

Supplemental Material for “Strain-Tuned Nodal Superconductivity in the Charge-Ordered Kagome Metal CsV_3Sb_5 ”

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(Dated: July 22, 2026)

S1. COMPARISON OF THE STRAIN DEPENDENCE OF T_c WITH PREVIOUS REPORT

To demonstrate that uniaxial strain is properly applied to the sample in our experimental setup, we compared the strain dependence of the superconducting transition temperature (T_c) obtained in this study with a previously reported macroscopic measurement.

Figure S1 shows the strain dependence of the normalized transition temperature, denoted here as $\Delta T_c = T_c(\varepsilon)/T_c(0)$, for $\varepsilon \parallel a$. The open squares represent our data evaluated from the onset of the ac susceptibility (χ^{onset}). For comparison, we plotted the data extracted from the electrical resistivity measurements (ρ^{mid}) by Qian *et al.* [35] as red crosses. As clearly seen in the figure, our ΔT_c data quantitatively agree well with the resistivity results over the entire strain range.

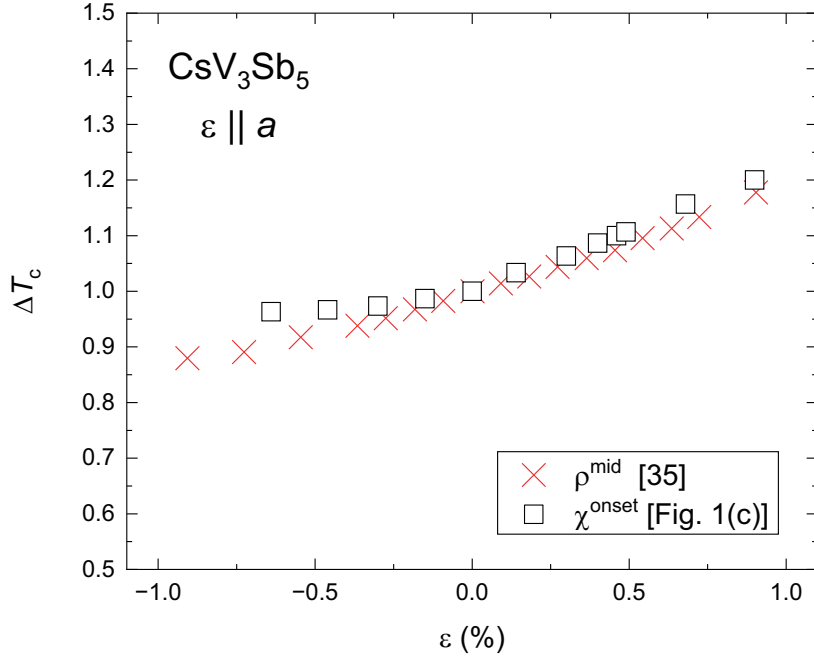


FIG. S1. **Strain dependence of the normalized superconducting transition temperature, $\Delta T_c = T_c(\varepsilon)/T_c(0)$, for $\varepsilon \parallel a$.** The open squares represent our data evaluated from the onset of the ac-susceptibility (χ^{onset}) taken from Fig. 1(c) of the main text. The red crosses represent the data extracted from the electrical resistivity measurements (ρ^{mid}) reported by Qian *et al.* [35].

S2. TEMPERATURE DEPENDENCE OF THE CDW ORDER PARAMETER UNDER UNIAXIAL STRAIN

To investigate the temperature dependence of the CDW state under uniaxial strain, we focused on the $^{121}\text{Sb}(1)$ -NQR spectrum. As clearly demonstrated by the zero-strain ($\varepsilon = 0$) data in Fig. S2(c), the resonance frequency of the Sb(1) site (solid diamonds) exhibits a massive shift below T_{CDW} compared to the almost temperature-independent behavior of the Sb(2) sites (blue circles). This stark contrast highlights that the Sb(1) site is exceptionally sensitive to the local CDW environment. Furthermore, as shown in Fig. S2(a), previous studies under hydrostatic pressure [17] have established a clear positive correlation between the CDW transition temperature, T_{CDW} , and the $^{121}\text{Sb}(1)$ -NQR resonance frequency. Due to this scaling and its high sensitivity, the Sb(1) resonance frequency serves as a robust, quantitative local probe for the CDW order parameter.

