

Supplemental Material: Spin correlations in the nematic quantum disordered state of FeSe

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Characterization of the uniaxial strain device

The uniaxial strain device shown in Fig. 1 of the main text is designed based on differential thermal expansion between an invar alloy frame and an aluminum sheet with a narrow bridge in the center [Figs. 1 and S1]. To validate the application of uniaxial strain and the homogeneity of strain distribution, we performed strain characterization on a similar, prototype uniaxial strain device [Fig. S1(a)] that can fit the chamber of a Physical Property Measurement System (PPMS), enabling temperature-dependent resistivity measurements. The aluminum sheet is fixed onto the invar alloy frame and pre-stretched at one end using screws.

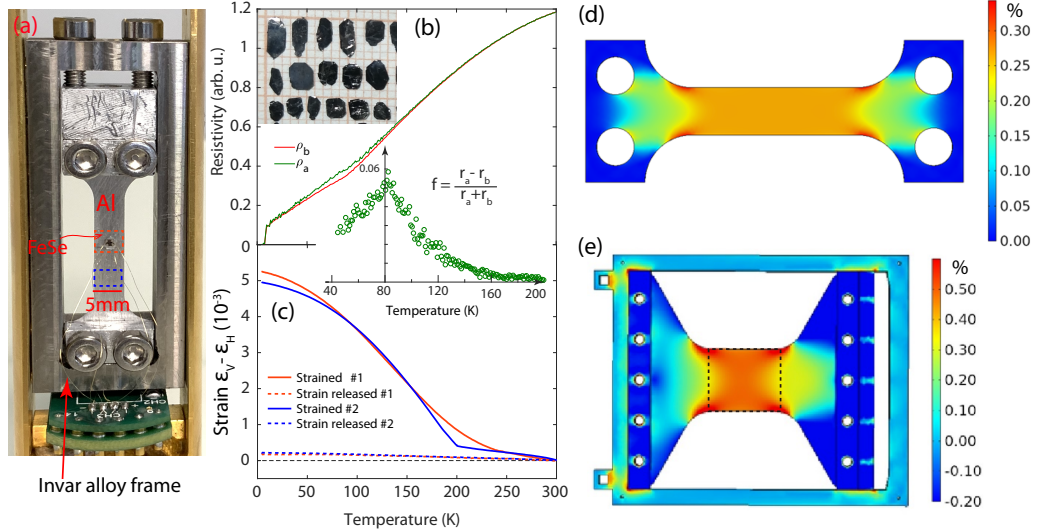


FIG. S1: (a) A prototype uniaxial strain device designed to evaluate the homogeneity of the applied strain and its effectiveness in detwinning FeSe single crystals. A thin FeSe crystal is glued onto the aluminum sheet for resistivity measurements. The red and blue dashed rectangles indicate the regions where anisotropic strain (vertical strain ε_V along the aluminum sheet, and horizontal strain ε_H perpendicular to it) was characterized using resistive strain gauges (the strain gauges are not shown here). (b) Anisotropic resistivity, ρ_a (green curve) and ρ_b (red curve), of the FeSe crystal shown in (a). The resistivity anisotropy, $\phi = (\rho_a - \rho_b) / (\rho_a + \rho_b)$, as shown in the middle inset, is comparable to that of a fully detwinned sample. The upper left inset displays a photograph of several FeSe single crystal pieces. (c) Anisotropic strain, $\varepsilon = \varepsilon_V - \varepsilon_H$, measured in the regions marked by the red and blue rectangles in (a). (d), (e) Finite element simulations of the uniaxial strain distribution for the device shown in (a) and the setup used in the INS experiment. The black dashed rectangle indicates the area where FeSe thin crystals were glued for the inelastic neutron scattering (INS) experiment.

To evaluate the distribution of uniaxial strain across the aluminum sheet, we measured the strains ε_H and ε_V simultaneously using a Tee-Rosette resistive strain gauge (part No. WK-05-062TT-350, Micro-Measurements) at two regions: one at the center of the bridge (red dashed rectangle) and the other near one end of the bridge (blue dashed rectangle). The results $\varepsilon = \varepsilon_V - \varepsilon_H$, shown as red and blue curves in Fig. S1(c), indicate that these regions exhibit similar temperature-dependent anisotropic strain, generated by the thermal expansion difference between the invar alloy frame and the aluminum sheet. This similarity demonstrates that the strain is homogeneous in the bridge region of the aluminum sheet. At low temperatures, ε reaches approximately 0.5% in these measurements. When the strain is released by loosening the screws, the anisotropic strain essentially disappears, leaving only a minor residual strain $\sim 0.02\%$ (blue and red dashed curves in Fig. S1(c)), likely due to slight plastic deformation of the aluminum sheet. These results confirm that the applied strain is predominantly elastic. The homogeneity of the strain in the bridge section is further validated by finite element simulations, as shown in Fig. S1(d).

Figure S1(b) presents the resistivity of a FeSe crystal glued to the aluminum sheet in Fig. S1(a). The tetragonal [110] direction of FeSe is aligned along the bridge, and the applied anisotropic strain aligns the orthorhombic a -axis along the vertical direction and the b -axis along the horizontal direction. This alignment is reflected in the resistivity measurements along the vertical and horizontal directions, which are consistent with the intrinsic resistivity along the orthorhombic a - and b -axes. The measured resistivity anisotropy ($\sim 0.06\%$) is also comparable to that reported for a fully detwinned sample, indicating that this configuration effectively detwins FeSe thin crystals.

For the uniaxial strain device used in our inelastic neutron scattering (INS) experiment, we also performed finite element simulations using COMSOL to verify the strain distribution. As shown in Fig. S1(e), the strain in the area where the FeSe crystals are glued (black dashed rectangle) is basically homogeneous. Some strain gradient at relative large scale does not affect detwinning as the FeSe single crystals are usually much smaller.

