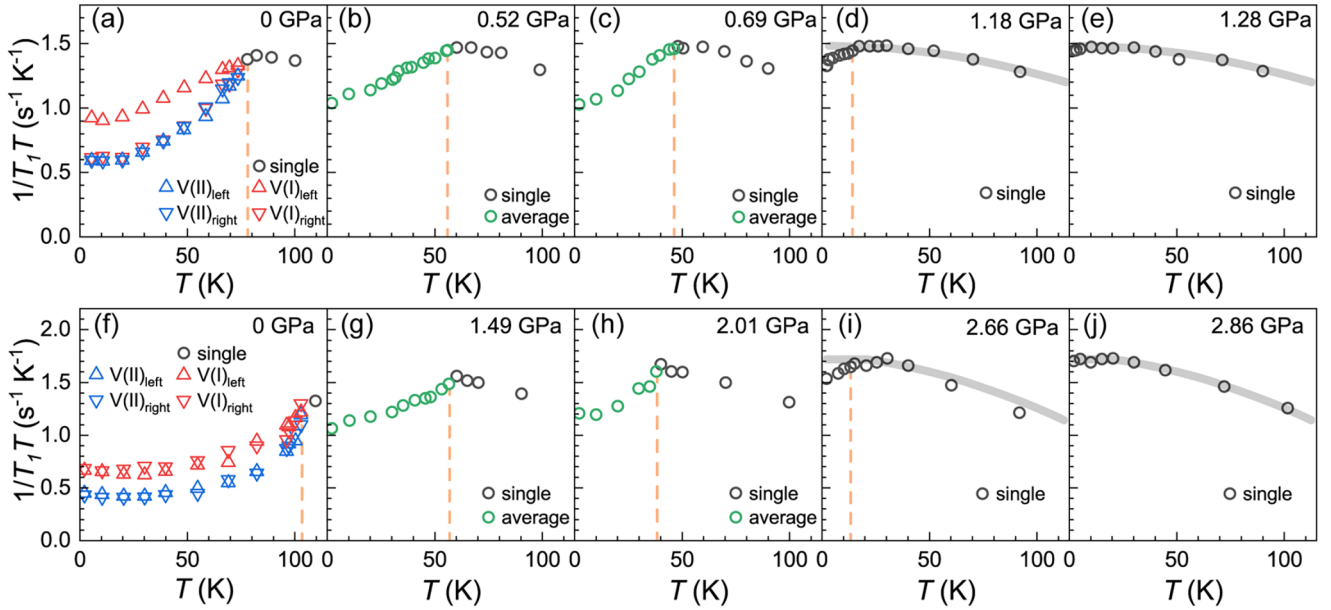


FIG. 4. Evidence for multiple CDW transitions from nuclear spin-lattice relaxation measurements. (a)–(e) Temperature-dependent $1/T_1T$ behaviors of ^{51}V NMR in KV_3Sb_5 under various pressures. (f)–(j) Temperature-dependent $1/T_1T$ behaviors of ^{51}V NMR in RbV_3Sb_5 under various pressures. The orange dashed lines delineate T_{CDW} , defined by the onset of the drop in $1/T_1T$. Near P_{c2} , where the drop in $1/T_1T$ becomes obscure, T_{CDW} is determined by the deviation from the high-temperature behavior, as indicated by the gray dashed lines in panels (d), (e), (i), and (j).

C. Identification of pressure-induced incommensurate CDW states

Characterizing the second CDW phases emerging between P_{c1} and P_{c2} remains a significant challenge. In contrast to the five discrete peaks observed in the striplike CDW state of CsV_3Sb_5 [23], the spectral profiles for KV_3Sb_5 and RbV_3Sb_5 in this regime manifest as broad, featureless “bell-like” shapes. Generally, in a commensurate CDW state, each characteristic NMR peak corresponds to a specific inequivalent nuclear site; consequently, the number of peaks and their intensity ratios provide a reliable basis for identifying discrete CDW models via spectral fitting. However, in an incommensurate CDW state, the loss of translational periodicity generates a continuum of inequivalent nuclear sites [95]. This absence of discrete sites likely accounts for the lack of distinct peaks and the pronounced spectral broadening observed in Figs. 2(a) and 2(b) between P_{c1} and P_{c2} .

Nevertheless, despite the absence of sharp discrete peaks, a closer inspection reveals subtle yet highly reproducible features within these broad profiles below T_{CDW} . For KV_3Sb_5 , the spectra at 0.69 GPa [Fig. 3(d)] and 0.77 GPa [Fig. S2(g) of the Supplemental Material [94]] consistently exhibit a small central protrusion. For RbV_3Sb_5 , the spectra below T_{CDW} at 2.01 GPa [Fig. 3(i)] display a central plateau flanked by two distinct inflection points. Remarkably, these line-shape characteristics bear a striking resemblance to the NMR profiles theoretically predicted for incommensurate systems governed by triple- Q modulation [95], as illustrated in Figs. 5(a) and 5(d). This close correspondence strongly suggests an incommensurate triple- Q CDW origin and motivates a systematic numerical simulation using various multiple- Q models to assess the consistency of the different models with the experimental data.

To investigate this further, we applied several incommensurate CDW models to simulate the spectral line shapes of KV_3Sb_5 and RbV_3Sb_5 (see Appendix for details). The single- Q and double- Q models considered here do not reproduce the key spectral features, yielding either a deep central gap or a disproportionate central weight (see Fig. 11 for details). Instead, triple- Q incommensurate CDW modulations incorporating distinct in-plane anisotropies provide the most consistent interpretation of the experimental data. As shown in Fig. 5(c), the simulated profile excellently reproduces the experimental spectrum of KV_3Sb_5 at 0.69 GPa. The optimal fit was achieved with a CDW amplitude ratio of 1:1:2, suggesting that while the macroscopic triple- Q symmetry is preserved, the local charge distribution exhibits pronounced anisotropy. Similarly, for RbV_3Sb_5 at 2.01 GPa, the experimental spectrum aligns with a simulated profile defined by an amplitude ratio of 2:1:1 [Fig. 5(f)].

Consequently, the bell-like line shape is consistent with, and supports, the proposed interpretation of an anisotropic triple- Q incommensurate CDW. In summary, our comprehensive analysis of the ^{51}V NMR spectra identifies this anisotropic incommensurate triple- Q CDW as the most plausible candidate for the second CDW phase in both KV_3Sb_5 and RbV_3Sb_5 . While NMR serves as a sensitive local probe for these charge modulations, determining the precise modulation period and detailed long-range crystallographic structure of these incommensurate phases will require future scattering measurements, such as high-pressure x-ray diffraction (XRD).

