

Supplementary Information:

Unusual competition of superconductivity and charge-density-wave state in  
a compressed topological kagome metal

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Supplementary Fig.1a shows the temperature dependence of resistivity up to room temperature, for sample 1 with PCC. Supplementary Fig.1b shows the temperature dependence of resistance up to room temperature, for sample 2 with DAC.

Supplementary Fig.2 shows the high-pressure magnetic susceptibility measurement for sample 3 with PCC. The  $T_c$  gradually increases with increasing the pressure below P1, consistent with the high-pressure resistivity measurements. With the pressure above P1, the superconducting volume fraction suddenly decreases. Further increasing the pressure, the bulk superconductivity  $T_c^{M2}$  emerges below 4 K around 1.1 GPa, while the magnetic susceptibility shows a weak reduction at higher temperature at  $T_c^{M1}$  as shown in Supplementary Fig.2b, indicating a filamentary superconductivity.

Supplementary Fig.3 shows the temperature dependence of resistivity under various magnetic fields with pressure of 1.06 GPa for sample 1 with PCC. The superconducting transition is very broad and  $T_c$  can be gradually suppressed by increasing the magnetic field.

Supplementary Fig.4 shows the magnetoresistance measured at 10 K under external magnetic field applied along the  $c$  axis and at various pressures, for sample 1 with PCC. The magnitude of magnetoresistance suddenly decreases above P1. The low-field magnetoresistance evolves from a “V”-shape to “U”-shape at P2. The different MR behavior can be related to the different electronic structure and electron scattering in the different pressure regions. The linear MR at the low field region below P2 can possibly arise from the linear band crossings<sup>1,2</sup> or/and the CDW state<sup>3</sup>. We also performed the MR measurements under various pressures at 1.02 GPa for sample 4 with PCC as shown in Supplementary Fig.5. The  $T^*$  is suppressed to 49 K in this sample as determined by  $d\rho_{xx}/dT$  in Supplementary Fig.5a. We find that the shape of MR suddenly changes from “V”-shape to “U”-shape above  $T^*$ , similar with the case by tuning using the pressure. Therefore, the observed linear MR is associated with the CDW order.

We performed magnetoresistance measurements with magnetic field up to 14 T at 2 K, at several pressures for sample 4 with PCC and the results are shown in Supplementary Fig.6. Clear Shubnikov-de Haas (SdH) quantum oscillations (QOs) are observed with pressure below P1 as shown in Supplementary Fig.6a. Above P1, the QOs become much weaker and cannot be resolved at pressure of 1.02 GPa. Analyzing the SdH QOs can provide insight on the Fermi surfaces. After subtracting a slowly changed polynomial background, the oscillation parts of resistivity  $\Delta\rho_{xx}$  as a function of  $1/(\mu_0 H)$  at various pressures are shown in Supplementary Fig.6b. The fast Fourier transform (FFT) spectra of the QOs reveal four principal frequencies (18, 26, 72 and 92 T) at

ambient pressure as shown in Supplementary Fig.6c. These frequencies remain the same as that at pressure of 0.36 GPa. With the pressure above P1, the QOs damping rapidly and we can hardly extract the reliable frequencies.

The effective mass can be extracted from the temperature dependence of the amplitude of FFT peak using the Lifshitz-Kosevich (LK) formula<sup>4</sup>. The oscillation amplitude at a fixed magnetic field is proportional to the thermal damping factor  $R_T$ :  $R_T = \frac{\alpha m^* T}{B \sinh(\alpha m^* T/B)}$ , where  $\alpha = 14.69$  T/K is a constant,  $B = \mu_0 H$  is the magnetic flux density (taking the average value of the field window used for the FFT of QOs), and  $m^* = m/m_e$  is the cyclotron mass ratio ( $m_e$  is the mass of free electron). The extracted fast Fourier transform frequency under various temperatures with pressure of 0.36 GPa are shown in Supplementary Fig.7. The temperature dependence of FFT amplitude of F1 and F2 can be fitted very well with LK formula as shown in the inset of Supplementary Fig.7, and yields effective mass to be  $0.025m_e$  and  $0.034m_e$  for F1 and F2 orbits, respectively. Comparing with the extracted effective mass of  $0.028 m_e$  and  $0.031m_e$  for F1 and F2 at ambient pressure<sup>5</sup>, the effective mass does not change much under pressure. The amplitude of the frequencies at 26 and 72 T dramatically increases at 0.36 GPa as shown in Supplementary Fig.6c. The amplitude of the QOs depends on  $R_T$  and Dingle damping factor  $R_D = \exp(-\alpha T_D m^*/B)$ , where  $T_D$  is the Dingle temperature which is associated with the scattering rate. Since the effective mass does not change much at 0.36 GPa, the enhancement of amplitude can be attributed to the reduction of the scattering rate for the two bands.

