

# Anisotropic spin fluctuations in detwinned FeSe

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Superconductivity in FeSe emerges from a nematic phase that breaks four-fold rotational symmetry in the iron plane. This phase may arise from orbital ordering, spin fluctuations, or hidden magnetic quadrupolar order. Here we use inelastic neutron scattering on a mosaic of single crystals of FeSe detwinned by mounting on a BaFe<sub>2</sub>As<sub>2</sub> substrate to demonstrate that spin excitations are most intense at the antiferromagnetic wave vectors  $\mathbf{Q}_{\text{AF}} = (\pm 1, 0)$  at low energies  $E = 611$  meV in the normal state. This two-fold ( $C_2$ ) anisotropy is reduced at lower energies 3-5 meV, indicating a gapped four-fold ( $C_4$ ) mode. In the superconducting state, however, the strong nematic anisotropy is again reflected in the spin resonance ( $E = 3.7$  meV) at  $\mathbf{Q}_{\text{AF}}$  with incommensurate scattering around 5-6 meV. Our results highlight the extreme electronic anisotropy of the nematic phase of FeSe and are consistent with a highly anisotropic superconducting gap driven by spin fluctuations.

High-transition temperature superconductivity in copper and iron based materials emerges from their antiferromagnetic (AF) ordered nonsuperconducting parent compounds [1]. While the parents of copper oxide superconductors are Mott insulators with a simple checkerboard AF structure [1], most iron pnictide parent materials exhibit a tetragonal-to-orthorhombic structural transition at  $T_s$  ( $< 295$  K) and form twin-domains before ordering antiferromagnetically at  $T_N$  ( $T_s \geq T_N$ ) [2]. Therefore, one must detwin iron pnictides in order to measure their intrinsic electronic properties below  $T_s$ . By applying a uniaxial pressure along one-axis of the orthorhombic lattice to detwin the sample, an in-plane resistivity anisotropy has been observed in strained iron pnictides BaFe<sub>2-x</sub>T<sub>x</sub>As<sub>2</sub> (where  $T$  is Co or Ni) above  $T_s$  [3, 4]. The resistivity anisotropy has been ascribed to an electronic nematic phase that spontaneously breaks the rotational symmetry while preserving the translation symmetry of the underlying lattice and is established in the temperature regime below  $T_s$  and above  $T_N$  [5, 6]. Below  $T_N$ , the AF structure is collinear, consisting of columns of antiparallel spins along the orthorhombic  $a_o$  axis and parallel spins along the  $b_o$  axis with an in-plane AF ordering wave vector  $\mathbf{Q}_{\text{AF}} = (\pm 1, 0)$  in reciprocal space [2].

The highly unusual iron-based superconductor FeSe exhibits an orthorhombic structural distortion and superconductivity without static AF order [Fig. 1(a)] [7–9]. Although the nematic phase in FeSe is established below  $T_s$  ( $\approx 90$  K) [8], it has been argued that nematic order and superconductivity are induced by orbital fluctuations [Fig. 1(b)] [10–14], forming a sign-preserving  $s^{++}$ -wave electron pairing and therefore would be fundamentally different from other iron-based superconductors [15]. Alternatively, the absence of static AF order in FeSe has been interpreted as evidence for a quantum paramagnet arising from the  $d$ -orbital spin-1 localized iron moments [16, 17]. Here, the nematic phase is driven by magnetic frustration due to competition between low-energy spin fluctuations associated with AF collinear order and those associated with various types of staggered order [18]. Third, the nematic superconductivity in FeSe without AF order may arise from a frustration-induced nematic quantum spin liquid state with melted AF order [19]. This model predicted a dramatic suppression of the magnetic spectral weight at  $\mathbf{Q} = (0, \pm 1)$  in a detwinned sample, and explained the observed superconducting gap anisotropy by angle resolved photo-emission spectroscopy (ARPES) [20–22] and scanning tunneling microscopy (STM) [23–25] experiments by an orbital dependent Hund's coupling [19]. Forth, the nematic order may arise from a hidden magnetic quadrupolar order [26, 27]. Finally, the nematic phase and superconductivity in FeSe has also been described by itinerant electrons interacting among quasi-nested hole-electron Fermi surfaces [28, 29], as in other iron based superconductors [30]. In this picture,

the electronic correlation effect is taken into account by orbital-dependent quasiparticle weights [24, 31]. Without electron correlation effects, spin fluctuations in the nematic phase below  $T_s$  exhibit only a minor  $C_4$  asymmetry. Including correlations in the theoretical calculations render the spin fluctuations highly  $C_2$  symmetric with negligible weight at  $(0, \pm 1)$ , and a neutron spin resonance exhibited only at  $\mathbf{Q}_{AF} = (\pm 1, 0)$  driven by the  $d_{yz}$  orbitals [31]. Approaches based on localized models with magnetic quadrupolar order have also predicted a strong suppression of low-energy  $(0, \pm 1)$  intensity [27].