D. Pressure-dependent phase diagram of the AV_3Sb_5 family

Figures 1(a)–1(c) summarize the pressure-dependent (P) phase diagrams of the AV_3Sb_5 family. On one hand, clear

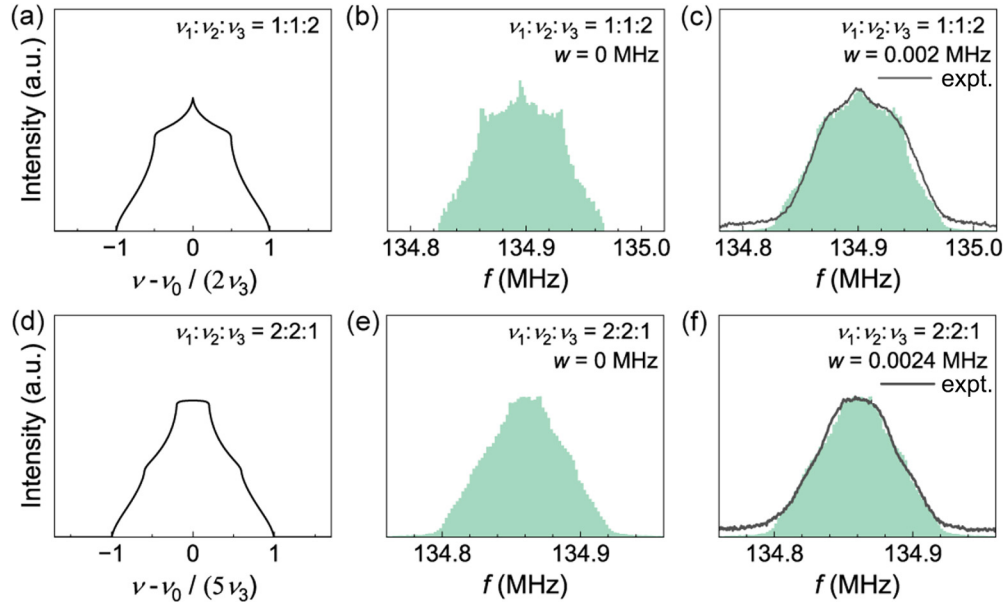


FIG. 5. Simulation of the pressure-induced incommensurate CDW states. (a) Theoretical NMR line shape for a triple- Q modulation with relative amplitudes $2\nu_1 = 2\nu_2 = \nu_3$. (b) The standard simulated NMR line shape as the same modulation in panel (a) without broadening. (c) Comparison between the simulated NMR line shape for triple- Q plane wave modulations (with parameters $\nu_0 = 134.901$ MHz and $2\nu_1 = 2\nu_2 = \nu_3 = 0.036$ MHz) and the experimental spectra of KV_3Sb_5 at 2 K and 0.69 GPa. (d) Theoretical NMR line shape for a triple- Q modulation with relative amplitudes $\nu_1 = \nu_2 = 2\nu_3$. (e) The standard simulated NMR line shape as the same modulation in panel (d) without broadening. (f) Comparison between the simulated NMR line shape for triple- Q plane wave modulations (with parameters $\nu_0 = 134.86$ MHz and $\nu_1 = \nu_2 = 2\nu_3 = 0.024$ MHz) and the experimental spectra of RbV_3Sb_5 at 2 K and 2.01 GPa. The theoretical NMR line shapes in panels (a) and (d) are cited from Ref. [95].

commonalities exist across all three compounds, most notably the direct competition between SC and the triple- Q CDW below P_{c1} , followed by the emergence of a second CDW phase between P_{c1} and P_{c2} . On the other hand, striking divergences emerge in the characterization of these second CDW phases and their respective interplay with superconductivity. This sharp contrast in phase evolution raises intriguing questions regarding the underlying physics governing these pressure-induced CDW states.

Previous theoretical analyses suggest that unstable phonon modes at the M and L points drive the triple- Q CDW order at ambient pressure [38,48,52,71,76–78]. Within this framework, a commensurate-to-incommensurate transition is expected if the minimum of the imaginary phonon frequency shifts away from these high-symmetry points. To test this, we performed first-principles calculations of the pressure-dependent phonon spectra. As shown in Fig. 12, the critical pressure at which the imaginary frequency disappears aligns closely with the suppression of the CDW in $(\text{K}, \text{Rb})\text{V}_3\text{Sb}_5$. However, the commensurate-to-incommensurate transition is notably absent in our calculations. Specifically, the minimum of the imaginary phonon frequency remains fixed at the M and L points until the instability vanishes, failing to reproduce the experimental observations. Furthermore, detailed characteristics of the incommensurate phase, such as the varying relative amplitudes of CDW modulation across different wave vectors, deviate significantly from standard weak-coupling scenarios. These discrepancies underscore that a pure EPC-driven mechanism is insufficient to explain the pressure-induced incommensurate CDW, identifying it instead as an

unconventional correlated state beyond the scope of standard DFT descriptions.

The limitations of the purely phonon-driven scenario discussed above lead us to consider the critical role of electronic correlations. Interestingly, the nearly plateaulike behavior of T_c observed within the incommensurate CDW phases of KV_3Sb_5 and RbV_3Sb_5 parallels observations in transition metal dichalcogenides, where a superconducting dome coincides with a commensurate-to-incommensurate transition [96,97]. Recent theoretical work on monolayer 1T-TiSe_2 has demonstrated that the suppression and incommensuration of the CDW cannot be accurately captured by phonon-driven scenarios alone but crucially require the inclusion of on-site electronic correlations [98]. Specifically, these correlations dominate the phonon-driven instability by renormalizing the momentum-dependent electron-phonon coupling, allowing for an accurate theoretical reproduction of the experimental phase diagram, including both commensurate and incommensurate regions. This suggests that the incommensurate CDW is an electronic state inherently governed by correlation effects, which may play a fundamental role in supporting the superconducting phase.