Quantum criticality may appear when the CDW transition is totally suppressed. In order to check the possibility of quantum criticality at P2, we replot the low-temperature resistivity as a function of  $T^2$  as shown in Supplementary Fig.8. The low-temperature resistivity above P2 follows  $T^2$  behavior below 35 K indicating the Fermi liquid nature in the normal state, in contrast with the quantum critical behavior. In addition, the residual resistivity does not show a sharp enhancement around P2. For these reasons, our results do not support the quantum criticality at P2. However, ultralow-temperature experiments are still highly required to clarify possible quantum criticality at lower temperature.

The upper critical field can be fitted well by using an effective two-band model<sup>5</sup>,

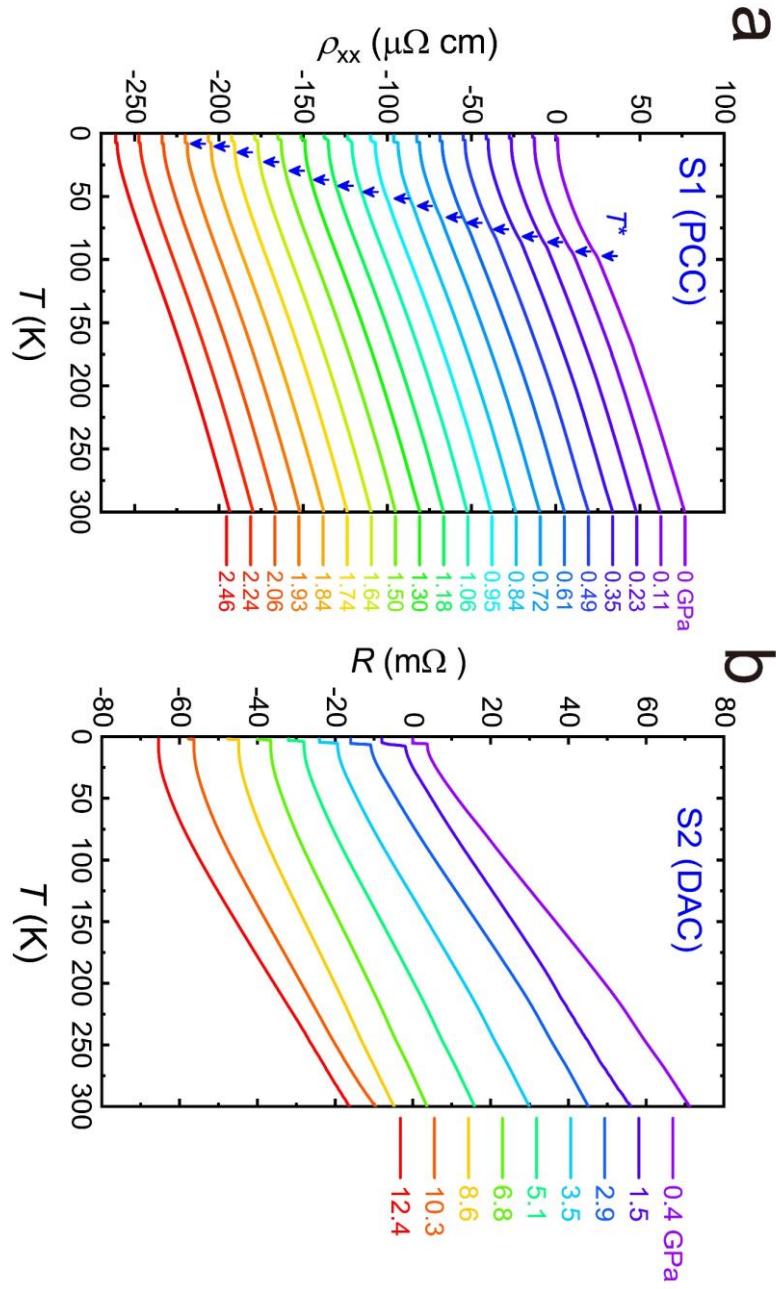
$$a_0 [\ln t + U(h)] [\ln t + U(\eta h)] + a_1 [\ln t + U(h)] + a_2 [\ln t + U(\eta h)] = 0$$