Detwinning efficiency

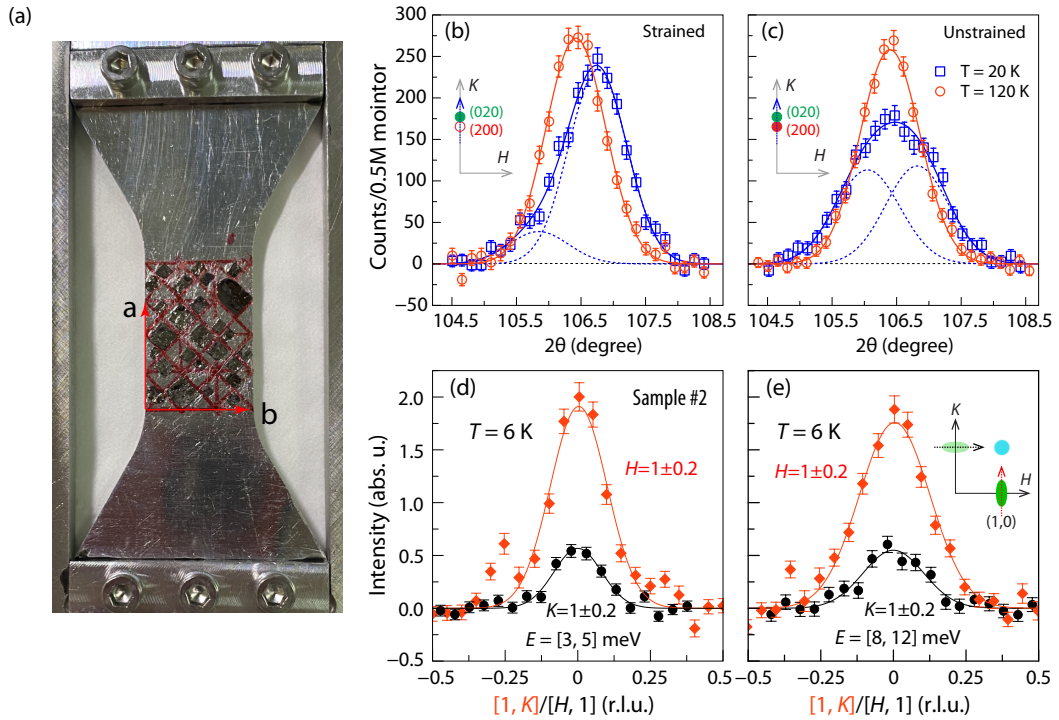


FIG. S2: (a) A uniaxial-strain device carrying 26 pieces of FeSe single crystals. (b), (c) $\theta - 2\theta$ scans of the nuclear (0, 2, 0) Bragg peak for strained (b) and unstrained (c) FeSe single crystals measured at temperatures (20 K and 120 K) well below and above the structural transition ($T_s = 90$ K). The solid and dashed curves are Gaussian fittings of the scans. (d), (e) Constant energy cuts of the spin excitations in uniaxially-strained Sample #2 across the (1, 0) and (0, 1) positions along the $[1, K]$ and $[H, 1]$ directions, with (d) $E = 4 \pm 1$ meV, and (e) 10 ± 2 meV. The data in (d) and (e) were collected on the 4SEASONS ToF spectrometer with $E_i = 21$ meV and 36 meV, respectively. The vertical bars indicate the statistical errors of one standard deviation.

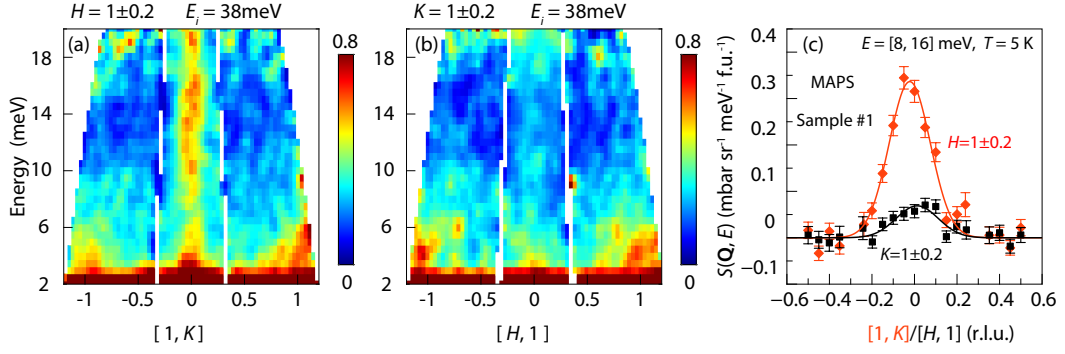


FIG. S3: (a), (b) Low energy spin excitations of sample #1 projected onto the $(\mathbf{Q}, E)$ plane with $\mathbf{Q}$ along the $[1, K]$ (a) and the $[H, 1]$ (b) directions. (c) Constant energy cuts of the spin excitations in sample #1 along the $[1, K]$ and $[H, 1]$ directions with $E = 12 \pm 4$ meV. The data were collected on the MAPS ToF spectrometer with $E_i = 38$ meV. The vertical bars indicate the statistical errors of one standard deviation.

To prove the effectiveness of the detwinning strategy and characterize the detwinning efficiency of the uniaxial-strain device, we glued ~ 30 pieces (~ 25 mg) of FeSe crystals using CYTOP on a single uniaxial-strain device [Fig. S2(a)] and performed neutron diffraction $\theta - 2\theta$ scans across the tetragonal (110) [orthorhombic (020)/(200)] nuclear Bragg peaks (Figs. S2(b) and S2(c)) on the Xingzhi cold neutron triple-axis spectrometer at the Chinese Advanced Research Reactor (CARR). During the measurements, the unstrained or strained states are achieved by loosening or tightening the screws at one end that can fix the aluminum sheets on the Invar alloy frame. For the unstrained case [Fig. S2(c)], the resolution limited tetragonal (110) peak at $T = 120$ K ($> T_s$) evolved to a broad peak consisting of orthorhombic (020) and (200) peaks because of the twinning effect. With the screws tightened slightly [Fig. S2(a)], the crystals were strained while cooling and detwinned across the T_s . The tetragonal (110) peak evolved to a strong orthorhombic (020) peak (integrated intensity $I_{(020)} \approx 264.9$), and a much weaker (200) peak (integrated intensity $I_{(200)} \approx 45.1$), indicating the samples are highly detwinned [$I_{(020)}/I_{(200)} \approx 5.9$] — 85.5% of the domains are aligned along one direction. The detwinning efficiency/ratio, defined as $\eta = \frac{I_{(020)} - I_{(200)}}{I_{(020)} + I_{(200)}}$, is $\approx 71\%$ for this sample set on a single device. This characterization demonstrates that such a detwinning strategy can provide a highly-detwinned sample set of FeSe for INS measurements.