Therefore, we propose that in $(\text{K}, \text{Rb})\text{V}_3\text{Sb}_5$, nonlocal electronic correlations promoted by VHS cooperate with the EPC to drive CDW evolution under pressure. Additionally, in CsV_3Sb_5 , the triple- Q CDW evolves into a unidirectional stripelike modulation; this transformation exceeds simple changes in band structure or phonon dispersion and likely reflects a regime dominated by more prominent electronic correlation effects [23]. Crucially, the interplay with

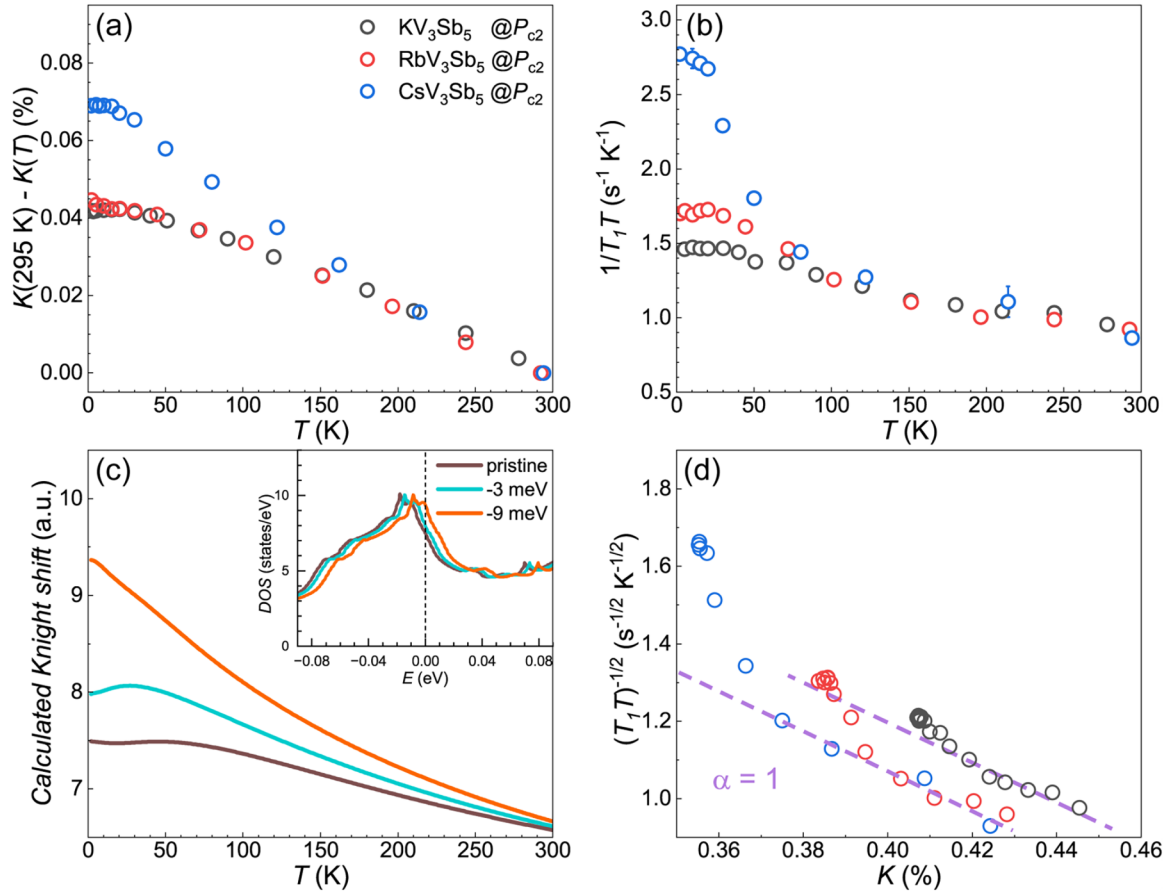


FIG. 6. Evidence for the role of VHS across the AV_3Sb_5 family, derived from Knight shift and $1/T_1T$ measurements at P_{c2} . (a) Temperature-dependent ^{51}V NMR Knight shifts in AV_3Sb_5 ($A = K, Rb, \text{ and } Cs$) at P_{c2} . The shifts for different compounds are normalized by subtracting the value at approximately 295 K. (b) Temperature-dependent $1/T_1T$ of ^{51}V NMR in AV_3Sb_5 ($A = K, Rb, \text{ and } Cs$) at P_{c2} . (c) Calculated temperature-dependent Knight shift, proportional to $\frac{1}{k_B T} \int (1-f(E))f(E)D(E)dE$, for different alignments of the VHS relative to E_F . Here, k_B is the Boltzmann constant, $f(E)$ is the Fermi-Dirac distribution function, and $D(E)$ is the density of states. (d) Korrington relation between $1/T_1T$ and the square of the Knight shift (K_{spin}^2) in AV_3Sb_5 ($A = K, Rb, \text{ and } Cs$) at P_{c2} . The relation is generally expressed as $T_1TK_{spin}^2 = (\hbar\gamma_e^2)/(4\pi k_B\gamma_n^2\alpha)$, where γ_e and γ_n are the electron and nuclear gyromagnetic ratios, respectively, and α is the Korrington enhancement factor. The violet dashed line indicates the standard Korrington relation with $\alpha = 1$, corresponding to the weak coupling limit.

superconductivity also shifts from the coexistence observed in KV_3Sb_5 and RbV_3Sb_5 to a regime of strong competition in CsV_3Sb_5 . Considering that the electronic structures of KV_3Sb_5 and RbV_3Sb_5 are more similar to each other than to CsV_3Sb_5 [82,89], we suggest that the divergent CDW evolution paths across the AV_3Sb_5 family are mediated by the VHS alignment, which acts as an effective knob to tune the correlation strength.

E. The role of VHS in electronic correlations

To explore the evolution of electronic correlations across the AV_3Sb_5 family, we systematically compared the temperature-dependent Knight shift [Fig. 6(a)] and $1/T_1T$ [Fig. 6(b)] behaviors at P_{c2} where the CDW phase is completely suppressed. Generally, the total Knight shift consists of a temperature-independent orbital term (K_{orb}) and a temperature-dependent spin term (K_{spin}). Since K_{orb} remains constant at a fixed pressure without structural or magnetic phase transitions, the observed temperature variation of the Knight shift is strictly governed by K_{spin} . This spin component

is proportional to the density of states (DOS) at the Fermi level, $N(E_F)$: $K_{spin} = A_{hf}\mu_B N(E_F)$ (where A_{hf} is the hyperfine form factor and μ_B is the Bohr magneton). In a weakly interacting Fermi liquid where additional spin fluctuations are negligible, $1/T_1T$ reflects $N(E_F)$ and follows the standard Korrington relation, $1/T_1T \propto K_{spin}^2 \propto N(E_F)^2$. Conversely, when quasiparticle interactions are significant, electronic instabilities can induce enhanced fluctuations that strongly renormalize $1/T_1T$.

As shown in Fig. 6(b), the temperature-dependent $1/T_1T$ behaviors in KV_3Sb_5 and RbV_3Sb_5 differ markedly from that in CsV_3Sb_5 , which displays a pronounced Curie-Weiss-like upturn upon cooling. According to our previous work, this Curie-Weiss-like behavior in CsV_3Sb_5 is pressure independent and originates from correlation-enhanced electronic fluctuations [23]. This clear discrepancy indicates that electronic correlation effects are relatively weaker in KV_3Sb_5 and RbV_3Sb_5 than in CsV_3Sb_5 .