where  $a_0 = 2(\lambda_{11}\lambda_{22} - \lambda_{12}\lambda_{21})$ ,  $a_1 = 1 + (\lambda_{11} - \lambda_{22})/\lambda_0$ ,  $a_2 = 1 - (\lambda_{11} - \lambda_{22})/\lambda_0$ ,

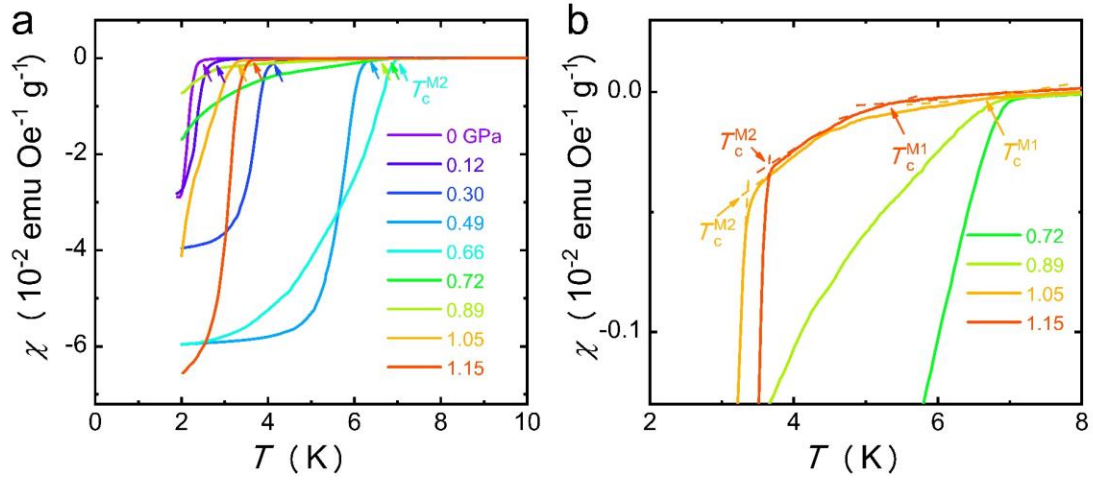
$\lambda_0 = [(\lambda_{11} - \lambda_{22})^2 + 4\lambda_{12}\lambda_{21}]^{1/2}$ ,  $\lambda_{11}$  and  $\lambda_{22}$  are the intraband BCS coupling constants, while  $\lambda_{12}$  and

$\lambda_{21}$  are the interband BCS coupling constants. The function  $U(x) = \psi(1/2+x) - \psi(1/2)$ , where  $\psi$  is the di-gamma function.  $t = T/T_c$ ,  $h = H_{c2}D_1/2\phi_0T$  and  $\eta = D_2/D_1$ , where  $\phi_0$  is the flux quantum and  $D_n$  is the electron diffusivity for the  $n$ th Fermi-surface sheet. The coherence lengths can be calculated as  $\varepsilon_n = \sqrt{D_n/2\pi T_c}$ , which are shown in Supplementary Fig.9. Both  $\varepsilon_1$  and  $\varepsilon_2$  show minimum values around P1 and P2.