In recent inelastic neutron scattering (INS) experiments on twinned FeSe [18, 32, 33], well-defined low-energy ( $E < 15$  meV) spin fluctuations are found at  $\mathbf{Q}_{AF} = (\pm 1, 0)$  and its twin-domain positions  $(0, \pm 1)$  in the nematic phase below  $T_s$ . On cooling below  $T_c$ , a neutron spin resonance, a key signature of unconventional superconductivity [1], appears at  $E \approx 4$  meV and sharply peaks at the  $(\pm 1, 0)$  and  $(0, \pm 1)$  positions [32, 33]. Figure 1(c) shows the energy dependence of the magnetic scattering  $S(E)$  integrated around  $\mathbf{Q}_{AF}$  obtained from our high-resolution INS experiments (see Methods). In the normal state, the magnetic scattering is gapless above  $E = 0.5$  meV and increases in intensity with increasing energy [Fig. 2(a)]. In addition to having a weak peak around  $E \approx 3.2$  meV, we find that the scattering changes from well-defined commensurate peaks centered around  $\mathbf{Q}_{AF}$  below  $E = 3.625 \pm 0.125$  meV [Figs. 2(b), 2(c)] to a peak with flattish top at  $E = 5.625 \pm 0.125$  meV [Fig. 2(d)]. Upon cooling to below  $T_c$  in the superconducting state, the spin excitation spectra open a gap below  $E \approx 2.5$  meV [Figs. 2(e), 2(f)], form a commensurate resonance at  $E = 3.6$  meV [Fig. 2(g)], and exhibit ring-like incommensurate scattering at  $E = 5.25 \pm 0.075$  meV [Fig. 2(f)]. The dispersive ring-like incommensurate resonance is also seen in hole-doped  $\text{Ba}_{0.67}\text{K}_{0.33}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  superconductors [34].

Although these results on twinned FeSe suggest that spin fluctuations play an important role in the superconductivity of FeSe, they provide no information on the possible orbital selective nature of the fluctuations that may lead to a highly anisotropic electron pairing state [19, 31, 35–38]. From STM quasiparticle interference measurements on a single domain (detwinned) FeSe, where the Fermi surface geometry of electronic bands can be determined in the nematic phase, sign-reversed superconducting gaps are found at the hole  $[\Gamma \text{ or } \mathbf{Q} = (0, 0)]$  and electron  $[X \text{ or } \mathbf{Q}_{AF} = (1, 0)]$  Fermi surface states derived from  $d_{yz}$  orbitals of the Fe atoms along the orthorhombic  $a_o$ -axis direction [Figs. 1(a) and 1(b)] [24]. Moreover, similar STM measurements show that the same orbital selective self-energy effects are present already in the normal state of FeSe above  $T_c$  [25].

If superconductivity in FeSe arises from the quasiparticle excitations between hole and electron pockets [Fig. 1(b)] that are indeed orbital selective [24, 25], detwinned crystals should exhibit a strong anisotropy of the low energy spin excitations. In particular, it is expected that the neutron spin resonance associated with  $s^\pm$  superconductivity [32, 33] should only occur along the orthorhombic  $a_o$ -axis direction at  $\mathbf{Q}_{AF} = (\pm 1, 0)$  in a detwinned FeSe, as the orbital selective superconducting gap with the  $d_{yz}$  orbital character is large for scattering vectors along the  $a_o$ -axis [24]. Similarly, orbital dependent Hund's coupling in a nematic quantum spin liquid of FeSe can also induce a large superconducting gap and spin excitation anisotropy [19]. To test these hypothesis, we used INS to study the low-energy spin fluctuations in detwinned FeSe [Figs. 1(d)-1(e)]. In the normal state, spin fluctuations from 6-11 meV are centered around  $\mathbf{Q}_{AF}$  with negligible intensity at  $(0, \pm 1)$ , thus exhibiting a pronounced  $C_2$  rotational symmetry as predicted by these theoretical approaches [19, 27, 31]. By contrast, for energies between 3-5 meV, the spin fluctuations have a  $C_4$  rotational symmetry magnetic component as shown in the schematic illustration in Figs. 1(f) and 1(g) which is based on combining experimental evidences from multiple instruments (Supplementary Fig. S10), possibly corresponding to a localized mode in both wave vector and energy. On cooling below  $T_c$ , the resonance only appears at  $\mathbf{Q}_{AF} = (1, 0)$  [Figs. 1(f) and 1(g)], consistent with the STM observation that superconducting gaps are extremely anisotropic with minima at the tips of the elliptical pockets. Therefore, while the normal state  $C_4$  rotational symmetry magnetic component in the 3-5 meV range is not anticipated, the anisotropic superconductivity-induced resonance is consistent with theoretical expectations [19, 24].

To detect anisotropic spin fluctuations by INS [32, 33], one needs to co-align hundreds of single crystal FeSe samples. These are grown by chemical vapor transport method and are about 1-3 mm<sup>2</sup> in size while few  $\mu\text{m}$  in thickness (see Methods) [9]. Therefore, the most difficult part of carrying out INS experiments on FeSe is to simultaneously detwin hundreds of samples. In previous work on iron pnictides, we were able to completely detwin large (on the order of 0.5-1 cm<sup>2</sup> by few mm in thickness) single crystals of  $\text{BaFe}_2\text{As}_2$  using a mechanical uniaxial pressure device [39, 40]. By gluing many oriented FeSe on uniaxial pressured  $\text{BaFe}_2\text{As}_2$  shown schematically in Fig. 1(d), we were able to simultaneously detwin many FeSe single crystals required for INS experiments (Supplementary Fig. S2). Figure 1(e) shows the temperature dependence of rocking scans along the  $[H, 0, 0]$  and  $[0, K, 0]$  directions on multiple FeSe on  $\text{BaFe}_2\text{As}_2$  assemblies. Below  $T_s \approx 90$  K, we see a clear splitting of the lattice constants. By comparing the scattering intensity of the  $(2, 0, 0)$  and  $(0, 2, 0)$  nuclear Bragg peaks, we find that the FeSe sample assembly has detwinning ratio of  $\eta = [I(2, 0, 0)_o - I(0, 2, 0)_o] / [I(2, 0, 0)_o + I(0, 2, 0)_o] \approx 50\%$  at 2 K [Fig. 1(e)], where  $I(2, 0, 0)_o$  and  $I(0, 2, 0)_o$  are

the observed Bragg peak intensity at  $(2, 0, 0)$  and  $(0, 2, 0)$ , respectively, below  $T_s$  (Supplementary Figs. S3 and S4).