Similarly, the Knight shift in CsV_3Sb_5 deviates remarkably from those in KV_3Sb_5 and RbV_3Sb_5 , exhibiting a strong temperature dependence that points to an intrinsically varying

$N(E_F)$ [Fig. 6(a)]. We attribute this deviation to the proximity of the VHS to the Fermi level (E_F) in CsV_3Sb_5 . By tuning the VHS alignment relative to E_F based on previous DFT calculations [23], we successfully reproduced the experimental Knight shift behaviors [Fig. 6(c)] (see Appendix for details). Prior DFT calculations suggest that while the Fermiology of the AV_3Sb_5 family shares similarities near the M point at ambient pressure, the high-order VHS—which exhibits flat dispersion and generates a power-law divergent DOS—is positioned significantly closer to E_F in CsV_3Sb_5 than in $(\text{K}, \text{Rb})\text{V}_3\text{Sb}_5$ [68,82,89]. Although recent ambient-pressure ARPES results have struggled to resolve the fine details of VHS alignment across the family, our NMR measurements provide a unique microscopic probe of the electronic structure in the high-pressure regime where the CDW phase is fully suppressed.

Crucially, our results highlight that the enhancement of electronic correlations in CsV_3Sb_5 transcends a simple DOS effect. While the proximity of the VHS typically increases $N(E_F)$ —which can theoretically enhance EPC—it further triggers a synergistic cooperation between the EPC and intrinsic electronic correlations that goes beyond a weak-coupling description. This conclusion is further corroborated by the Korringa plots in Fig. 6(d). While KV_3Sb_5 closely follows the standard Korringa behavior and RbV_3Sb_5 exhibits a moderate deviation below about 101 K, CsV_3Sb_5 shows the largest departure from linearity below about 80 K. This anomalous scaling serves as a clear experimental signature of enhanced electronic fluctuations and the breakdown of the simple quasiparticle picture. Therefore, these results reveal a progressive migration of the VHSs toward E_F as one moves from $(\text{K}, \text{Rb})\text{V}_3\text{Sb}_5$ to CsV_3Sb_5 , establishing VHS tuning as the decisive effective knob for scaling the electronic correlation strength across the AV_3Sb_5 family.

III. DISCUSSION

Our discovery of emergent high-pressure CDW phases uncovers a novel regime that defies a simple weak-coupling paradigm, emphasizing the pivotal synergistic interplay between EPC and electronic correlations. Although on-site correlations in AV_3Sb_5 systems remain relatively modest, theoretical works [34,39,69,73,74,99–101] have implicated nonlocal electronic correlations, facilitated by VHSs, as critical stabilizers of charge order. Notably, the present NMR work reveals an intermediate, anisotropic incommensurate CDW phase preceding the complete suppression of CDW order in $(\text{K}, \text{Rb})\text{V}_3\text{Sb}_5$. This phase exhibits distinct modulation amplitudes across the three wave vectors, approximating ratios of 2:1:1 in KV_3Sb_5 and 2:2:1 in RbV_3Sb_5 . These ratios signify an emergent in-plane anisotropy—dissociated from interlayer stacking-induced anisotropy observed at ambient pressure—that prefigures the unidirectional stripelike modulation inherent to the AV_3Sb_5 family. While nonlocal correlations [23,31] can potentiate this stripelike tendency, temperature-dependent Knight shift measurements indicate insufficient proximity of the VHS to the E_F in these compounds. Consequently, electronic correlations lack the requisite strength to rigidify unidirectional stripe distortions, culminating instead in an incommensurate CDW phase.

Furthermore, while standard DFT calculations accurately predict the critical pressure for imaginary phonon frequency disappearance aligning with CDW suppression in $(\text{K}, \text{Rb})\text{V}_3\text{Sb}_5$, the observed incommensurability phenomenon eludes explanation by pure EPC models. Therefore, we posit that CDW evolution in $(\text{K}, \text{Rb})\text{V}_3\text{Sb}_5$ is sculpted by cooperative EPC and finite electronic correlations, collectively driving the system away from commensurability.

In stark contrast, the CDW evolution in CsV_3Sb_5 deviates fundamentally from phonon-centric scenarios. While standard DFT calculations (Fig. 12) project a CDW instability persisting up to 4 GPa in CsV_3Sb_5 , experiments establish its full suppression near 2 GPa. This striking disparity implicates a correlation-dominated regime. Notably, CsV_3Sb_5 exhibits the greatest departure from the Korringa relation, indicating that its VHS's closer proximity to E_F triggers intrinsic correlations surpassing mere DOS enhancement. These intensified correlations effectually suppress the initial triple- Q CDW order while stabilizing a unidirectional stripelike CDW state—a correlated electronic phase emergent from synergistic EPC and VHS-tuned correlations. It should be noted that recent first-principles calculations also reveal significant lattice anharmonicity in CsV_3Sb_5 [62,65], which might also be related to the VHS and important for the pressure-induced unconventional CDW phases.

Furthermore, VHS-tuned correlations mediate contrasting CDW-superconductivity interplays across the family. In $(\text{K}, \text{Rb})\text{V}_3\text{Sb}_5$, the transition to an incommensurate phase often coincides with the emergence of a superconducting plateau. This coincidence may simply suggest that the pressure-induced CDW and superconductivity in these compounds are only weakly coupled or largely independent. Alternatively, as a possible scenario, analogous phenomena might occur here as in TiSe_2 [96,97], where electronic correlations renormalize momentum-dependent EPC, stabilizing incommensurate domain walls compatible with Cooper pairing [98]. Conversely, in CsV_3Sb_5 , stripe order emergence precipitates a sharp T_c suppression. Recent investigations of emergent superconducting fluctuations [25] report enhanced pairing gaps under pressure in CsV_3Sb_5 , yet stripe order violently competes with superconducting coherence—echoing cuprate stripe phases, where phase fluctuations robustly suppress macroscopic superconductivity [102]. Moreover, ARPES studies [68] have unveiled a common normal-state electronic structure in AV_3Sb_5 deviating from standard DFT predictions, identifying multiple sublattice-pure VHS near E_F , facilitated by significant $V-d/\text{Sb}-p$ hybridization. Theoretically, this configuration enhances charge and spin bond fluctuations through Fermi surface nesting, favoring bond order or unconventional superconductivity [68], which might be also important for the formation of stripe order in pressurized CsV_3Sb_5 .