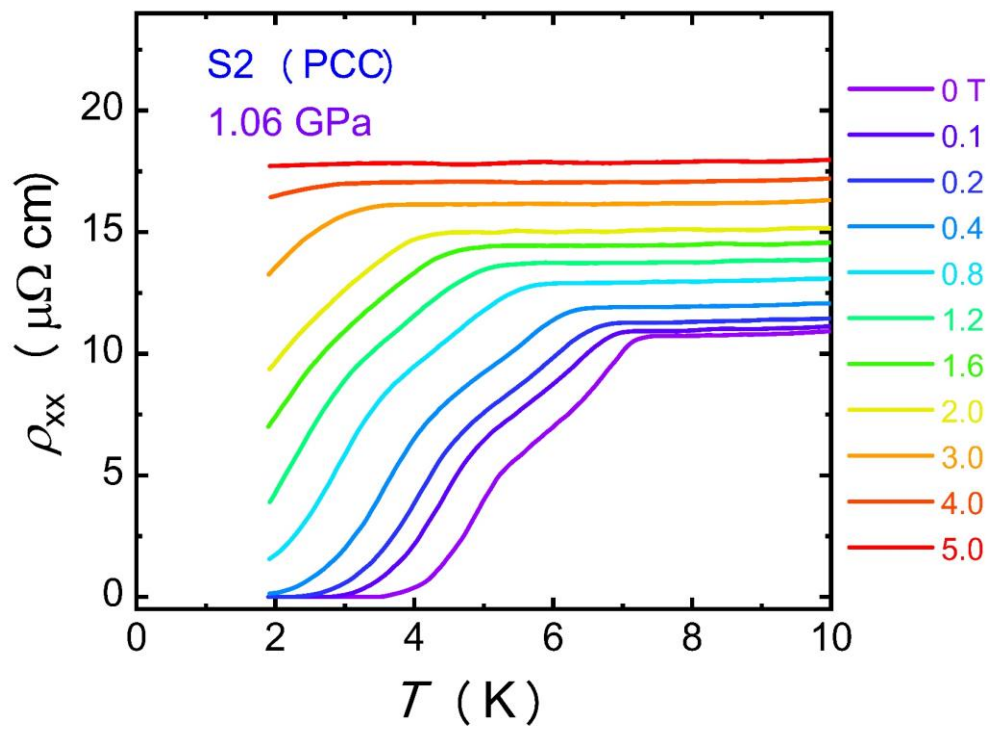
Supplementary Fig.10 shows the expanded low-pressure phase diagram with  $T_c$  measured by various methods.



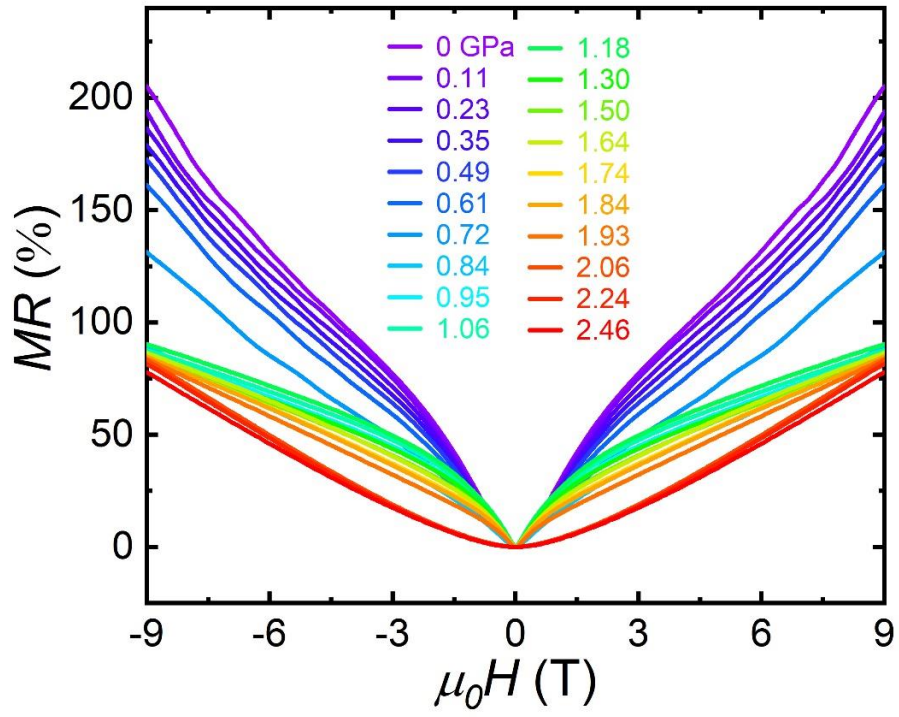
Supplementary Fig.1. (a): Temperature dependence of resistivity in the temperature range from low temperature to room temperature at various pressures for sample 1 measured with PCC. (b): Temperature dependence of resistance in the temperature range from low temperature to room temperature for sample 2 with DAC. All the curves were shifted vertically for clarity.



