Magnetic ground state of FeSe

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Elucidating the nature of the magnetism of a high-temperature superconductor is crucial for establishing its pairing mechanism. The parent compounds of the cuprate and iron-pnictide superconductors exhibit Neél and stripe magnetic order, respectively. However, FeSe, the structurally simplest iron-based superconductor, shows nematic order ($T_s = 90$ K), but not magnetic order in the parent phase, and its magnetic ground state is intensely debated. Here we report inelastic neutron-scattering experiments that reveal both stripe and Neél spin fluctuations over a wide energy range at 110 K. On entering the nematic phase, a substantial amount of spectral weight is transferred from the Neél to the stripe spin fluctuations. Moreover, the total fluctuating magnetic moment of FeSe is $\sim 60\%$ larger than that in the iron pnictide BaFe$_2$As$_2$. Our results suggest that FeSe is a novel $S=1$ nematic quantum-disordered paramagnet interpolating between the Neél and stripe magnetic instabilities.

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Recently, FeSe has attracted considerable interest because of its atypical magnetism\(^7\)-\(^6\) and fascinating superconducting properties\(^7\)-\(^10\). Although the superconducting transition temperature \(T_c\) of bulk FeSe (\(T_c \approx 8 \text{ K}\)) is low, it increases drastically under pressure (\(T_c \approx 37 \text{ K}\); ref. 7), by carrier doping (\(T_c \approx 40-48 \text{ K}\); refs 8, 9), or in the mono layer limit (\(T_c \sim 65-109 \text{ K}\), ref. 10). The unique superconducting properties of FeSe are argued to be the consequence of stripe magnetic order, and both temperature \(T\) and \(E\) are roughly the same. Symmetry equivalent data were pooled to enhance statistical accuracy. The scale as those acquired at 110 K. The measurements in (a) Schematic representation of the stripe and Néel spin fluctuations in the \((H, K)\) plane. (b-j) Constant-energy images acquired at 110 K at indicated energies. (k-s) Constant-energy images obtained at 4 K at the same intensity scale as those acquired at 110 K. The measurements in (b-d,k-m) and (e-j,n-s) were carried out on ARCS with the incident neutron energy of 79 meV and 294 meV, respectively. The sample has two equally populated orthogonal twin domains in the \(ab\) plane at 4 K and the intensities near (1, 0) and (0, 1) are roughly the same. Symmetry equivalent data were pooled to enhance statistical accuracy. The \(|Q|\)-dependent background is subtracted for the data (b-d,k-m) below the aluminium phonon cutoff energy of \(\sim 40 \text{ meV}\). Above 40 meV, raw data are presented (e-j,n-s). The colour bars indicate intensity in unit of mbar sr\(^{-1}\) meV\(^{-1}\) f.u.\(^{-1}\). The dashed ellipses and circles mark the stripe and Néel wavevectors, respectively.

**Figure 1** | Momentum dependence of the spin fluctuations in FeSe at 4 and 110 K. (a) Schematic representation of the stripe and Néel spin fluctuations in the \((H, K)\) plane. (b-j) Constant-energy images acquired at 110 K at indicated energies. (k-s) Constant-energy images obtained at 4 K at the same intensity scale as those acquired at 110 K. The measurements in (b-d,k-m) and (e-j,n-s) were carried out on ARCS with the incident neutron energy of 79 meV and 294 meV, respectively. The sample has two equally populated orthogonal twin domains in the \(ab\) plane at 4 K and the intensities near (1, 0) and (0, 1) are roughly the same. Symmetry equivalent data were pooled to enhance statistical accuracy. The \(|Q|\)-dependent background is subtracted for the data (b-d,k-m) below the aluminium phonon cutoff energy of \(\sim 40 \text{ meV}\). Above 40 meV, raw data are presented (e-j,n-s). The colour bars indicate intensity in unit of mbar sr\(^{-1}\) meV\(^{-1}\) f.u.\(^{-1}\). The dashed ellipses and circles mark the stripe and Néel wavevectors, respectively.
crystal\cite{12} and powder samples\cite{13}. However, because of the limitations of the q-space information that can be obtained for powder measurements and the relatively narrow energy range probed previously, the precise nature of the magnetic ground state remains undetermined; this elucidation requires measurements of the momentum structure of the spin-fluctuation spectrum from low energy to the zone boundary over the entire Brillouin zone.

