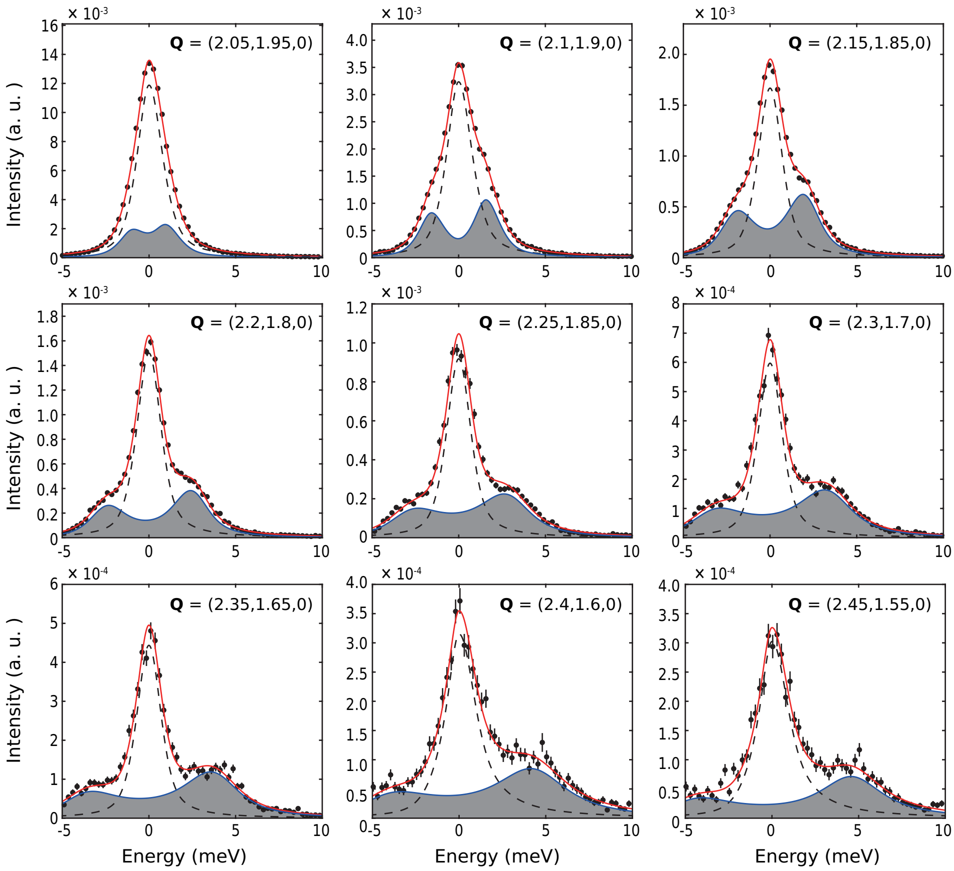
Supplementary Materials: Antiferromagnetic Dimers in the Parent Phase of a Correlated Kagome Superconductor

SUPPLEMENTARY NOTE 1: RAW AND ADDITIONAL IXS DATA

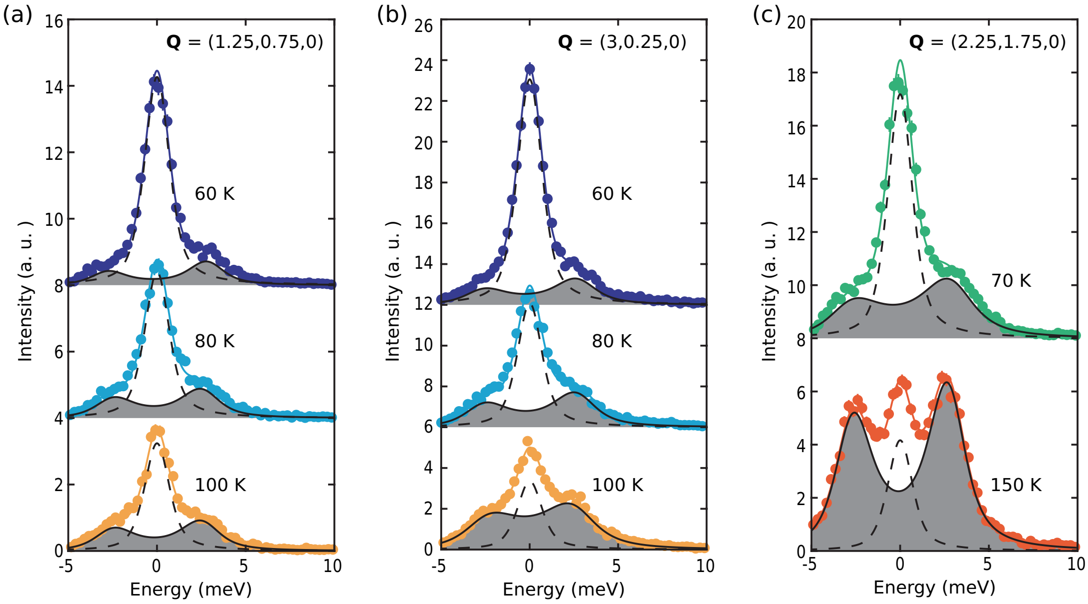
The raw data for Figs. 3(c) and 3(d) of the main text are respectively shown in Supplementary Figs. 1 and 2, with the fitted acoustic phonons shown as gray shaded areas. To highlight the dispersion of acoustic phonons along $(2+q, 2-q, 0)$, the fitted elastic peaks (dashed lines in Supplementary Fig. 1) were subtracted, and the resulting data are shown as a color map in Fig. 3 (c) of the main text.