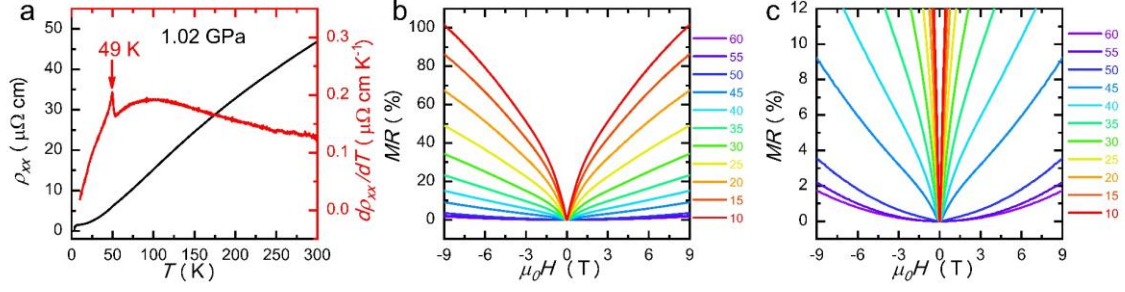
Supplementary Fig.2. (a): High-pressure magnetic susceptibility measurement for sample 3 with PCC. (b): Enlarged area of magnetic susceptibility around  $T_c$  with pressure above P1.  $T_c^{M2}$  represent the bulk superconducting temperature and  $T_c^{M1}$  represents the transition of filamentary superconductivity.



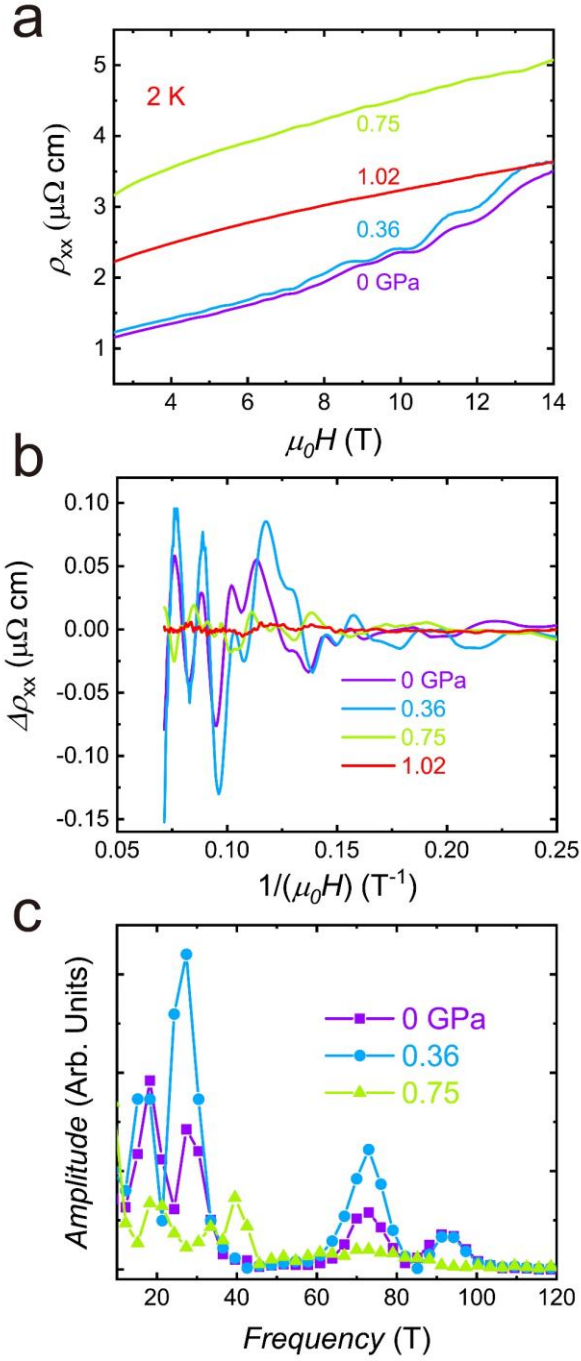
Supplementary Fig.3. Temperature dependence of resistivity under various magnetic fields with pressure of 1.06 GPa.