Figure S2(b) shows the $^{121}\text{Sb}(1)$ -NQR spectra measured at various temperatures up to 60 K under a constant tensile strain of $\varepsilon = +0.90\%$. As the temperature increases, the Sb(1) peak shifts to lower frequencies. The measurement under strain was limited to 60 K due to the reduced signal-to-noise (S/N) ratio inherent to the small single crystal required for the strain cell.

In Fig. S2(c), the temperature dependence of the evaluated Sb(1) resonance frequency under strain (red open stars) is plotted alongside the zero-strain data extracted from a previous study by Luo *et al.* [14]. The temperature evolution of the Sb(1)-NQR frequency under the $+0.90\%$ strain closely tracks the zero-strain curve. This coincidence over the measured temperature range firmly indicates that the CDW order parameter is virtually independent of the applied uniaxial strain.

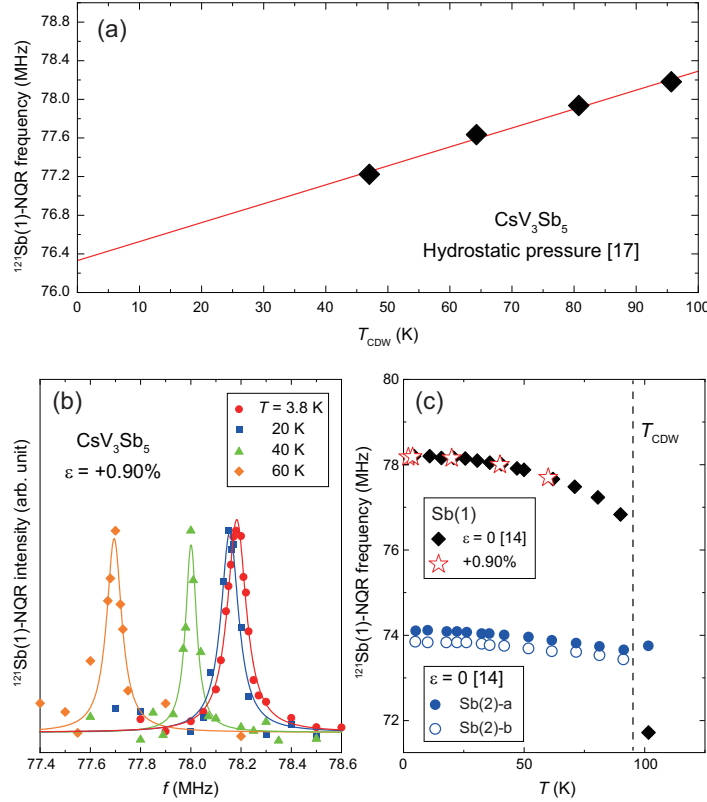


FIG. S2. **Temperature dependence of the CDW order parameter under a tensile strain of $\varepsilon = +0.90\%$.** (a) Positive correlation between the CDW transition temperature T_{CDW} and the $^{121}\text{Sb}(1)$ -NQR resonance frequency, extracted from a previous NQR study under hydrostatic pressure [17]. (b) $^{121}\text{Sb}(1)$ -NQR spectra measured at various temperatures between 3.8 K and 60 K under a tensile strain of $\varepsilon = +0.90\%$. The solid curves represent the best fits using a single Lorentzian function. (c) Temperature dependence of the NQR resonance frequencies. The red open stars denote our experimental data for the Sb(1) site at $\varepsilon = +0.90\%$. The black solid diamonds and blue circles represent the ambient pressure ($\varepsilon = 0$) data for the Sb(1) and Sb(2) sites, respectively, extracted from Luo *et al.* [14]. The massive shift of the Sb(1) frequency below T_{CDW} , in sharp contrast to the nearly temperature-independent behavior of the Sb(2) sites, highlights the exceptional sensitivity of the Sb(1) site to the local CDW environment. The excellent agreement between the strained and zero-strain Sb(1) data, together with the correlation shown in (a), indicates that T_{CDW} is effectively unaffected by the applied strain.

