nature portfolio

Peer Review File

Spin correlations in the nematic quantum disordered state of FeSe

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This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Nature Communications.

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #2

(Remarks to the Author)
After seeing the reviews from the other referees, I still very much like this paper. I think a lot of work went into revising it for this journal.

It is a great paper, and the authors can be proud of it. I recommend publishing as is.

Reviewer #4

(Remarks to the Author)
see comments to EDITORS

Reviewer #5

(Remarks to the Author)
The authors have performed inelastic neutron scattering (INS) experiments on iron-based superconductor FeSe while using a thermally driven strain device to detwin the single crystals. While the system in question does not exhibit magnetic long-range order, strong correlations exists and the authors observe spin waves emerging from stripe and Néel order with C2 and C4 symmetry, respectively. The difference in the neutron intensities at (1,0) and (0,1) is used as a measure for the nematic spin correlation, suggesting that nemacity in FeSe may be spin driven. By fitting a Heisenberg model to the excitations they determine the magnetic exchange interaction parameters and find that FeSe resides in the antiferroquadrupolar regime but close to both the stripe and Néel parts of the phase diagram. However, the model seems to describe only the stripe part of the correlations and the role of the Néel fluctuations is not clear from the manuscript.

The thermal strain device used in this work allows for highly improved data quality compared to previous INS studies which demonstrates the utility of this technique to detwin samples and which may be applied to other systems with similar twinning issues.

Although the results are convincing and of high quality they primarily consolidate previous findings rather than bring novel results. Nevetheless, the higher quality data enable better distinguishing between proposed models. The novelty in this work lies more in the method for detwinning which will be of interest for a broad audience. I therefore support its publication in Nature Communications after a few clarifications.

A few minor comments:

The results are generally well presented and the only overall comment that I have is that it would be nice with larger and more readable figure panels in general.

Figure 4(m), please explain the white and and pink symbols in the caption. Does the Néel spin wave have a gap?

What are the uncertainties on all the fitted model parameters?

How are the phase boundaries determined in Fig. 1(h)? The previous reviewer reports also asked about this but I was still in doubt when reading the revised manuscript.

Why are sample #1 and #2 too large to estimate the detwinning ratio using nuclear Bragg peaks? Normally the beamsize can be regulated to fit the sample and yours doesn't seem too large.

Reviewers' comments and our replies:

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Reviewer #3 (Remarks to the Author):

The authors have performed inelastic neutron scattering (INS) experiments on iron-based superconductor FeSe while using a thermally driven strain device to detwin the single crystals. While the system in question does not exhibit magnetic long-range order, strong correlations exists and the authors observe spin waves emerging from stripe and Néel order with C_2 and C_4 symmetry, respectively. The difference in the neutron intensities at (1,0) and (0,1) is used as a measure for the nematic spin correlation, suggesting that nemacity in FeSe may be spin driven. By fitting a Heisenberg model to the excitations they determine the magnetic exchange interaction parameters and find that FeSe resides in the antiferroquadrupolar regime but close to both the stripe and Néel parts of the phase diagram. However, the model seems to describe only the stripe part of the correlations and the role of the Néel fluctuations is not clear from the manuscript.

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Response to Reviewer #3 (1):

We sincerely appreciate the Reviewer's thoughtful evaluation and constructive feedback on our work. We are particularly encouraged by their recognition of the high-quality inelastic neutron scattering data abtained through our novel thermal strain device. Below we provide detailed responses to specific comments to further improve the manuscript.

Reviewer #3 (Remarks to the Author):

A few minor comments:

1. The results are generally well presented and the only overall comment that I have is that it would be nice with larger and more readable figure panels in general.

Response to Reviewer #3 (2):

We thank the Reviewer for the constructive suggestion regarding figure presentation. In response, we have carefully revised figures to optimize readability.

Reviewer #3 (Remarks to the Author):

2. Figure 4(m), please explain the white and and pink symbols in the caption. Does the Néel spin wave have a gap?

Response to Reviewer #3 (3):

We thank the Reviewer for raising these important points regarding Fig. 4(m). Please find our point-by-point responses below:

(1) Explanation of symbols in Figure 4(m)：

The white square and pink diamond symbols in Fig. 4(m) correspond to two key parameters from the damped harmonic oscillator fitting in Fig. 4(k). Specifically, white square and pink diamonds represent the damping factor [γ (q)/2] and the undamped energy (E_0), respectively. The observed results E_0 > γ (q)/2 confirms that the intrinsic spin excitations in FeSe are underdamped. We have revised the caption to explicitly clarify this.