At $\mathbf{q}_{\mathrm{CDW}}$ and $2\mathbf{q}_{\mathrm{CDW}}$, the elastic peak exhibits an increase upon cooling towards T_{CDW} from above, as can be seen for $\mathbf{Q} = (1.5, 0, 2)$ [Supplementary Fig. 3(a)]. The fitted elastic intensity c and phonon intensity A_{ph} are shown in Supplementary Fig. 3(b), showing that whereas A_{ph} is essentially temperature-independent,the elastic intensity c clearly increases upon cooling. Such an effect may result from an order-disorder character of the CDW transition, with the increase in c above T_{CDW} due to the nucleation of ordered regions.

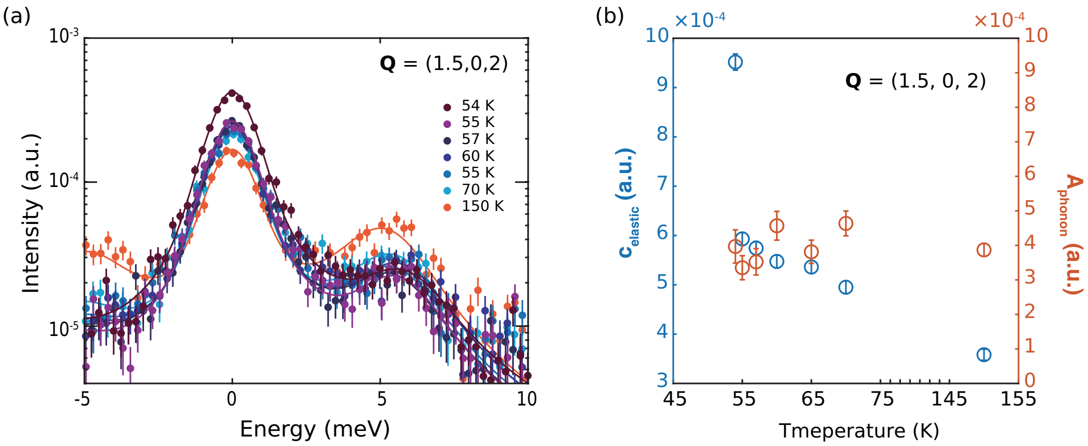


Supplementary Figure 1: Phonon spectra at 70 K for various momenta $\mathbf{Q} = (2+q, 2-q, 0)$. The corresponding color map, with the fitted elastic peak subtracted, is shown in Fig. 3(c) of the main text, together with the fitted phonon energies E_{ph}.

Supplementary Fig. 4 compares the experimentally determined CDW structure with the calculated ground state, with the latter constrained by the experimentally determined space group. As can be seen, the experimental and calculated structures are in excellent agreement, with the Cr layer showing Cr-Cr dimers and chains, and the Cs layer showing Cs dimers.



Supplementary Figure 2: Phonon spectra at $\mathbf{q}_{\rm CDW}$ in different Brillouin zones. The corresponding fitted phonon energies $E_{\rm ph}$ are shown in Fig. 3(d) of the main text.