S3. HYSTERETIC BEHAVIOR OF THE NQR LINEWIDTH

To investigate the effect of uniaxial strain on the CDW state, we measured the strain history dependence of the $^{121}\text{Sb}(1)$ -NQR spectra.

Figures S3(a) and S3(b) show the $^{121}\text{Sb}(1)$ -NQR spectra at $T = 3.8$ K under different strain application sequences: compressive ($\varepsilon = -0.46\%$) and tensile ($\varepsilon = +0.90\%$), respectively. Upon cooling under zero strain, the CDW transition breaks the sixfold rotational symmetry of the kagome lattice, leading to the formation of twinned domains [38]. The presence of these twin boundaries naturally introduces a spatial distribution of the local electric field gradient (EFG), which broadens the NQR spectra in the virgin zero-strain state ($\varepsilon = 0$). When either compressive or tensile uniaxial strain is applied, the NQR peak sharpens. This reduction in the linewidth clearly indicates that the applied strain induces macroscopic alignment of the CDW domains (detwinning), thereby eliminating the twin boundaries and homogenizing the local EFG.

As summarized in Fig. S3(c), when the applied strain is subsequently released back to $\varepsilon = 0$, the full width at half maximum (FWHM) recorded immediately after the strain release does not revert to its initial value but maintains the reduced linewidth. This hysteretic behavior indicates that the detwinned, single-domain state is preserved at this low temperature even after the strain is removed. The distinct contrast between the initial and final $\varepsilon = 0$ spectra clearly demonstrates the metastable nature of this strain-induced detwinning at $T = 3.8$ K.