In order to understand the effect of detwinning FeSe, we first need to determine the wave vector and energy dependence of the magnetic scattering  $S(\mathbf{Q}, E)$  in twinned samples (See Methods and supplementary Fig. S5). Figures 2(a) and 2(e) show the energy dependence of the magnetic scattering along the  $[1, K, 0]$  direction above and below  $T_c$ , respectively. In the normal state at  $T = 10$  K, the scattering is gapless above  $E = 0.5$  meV and exhibits a weak peak around  $E = 3.2$  meV [Fig. 2(a)]. The spin excitations are centered around  $\mathbf{Q}_{AF} = (1, 0)$  at  $E = 1 \pm 0.25$  meV [Fig. 2(b)] and  $3.625 \pm 0.125$  meV [Fig. 1(c)]. At  $E = 5.625 \pm 0.125$  meV, the spin excitations have a flattish top as revealed by wave vector cuts along the  $[H, 0]$  and  $[1, K]$  directions [Fig. 1(d)]. In the superconducting state at  $T = 2$  K, a superconductivity-induced spin gap opens below  $E \approx 2.5$  meV and a resonance forms around  $E = 3.6$  meV [Figs. 1(c), 2(e)]. This is confirmed by the vanishing signal at  $E = 1 \pm 0.25$  meV [Fig. 2(f)] and enhanced magnetic scattering at  $3.625 \pm 0.125$  meV [Fig. 2(g)]. In addition, the resonance is clearly centered at the commensurate  $\mathbf{Q}_{AF} = (1, 0)$  position [Fig. 2(g)]. However, on increasing energy to  $E = 5.25 \pm 0.075$  meV, we see clear incommensurate ring-like magnetic scattering centered around  $\mathbf{Q}_{AF} = (1, 0)$ , as confirmed by wave vector cuts along the  $[H, 0]$  and  $[1, K]$  directions [Fig. 2(h)]. The incommensurate scattering intensity in the superconducting state is higher than that of the normal state, suggesting it is a part of the dispersive resonance. In previous work, a dispersive ring-like neutron spin resonance has been seen in the hole-doped  $\text{BaFe}_2\text{As}_2$  family of materials, where the incommensurate scattering has been ascribed to quasiparticle excitations from mismatched hole and electron Fermi surfaces [34].

Figure 3 summarizes the energy evolution of the normal state spin fluctuations at  $\mathbf{Q}_{AF} = (1, 0)$  and  $(0, 1)$  in the  $(H, K)$  plane in partially detwinned FeSe. Since our FeSe single crystals are mounted on surfaces of  $\text{BaFe}_2\text{As}_2$ , one should also see spin fluctuations from  $\text{BaFe}_2\text{As}_2$  at approximately the same position in reciprocal space. However, the spin waves in  $\text{BaFe}_2\text{As}_2$  are gapped below  $\sim 10$  meV in the low-temperature AF ordered state [41, 42], meaning that spin fluctuations at  $\mathbf{Q}_{AF} \approx (1, 0)$  and  $(0, 1)$  below 10 meV must originate from FeSe. Figures 3(a) and 3(b) show constant-energy cuts in the  $(H, K)$  plane for energy transfers of  $E = 3.5 \pm 0.5$  and  $4.5 \pm 0.5$  meV, respectively, in the normal state at  $T = 12$  K. We see clear evidence for magnetic scattering at  $\mathbf{Q}_{AF} \approx (1, 0)$  and  $(0, 1)$  with about the same strength (Supplementary Figs. S6a-S6d), suggesting a possible mode that has  $C_4$  rotational symmetry in the normal state. On increasing energies to  $E = 6 \pm 1$  and  $8 \pm 1$  meV, the scattering at  $\mathbf{Q}_{AF} \approx (1, 0)$  becomes much stronger than those at  $(0, 1)$ , suggesting that spin fluctuations become highly  $C_2$  symmetric at these energies [Figs. 3(c) and 3(d)]. To confirm these results, we carried out energy scans at  $\mathbf{Q}_{AF} \approx (1, 0)$  and  $(0, 1)$  from 2.5 meV to 11 meV as shown in Fig. 3(e) (Supplementary Fig. S6e). From 6 meV to 11 meV, magnetic scattering at  $(1, 0)$  increase in intensity with increasing energy approximately two times faster than the increase of magnetic scattering at  $(0, 1)$ . Figure 3(f) shows wave vector scans approximately along the  $[1, K]$  and  $[H, 1]$  directions at  $E = 8$  meV (see  $E = 3.6$  meV data in supplementary Fig. S6f). The scattering intensity at  $(1, 0)$  dominated the signal while spin fluctuations at  $(0, 1)$  are only 1/3 of that at  $(1, 0)$ . After taking into account the finite detwinning ratio  $\eta$  of the FeSe samples (see supplementary information), there is almost no magnetic scattering at  $(0, 1)$  above the background. These results are consistent with Figs. 3(c) and 3(d), suggesting that the spin fluctuations between 6-10 meV are strongly  $C_2$  symmetric.