In this paper, we used inelastic neutron scattering to map out the spin-fluctuation spectra over the entire Brillouin zone in single-crystalline FeSe (Methods). Our data reveal the coexistence of the stripe and Néel spin fluctuations, both of which are coupled with nematicity. In addition, although the spin-fluctuation bandwidth is lower, the total fluctuating magnetic moment \((\langle m^2 \rangle) = 5.19 \mu_B^2/\text{Fe}\) of FeSe is \(\approx 60\%\) larger than that in the iron pnictide BaFe\(_2\)As\(_2\) (ref. 14). These findings suggest that FeSe is an \(S = 1\) nematic quantum-disordered paramagnet, interpolating between the Néel and stripe magnetic instabilities\cite{15}.

**Results**

**Momentum and energy dependence of spin fluctuations.** Figure 1 shows the constant-energy images of spin fluctuations in the \((H, K)\) plane. Here, in the high-temperature tetragonal phase (110 K), the spin response is strongest at \(Q = (1, 0)\) (marked by dashed ellipse) at 15 meV (Fig. 1b), which is consistent with previous low-energy measurements\cite{12,13}. With an increase in energy (Fig. 1c–j), the spin fluctuations show anisotropic dispersion and counter-propagate mainly along the \(K\) direction. This is analogous to the stripe spin fluctuations detected in other iron-based superconductors\cite{11,14,15}. Most notably, in addition to the stripe spin fluctuations, comparatively weaker, but clear scattering appears near \((1, 1)\) (Fig. 1b, dashed circle), which implies the presence of spin fluctuations associated with the Néel magnetic instability (Methods); different from the anisotropic stripe spin fluctuations, the Néel spin fluctuations are nearly isotropic in the transverse and longitudinal directions (Fig. 1b–g). With an increase in the energy up to \(\approx 150\) meV, the Néel and stripe spin fluctuations overlap and cover a broad area centred at \((1, 1)\) (Fig. 1i). On cooling to within the nematic phase \((T = 4\) K\), the Néel-spin-fluctuation signal weakens considerably and is almost undetectable \(< 35\) meV (Fig. 1k,l). On the other hand, the momentum structure of the stripe spin fluctuation is essentially unchanged above and below \(T_c\) (Fig. 1k–s).

To further elucidate the spin fluctuations in \(E-Q\) space, we projected the spin fluctuations along the \(K\) direction near \((1, 0)\) and \((1, 1)\) (Fig. 2). Because the incident neutron beam was parallel to the \(c\) axis, the energy transfer was coupled with \(L\). No \(L\) modulations were observed from the scattering near \((1, 0, L)\) and \((1, 1, L)\), which indicates a two-dimensional nature of the magnetism. As shown in Fig. 2a, the stripe spin fluctuations stem from \((1, 0)\), split into two branches at \(\approx 35\) meV and extend up to above \(\approx 150\) meV at 110 K. The steeply dispersive Néel spin fluctuations are also visible (green arrowheads). As the temperature is lowered to 4 K, the Néel spin fluctuation exhibits a \(\approx 30\) meV gap, while the stripe spin fluctuations \(< 70\) meV are clearly enhanced (Fig. 2b).