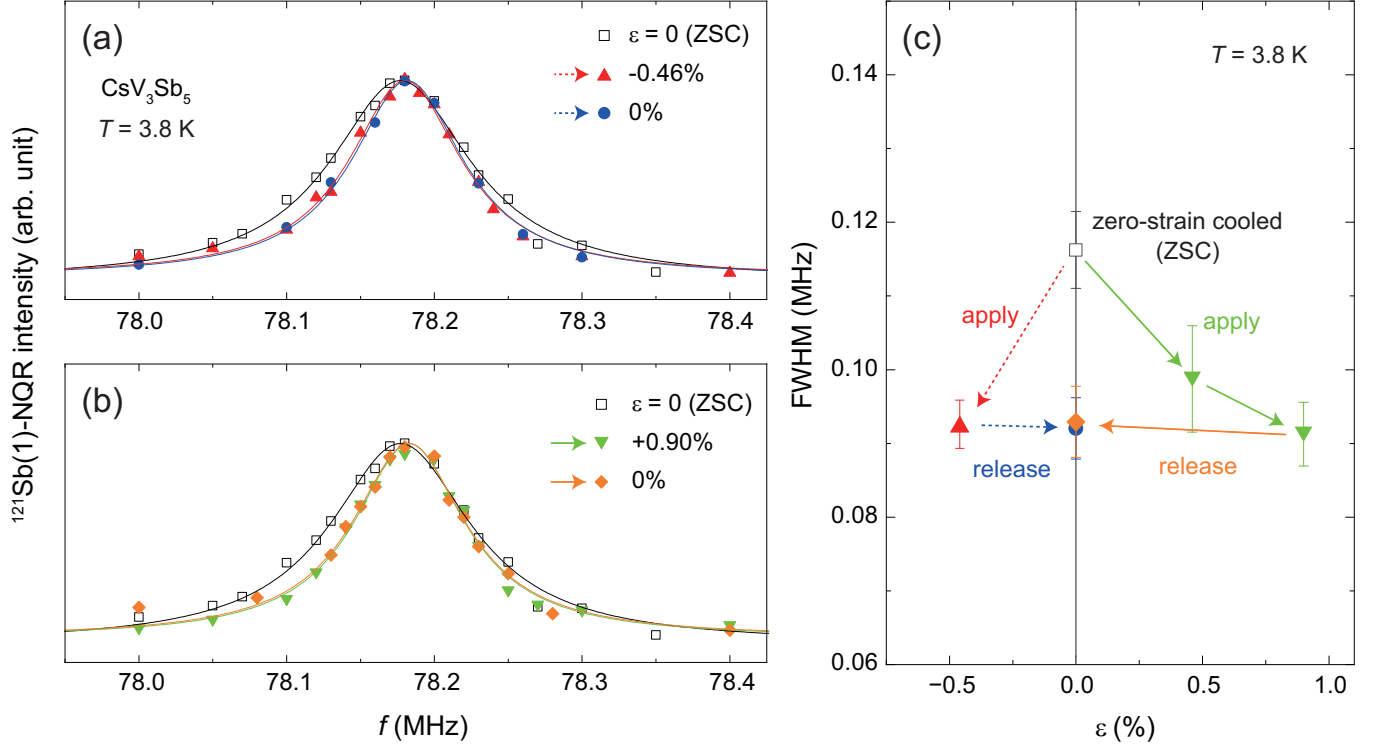


FIG. S3. **Strain history dependence of the $^{121}\text{Sb}(1)$ -NQR spectra in CsV_3Sb_5 at $T = 3.8$ K.** (a), (b) $^{121}\text{Sb}(1)$ -NQR spectra obtained during the (a) compressive and (b) tensile strain cycles. The initial measurements were performed in the zero-strain cooled (ZSC) state ($\varepsilon = 0$, open squares). Spectra were then recorded under applied strain (triangles) and immediately after releasing the strain back to $\varepsilon = 0$ (circles and diamonds). Solid lines represent the fitting curves. (c) FWHM of the $^{121}\text{Sb}(1)$ -NQR spectra as a function of the applied strain ε . The arrows denote the sequence of the strain application and release. The reduced FWHM is maintained even after the external strain is removed.

S4. SINGLE-COMPONENT NUCLEAR SPIN RELAXATION AND MACROSCOPIC HOMOGENEITY UNDER UNIAXIAL STRAIN

Figure S4(a) replots the temperature dependence of $1/T_1$ at $\varepsilon = +0.90\%$ and the total $s+d$ wave fitting curve (blue solid curve) presented in Fig. 4 of the main text. To explicitly show the contribution from each gap, the individual s -wave and d -wave components deduced from the two-component model are additionally plotted as black and green solid curves, respectively.

To critically verify the macroscopic homogeneity of the sample under uniaxial strain and the microscopic nature of the two superconducting transitions, we carefully examined the nuclear magnetization recovery curves across the relevant temperature ranges. Figures S4(b), S4(c), and S4(d) display the recovery curves measured at representative temperatures: $T = 3.8$ K in the normal state ($T > T_{c1} = 3.6$ K), $T = 3.2$ K in the intermediate state precisely between the two transitions ($T_{c2} = 3.0$ K $< T < T_{c1} = 3.6$ K), and $T = 2.0$ K in the fully superconducting state ($T < T_{c2} = 3.0$ K), respectively.

Crucially, at all measured temperatures, the recovery curves are excellently described by a single spin-lattice relaxation time T_1 (red solid curves). This single-component behavior is naturally expected in the normal state [Fig. S4(b)], where the density of states at the Fermi level, $N(E_F)$, is uniform. The strict persistence of this single-component relaxation at $T = 3.2$ K [Fig. S4(c)] and $T = 2.0$ K [Fig. S4(d)] provides profound physical implications.

If the observed double transition (T_{c1} and T_{c2}) were caused by macroscopic strain inhomogeneity or real-space phase separation (e.g., a spatial mixture of highly strained d -wave regions and weakly strained s -wave regions), the Sb nuclei in different domains would experience distinctly different electronic environments. This would inevitably result in a multi-component or stretched-exponential recovery curve below T_{c1} .