To confirm that spin fluctuations in FeSe for energies below 5 meV have a  $C_4$  component as suggested in Figs. 3(a) and 3(b) and determine the impact of superconductivity (supplementary Figs. S7 and S8), we carried out constant-energy and constant-wave-vector scans at  $(1, 0)$  and  $(0, 1)$  using a cold neutron triple axis spectrometer (see Methods). Figures 4(a) and 4(b) show temperature difference plot below ( $T = 2$  K) and above ( $T = 12$  K)  $T_c$  as a function of energy at  $(1, 0)$  and  $(0, 1)$ , respectively. In previous work on twinned samples, superconductivity is found to induce a neutron spin resonance appearing below  $T_c$  at  $(1, 0)$  and  $(0, 1)$  around  $E \approx 3.6$  meV [Fig. 1(c)] [32, 33]. While Figure 4(a) shows clear evidence for the resonance at  $E \approx 3.6$  meV with intensity reduction (negative scattering) below the mode indicating opening of a spin gap [32, 33], an identical temperature difference plot at  $(0, 1)$  in Fig. 4(b) yields no observable temperature difference across  $T_c$ , and therefore no superconductivity-induced resonance and spin gap. Figures 4(c) and 4(d) show wave vector scans along the  $[H + 1, 0]$  and  $[0, K + 1]$  directions, respectively, at  $E = 3.6$  meV. In the normal state ( $T = 12$  K), we see well defined peaks centered at  $(1, 0)$  and  $(0, 1)$ , consistent with Figs. 3(a) and 3(b). On cooling below  $T_c$ , the scattering at  $(1, 0)$  increases in intensity and forms a resonance [Fig. 4(c)], while it does not change across  $T_c$  at  $(0, 1)$  [Fig. 4(d)]. Figures 4(e) and 4(f) show the same data after correcting for background scattering and detwinning ratio  $\eta$ . Similar to Figs. 4(c) and 4(d), we again find that superconductivity induces a  $C_2$  symmetric resonance on a background of approximately  $C_4$  symmetric normal state magnetic scattering (supplementary Figs. S9). Thus, it is the highly anisotropic pairing state of FeSe that drives the  $C_2$  symmetric magnetic scattering at these energies below  $T_c$ . Figures 1(f) and 1(g) summarize the key results of our INS experiments on detwinned FeSe. The deviation of magnetic scattering intensity ratio at  $(1, 0)$  and  $(0, 1)$  from 3 : 1 provides convincing evidence for the existence of an unexpected mode. In the normal state, spin fluctuations

have approximate  $C_4$  symmetry near the resonance energy but become  $C_2$  symmetric for energies above 6 meV. Upon entering the superconducting state, a resonance with  $C_2$  symmetry is formed at  $\mathbf{Q}_{\text{AF}}$  [Fig. 1(f)] (supplementary Figs. S10).

In order to achieve a theoretical understanding of the experimental results presented above, we start from an itinerant five-band model that quantitatively matches the low-energy electronic structure of FeSe in its nematic state [9, 21, 24, 36] and compute the magnetic scattering  $S(\mathbf{Q}, E) \propto \chi''(\mathbf{Q}, E)/(1 - e^{-E/k_B T})$  within a standard random phase approximation formulation [28, 29, 31, 36]. The spectral function at the Fermi level is presented in Fig. 5(a). As illustrated in Figs. 5(c) and 5(e), this “plain vanilla” approach completely fails as is evident, e.g., from the presence of scattering close to (1, 1), and a negligible (1, 0) - (0, 1) anisotropy. The latter properties can be traced to an improper balance of the three most important scattering channels [see Fig. 5(a)]. However, electronic interactions and associated self-energy effects are known to be important in FeSe, constituting an example of a Hund’s metal [25]. Important properties of Hund’s metals include the existence of orbital dependent mass renormalizations [43–45], and an associated redistribution of the relative importance of different orbital dependent scattering channels in the spin susceptibility [46].

A simple means to incorporate the important effects of such orbital selectivity is through the introduction of orbital-dependent quasiparticle weights  $Z_\ell < 1$  [31, 36] leading to a modified bare susceptibility  $\tilde{\chi}_{\ell_1 \ell_2 \ell_3 \ell_4}^0(\mathbf{Q}, E)$  given by

$$\tilde{\chi}_{\ell_1 \ell_2 \ell_3 \ell_4}^0(\mathbf{Q}, E) = \sqrt{Z_{\ell_1} Z_{\ell_2} Z_{\ell_3} Z_{\ell_4}} \chi_{\ell_1 \ell_2 \ell_3 \ell_4}^0(\mathbf{Q}, E). \quad (1)$$

In agreement with theoretical expectations [43–45, 47], and earlier detailed studies of tunneling spectroscopy [24, 25], we apply the hierarchy  $Z_{xy} < Z_{xz} < Z_{yz}$ , which shifts the relative importance of the dominant scattering vectors, as illustrated in Fig. 5(b), and thereby modifies the magnetic scattering. As seen in Fig. 5(d), the  $d_{xy}$ -dominated (1, 1)-scattering is strongly reduced (because  $Z_{xy}$  is the smallest), and the degree of  $C_4$ -symmetry breaking as seen by the difference in the scattering intensities at  $(\pm 1, 0)$  versus  $(0, \pm 1)$  is strongly enhanced (because  $Z_{xz} < Z_{yz}$ ), as seen explicitly by the dashed lines in Fig. 5(f) (Supplementary Fig. S1).

In the superconducting state, we employ the gap structure identical to the one of Refs. [24, 36] which is known to faithfully describe the gap in FeSe, and modify the bare susceptibility accordingly [28, 36]. When entering the highly anisotropic superconducting state, generated by the orbital-selective spin fluctuations [24, 36], a neutron resonance is exhibited solely at the  $(\pm 1, 0)$  position as seen from Figs. 5(f) and 5(g), in agreement with experiments. The associated neutron resonance is highly orbital-selective with predominant  $d_{yz}$  character, as seen by the orbital-resolved spin susceptibilities plotted in Fig. 5(h). Therefore, both the very strong  $C_4$ -symmetry breaking in the 5-10 meV range and the unidirectional neutron resonance observed experimentally are captured by the itinerant orbital-selective scenario.