For INS experiments, we have prepared two sample sets, Sample #1 (1.61 grams) and Sample #2 (1.24 grams). As the samples are too large to do neutron diffraction measurements on regular triple-axis spectrometers, and the time-of-flight (ToF) spectrometers we used do not have enough momentum resolution for accurate refinements, it is difficult to determine the detwinning efficiency for the whole sample set in Fig. 1(b) (in the main text) with neutron diffraction. We use the low-energy spin excitations at $(1, 0)$ and $(0, 1)$ measured at base temperature to estimate the detwinning ratio. Figure S2(d) and S2(e) are the constant-energy transverse cuts of the spin excitations across $(1, 0)$ and $(0, 1)$ with $E = 4 \pm 1$ meV, and 10 ± 2 meV, respectively. The data were collected on Sample #2 at the 4SEASONS ToF spectrometer with $T = 6$ K. The ratio between the excitation intensity at $(1, 0)$ (I_{10}) and $(0, 1)$ (I_{01}) is $I_{10}/I_{01} = 3.79$ for $E = [3, 5]$ meV and 3.74 for $E = [8, 12]$ meV, resulting in a detwinning ratio $\eta = [I_{10} - I_{01}]/[I_{10} + I_{01}] \approx 58\%$ — 79% of the domains are aligned along one direction. The same estimate on the low-energy spin excitations of Sample #1 (Fig. S3) gives a similar detwinning ratio $\eta \approx 60\%$.

Strain transfer, partial detwinning, and additional strain effects

The nominal strain applied onto the aluminum bridge are typically $\varepsilon \approx 0.5\% - 0.7\%$, which is enough for detwinning FeSe single crystal. However, we argue that, 100% detwinning could not be achieved when applying strain using substrate. Here we explain why we have only partial detwinning in neutron diffraction [Fig. S2(a)-(c)] and INS measurements [Fig. S2(d)-(e) and S3]. The orthorhombic lattice distortion in FeSe, defined as $\delta = (a - b)/(a + b)$, is $\sim 0.27\%$ at $T \ll T_s$ measured by high-resolution XRD [1]. Note that the definition of the anisotropic strain in our study is $\varepsilon = \varepsilon_V - \varepsilon_H = (a - a_0)/a_0 - (b - b_0)/b_0 \approx 2(a - b)/(a_0 + b_0) \approx 2\delta$. Thus, the orthorhombicity of FeSe corresponds to anisotropic strain $\varepsilon \approx 2\delta \approx 0.54\%$, which is still within the range of the applied strain in our experiment.

The discrepancy between large nominal strain and partial detwinning ($\eta = 71\%$ for neutron diffraction and 58% for INS) arises due to several factors related to the geometry and physical properties of the FeSe crystals, as well as the mechanics of strain transfer.

Firstly, while our uniaxial strain device is capable of applying a large nominal strain, the actual strain experienced by the sample is reduced due to the load transfer mechanism and strain relaxation effects. On the verge of the FeSe thin crystals,

there is a "transfer length" within which the applied strain cannot be fully transferred to the sample [2, 3]. This transfer length depends on the stiffness and bonding properties of CYTOP used to mount the sample, as well as the aspect ratio of the crystal. Therefore, the strain near the edges of the sample is lower than the nominal applied strain, reducing the effective detwinning ratio. Secondly, resistivity measurements suggest that thinner samples ($\sim 20 \mu\text{m}$) achieve better strain transfer due to reduced strain relaxation effects [4]. However, our FeSe crystals were typically $\sim 50 \mu\text{m}$ thick to ensure sufficient sample volume for neutron scattering. In thicker samples, strain relaxation occurs across the thickness, resulting in non-uniform strain distribution and further reducing the effective strain transferred to the entire sample.

In our neutron diffraction measurements, we selected large, thin, and flat crystals and achieved a higher detwinning ratio $\eta = 71\%$ (e.g., 85.5% domains co-aligned along one direction). For INS experiment, we used more than 1000 pieces of crystals. Some of them are smaller or thicker than those in the neutron diffraction measurements. In this case, a compromise between sample volume and detwinning ratio was achieved — a relative smaller detwinning ratio $\eta = 58 - 60\%$ (e.g., $\sim 79 - 80\%$ domains co-aligned along one direction) for $m = 1.24 - 1.61$ grams.

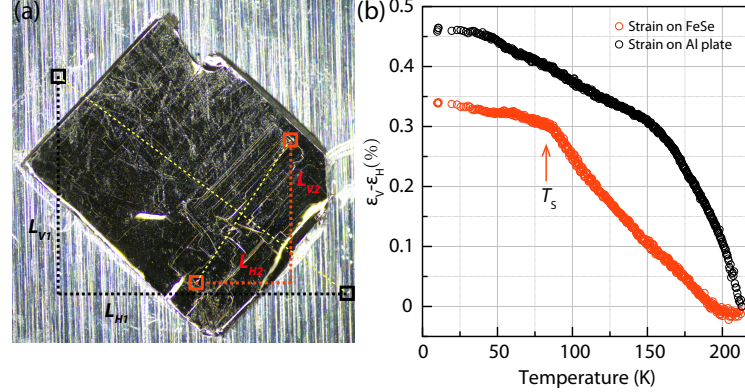


FIG. S4: (a) A photo of a FeSe crystal glued on an aluminum sheet for strain characterization while cooling. The aluminum sheet was installed on an invar alloy frame and was pre-scratched for strain application. The black and red squares are feature spots used to track the relative strains on aluminum bridge and the FeSe crystal, respectively. (L_{H1}, L_{V1}) and (L_{H2}, L_{V2}) represent the distances between the feature spots. (b) Temperature dependent anisotropic strain ϵ on aluminum (black circles) and on the surface of FeSe crystal (red circles), calculated from the relative changes of the displacements between the feature points. To show the $\epsilon(T)$ across T_s clearly, the measurements were performed from $T \approx 200$ K to $T \approx 10$ K. The red arrow marks the structural transition $T_s \approx 90$ K.