Finally, we propose that the positioning of VHS relative to E_F functions as a pivotal tuning parameter, governing electronic correlation strength and dictating the selection of dominant ground states—whether loop currents [41,74,99,103–105], chiral excitonic states [106], or correlated stripe phases [23,31]—across the AV_3Sb_5 family. This framework unifies disparate phase diagrams in kagome superconductors, highlighting the central role of VHS in sculpting intertwined electronic orders.

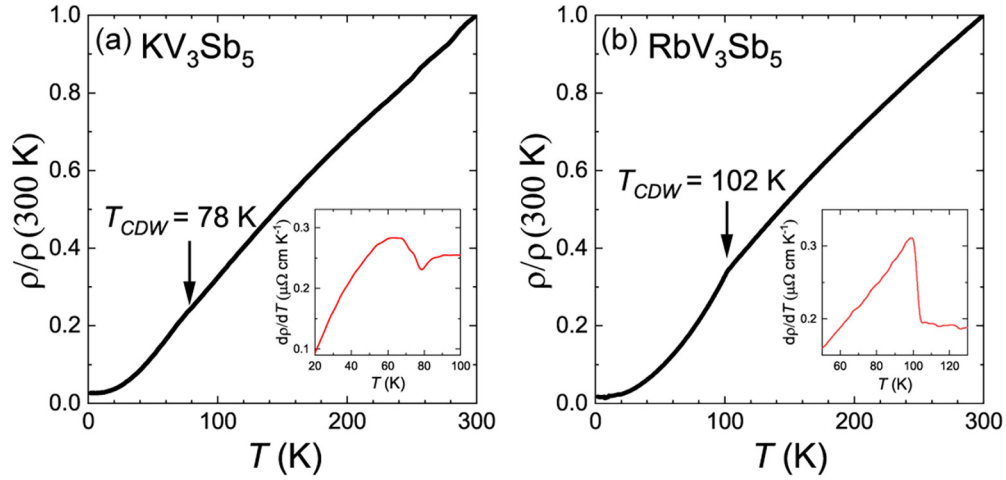


FIG. 7. T -dependent resistivity of KV_3Sb_5 and RbV_3Sb_5 . The inset plots are the temperature derivative of the in-plane resistivity showing the CDW transitions.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

APPENDIX: MATERIALS AND METHODS

1. Samples' growth

High-quality single crystals of KV_3Sb_5 and RbV_3Sb_5 were synthesized via self-flux growth based on previous reports [8]. The high quality of the samples has been verified by the resistivity data in Fig. 7, which were collected on a quantum design physical properties measurement system. The NMR measurement at 0 and 2.02 GPa is carried on the high-quality single crystal CsV_3Sb_5 provided by Dong Chen. All measurements were conducted on the same single crystals to maintain consistency.

2. NMR measurements

A commercial NMR spectrometer from Thamway Co. Ltd. was used for the NMR and nuclear quadrupole resonance

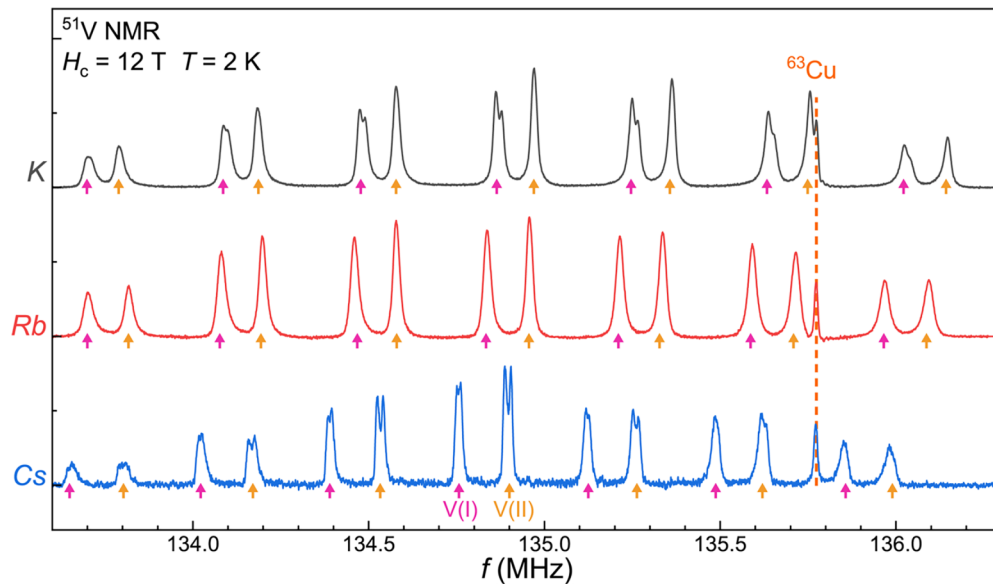


FIG. 8. The typical ^{51}V NMR full spectra of AV_3Sb_5 ($A = K, Rb$, and Cs) at ambient pressure below T_{CDW} . The pink and orange arrows, respectively, mark for the vanadium sites in the hexagon cluster and the triangle cluster in Fig. 1(b).