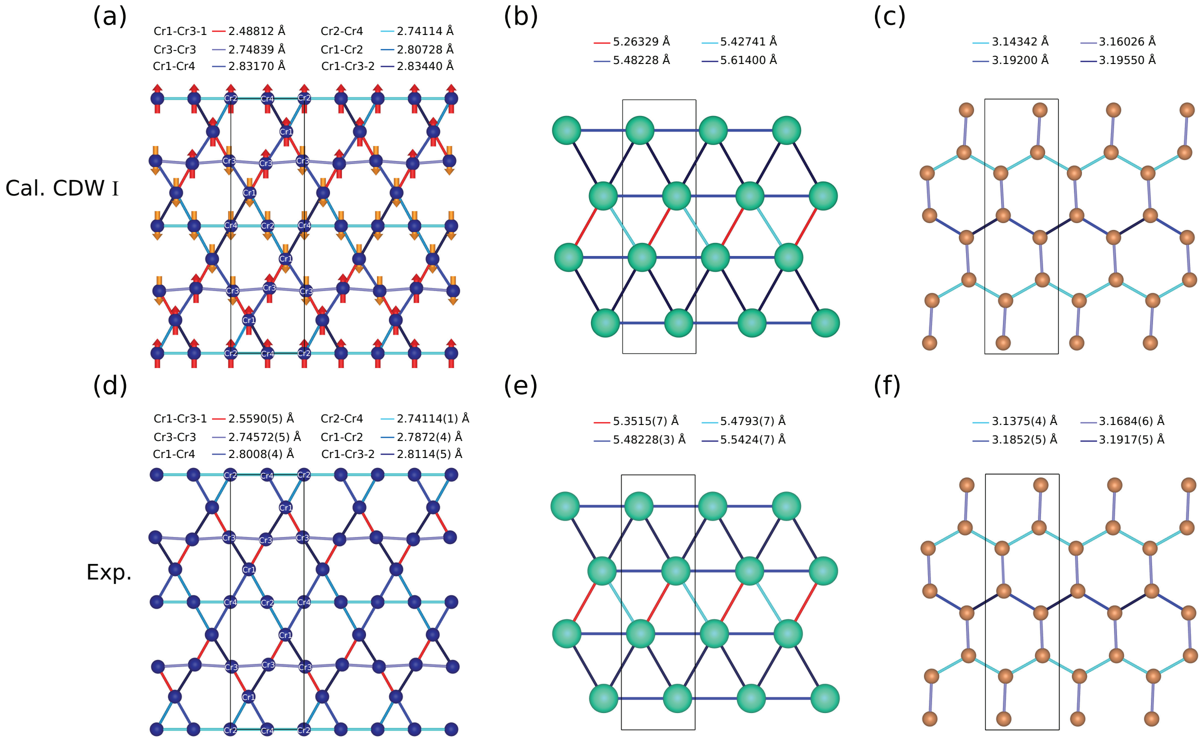
Supplementary Figure 3: (a) Phonon spectra at $\mathbf{Q} = (1.5, 0, 2)$ at various temperatures, shown in log scale. (b) Temperature dependence of elastic intensity c and phonon intensity $A_{\rm ph}$, from fits in (a).

SUPPLEMENTARY NOTE 2: CALCULATED 4×1 Pbam STRUCTURES

By assuming collinear magnetic structures without net moments, an exhaustive search of magnetic structures was performed for $CsCr_3Sb_5$ in Ref. [1]. Of these, 7 distinct magnetic configurations were found to have CDW structures consistent with the experimentally determined 4×1 *Pbam* supercell. These calculated CDW structures (Cal. CDWs I-VII) were then relaxed by constraining the lattice parameters to experimental values at 40 K, and are shown in Supplementary Fig. 5, with the corresponding Cr-Cr bond lengths presented in Table I.

The 4×1 *Pbam* structure with the lowest energy [Fig. 4(a) of the main text and Supplementary Fig. 4(a)] is in good agreement with the experimentally determined CDW structure, and has Cr1-Cr3-1 bonds that are significantly shorter than other Cr-Cr bonds, as discussed in the main text.

Interestingly, Cr-Cr dimers are also observed in several other 4×1 *Pbam* structures [Supplementary Figs. 5(b)-(g)], and suggests that Cr-Cr dimerization is a robust tendency in $CsCr_3Sb_5$.