The unambiguous observation of a single-component relaxation at $T = 3.2$ K definitively rules out such macroscopic real-space phase separation. It demonstrates that every relevant Sb nucleus in the sample simultaneously probes the exact same total electronic environment. This perfectly supports our proposed scenario: the crystal is uniformly deformed, and the two distinct superconducting gaps reside on different band. In this multi-band picture, the total density of states felt by the nuclei is a uniform superposition of the distinct bands, $N_{\text{tot}}(E) = wN_d(E) + (1-w)N_s(E)$, naturally yielding the single T_1 component that precisely follows the theoretical $s+d$ wave fitting in Fig. S4(a).

Therefore, these relaxation dynamics provide compelling microscopic evidence that the remarkable two-stage superconducting transition under uniaxial strain is a genuine, macroscopically homogeneous bulk phenomenon.

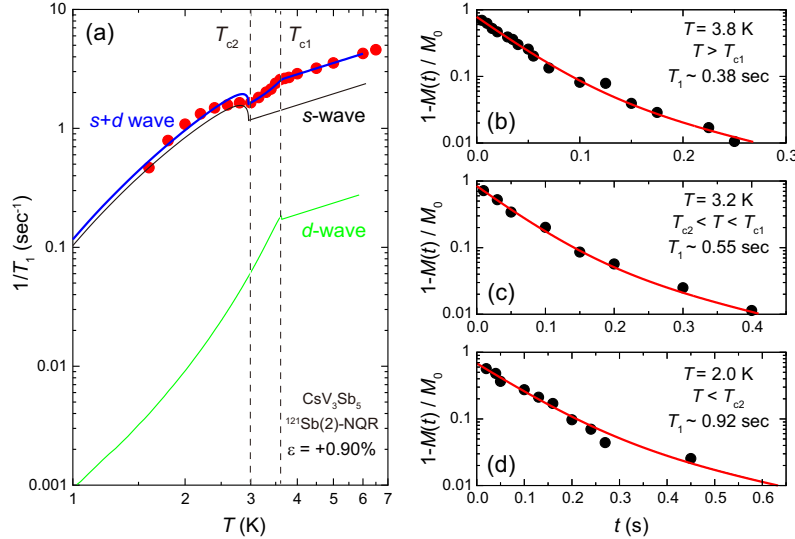


FIG. S4. **Nuclear magnetization recovery curves under uniaxial strain.** (a) Temperature dependence of $^{121}\text{Sb}(2)\text{-NQR}$ $1/T_1$ under a tensile strain of $\varepsilon = +0.90\%$. The blue solid curve represents the total $1/T_1$ calculated using the two-component ($s+d$ wave) model, replotted from Fig. 4 of the main text. The calculated individual contributions from the s -wave and d -wave components are shown as black and green solid curves, respectively. The red circles denote the experimentally obtained relaxation rates. The vertical dashed lines indicate the superconducting transition temperatures T_{c1} and T_{c2} . (b)-(d) Nuclear magnetization recovery curves measured at (b) $T = 3.8$ K ($T > T_{c1}$), (c) $T = 3.2$ K ($T_{c2} < T < T_{c1}$), and (d) $T = 2.0$ K ($T < T_{c2}$). The red solid curves represent strictly single-component fits using the standard theoretical recovery function for the $\pm 1/2 \leftrightarrow \pm 3/2$ NQR transition of a spin $I = 5/2$ nucleus (^{121}Sb) [34], $1-M(t)/M_0 = (3/28)\exp(-3t/T_1) + (25/28)\exp(-10t/T_1)$. These fits consistently reproduce the data across all temperature regions.

S5. REPRODUCIBILITY AND STRAIN-DIRECTION INDEPENDENCE OF THE T_c ENHANCEMENT

To verify the reproducibility and examine the effect of the in-plane strain direction, we performed additional measurements using another CsV_3Sb_5 single crystal from a different batch. In this setup, the uniaxial strain was applied perpendicular to the a -axis ($\varepsilon \perp a$).