Since a strain gradient is present near the edges of the FeSe crystals, and the applied anisotropic strain ϵ does not fully correspond to the orthorhombic lattice distortion, it is essential to clarify any additional strain effects beyond detwinning. Previous studies on underdoped BaFe_2As_2 have shown that the Young's modulus along the tetragonal $[110]$ direction ($Y_{[110]} \approx C_{66}$) decreases monotonically while cooling toward T_s . This reflects the structural instability associated with nematic fluctuations in the fluctuating regime above T_s . Around T_s , a spontaneous orthorhombic lattice distortion δ appears without the application of external pressure P . However, due to the twinning effect in most crystals, the macroscopic relative changes in sample dimensions $\Delta L/L$ across T_s are much smaller than the spontaneous distortion.

When a small pressure $P \sim 5$ MPa is applied along the tetragonal $[110]$ direction, the twin domains are detwinned, aligning the orthorhombic b -axis of the twin domains along the pressure direction. This is accompanied by a relative change in strain, $\epsilon_b = \Delta L_b/L_b$, which consists of two components: the spontaneous distortion ϵ_{b0} , generated by the structural transition, and the intrinsic strain on the orthorhombic b -axis, $\epsilon_{b'}$. Using the definition of the Young's modulus, we have $Y_{[110]} = Y_b = \frac{P}{\epsilon_{b0} + \epsilon_{b'}}$, where ϵ_{b0} remains constant and $\epsilon_{b'}$ depends on the applied pressure. In underdoped BaFe_2As_2 , previous studies have demonstrated that $P \sim 20$ MPa induces significant lattice distortions in the nematic fluctuating regime. However, these distortions become negligible ($\epsilon_{b'} \sim 0.02\% - 0.05\%$) at lower temperatures, indicating that the intrinsic Young's modulus of the orthorhombic lattice approaches the value of $Y_{[110]}$ at $T \sim 300$ K [5].

In FeSe, the situation is similar. While the applied anisotropic strain in our experiments detwins the sample, the magnitude of the strain-induced lattice distortions beyond detwinning is much smaller, particularly at low temperatures where the structural instability is suppressed. As shown in Fig. S4, the strain on the aluminum sheet is constantly larger than that on the FeSe crystal indicating the strain is to some extent relaxed. While the strain on aluminum increase continuously while cooling, ϵ on FeSe increases quickly toward T_s but slow down across T_s , indicating additional strain beyond detwinning is much smaller.

The absence of significant strain-induced effects is further supported by our INS measurements, which show no deviations in the spin excitation dispersion compared to previous studies on twinned FeSe crystals. These results confirm that the strain effects at $T < T_s$ in our experiments are primarily limited to detwinning, without introducing additional artifacts or altering the

intrinsic magnetic properties of FeSe.

Background subtraction and the significance of the clear spin-excitation spectrum

The uniaxial-strain device for INS experiments contains several invar-alloy frames and aluminum sheets. To reduce possible background scattering from invar alloy, the frames were wrapped in cadmium foil, leaving the samples glued on the middle part of the aluminum sheets ($22 \text{ mm} \times 20 \text{ mm} \times 2 \text{ sides} \times 5 \text{ sheets}$) open to the beam. On SEQUOIA, we measured the neutron scattering signal of an empty strain device (containing CYTOP) with the same conditions as the measurements of the sample set #1 ($T = 5, 80, 105, 120, 150 \text{ K}$ and $E_i = 147.5 \text{ meV}$). The background scattering signals from the empty device were subtracted from the raw data of the sample to obtain clean magnetic scattering signals, as shown in Fig. S5. A $|\mathbf{Q}|$ -dependent background was subtracted from the data measured on 4SEASONS with $E_i = 21, 36$ and 80 meV and on MAPS with $E_i = 38$ and 80 meV , which was estimated according to the magnetic excitations signals at $\mathbf{Q} = (1, 0)$, $(0, 1)$, and $(1, 1)$. This method has been described [6]. Energy-dependent (and $\mathbf{Q}$ -independent) flat backgrounds were subtracted from the data measured with $E_i = 295 \text{ meV}$ collected on 4SEASONS.

The raw data of two-dimensional energy-vs-momentum slices across $\mathbf{Q} = (1, 0)$, $(0, 1)$ and $(1, 1)$ and constant-energy magnetic excitations ($E_T = 55 \pm 7 \text{ meV}$) collected on SEQUOIA with $E_i = 147.5 \text{ meV}$ at $T = 5 \text{ K}$ are shown in Fig. S5 (a)-(e). The corresponding neutron scattering signals of the empty strain device with CYTOP are shown in Fig. S5 (f)-(j). The scattering signals at $E \lesssim 40 \text{ meV}$ and $E \approx 90 \text{ meV}$ are contaminated by the signal from aluminum sheets and CYTOP. Background-subtracted two-dimensional slices display much clearer spin-excitation dispersions below $E = 100 \text{ meV}$ at $T = 5 \text{ K}$ in Fig. S5 (k)-(m) than previously reported [6]. The spin excitations with $E_T = 55 \pm 7 \text{ meV}$ in the $[H, K]$ plane, confirming again the C_2 and C_4 symmetry of the stripe and the Néel excitations in Figure S5(o).