This approach, however, does not provide an explanation of the emergence of the localized approximately  $C_4$ -symmetric spin excitations near  $E = 3$  meV as shown in Figs. 1(f), 1(g) and 3(a), 3(b). There are several possible scenarios for this remarkable discovery. First, it is possible that self-energy effects in FeSe have a significantly more complicated functional form that cannot be simply captured by including energy- and momentum-independent  $Z$ -factors. Second, there is a possibility of impurity-generated low-energy spectral weight similar to the case of cuprates where vortices and disorder have been shown to generate localized modes in a restricted low-energy regime [48–50]. A counter-argument to disorder-based scenario, however, is the high quality of the FeSe crystals used in the current experiment (Supplementary Fig. S2).

Finally, if part of the spin excitations in FeSe arise from a local moment quantum paramagnet [16, 17, 19], the  $C_2$  symmetric AF collinear order competes with the  $C_4$  symmetric Néel order across the nematic ordering temperature  $T_s$  [18]. In this picture, the  $C_4$  symmetric low-energy magnetic excitations with spin-wave ring-like features in detwinned FeSe may simply be the remnant of the localized moment not directly associated with Fermi surface nesting and itinerant electrons.

Regardless of the microscopic origin of the  $C_4$  spin excitations, our data support the notion that the spin fluctuations in the nematic phase of FeSe are, generally, highly anisotropic, and is consistent with superconductivity being driven by spin fluctuations arising mainly from the  $d_{yz}$  orbital states. Our measurements highlight the need for a quantitative understanding of both the extreme spin anisotropy, as well as the emergence of  $C_4$ -symmetric magnetic excitations at the very lowest energies. Progress in this direction may well shed new light on the role of electronic correlations in FeSe, in particular, and the origin of unconventional superconductivity in interacting systems, in general.

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## Author contributions

X.Y.L., T.C., and P.D. conceived the project. T.C. prepared all the FeSe single crystal samples.  $\text{BaFe}_2\text{As}_2$  single crystals are prepared by T.C., X.Y.L., R.Z., Y.L., Y.R., Y.W. Neutron scattering experiments on twinned samples are carried out by T.C., Y.C., Y.M.Q., C.B., and P.D. at NCNR. Neutron scattering experiments on detwinned samples are carried out by T.C., J.P., T.G.P., J.R.S., H.B.C., and P.D. at ORNL, ISIS, and MLZ. Theoretical analysis was performed by A.G., B.M.A., and P.J.H. The entire project was supervised by P.D. The manuscript is written by P.D., T.C., A.K., B.M.A., and P.J.H. All authors made comments. The authors declare no competing financial interests.

## Additional information

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## Methods

**Experimental Setups** Elastic neutron experiments were carried out on the HB-3A four-Circle diffractometer at the High-Flux-Isotope Reactor (HFIR), Oak Ridge National Laboratory (ORNL), United States to first check if the method works well in detwinning FeSe on a single piece of  $\text{BaFe}_2\text{As}_2$  (Supplementary Fig. S3). HB-3A uses a silicon monochromator and a scintillator-based 2D Anger Camera. We define  $(H, K, L) = (q_x a/2\pi, q_y b/2\pi, q_z c/2\pi)$

in reciprocal lattice units (r.l.u.) using the orthorhombic lattice notation for FeSe, where  $a \approx 5.33$  Å,  $b \approx 5.31$  Å,  $c = 5.486$  Å [9].

Our INS experiments on twinned samples were done on MACS cold triple axis spectrometer at NIST center for neutron scattering at Gaithersburg, Maryland. MACS spectrometer has a double focusing pyrolytic graphite [PG(002)] monochromator and multi detectors. We used  $E_f = 3.7$  meV with a BeO filter after the sample and a Be filter before the monochromator for energy transfers below  $E = 1.5$  meV.

Our INS experiments on detwinned samples were carried out on the PANDA cold neutron and PUMA thermal neutron triple-axis spectrometers, at MLZ, Garching, Germany [39], and on the MAPS time-of-flight chopper spectrometer, at ISIS, Rutherford-Appleton laboratory, Didcot, United Kingdom [40].

For PANDA experiments, a double-focused pyrolytic graphite [PG(002)] monochromator and analyzer with fixed scattered neutron energy  $E_f = 5.1$  meV were used with collimations of none-40'-40'-none for inelastic measurements. For elastic measurements, we used  $E_f = 4.39$  meV with collimations of 80'-80'-80'-80'. For thermal neutron measurements on PUMA, we used  $E_f = 14.69$  meV with double focusing monochromator and analyzer and no collimators. For MAPS neutron time-of-flight measurements, we used an incident beam energy of  $E_i = 38$  meV with the incident beam along the  $c$ -axis of the crystal.

**Sample growth and preparation** The high quality FeSe single crystals used in the experiments are grown by a chemical vapor transport method. Fe and Se powder are sealed in quartz tubes with KCl-AlCl<sub>3</sub> flux. The growth takes 28 days in a temperature gradient from 330°C to 400°C. Typical samples are  $1 \times 1$  mm<sup>2</sup> in area and  $< 0.1$  mm in thickness. The square-shaped BaFe<sub>2</sub>As<sub>2</sub> crystals are grown using flux method [40]. They are aligned using a Laue camera and cut along the tetragonal  $[1, 1, 0]$  and  $[1, -1, 0]$  directions by a high-precision wire saw. Since single crystals of FeSe have one natural edge 45° rotated from the orthorhombic  $a_o$  direction, we can use an optical method to co-align FeSe on the surface of BaFe<sub>2</sub>As<sub>2</sub>. Given our intent to measure spin excitations in detwinned FeSe, we aligned and glued (with CYTOP type-M) about 300 small pieces FeSe single crystals on many pieces of big BaFe<sub>2</sub>As<sub>2</sub> single crystals (Supplementary Figs. S1-S3).