Figure S5(a) displays the temperature dependence of the ac-susceptibility under various strains for $\varepsilon \perp a$. The superconducting transition shifts systematically with strain. In Fig. S5(b), we plot the strain dependence of T_c^{onset} for $\varepsilon \perp a$ (solid red triangles) alongside the $\varepsilon \parallel a$ data (open squares) extracted from Fig. 1(c) of the main text. The two datasets collapse onto a single curve, demonstrating that the strain-induced change in T_c is independent of the in-plane strain direction.

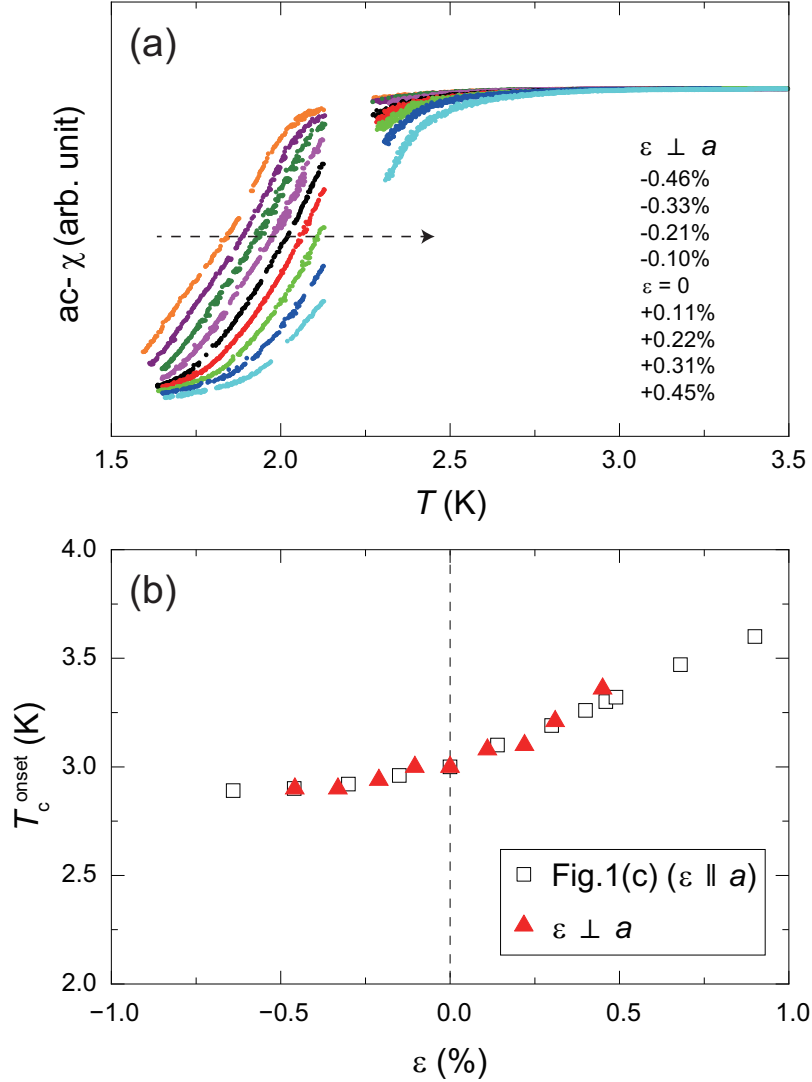


FIG. S5. **Reproducibility and strain-direction independence of the superconducting transition in CsV_3Sb_5 .** (a) Temperature dependence of the ac-susceptibility (ac- χ) under various uniaxial strains applied perpendicular to the a -axis ($\varepsilon \perp a$). (b) Strain dependence of T_c^{onset} . The solid red triangles represent the data for $\varepsilon \perp a$ obtained from panel (a), and the open squares denote the data for $\varepsilon \parallel a$ taken from Fig. 1(c) of the main text.