Previous studies on the spin-excitation spectrum of FeSe [6] revealed a broad excitation continuum spanning $\sim 70 - 160 \text{ meV}$ around $(1, 1)$, which was subsequently used as a key reference for developing theoretical models of the magnetic ground state in FeSe [7–9]. However, the lack of clarity in these measurements raised questions about the intrinsic nature of the observed excitations and their connection to the underlying spin interactions. In contrast, our new results, obtained with a significantly improved experimental setup, reveal a much sharper and more detailed spin-excitation spectrum. These results disprove the existence of a high-energy excitation continuum at $(1, 1)$ and challenge the validity of previous theoretical models that were built around such features.

The background subtracted, clear spin-excitation spectrum obtained in detwinned FeSe not only provides insight into the magnetic dynamics of FeSe but also serves as a benchmark for developing more accurate theoretical models in this system. The high resolution and precision of the experimental data enable a detailed comparison with theoretical predictions, allowing researchers to refine existing models and better capture the interplay between nematicity, magnetism, and superconductivity in FeSe. By resolving the intrinsic magnetic excitations with unprecedented clarity, this work lays the groundwork for a deeper theoretical understanding of the complex electronic and spin interactions in iron-based superconductors.

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- [1] Carr, S. V. Electron doping evolution of the magnetic excitations in $\text{NaFe}_{1-x}\text{Co}_x\text{As}$. *Phys. Rev. B* **93**, 214506 (2016).
- [2] C. W. Hicks, M. E. Barber, S. D. Edkins, D. O. Brodsky, and A. P. Mackenzie, Piezoelectric-based apparatus for strain tuning, *Rev. Sci. Instrum.* **85**, 065003 (2014)
- [3] J. Park, J. M. Bartlett, H. M. L. Noad, A. L. Stern, M. König, S. Hosoi, T. Shibauchi, A. P. Mackenzie, A. Steppke, and C. W. Hicks, Rigid platform for applying large tunable strains to mechanically delicate samples, *Rev. Sci. Instrum.* **91**, 083902 (2020)
- [4] J. M. Bartlett, A. Steppke, S. Hosoi, H. Noad, J. Park, C. Timm, T. Shibauchi, A. P. Mackenzie, and C. W. Hicks, Relationship between Transport Anisotropy and Nematicity in FeSe, *Phys. Rev. X* **11**, 021038 (2021)
- [5] Lu, X. et al. Impact of uniaxial pressure on structural and magnetic phase transitions in electron-doped iron pnictides. *Phys. Rev. B* **93**, 134519 (2016).
- [6] Wang, Q. et al. Magnetic ground state of FeSe. *Nat. Commun.* **7**, 12182 (2016).
- [7] A. Kreisel, B. M. Andersen, and P. J. Hirschfeld, Itinerant approach to magnetic neutron scattering of FeSe: Effect of orbital selectivity, *Phys. Rev. B* **98**, 214518 (2018)
- [8] A. Kreisel, P. J. Hirschfeld, and B. M. Andersen, Theory of spin-excitation anisotropy in the nematic phase of FeSe obtained from RIXS measurements, *Front. Phys.* **859**, 424 (2022).
- [9] J.-H. She, M. J. Lawler, and E.-A. Kim, Quantum Spin Liquid Intertwining Nematic and Superconducting Order in FeSe, *Phys. Rev. Lett.* **121**, 237002 (2018)

