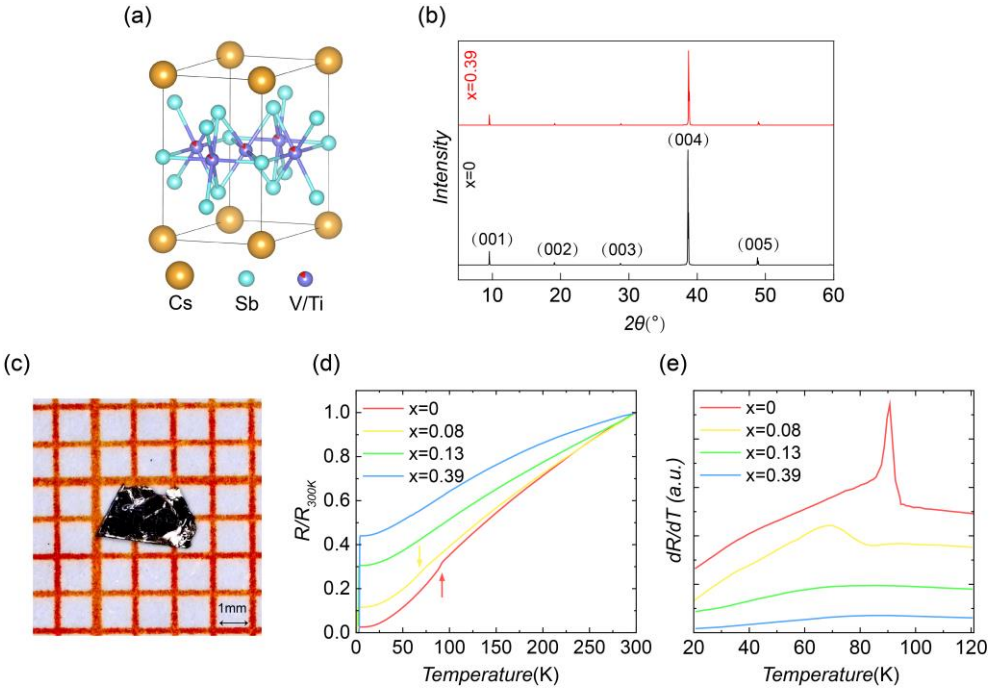
Supplementary Information

Supplementary Note 1

Single crystals of $CsV_{3-x}Ti_xSb_5$ were grown by self-flux growth method similar to that in the previous report [1]. The chemical composition was determined by inductively coupled plasma atomic emission spectroscopy (ICP). Electrical transport measurements were carried out by using a Quantum Design physical property measurement system (PPMS-9T). The electrical transport measurements in a conventional four-lead configuration were realized by attaching four platinum wires to the samples. Contacts were made by using DuPont 4929N conductor paste. Single-crystal X-ray diffraction measurements were carried out on a XtaLAB AFC12 (RINC).

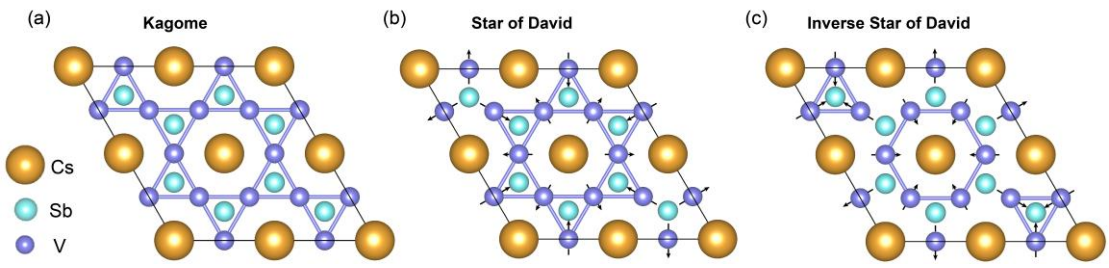


Supplementary Figure 1: Sample characterization of $CsV_{3-x}Ti_xSb_5$. (**a**) Crystal structure of $CsV_{3-x}Ti_xSb_5$. (**b**) XRD results of the $CsV_{3-x}Ti_xSb_5$ samples. (**c**) Image of the as-grown $CsV_{3-x}Ti_xSb_5$ single crystal. (**d**) Temperature dependence of the ab-plane resistivity for the $CsV_{3-x}Ti_xSb_5$ samples measured by ARPES (x=0, 0.08, 0.13, 0.39). (**e**) Second derivative plots of the resistivity results in **d**, which enhances the feature induced by the CDW order. Apparently, the CDW order persists in samples with x=0 and 0.08, but disappears in the samples with x=0.13 and 0.39.

Supplementary Note 2

First-principles calculations were performed by using the projected augmented-wave method [2] as implemented in the Vienna ab initio simulation package (VASP) [3,4]. The exchange-correlation interaction was addressed using the Perdew-Burke-Ernzerhof type of generalized gradient approximation [5]. The kinetic energy cutoff and energy threshold for convergence were set to be 520 eV and 10^{-6} eV, respectively. The van der Waals correction was applied using the zero-damping DFT-D3 method [6].

In order to model the Ti-doped CsV_3Sb_5, we constructed a 2×2×2 supercell and employed the experimental lattice constants with a = b = 5.4949 Å and c = 9.3085 Å. To assess the energy variation concerning structural distortion in $CsV_{3-x}Ti_xSb_5$, we utilized the pristine Star of David (SD), Inverse Star of David (ISD) and Kagome geometries as the initial structures (Supplementary Figure 2), and we further generated several interpolated structures between these initial configurations. The calculated doping levels were selected to mimic those in the experiments. We note that the above calculation approach focuses on the evolution between SD, ISD and Kagome structures and ignores the small lattice distortion induced by Ti atoms. This is different from the method used in our previous work [7], where an optimized geometry of Ti-doped CsV_3Sb_5 (slightly distorted Kagome lattice) was used as the initial structure to calculate the samples with low Ti doping levels. However, it is important to note that the qualitative trend of the doping dependent total energy remains robust, regardless of the different approaches and different doping levels applied in the calculations.



Supplementary Figure 2: Top-view of the supercells for Kagome structure, Star of David structure and Inverse Star of David structure. The black arrows indicate the lattice distortion due to the breathing mode.

Supplementary References

1. Wu, P. et al. Unidirectional electron–phonon coupling in the nematic state of a kagome superconductor. Nat. Phys. **19,** 1143–1149 (2023).
2. Blöchl, P. E. Projector augmented-wave method. Phys. Rev. B **50**, 17953-17979 (1994).
3. Kresse, G. & Furthmuller, J. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. Phys. Rev. B **54**, 11169 (1996).
4. Kresse, G. & Joubert, D. From ultrasoft pseudopotentials to the projector augmented-wave method. Phys. Rev. B **59**, 1758 (1999).
5. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized Gradient Approximation Made Simple. Phys. Rev. Lett. **77**, 3865 (1996).
6. Grimme, S., Antony, J., Ehrlich, S. & Krieg, H. A consistent and accurate ab initio parametrization of density functional dispersion correction (DFT-D) for the 94 elements H-Pu. J. Chem. Phys. **132**, 154104 (2010).
7. Liu, B. et al. Tunable van Hove singularity without structural instability in kagome metal $CsTi_3Bi_5$. Phys. Rev. Lett. **131**, 026701 (2023).