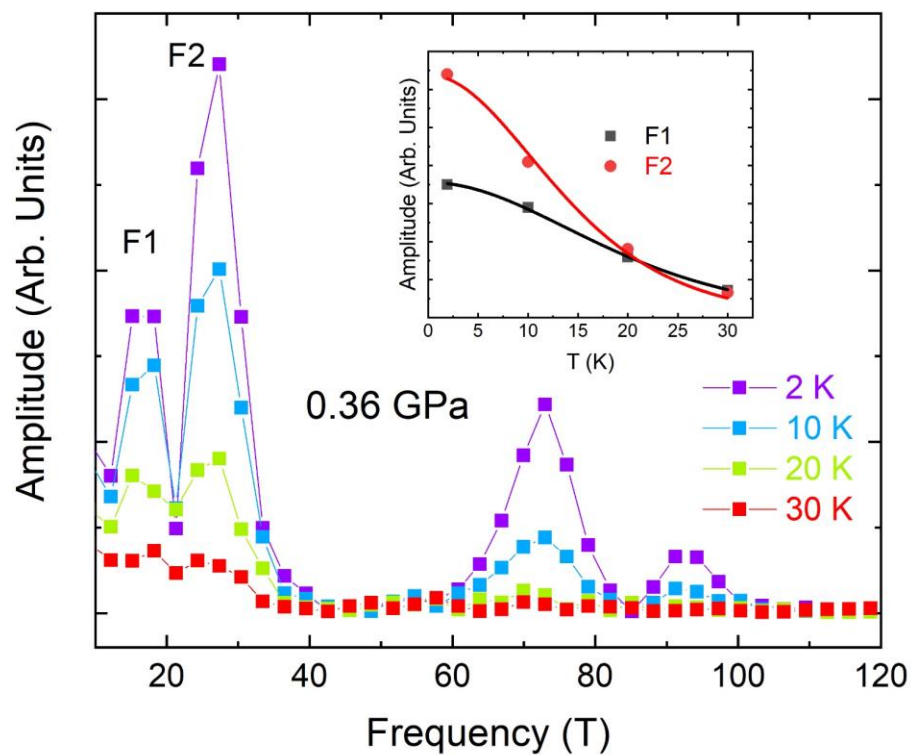
Supplementary Fig.4. Magnetoresistance measured at 10 K under external magnetic field applied along  $c$  axis and at various pressures for sample 1 with PCC. The magnitude of magnetoresistance suddenly decreases above P1. The low-field magnetoresistance evolves from “V”-shape to “U”-shape at P2.