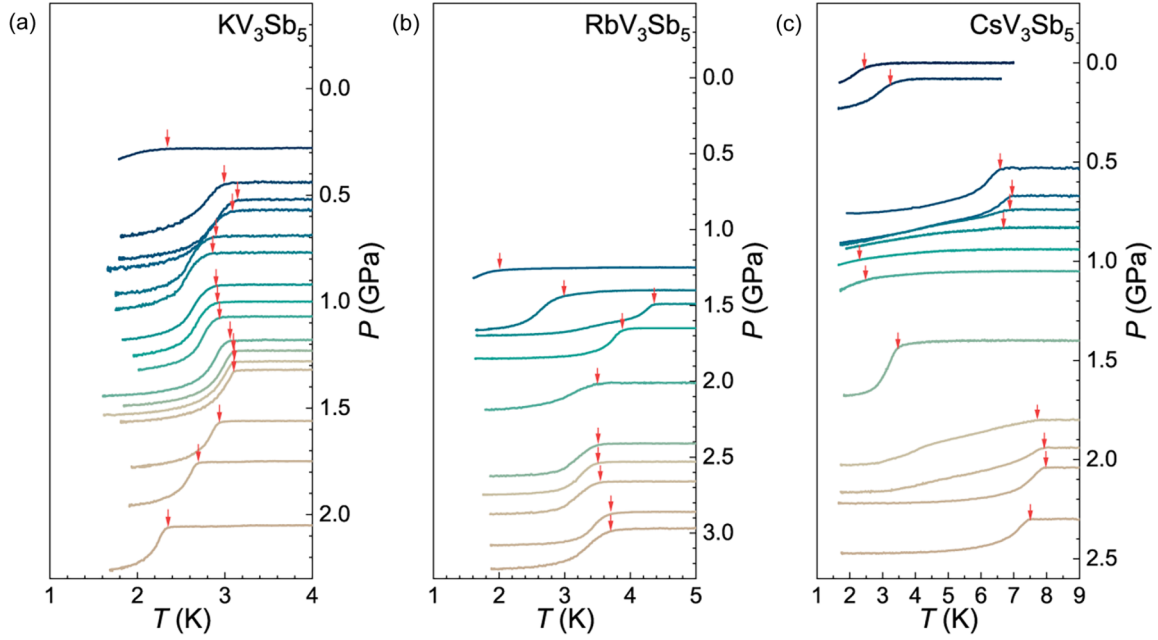


FIG. 9. The T -dependent resonating frequency of the NMR tank circuit under different pressures in (a) KV_3Sb_5 , (b) RbV_3Sb_5 , and (c) CsV_3Sb_5 . The red arrows label the onset temperatures of superconducting transition, where the data and arrows in panel (c) are given by Ref. [23].

(NQR) measurements. An NMR-quality magnet from Oxford Instruments offers a uniform magnetic field of up to 12 T. The external magnetic field applied on the sample is calibrated by ^{63}Cu NMR of the NMR coil. All NMR measurements were performed under an external magnetic field at 12 T along the c axis. The nuclear spin-lattice relaxation times (T_1) of ^{51}V NMR and ^{121}Sb NQR were measured via the inverse recovery method. The fitting function of the recovery of the nuclear magnetization $M(t)$ for ^{51}V NMR is $1 - \frac{M(t)}{M(\infty)} = I_0 \left(\frac{1}{84} \times \exp\left(-\left(\frac{t}{T_1}\right)^\beta\right) + \frac{3}{44} \times \exp\left(-\left(\frac{6t}{T_1}\right)^\beta\right) + \frac{75}{364} \times \exp\left(-\left(\frac{15t}{T_1}\right)^\beta\right) + \frac{1225}{1716} \times \exp\left(-\left(\frac{28t}{T_1}\right)^\beta\right) \right)$ [107], whereas that for ^{121}Sb NQR on the $(\pm\frac{5}{2} \leftrightarrow \pm\frac{3}{2})$ transition is $1 - \frac{M(t)}{M(\infty)} = I_0 (0.571 \times \exp\left(-\left(\frac{10t}{T_1}\right)^\beta\right) + 0.428 \times \exp\left(-\left(\frac{3t}{T_1}\right)^\beta\right))$ [108].

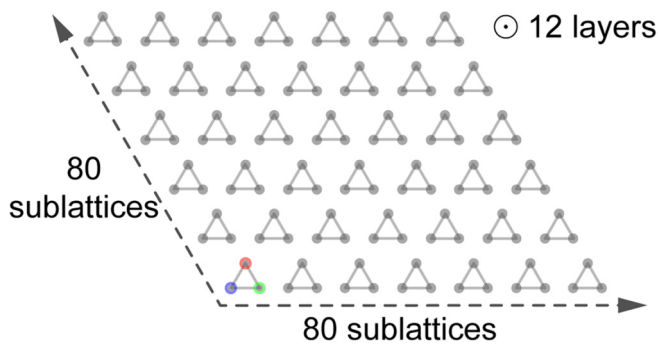


FIG. 10. Sketch plot of the kagome lattice model in the simulation. This model consists of 12 layers of vanadium atoms. Each layer contains an 80×80 grid of triangle sublattices with three V sites, which are highlighted in one sublattice with the three primary colors.

3. Calibration of the hydrostatic pressure

The hydrostatic pressure is realized by self-clamped BeCu piston-cylinder cell. The pressure transmitting medium is Daphne 7373 oil. The pressure was calibrated through the ^{63}Cu NQR frequency of Cu_2O powder in Fig. S1 of the Supplemental Material [94]. To ensure accurate pressure determination, we employed a dual-coil configuration inside the sample chamber. The pressure manometer (Cu_2O NQR coil) was positioned just below the sample coil, with a vertical spatial separation of less than 2 mm. This minimal distance eliminates any significant spatial pressure gradient between the sample and the pressure sensor, which has also been elucidated by our previous NMR experiment [23]. The relation between the pressure and the central frequency (f) of the NQR spectra of Cu_2O powder at different temperatures (T) and pressures (P) has been determined in Ref. [109] with the formula: $f(P, T) = A + BP + CP^2$, where $A = 27.06 - 0.4762\rho(T)$, $B = 0.4154 - 0.02682\rho(T)$, and $C = -2.992 \times 10^{-4}\rho(T)$. In this formula, $\rho(T)$ is the T -dependent phonon distribution function, which is given by $\rho(T) = \frac{1}{2} \coth\left(\frac{h\nu_l}{2k_B T}\right)$, and ν_l is the phonon frequency, which is approximately 2.82×10^{-4} MHz.

4. ^{51}V NMR full spectra below T_{CDW} at ambient pressure in AV_3Sb_5 ($A = \text{K, Rb, and Cs}$)

In AV_3Sb_5 ($A = \text{K, Rb, and Cs}$), the ^{51}V NMR spectrum exhibited seven transition lines with the magnetic field applied along the c axis above T_{CDW} . Below T_{CDW} , as the kagome lattice reconstructs into a staggered trihexagonal superlattice, two inequivalent vanadium sites are generated within the hexagon cluster [V(I)] and the triangle cluster [V(II)]. As shown in Fig. 8, the seven ^{51}V NMR transition lines split into two sets of peaks with equal intensity.