To quantify the dispersions and intensities of the stripe and Néel spin fluctuations, we made constant-energy cuts at distinct energies (Fig. 3). As Fig. 3d–i show, at \(T = 110\) K, the single peak centred at \((1, 0)\) at 15 meV evolves into a pair of peaks along the \(K\) direction at \(E \geq 35\) meV. By contrast, the peak position of the Néel spin fluctuation (see green arrowheads) shows little change; it only broadens gradually in wavevector with increasing energy. The Néel spin fluctuation is more clearly visible along the transverse direction (Fig. 3m–r) because of the comparatively weaker influence of the stripe spin fluctuations. Here, double peaks formed due to the dispersion are not seen \(< 68\) meV, because the Néel spin fluctuations are commensurate and steeply dispersive (Fig. 3m–r). At higher energies, the Néel and stripe spin-fluctuation spectra merge with each other, and their dispersions cannot be determined unambiguously (Fig. 3a–c, j–l). The Néel spin fluctuation becomes featureless at low energies (Fig. 3i,r) at 4 K, which agrees with the results shown in the constant-energy and \(E-Q\) images (Figs 1k and 2b). We attempted to fit both types of spectra concurrently using a linear spin-wave theory for the two-neighbour \((J_{1(a/b)}, J_2-J_3)\) or three-neighbour \((J_{1(a/b)}, J_2, J_3)\) Heisenberg model, where \(J_{1(a/b)}\), \(J_2\) and \(J_3\) are nearest neighbour (in the \(a/b\) direction), next-nearest neighbour and next next-nearest neighbour exchange coupling constants, respectively; but this was unsuccessful mainly because this theory cannot account for the observed strong low-energy spin excitations at both \((1, 0)\) and \((1, 1)\).

**Figure 2 | Dispersions of the stripe and Néel spin fluctuations in FeSe at 4 and 110 K.** Background-subtracted \(E-K\) slice of the spin fluctuations at various incident energies: (a) \(T = 110\) K; (b) \(T = 4\) K. The data at \(E \geq 40\) and \(E \leq 40\) meV were collected on ARCS by using incident energy of 294 and 79 meV, respectively. The isotropic Fe\(^{2+}\) magnetic form factor is corrected for both sets of the data. The spectral weight transfer from the Néel \((1, 1)\) to stripe \((1, 0)\) wavevector below \(\approx 70\) meV on cooling to 4 K can be clearly seen. The open circles are dispersions obtained from the constant-energy cuts \(< 68\) meV in Fig. 3. The colour bar indicates intensity in unit of mbar sr\(^{-1}\) meV\(^{-1}\) sr\(^{-1}\) f.u.\(^{-1}\). The vertical bars indicate the energy integration range. The horizontal bars at 15 meV indicate the full-width at half-maximum of the Gaussian fittings in Fig. 3i. The horizontal bars at other energies are the errors derived by least-square fittings. The dashed lines are a guide to the eye.
Momentum integrated local susceptibility above and below $T_s$.

More insight into the nature of the underlying magnetic ground state and its interaction with the nematicity could be acquired by calculating in absolute units the energy dependence of the momentum integrated local susceptibility $\chi'\omega$ above and below $T_s$; as Fig. 4a,b show, at $T = 110$ K, the Néel spin-fluctuation spectral weight is roughly 26% of that of the stripe spin fluctuation <52 meV, where the two signals are well separated in $q$-space. Upon cooling to $T = 4$ K, the spectral weight loss for the Néel spin fluctuations is approximately recovered by the enhanced stripe spin fluctuations (red shaded areas), and thus the total local susceptibility $\chi'\omega$ does not show a marked change across $T_s$ (Fig. 4c). Moreover, the detailed temperature dependence of the stripe and Néel spin fluctuations show that the spectral weight transfer is clearly coupled with the development of the nematic phase (Fig. 4d). At both 4 and 110 K, the total $\chi'\omega$ exhibits a high maximum at $\sim 105$ meV and extends up to 220 meV (Figs 2a,b and 4c). This bandwidth is considerably lower than that ($\sim 340$ meV) of the stripe ordered BaFe$_2$As$_2$ (ref. 14), which is very likely due to the competition between the Néel and stripe magnetic instabilities. Clearly, this type of competition also prevents the long-range magnetic order in FeSe. By integrating the spectral weight from low energy to the zone boundary, we determined that the total fluctuating moment at 4 and 110 K are $\langle m^2 \rangle = (g\mu_B)^2 S(S + 1) = 5.19 \pm 0.32$ and $5.12 \pm 0.27 \mu_B^2$/Fe, respectively, which are larger than those in the superconducting BaFe$_{1.9}$Ni$_{0.1}$As$_2$ ($\langle m^2 \rangle = 3.2 \mu_B^2$/Fe) and stripe-ordered BaFe$_2$As$_2$ ($\langle m^2 \rangle = 3.17 \mu_B^2$/Fe) (ref. 14). Accordingly, this yields an effective spin of $S = 0.74$ in FeSe, which likely corresponds to an $S = 1$ ground state in the presence of itinerant electrons.