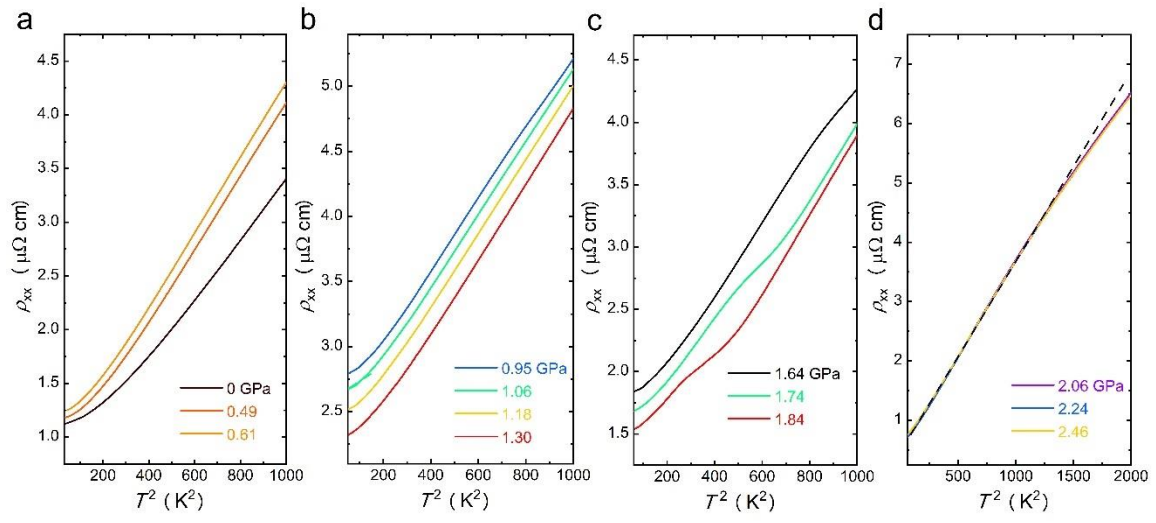
Supplementary Fig.5. (a): Temperature dependences of resistivity and the derivative  $d\rho_{xx}/dT$  curve for sample 4 with the pressure at 1.02 GPa.  $T^*$  is determined to be 49 K. (b) and (c): Magnetoresistance measured at various temperatures with external magnetic field applied along  $c$  axis. Magnetoresistance suddenly changes around  $T^*$ .



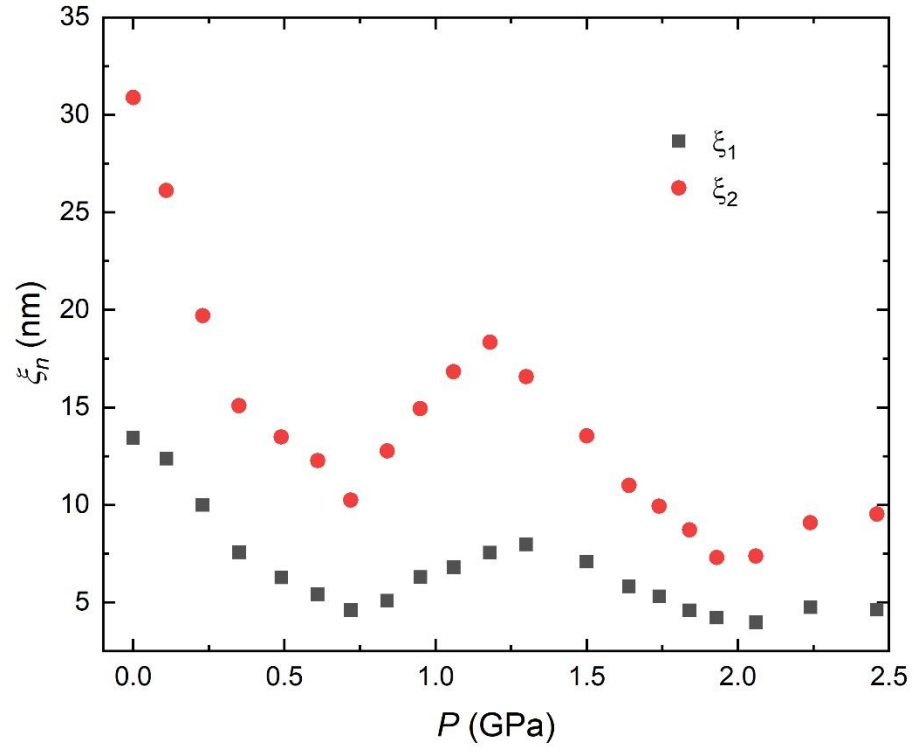
Supplementary Fig.6. (a): The magnetoresistance measured under external magnetic field applied up to 14 T along  $c$  axis at 2 K and at several pressures for another sample with PCC. (b): The oscillation parts of resistivity  $\Delta\rho_{xx}$  after subtracting the background as a function of  $1/(\mu_0 H)$  at various pressures. (c): Extracted fast Fourier transform spectra at various pressures.