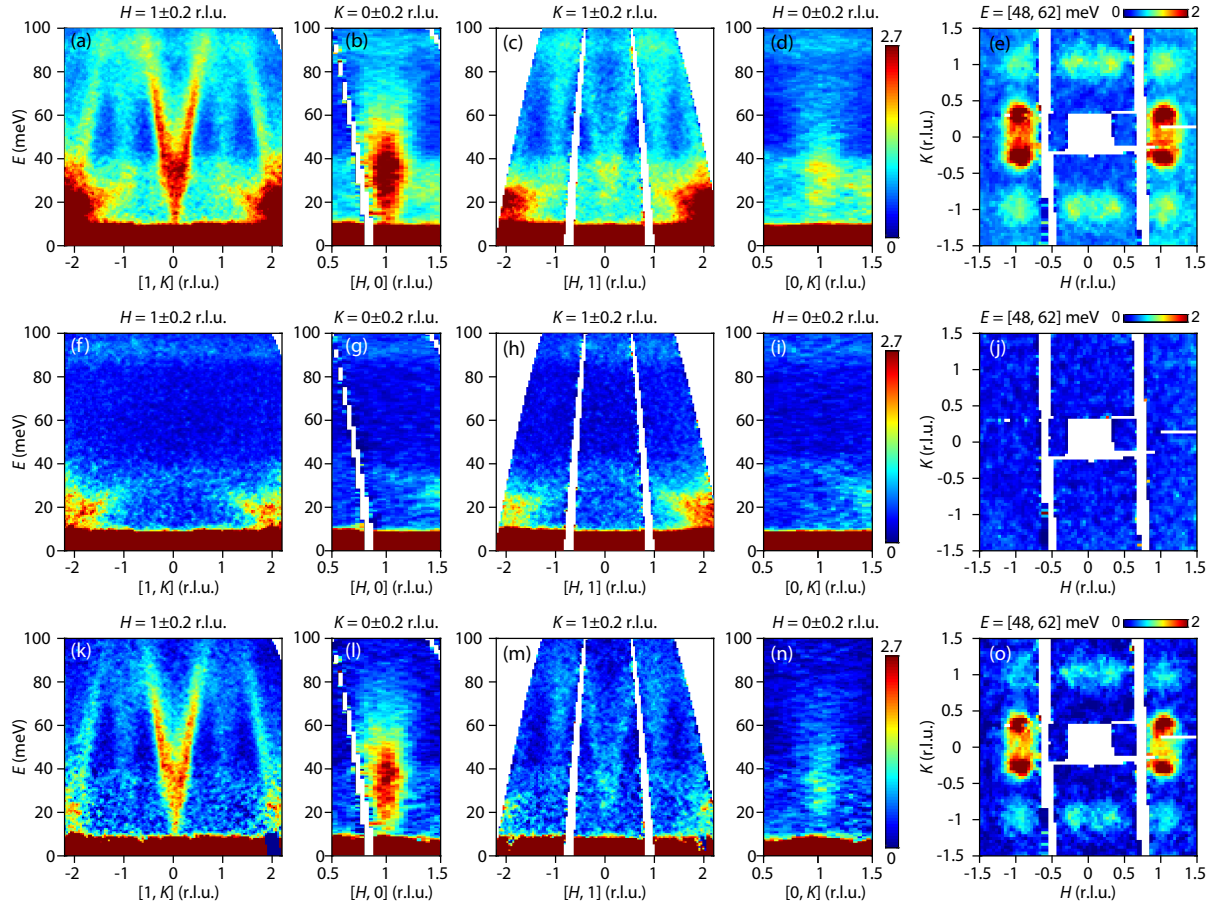


FIG. S5: **Two-dimensional energy-vs-momentum slices across $Q = (1, 0)$, $(0, 1)$, and $(1, 1)$.** The results were measured on Sample #1 with $E_i = 147.5$ meV at $T = 5$ K of raw data (a)-(d), empty strain device with CYTOP as the background signal (f)-(i) and background-subtracted data (k)-(n). Constant-energy slices with $E_T = 55 \pm 7$ meV at $T = 5$ K of raw data (e), empty strain device with CYTOP as the background signal (j) and background-subtracted data (o).

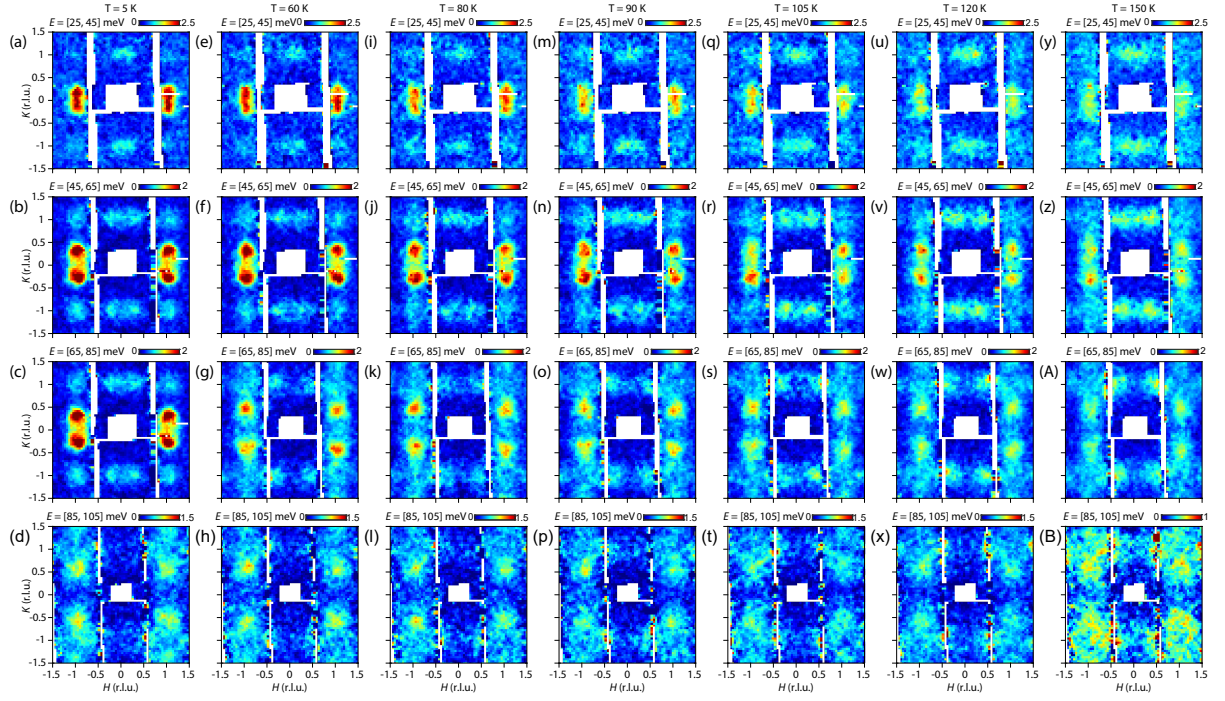


FIG. S6: **Temperature dependence of the spin excitations in detwinned FeSe.** (a)-(z), (A), (B) The constant energy magnetic scattering intensity maps in $[H, K]$ plane measured with $E_i = 147.5$ meV at $T = 5$ K (a-d), $T = 60$ K (e-h), $T = 80$ K (i-l), $T = 90$ K (m-p), $T = 105$ K (q-t), $T = 120$ K (u-x), $T = 150$ K (y-B). INS signals of an empty strain device with CYTOP measured at various temperatures have been subtracted from the data. The stripe spin excitation anisotropy gradually decreases with temperature increases. It persists to a higher temperature at $T = 150$ K far above $T_s = 90$ K.