#### **Data availability**

The data that support the plots in this paper and other findings of this study are available from the corresponding authors upon reasonable request.

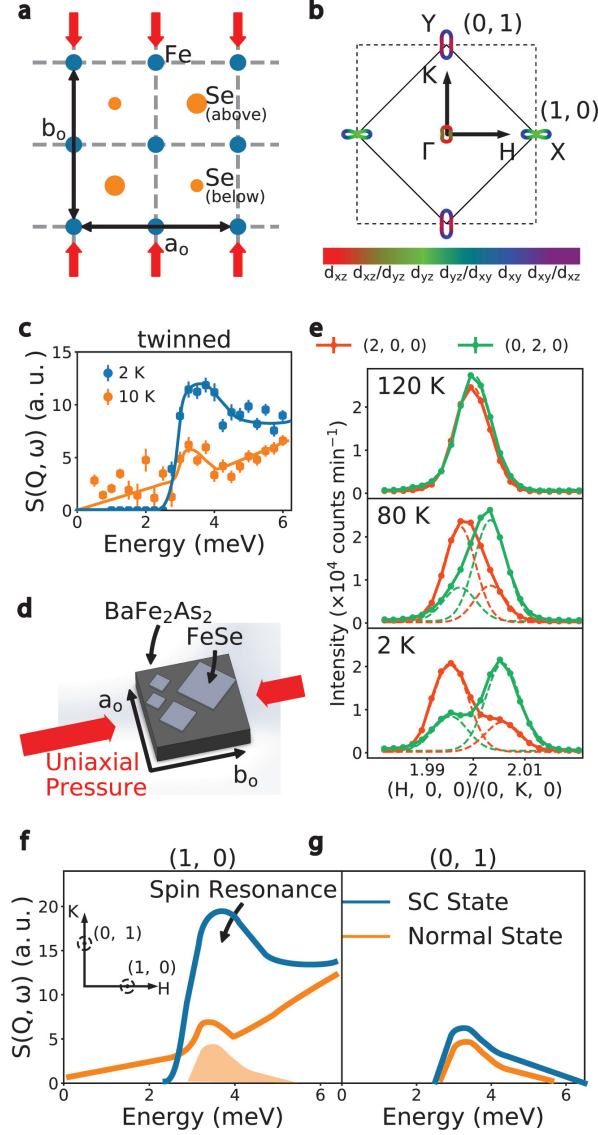


FIG. 1: **Crystal structure, Fermi surface and neutron scattering of FeSe** (a) Crystal structure of FeSe, where blue and orange colors mark Fe and Se positions, respectively. The red arrows indicate the uniaxial strain direction applied through detwinned  $\text{BaFe}_2\text{As}_2$ .  $a_0$  and  $b_0$  are the orthorhombic lattice parameters (double-sided black arrows) in the nematic phase. Gray dashed lines are guides for the eye. (b) Hole/electron Fermi surfaces of the tight binding model for FeSe [24]. Color indicates orbital character of the Fermi surfaces, where red, green, and blue indicate  $d_{xz}$ ,  $d_{yz}$ , and  $d_{xy}$  orbitals of the Fe atom. Fermi surface nesting of  $\Gamma \rightarrow X$  and  $\Gamma \rightarrow Y$  corresponds to  $(1, 0)$  and  $(0, 1)$  in reciprocal lattice units (r.l.u.), where  $(H, K) = (q_x a_0 / 2\pi, q_y b_0 / 2\pi)$  are in-plane Miller indices of the orthorhombic lattice, respectively (see Methods). (c) Energy dependence of the measured magnetic scattering ( $S(Q, E)$ ) integrated around  $(1, 0)$  above ( $T = 10$  K) and below ( $T = 2$  K)  $T_c$  ( $\approx 8$  K) on twinned FeSe (Supplemental Fig. S5). The vertical error bars indicate the statistical errors of one standard deviation. (d) Schematic diagram of the sample arrangement, FeSe samples are glued on large single crystals of  $\text{BaFe}_2\text{As}_2$  under uniaxial pressure of  $\sim 20$  MPa [39, 40], where red arrows indicate pressure direction. (e) Wave vector scans of nuclear  $(2, 0, 0)$  and  $(0, 2, 0)$  Bragg peaks of FeSe on an assembly of  $\text{BaFe}_2\text{As}_2$  single crystals at different temperatures. The dashed lines indicate the single Gaussian components of the fitting. (f,g) Schematic illustration of the magnetic scattering at  $(1, 0)$  and  $(0, 1)$  in the normal and superconducting (SC) states estimated from the twinned and detwinned samples (Supplemental Fig. S10). Shaded region in (f) is  $(0, 1)$  data from (g).