**Discussion**

The coexistence of the Néel and stripe spin fluctuations is unexpected because FeSe contains only one type of magnetic ions. This differs from the Mn-doped nonsuperconducting iron-nitride compound BaFe$_{2-x}$Mn$_x$As$_2$, where the Nél magnetic correlation is induced by the local moments of Mn, while the stripe magnetic correlation is induced by Fe, given that pure BaMn$_2$As$_2$ is a local-moment Nél type antiferromagnet$^{16}$. Although density functional theory and dynamical mean-field

| Figure 3 | Constant-energy cuts of the stripe and Néel spin fluctuations in FeSe at 4 and 110 K. (a–i) Constant-energy cuts through the stripe and Néel magnetic wavevectors along the $K$ direction at 4 and 110 K. (j–r) Constant-energy cuts through the Néel magnetic wavevector $\mathbf{Q} = (1, 1)$ along the transverse direction. The scan directions are marked by the arrows in the insets. The peak positions (dispersions) are determined by fitting with Gaussian profiles convoluted with the instrumental resolution, with the Fe$^{2+}$ magnetic form factor corrected. The fitted dispersions are shown in Fig. 2. The error bars indicate 1 s.d. |
Figure 4 | Energy dependence of the local susceptibility $\chi''(\omega)$ for FeSe at 4 and 110 K. (a–c) Energy dependence of $\chi''(\omega)$, at 4 and 110 K, calculated for (a) the Néel spin fluctuations, (b) the stripe spin fluctuations, and (c) the sum of the stripe and Néel spin fluctuations. A resonance mode is clearly observed at $\sim 4$ meV and 4 K, whose intensity is consistent with the previous low-energy measurements normalized with acoustic phonons12. (d) Temperature dependence of the intensities of the stripe and Néel spin fluctuations. The data in a–c were collected on ARCS (incident energy $E_i = 294$, 79, 40 meV) and 4SEASONS ($E_i = 21$, 13.6 meV). The data in d were collected on MERLIN ($E_i = 123.4$ meV). The horizontal and vertical bars indicate the energy integration range, and the statistical errors of 1 s.d., respectively. The solid and dashed lines are a guide to the eye.

theory failed to reproduce the observed band structure of FeSe (refs 17,18), a random-phase approximation calculation with an engineered tight-binding band structure19 predicted the spin fluctuations near $(1, 0)$ and $(1, q)$. However, in this phenomenological model19, the $(1, q)$ spin fluctuation is incommensurate and gapless below $T_s$, which is inconsistent with our data. The relatively small spin-fluctuation bandwidth and large fluctuating moment together with the low-carrier density17,18 indicate that the magnetic moments in FeSe are more localized than in iron pnictides.

In the more localized case, we can exclude the previously proposed competing staggered dimer/trimers/tetramers magnetic order3 and pair-checkerboard order9. Here, the competition between the Néel and stripe magnetic instabilities could be instead understood within the framework of a frustrated $J_1−J_2$ model. In this model, for an $S = 1$ system, the Néel order is stable for $J_2/J_1 < 0.525$, whereas the stripe order is the ground state for $0.555 < J_2/J_1$ (refs 20,21). It was predicted that FeSe would be an $S = 1$ nematic quantum paramagnet in the intermediate coupling region ($0.525 < J_2/J_1 < 0.555$), which is characterized by gapped stripe and Néel spin fluctuations2,21. This agrees with our data that the majority of the spectral weight is concentrated at relatively high energies ($\sim 100$ meV) even in the presence of itinerant electrons (Figs 2a,b and 4c). Furthermore, in this scenario, the nematic order is viewed as a vestigial order that is retained when the static stripe order is suppressed by quantum fluctuations2. The finding that the stripe spin fluctuation carries considerably more spectral weight than the Néel spin fluctuation suggests that the system is indeed closer to stripe rather than to Néel order. On this basis, the temperature evolution of the spin fluctuations that we observed can be explained. As the stripe magnetic order breaks the C4 lattice symmetry, while the Néel order preserves it, the orthorhombic-/nematic-phase transition might partially lift the magnetic frustration and drive the system towards the stripe-ordered phase. Thus, the stripe spin fluctuations are enhanced, while the Néel spin fluctuations are suppressed and gapped in the nematic phase. These considerations lead to a natural understanding of the paramagnetic nematic phase in FeSe.