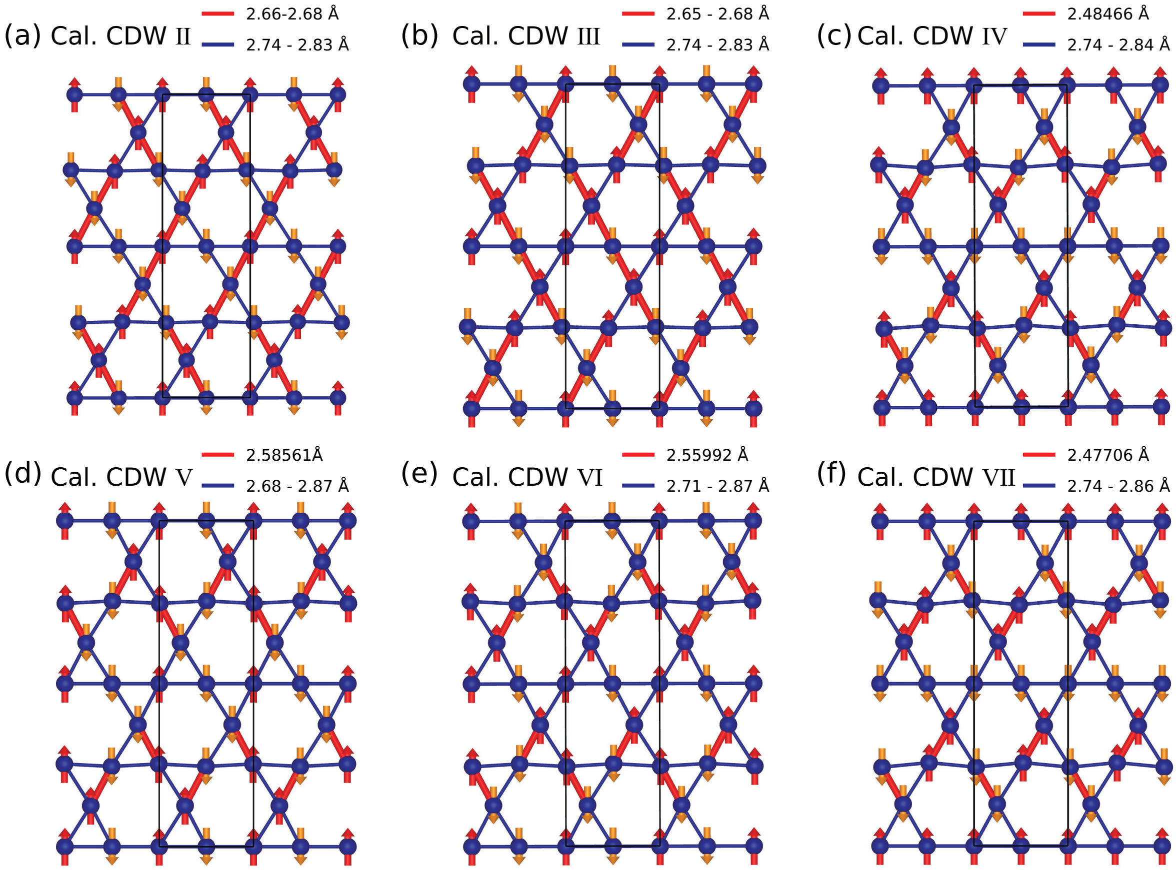
Supplementary Figure 4: Comparison between the experimental and calculated CDW structures of $CsCr_3Sb_5$. Calculated 4×1 *Pbam* ground state structure of $CsCr_3Sb_5$, for (a) Cr atoms in the $z = 0.5$ plane, (b) Cs atoms in the $z = 0$ plane, and (c) Sb atoms in the $z = 0.25$ plane. (d)-(f) shows corresponding plots for the experimentally determined structure.

Table I: Comparison between experimentally determined and calculated 4×1 *Pbam* crystal structures. The calculated CDW structures (Cal. CDW I - Cal. CDW VII) were obtained by relaxing the the atomic coordinates while fixing the lattice parameters to experimentally determined values at 40 K. This constrains the Cr2-Cr4 bond length to 2.74114 Å, half of the lattice parameter a.

Type	Energy*	Cr1-Cr3-1 (Å)	Cr3-Cr3 (Å)	Cr1-Cr2 (Å)	Cr1-Cr4 (Å)	Cr1-Cr3-2 (Å)
Experiment	-	2.5590(5)	2.74572(5)	2.7872(4)	2.8008(4)	2.8114(5)
Cal. CDW I	0	2.48812	2.74839	2.80728	2.83170	2.83440
Cal. CDW II	4.94	2.66267	2.74217	2.80578	2.67012	2.82447
Cal. CDW III	5.09	2.82523	2.74225	2.67108	2.80805	2.65884
Cal. CDW IV	11.26	2.48812	2.74839	2.80728	2.83440	2.83170
Cal. CDW V	33.94	2.86973	2.74483	2.82032	2.68947	2.58561
Cal. CDW VI	34.89	2.55992	2.74577	2.71684	2.82596	2.86093
Cal. CDW VII	188.40	2.47706	2.75422	2.80050	2.82651	2.85505

* Total energy (in meV/f.u.) relative to the calculated ground state (Cal. CDW I).

[1] C. Xu, S. Wu, G.-X. Zhi, G. Cao, J. Dai, C. Cao, X. Wang, and H.-Q. Lin, Nature Communications **16**, 3114 (2025), ISSN 2041-1723, URL https://www.nature.com/articles/s41467-025-58446-6.



Supplementary Figure 5: Other 4×1 *Pbam* supercell structures of $CsCr_3Sb_5$. The shortest Cr-Cr bonds are shown in red, whereas the other Cr-Cr bonds are shown in blue.