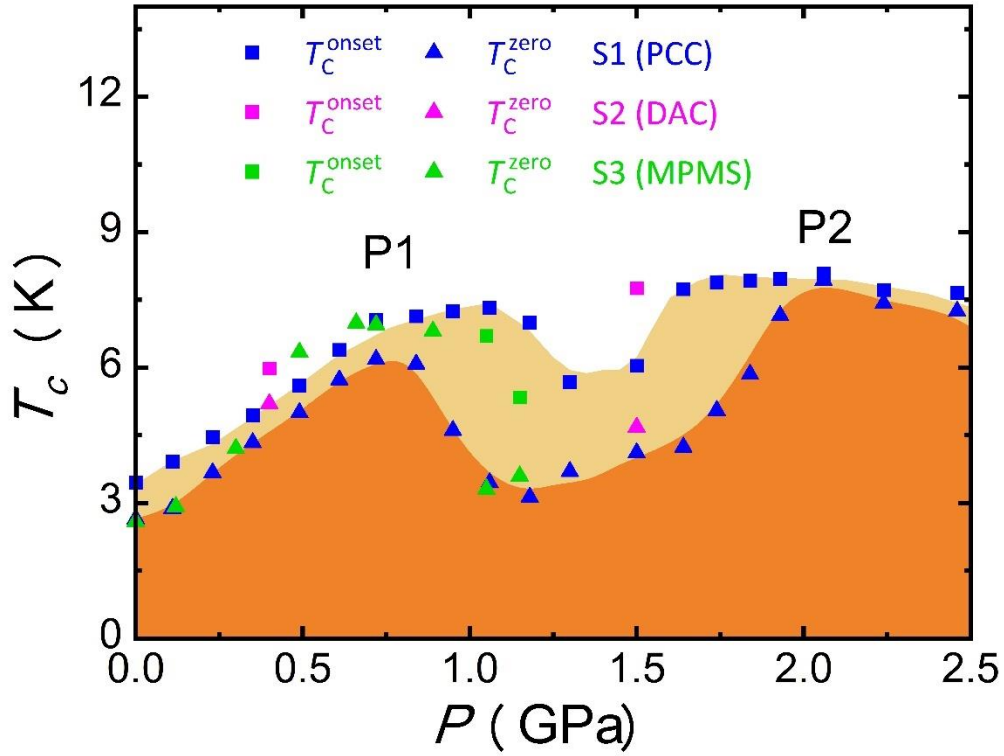
Supplementary Fig.7. Extracted fast Fourier transform frequency of  $\text{CsV}_3\text{Sb}_5$  measured at various temperatures with pressure of 0.36 GPa. The inset shows the Lifshitz-Kosevich fit of the F1 and F2 orbits to extract the effective mass.



Supplementary Fig.8. Low-temperature resistivity as a function of  $T^2$  under pressure. The resistivity curves above P2 are nearly the same and follow the  $T^2$  behavior below 35 K, indicating the Fermi liquid nature for  $\text{CsV}_3\text{Sb}_5$  at P2. The anomaly for resistivity between 1.64 and 1.84 GPa is related to the CDW transition.



Supplementary Fig.9. Temperature dependence of coherence length  $\varepsilon_1$  and  $\varepsilon_2$  derived from the two-band model fitting of the upper critical field. Both  $\varepsilon_1$  and  $\varepsilon_2$  show minimum values around P1 and P2.



Supplementary Fig.10. Pressure dependence of superconducting transition temperatures  $T_c^{\text{onset}}$ ,  $T_c^{\text{zero}}$ ,  $T_c^{M1}$  and  $T_c^{M2}$  measured on various samples below 2.5 GPa.

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- 5 Yu, F. H. *et al.* Concurrence of anomalous Hall effect and charge density wave in a superconducting topological kagome metal. *arXiv:2102.10987* (2021).
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