We now discuss the evolution of the magnetism, nematicity and superconductivity in FeSe, and its derivatives. Since the Néel spin fluctuation is gapped in the nematic phase, it is unlikely responsible for the electron pairing in bulk FeSe at ambient pressure. However, the competition between the Néel and stripe magnetic instabilities across $T_s$ suggests that the magnetic ground state and superconductivity could be highly tunable. Indeed, it has been shown that high pressure not only enhances superconductivity, but also induces static magnetic order in FeSe...
where only one type of magnetic order is observed. Our findings relate the cuprate and iron pnictide high-temperature superconductors, magnetic instabilities. This differs from the parent compounds of the Neél and stripe magnetic instabilities, which indicates a need for a comprehensive understanding of the cuprate and iron pnictide high-temperature superconductors, where only one type of magnetic order is observed. Our findings agree with a theoretical prediction that FeSe is a novel S=1 nematic quantum-disordered paramagnet interpolating between the Neél and stripe magnetic instabilities, which indicates a competition between the magnetic fields of the cuprate and iron pnictide high-temperature superconductors. The experimental determination of the nematic magnetic ground state of FeSe will be extremely valuable in identifying the microscopic mechanism of superconductivity in FeSe-based materials.

Methods

Sample growth and characterizations. Our FeSe single crystals were grown under a permanent gradient of temperature (~400–330 °C) in the KCl–AlCl3 flux. The single-crystal X-ray and neutron diffraction refinements on our samples indicated a stoichiometric chemical composition to within the error bars, and no interstitial atoms or impurity phases were observed (Supplementary Note 1; Supplementary Figs 1–3; Supplementary Table 1). The specific heat, magnetic susceptibility and resistivity measurements performed on randomly selected FeSe single crystals further demonstrate that our sample is a bulk superconductor without detectable impurities. (Supplementary Note 1; Supplementary Figs 4 and 5).

Neutron-scattering experiments. Our inelastic neutron-scattering measurements were carried out on the ARCS time-of-flight spectrometer at the Spallation Neutron Source of Oak Ridge National Laboratory, USA, and 4SEASONS chopper spectrometer at the Japan Proton Accelerator Research Complex and MERLIN chopper spectrometer at the Rutherford Appleton Laboratory, Didcot, UK. The large detector arrays on these instruments allowed us to measure spin excitations over a wide range of energy and momentum. The Q-dependent background is subtracted for the data below the aluminium phonon cutoff energy of ~40 meV (Supplementary Note 2; Supplementary Fig. 7). To facilitate comparison with theory and previous measurements, our data were normalized into absolute units by using the elastic incoherent scattering of a standard vanadium sample. The absolute intensity of the resonance mode is consistent with previous low-energy inelastic neutron-scattering measurements.

Data availability. All relevant data that support the findings of this study are available from the corresponding author on request.

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Author contributions

I.Z. conceived the research. Q.W., X.Z., D.A.C. and A.N.V. synthesized the sample. Q.W., Y.S., B.P. and X.C. characterized the sample. Q.W., Y.S. and B.P. conducted the neutron experiments with experimental assistance from K. Ikeshi, K. Iida, A.D.C.,...
D.T.A. and H.C.W. J.Z., Q.W., Y.S. and B.P. analysed the data and wrote the paper. All authors provided comments for the paper.

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