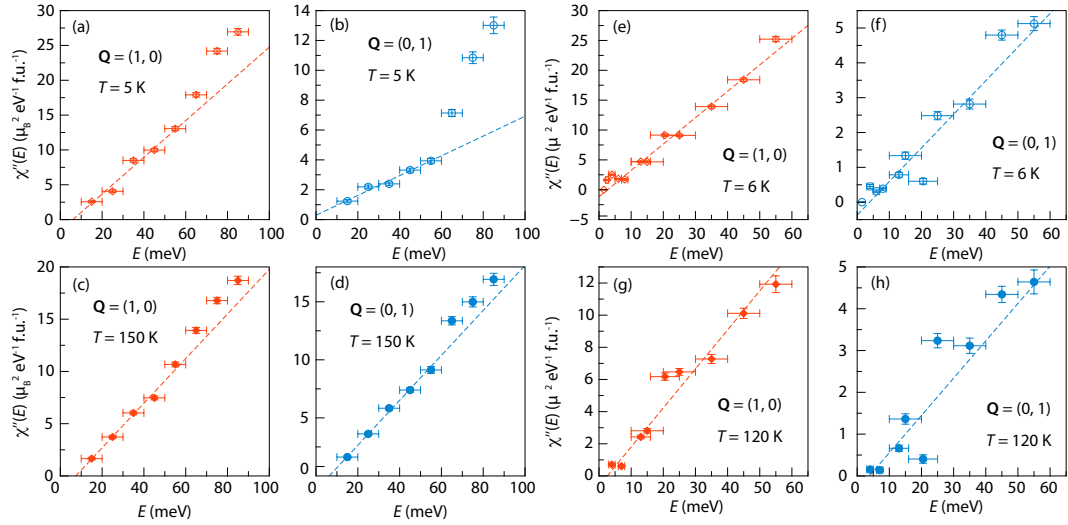


FIG. S7: **The linear energy dependence of the low-energy local susceptibility $\chi''(E)$.** The results (a)-(d) were measured on Sample #1 at the SEQUOIA spectrometer and (e)-(h) on Sample #2 at the 4SEASONS spectrometer, respectively. The linear-in- E spectral weight of the low-energy spin excitations in FeSe is a prediction from the antiferroquadrupolar (AFQ) regime. In the local moment model with AFQ order, the linear-in-energy behavior $\chi''(E) \propto E$ is a result of the coherent factor associated with localized spin excitations. In contrast, such a behavior is absent in BaFe_2As_2 with stripe order ($\langle m \rangle \approx 0.87\mu_B$) in which the spin waves open a $E \sim 10 - 15$ meV energy gap and carrier substantial spectral weight at low-energy range, resulting in a non-linear $\chi''(E)$ at $E \lesssim 80$ meV. In NaFeAs where stripe order is weak ($\langle m \rangle \approx 0.17\mu_B$), the spin wave energy gap remains ($E \approx 10$ meV). The spectral weight $\chi''(E)$, however, is close to have this linear-in-energy behavior. This could be attributed to that NaFeAs is close to FeSe (and the AFQ phase) in the spin-interaction phase diagram. The horizontal and vertical bars indicate the energy integration range, and the statistical errors of 1 s.d., respectively.

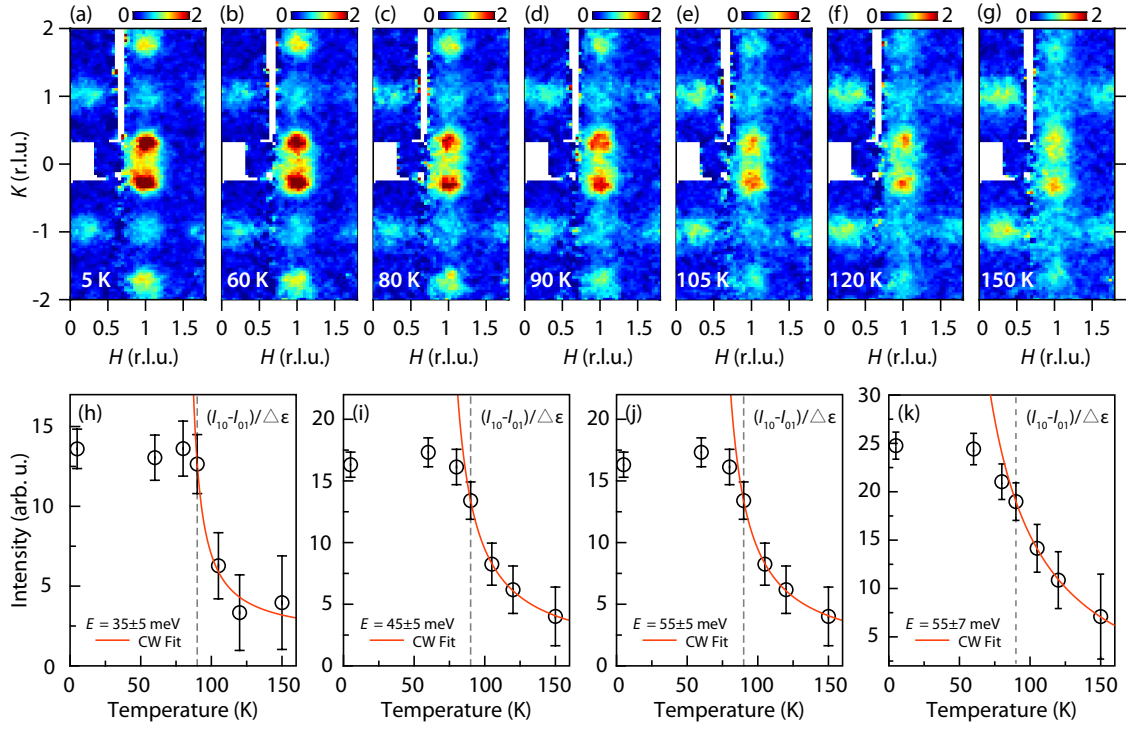


FIG. S8: Temperature dependence of the magnetic excitations in detwinned FeSe. Constant energy slices of the magnetic excitations for detwinned FeSe single crystals with $E = 55 \pm 7$ meV at $T = 5, 60, 80, 90, 105, 120$ and 150 K. The background scattering collected from an empty strain device with CYTOP has been subtracted from the data (a)-(g). Theoretically (ref. [3] in the main text), the spin-excitation difference between $(1, 0)$ and $(0, 1)$, termed the nematic spin correlation, reflects electronic nematicity in the spin-spin correlation channel. In the nematic fluctuation regime at $T > T_s$, it could serve as a measure of nematic susceptibility. (h)-(k) Strain normalized nematic spin correlation $\Delta\psi(E, T)/\Delta\epsilon = (I_{10} - I_{01})/\Delta\epsilon$ with $E = 35 \pm 5, 45 \pm 5, 55 \pm 5$, and 55 ± 7 meV. The red curves are Curie-Weiss fittings of the data at $T = 90 - 150$ K with $\Delta\psi(E)/\Delta\epsilon = \lambda/[a_0(T - T^*) + b_0] + \chi_0$. The vertical dashed lines in (h)-(k) mark the unstrained $T_s = 90$ K. Note that the applied strains are temperature-dependent (Fig. S4), ranging from $\epsilon \approx 0.2\% - 0.4\%$, which may exceed the linear regime necessary for rigorous nematic susceptibility measurements. Thus, the fitting and analysis of nematic susceptibility in the spin-spin correlation channel in (h)-(k) are not precise. Nonetheless, the results indicate the establishment of a nematic quantum disordered state. As the energy scale of the nematic spin correlations in (h)-(k) is $\lesssim 50$ meV—similar to the energy scale of orbital ordering in FeSe—and the spin and orbital degrees of freedom are entangled in the low-energy range, the results in (h)-(k) do not provide definitive evidence for spin-driven electronic nematicity in FeSe. The vertical bars indicate the statistical errors of one standard deviation.