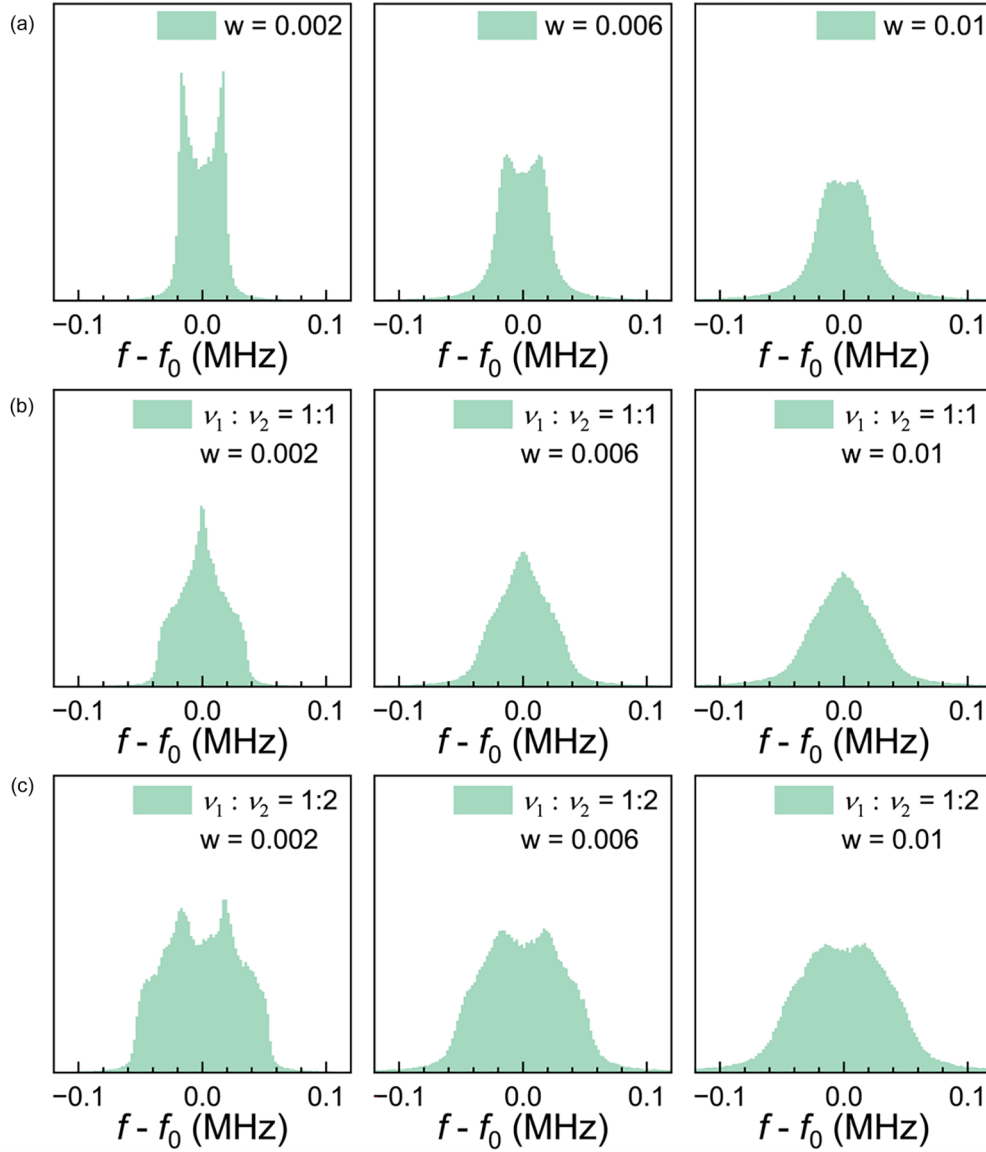


FIG. 11. Single- Q and double- Q simulations of the ^{51}V NMR spectra line shapes. (a) Single- Q incommensurate CDW with linewidth $w = 0, 0.006$, and 0.01 MHz. (b) Double- Q incommensurate CDW with $\nu_1 : \nu_2 = 1:1$ and linewidth $w = 0.006$, and 0.01 MHz. (c) Double- Q incommensurate CDW with $\nu_1 : \nu_2 = 1:2$ and linewidth $w = 0, 0.006$, and 0.01 MHz.

5. Pressure-dependent evolution of superconductivity in AV_3Sb_5 ($\text{A} = \text{K, Rb, and Cs}$)

The NMR tank circuit can be used to detect superconducting transition by measuring the change in the resonant frequency of the NMR tank circuit as shown in Fig. 9. In our experiment, the NMR coil functions as a key inductive component within an LC resonant circuit. When a sample inserted inside the coil cools down and enters the superconducting state, the diamagnetic shielding effect repels the magnetic flux, which effectively reduces the overall inductance of the NMR coil. As the resonant frequency of the LC tank circuit is governed by the relationship $f = \frac{1}{2\pi\sqrt{LC}}$, this physical reduction in inductance directly causes the resonant frequency to increase. Mathematically, this relative change in frequency ($-\Delta f/f$) is directly proportional to the change in the sample's magnetic penetration depth $\Delta\lambda$. Consequently, by continuously

monitoring this background-subtracted shift, the onset of the onset temperatures of superconducting transition is explicitly defined as the temperature where $-\Delta f/f$ deviates from zero.

6. Simulation of the NMR spectra in the incommensurate CDW phases

Assuming that the relation between the NMR frequency shift and the nuclear displacements is purely local and linear [95], the triple- Q CDW modulation at a V atom on the coordinate $\mathbf{r}$ can be denoted as

$$\begin{aligned} \nu(\phi_1, \phi_2, \phi_3) &= \nu_0 + \sum_{i=1,2,3} \nu_i \cos(\phi_i) \\ &= \nu_0 + \sum_{i=1,2,3} \nu_i \cos(\mathbf{q}_i \mathbf{r} + \varphi_i). \end{aligned}$$