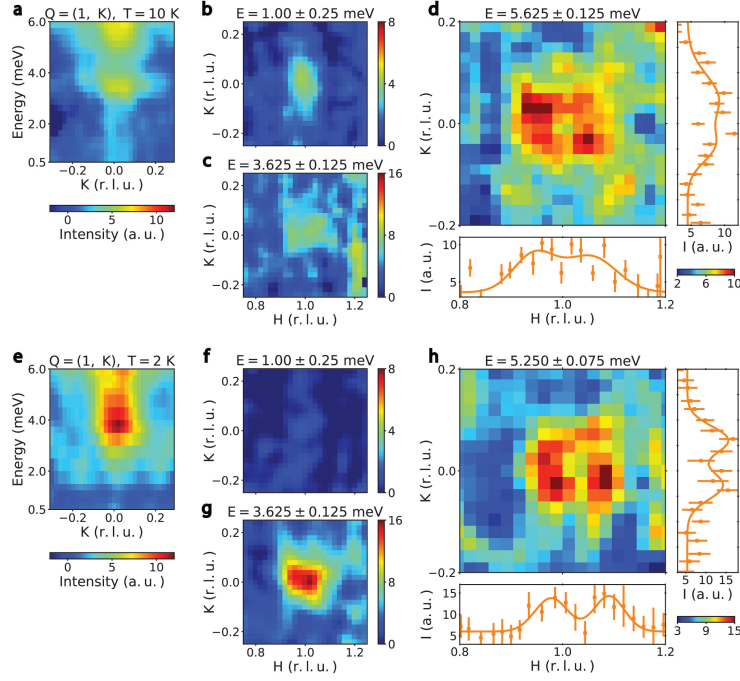


FIG. 2: **Low-energy spin fluctuations in twinned FeSe below and above  $T_c$ .** Two-dimensional images of wave vector and energy dependence of spin fluctuations at (a)  $T = 10$  K, (e)  $T = 2$  K. Wave vector dependence of spin fluctuations in the  $(H, K)$  plane at energies (b,f)  $E = 1 \pm 0.25$  meV, (c,g)  $E = 3.6 \pm 0.125$  meV, (d)  $E = 5.62 \pm 0.125$  meV. Cuts along the  $[H, 0]$  and  $[1, K]$  directions with a width of  $\pm 0.04$  r.l.u. show flattish top scattering near  $(1, 0)$ . (h)  $E = 5.25 \pm 0.075$  meV. Incommensurate scattering is clearly seen through the identical cuts along the  $[H, 0]$  and  $[1, K]$  directions. This feature is missed in previous work [18, 32, 33] due to the small incommensurability and narrow energy range. Panels (a,b,c,d) and (e,f,g,h) are at  $T = 10$  K and  $T = 2$  K, respectively. Solid lines are fitting with the sum of two Gaussians to the data. The vertical error bars indicate the statistical errors of one standard deviation.

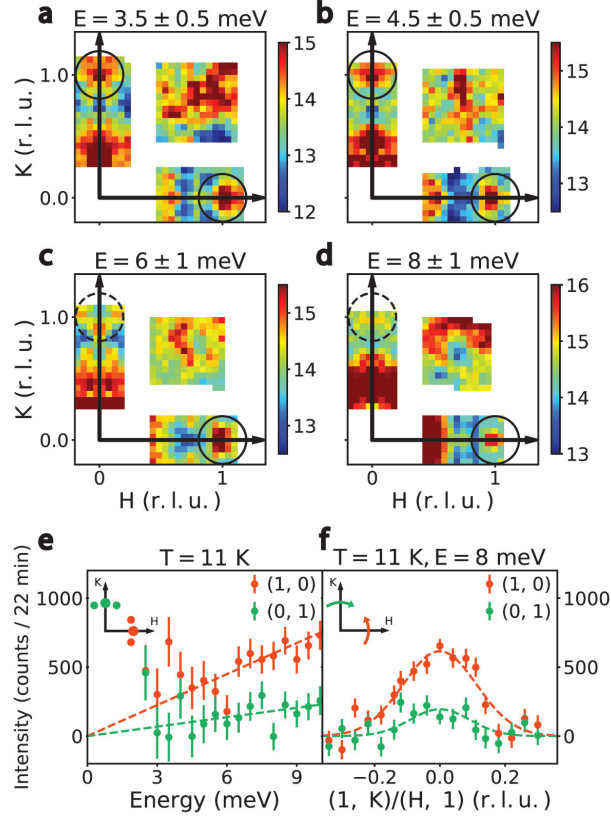


FIG. 3: **Normal state spin fluctuations in detwinned FeSe.** The two-dimensional images of spin fluctuations at (a)  $E = 3.5 \pm 0.5$  meV, (b)  $E = 4.5 \pm 0.5$  meV, (c)  $E = 6 \pm 1$  meV, and (d)  $E = 8.5 \pm 1$  meV. The data are collected at  $T = 12$  K using MAPS chopper spectrometer with incident neutron energy of  $E_i = 38$  meV along the  $c$  axis and are folded to improve statistics. The scattering near wave vector  $(1,1)$  is background and not magnetic in origin (Supplementary Figs. S6-S8). (e) Energy dependence of the scattering  $(1,0)$  and  $(0,1)$  above background at  $T = 11$  K. The positions of signal and background are marked as large and small spots in the inset. (f) Wave vector scans at  $E = 8$  meV along the  $[1, K]$  (green) and  $[H, 1]$  direction at  $T = 11$  K. Linear backgrounds have been subtracted from the data. The vertical error bars indicate the statistical errors of one standard deviation.