(2) Néel spin wave have a gap

Our experimental data reveal a finite gap (~40 meV) in Néel spin excitations at **Q** = ($\pm$1, $\pm$1). This is supported by the absence of Néel spin excitations signals below *E* ~40meV in Figs. 4(e)-(h).

Reviewer #3 (Remarks to the Author):

3. What are the uncertainties on all the fitted model parameters?

Response to Reviewer #3 (4):

We appreciate the Reviewer's reminder. The uncertainties were estimated according to the 68% confidence interval of the fittings. We have added the fitting errorbars in the main text and markede the uncertainties as errorbars in Fig. 1(h).

Reviewer #3 (Remarks to the Author):

How are the phase boundaries determined in Fig. 1(h)? The previous reviewer reports also asked about this but I was still in doubt when reading the revised manuscript.

Response to Reviewer #3 (5):

We thank the Reviewer for raising this important question. In the previous version of the manuscript, we sketched the phase boundaries in Fig. 1(h) by referring to the calculated phase diagram from Ref. [49] and by emphasizing the approximate positions of FeSe, NaFeAs, and $BaFe_2As_2$.

In the revised manuscript, we have improved the accuracy of the phase boundaries by explicitly calculating them using the variational method described in Ref. [49] (Phys. Rev. Res. **2**, 023359 (2020)). For clarity, we have now included only the dominant phases—Néel AFM (NAFM), collinear AFM (CAFM/stripe), and antiferroquadrupolar (AFQ)—and omitted minor or potentially disordered phases arising from strong fluctuations. The regions near the phase boundaries are left white to denote crossover regimes.

The phase boundaries are determined using the following relations:

- **NAFM/CAFM**: $K = (4J_2 - 2) / (\alpha_2 - 2\alpha_1)$
- **CAFM/AFQ**: $K = J_2 / (\alpha_1 - 2\alpha_3)$
- **AFQ/NAFM**: $K = (4J_2 - 2) / (\alpha_2 - \alpha_3)$
- $\mathbf{K_i = \alpha_i K, \quad i = 1, 2, 3}$

Here, the α_i are numerical parameters defined in Ref. [49], and the three phase boundaries intersect at a single point determined by J_2 and α_3. For reference:

- Line 1 (NAFM/CAFM) passes through (½, 0),
- Line 2 (CAFM/AFQ) passes through (0, 0), and
- Line 3 (AFQ/NAFM) passes through (1, 0).

In the updated figure, we have replotted the phase boundaries and retained FeSe and NaFeAs in the white crossover region where the stripe, Néel, and AFQ phases meet.

Reviewer #3 (Remarks to the Author):

5. Why are sample #1 and #2 too large to estimate the detwinning ratio using nuclear Bragg peaks? Normally the beamsize can be regulated to fit the sample and yours doesn't seem too large.

Response to Reviewer #3 (6):

We sincerely thank the Reviewer for the insightful comment regarding the determination of the detwinning ratio. While we agree that nuclear Bragg peaks are typically a reliable method for such estimation, in our case, technical and logistical challenges prevented us from conducting full elastic diffraction measurements on Samples #1 and #2.

Specifically, there were two main reasons:

1. **Instrumental Constraints**: Our uniaxial strain device, with dimensions of 104 × 80 × 37 mm^3, imposes strict limitations on sample rotation during neutron diffraction experiments. As a result, only the (040) Bragg peak was marginally accessible. The (400) peak was out of the scattering plane, and even the (040) peak could sometimes be blocked by the invar alloy frames or by the cadmium foil shielding used to suppress background scattering.

2. **COVID-19 Disruptions**: Our planned beamtime proposals for diffraction-based detwinning analysis were unfortunately not approved, and further attempts were delayed due to pandemic-related restrictions, which significantly disrupted offline experimental scheduling and operations.

As an alternative and validated approach (Ref. [13]), we estimated the detwinning ratio from the anisotropy of low-energy magnetic excitations at (1,0) and (0,1) at base temperature (5 K), which reliably reflects domain population in detwinned Fe-based superconductors.