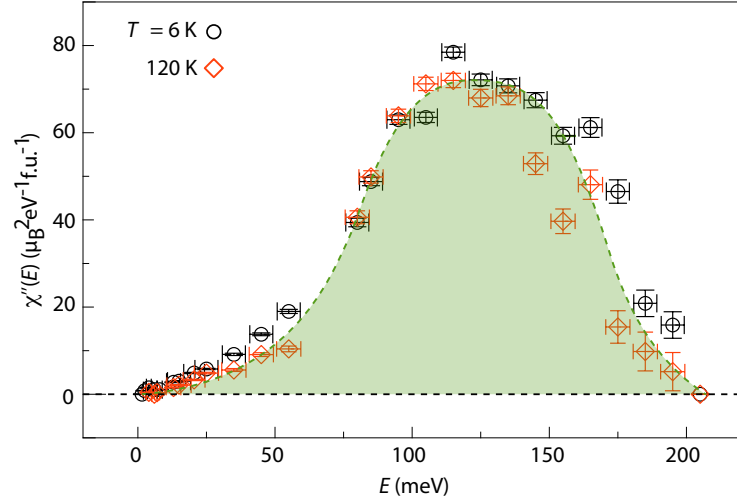


FIG. S9: Energy dependence of local dynamic susceptibility $\chi''(E)$ for detwinned FeSe single crystals at $T = 6$ K (black open circles) and 120 K (red open diamond). The horizontal and vertical error bars indicate the energy integration range for calculating $\chi''(E)$ and the statistical errors of one standard deviation. The data was collected on 4SEASONS with $E_i = 21, 36, 80$ and 295 meV). The peak intensity of $\chi''(E)$ here is $\sim 30\%$ higher than that measured in the twinned sample (ref. [20] in the main text). This difference arises due to several factors. First, in INS experiments, converting the scattering signal into absolute intensity units using a vanadium standard can introduce systematic errors of up to 20% – 30%. These errors stem from differences in neutron illumination between the vanadium and the sample, as well as systematic uncertainties in time-of-flight instruments (see, e.g., ref. [13] in the main text). Second, for the present study, we used selectively chosen thin and flat single crystals with minimal impurities to ensure a high detwinning efficiency. In contrast, the twinned samples used in Ref. [20] were bulkier and potentially contained more impurities, which could slightly suppress the measured intensity. The horizontal and vertical bars indicate the energy integration range, and the statistical errors of 1 s.d., respectively.

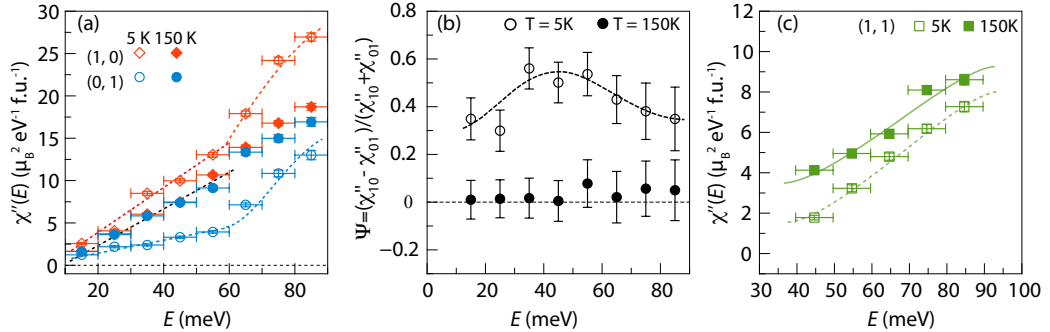


FIG. S10: **Energy-dependent local dynamic susceptibility $\chi''(E)$ and nematic spin correlations for detwinned FeSe single crystals.** (a) Comparison of $\chi''(E)$ at $\mathbf{Q} = (1,0)$ and $(0,1)$, measured at $T = 5$ K (open symbols) and 150 K (filled symbols). The data is corrected with the magnetic form factor and Bose factor. (b) Nematic spin correlations $\Psi(E) = [\chi''_{10}(E) - \chi''_{01}(E)] / [\chi''_{10}(E) + \chi''_{01}(E)]$ calculated from the data shown in (a). (c), Local dynamic susceptibility $\chi''(E)$ of the Néel spin excitations measured at $T = 5$ K and 150 K. The $\chi''(E)$ is slightly enhanced at $T = 150$ K compared with that at $T = 5$ K. The solid and dashed curves are guides to the eyes. With $\sim 80\%$ of the domains co-aligned along one direction, we obtained the clear spin-excitation spectrum as shown in the main text. Regarding the contribution of the $\sim 20\%$ twin domains along the other direction, we note that the spin excitations at $(0,1)$ exhibit a similar energy dispersion [Fig. 2(c)] to those at $(1,0)$, which suggests that the signal at $(0,1)$ primarily arises from the residual contribution of the minor domains. Consequently, due to the presence of these minor domains, we cannot definitively determine whether intrinsic spin excitations exist at $(0,1)$ or whether the excitations at $(0,1)$ are largely gapped. For this reason, we have refrained from emphasizing conclusions about the spin excitations at $(0,1)$ in the manuscript. The energy dependence of the spin-excitation anisotropy in (b), which does not decrease gradually at $E \lesssim 80$ meV, could mean that the intrinsic excitations at $(0,1)$ are weak and could be gapped. The horizontal and vertical bars indicate the energy integration range, and the statistical errors of 1 s.d., respectively.