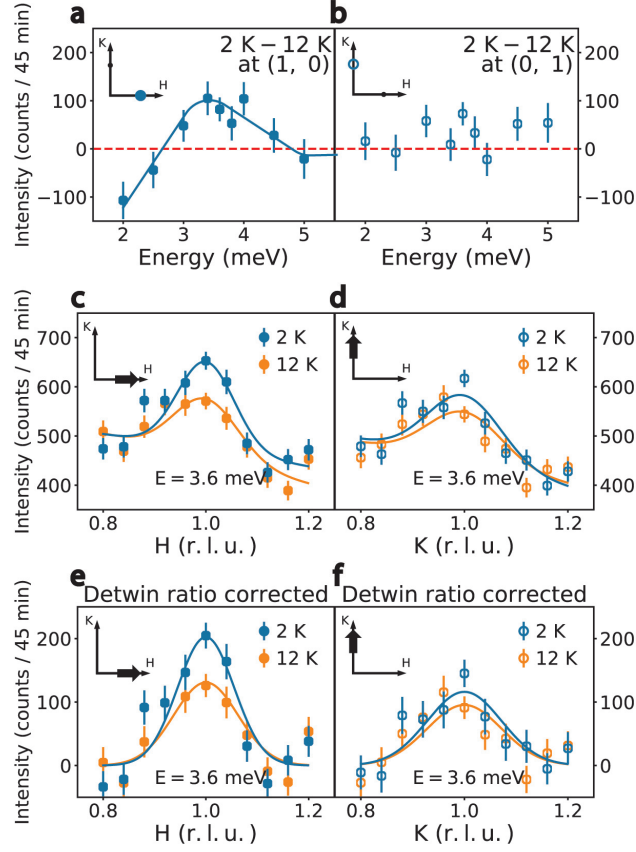


FIG. 4: **Effect of superconductivity on low-energy spin fluctuations of detwinned FeSe.** Difference of the scattering in the superconducting state (below  $T_c$ ) and the normal state plotted as function of energy for the momentum transfer (a) (1,0) and (b) (0,1). The peak seen at  $E \approx 3.6$  meV in (a) marks the neutron spin resonance. The solid blue line and dashed red lines are guides to the eye. (c) Wave vector scans below and above  $T_c$  at  $E = 3.6$  meV and (1,0). (d) Similar scans at (0,1). (e) (1,0) and (f) (0,1) scans with background subtracted and detwinning ratio corrected (Supplemental Fig. S9). The solid lines in (c,d) and (e,f) are Gaussian fits to the data before and after linear background subtraction. The vertical error bars indicate the statistical errors of one standard deviation.

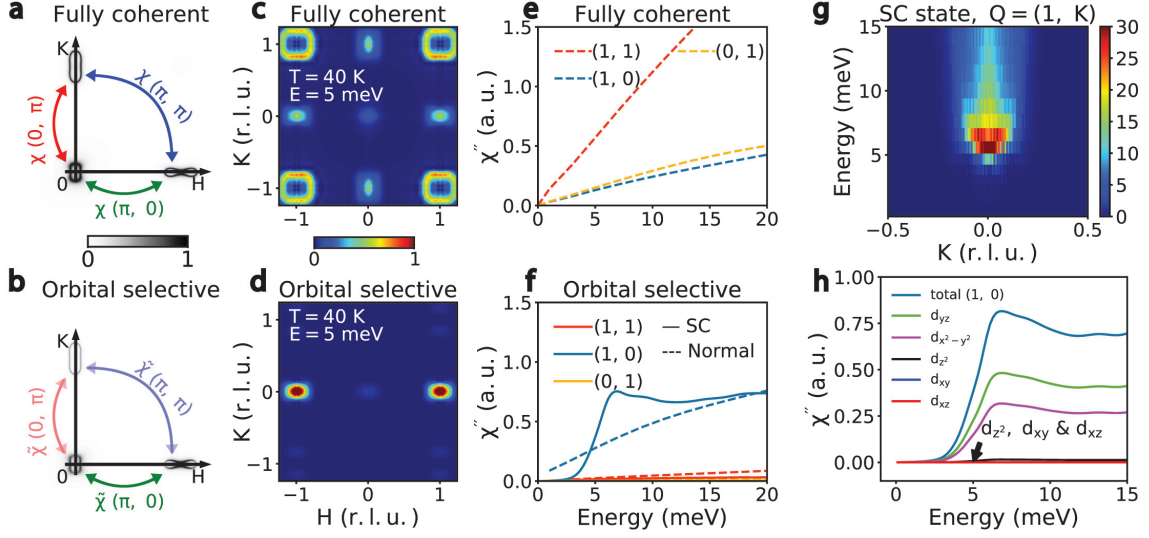


FIG. 5: **Theoretical calculations of the spin fluctuations in detwinned FeSe.** Map of the spectral function at zero energy for our model of the electronic structure of FeSe in (a) the fully coherent case with quasiparticle weights ( $Z_\ell = 1$ ) and (b) the orbital-selective case where reduced quasiparticle weights ( $Z_\ell < 1$ ) weaken the spin-fluctuations at certain momentum transfer (blue and red arrows) while the spin fluctuations stemming mostly from the  $d_{yz}$  orbital become dominant (green arrow). (c) For the fully coherent model, one obtains in the normal state large contributions to the dynamical structure factor close to  $(\pm 1, \pm 1)$  from the  $d_{xy}$  orbital and an almost identical but weaker contribution at  $(\pm 1, 0)$  and  $(0, \pm 1)$ . (e) The susceptibility integrated around these momentum transfer vectors, shows the same trend at low energies. (d,f) In contrast, the orbital selective model yields spin fluctuations at low energies which are dominated by peaks at  $(\pm 1, 0)$  in the low energy range shown. The enhancement of the spin fluctuations at  $(\pm 1, 0)$  in the superconducting state is clearly seen when plotted (f) as function of energy and (g) as an intensity map in momentum-energy space. (h) The spin fluctuations at  $(1, 0)$  are dominated by the contributions of the  $d_{yz}$  orbital.