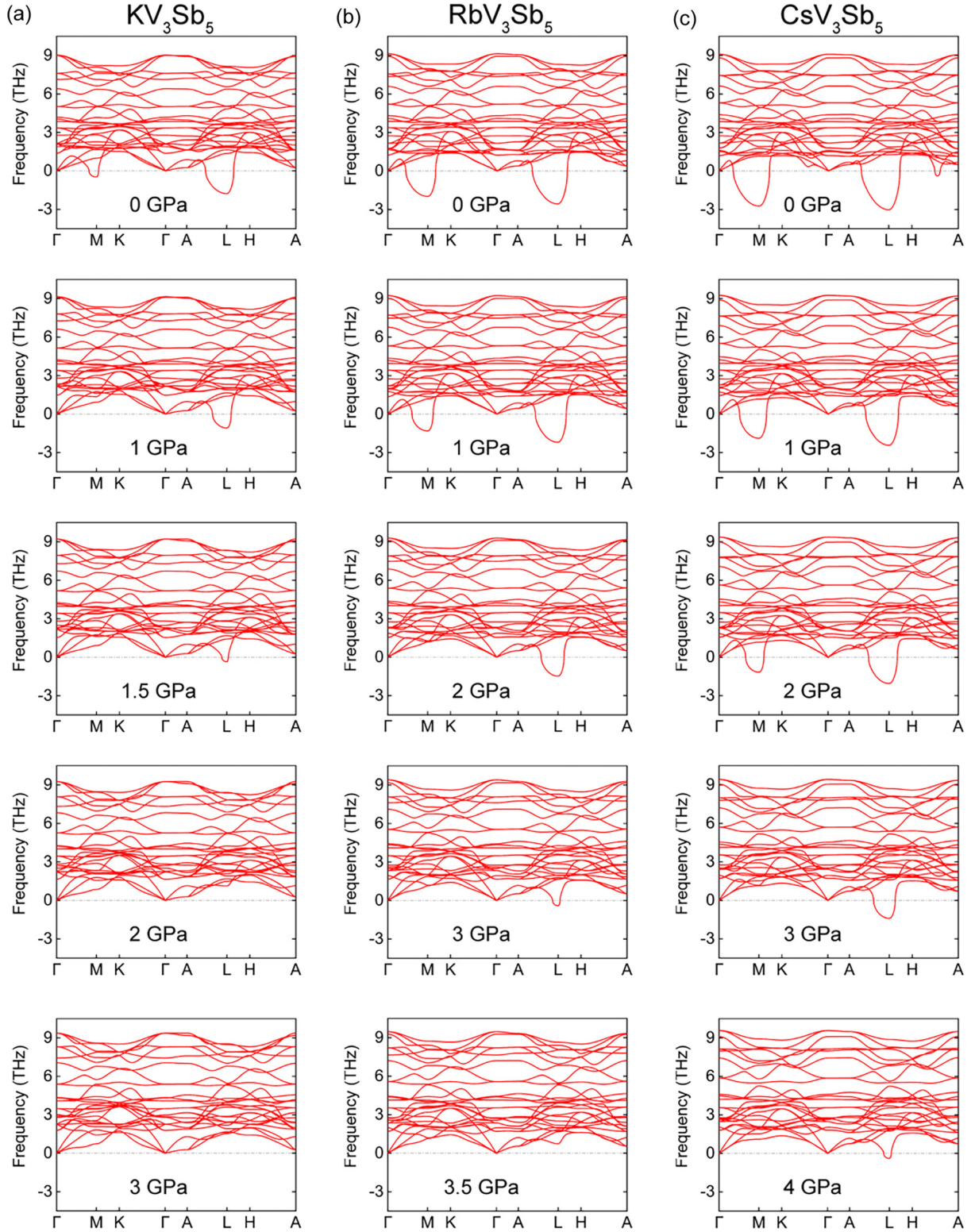


FIG. 12. P -dependent phonon spectra of (a) KV_3Sb_5 , (b) RbV_3Sb_5 , and (c) CsV_3Sb_5 . In KV_3Sb_5 and RbV_3Sb_5 , the phonon imaginary frequency, respectively, persists up to 1.5 and 3.0 GPa, which basically collapse into the critical pressure of the suppression of CDW. However, the phonon imaginary frequency persists up to 4.0 GPa in CsV_3Sb_5 , which strikingly differs from the experimental result.

Here, $\mathbf{q}_i$ represents the various modulation wave vectors; ν_0 is the center frequency; and ν_i is the modulation amplitude, which is proportional to the CDW amplitude. As shown in Fig. 1(b), the standard staggered trihexagonal lattice is

described using the (MLL) basis [71]:

$$\mathbf{q}_1^{(L)} = \left(\frac{1}{2} \ 0 \ \frac{1}{2}\right), \quad \mathbf{q}_2^{(L)} = \left(0 \ \frac{1}{2} \ \frac{1}{2}\right), \quad \mathbf{q}_3^{(M)} = \left(-\frac{1}{2} \ -\frac{1}{2} \ 0\right),$$

in the basis ($\mathbf{G}_1, \mathbf{G}_2, \mathbf{G}_3$):

$$\mathbf{G}_1 = \frac{2\pi}{a_0} \begin{bmatrix} 1 \\ 1/\sqrt{3} \\ 0 \end{bmatrix}, \quad \mathbf{G}_2 = \frac{2\pi}{a_0} \begin{bmatrix} 0 \\ -2/\sqrt{3} \\ 0 \end{bmatrix}, \quad \mathbf{G}_3 = \frac{2\pi}{c_0} \begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix},$$

with $v_1 = v_2 \approx v_3$, and $\varphi_1 = 0$, $\varphi_2 = \frac{\pi}{2}$, and $\varphi_3 = \frac{\pi}{2}$.

To simulate the incommensurate CDW modulation observed under pressure, the wave vectors are modified to $\mathbf{q}_i^* = \mathbf{q}_i(1 - \delta)$, where δ constitutes the incommensurability parameter of the charge density waves. The simulation was performed on a kagome lattice containing 230 400 ($12 \times 80 \times 80 \times 3$) vanadium atoms as shown in Fig. 10. We utilized $\delta = 0.005$, which provides sufficient scale to achieve stable incommensuration in the numerical results.

The spectral intensity $I(\nu)$ in the CDW phase is calculated by summing the frequency contributions of all N atoms:

$$I(\nu) = \sum_j^N \left[\int \delta(\nu - \nu_j(\mathbf{r})) w(\nu) d\nu \right],$$

where $N = 230\,400$ and $w(\nu)$ is a Lorentzian broadening function:

$$w(\nu) = \frac{2A}{\pi} \frac{W}{4(\nu - \nu_0)^2 + W^2},$$

where A is the intensity and W is the width of peak.

We first evaluated the single- Q and double- Q CDW modulations through spectral line shape simulations. As shown in

Fig. 11(a), the line shape predicted by the single- Q model fundamentally deviates from the experimental profile, manifesting as a deep spectral gap at the center. Although the double- Q model with $\nu_1 = \nu_2$ does yield a central protrusion [Fig. 11(b)], its spectral weight is disproportionately pronounced compared to the subtle feature observed in KV_3Sb_5 . Furthermore, under the condition $2\nu_1 = \nu_2$, the characteristic central plateau is entirely absent, replaced instead by a pronounced central depression. Because these simpler modulations fail to reproduce the key spectral features, we conclude that the experimental spectra are best described by the distinct triple- Q incommensurate CDW modulations presented in Fig. 5.

7. First-principles calculations

The temperature-dependent $N(E_F)$ is calculated with the formula $N(E_F) = \frac{1}{k_B T} \int (1 - f(E)) f(E) D(E) dE$. Here, k_B denotes the Boltzmann constant, $f(E)$ denotes the Fermi-Dirac distribution, and $D(E)$ denotes the DOS. The T -dependent $N(E_F)$ values are calculated with the VHS point located at different places relative to the Fermi surface in the kagome lattice. Here, we take the DFT calculation of the electronic structure of CsV_3Sb_5 at ambient pressure as example and adjust the alignment of VHS at Fermi surface [23]. The DFT calculations were performed as implemented in the Vienna *ab initio* simulation package [110]. The phonon dispersions are obtained by using the PHONOPY code [111], and the results are shown in Fig. 12.

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