Low-energy spin fluctuations in $FeSe_{0.95}S_{0.05}$

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We report inelastic neutron scattering measurements of low-energy spin fluctuations in $FeSe_{0.95}S_{0.05}$ ($T_c =$ 10 K). Our experiments revealed a resonance mode at 3.5 meV, accompanied by a spin gap below 3 meV at the stripe wave vector in the superconducting state. The resonance mode is sharp in energy and appears at an energy level considerably lower than the superconducting gap, implying that the mode is a bound spin exciton below the superconducting gap rather than an enhanced paramagnon. An abrupt enhancement of stripe spin fluctuations is found below a nematic ordering temperature of $T_s = 80$ K. These results indicate that the direct coupling between the stripe spin fluctuations, nematicity, and superconductivity persists in sulfur doped FeSe, where superconductivity is enhanced and nematicity is partially suppressed.

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I. INTRODUCTION

Unconventional superconductors typically display a complex phase diagram with multiple symmetry breaking states [1–3]. Whether superconductivity and normal state instabilities are described by similar interactions has been the theme of many studies. One salient feature of iron-based superconductors is the existence of a nematic order, which spontaneously breaks the rotational symmetry of electrons and the crystal lattice [4–7]. Nematicity in iron-based compounds is typically accompanied by an antiferromagnetic order and complex orbital ordering [8–11]. This has generated theories that propose both spin [12–14] and orbital [15–17] fluctuations to be the dominant interaction accounting for the nematic order. In the two respective scenarios, superconducting order parameters with sign-reversed or sign-preserved symmetry have been obtained [18–25], assuming that the same interaction mediates the Cooper pairing. Thus, elucidating the interplay between nematicity, superconductivity, and the underlying interactions or fluctuations is crucial for understanding iron-based superconductivity.

Recently, the structurally simplest iron-based superconductor FeSe has received considerable attention because of its intriguing superconducting and normal state properties. On one hand, FeSe exhibits superconductivity with a highly tunable transition temperature that ranges from $T_c \sim 9$ K in the bulk form to ~65 K in the monolayer limit [26–31]. On the other hand, it displays exotic normal state properties unprecedented in other iron-based materials [32–39]. Bulk FeSe undergoes a nematic transition at $T_s \approx 90$ K, signaled by a tetragonal-to-orthorhombic structural transition, but exhibits no static magnetic order down to the lowest temperature measured at ambient pressure [40]. Although there is no long-range magnetic order, inelastic neutron scattering measurements reveal substantial spin fluctuations that are coupled with nematicity and superconductivity in FeSe [41,42]. The intimate coupling between spin fluctuations and nematicity is also manifested by the spin-excitation anisotropy that persists to very high energy (~200 meV), as is revealed by a recent resonant inelastic x-ray scattering study of detwinned FeSe [43,44]. These observations highlight the importance of spin fluctuations in the mechanism of superconductivity and nematicity in this system.

Isovalent sulfur doping can gradually suppress nematicity and enhance superconductivity, resulting in a nematic quantum critical point in $FeSe_{1-x}S_x$ at $x \sim 0.17$ [45,46]. These observations naturally raise a critical question as to how the spin degree of freedom is influenced by the tuning of nematicity and superconductivity. A recent study combining nuclear magnetic resonance (NMR) and muon spin rotation/relaxation (μSR) reported that critical spin fluctuations emerge at a temperature significantly lower than the nematic transition temperature by ~10–30 K in sulfur-doped FeSe based on which a lack of direct coupling of low-energy spin fluctuations to the nematic order was concluded [47]. However, these measurements primarily focused on momentum-integrated spin fluctuations in the zero-energy limit, and momentum-resolved spin fluctuations of the energy scale comparable to electronic nematic instability (in the order of 1–10 meV [46,48]) remain undetermined.

Here, we use inelastic neutron scattering to investigate low-energy spin fluctuations in sulfur-doped $FeSe_{1-x}S_x$ ($x = 0.05$, $T_c = 10$ K, $T_s = 80$ K) single crystals. In the normal state at $T = 12$ K, commensurate spin fluctuations are observed

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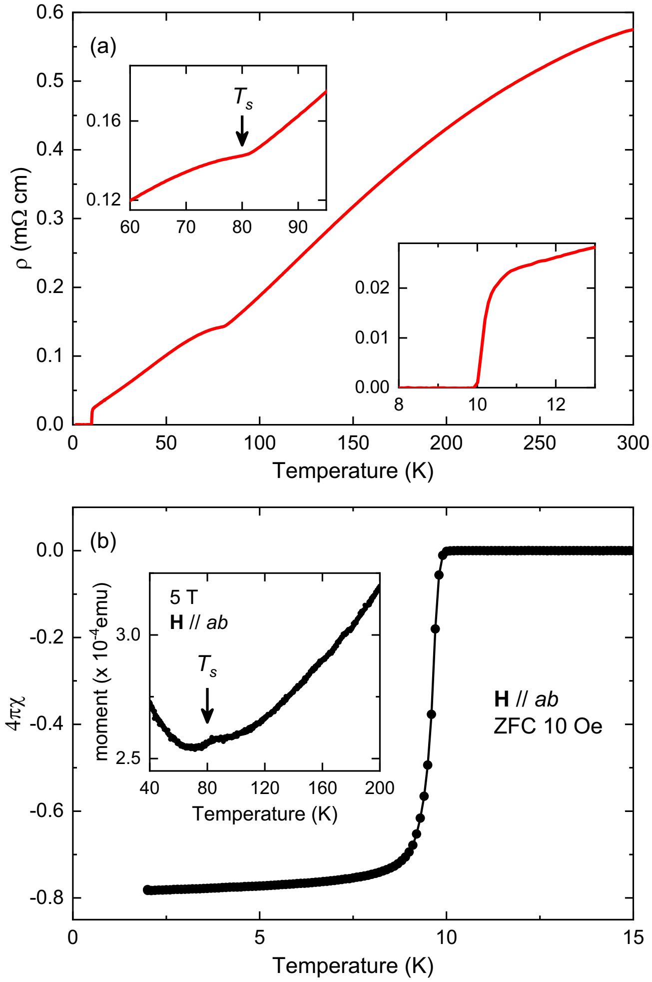

FIG. 1. (a) In-plane resistivity as a function of temperature of $FeSe_{0.95}S_{0.05}$ single crystal. The upper and low insets show data around $T_s = 80$ K and $T_c = 10$ K on an enlarged scale, respectively. (b) dc magnetic susceptibility of $FeSe_{0.95}S_{0.05}$ single crystal. A sharp superconducting transition is observed at $T_c = 10$ K in zero-field-cooled (ZFC) measurements in a magnetic field of $H = 10$ Oe. The inset shows susceptibility measured in a field of $H = 5$ T, revealing a nematic transition at T_s. Magnetic fields are applied perpendicular to the c axis.

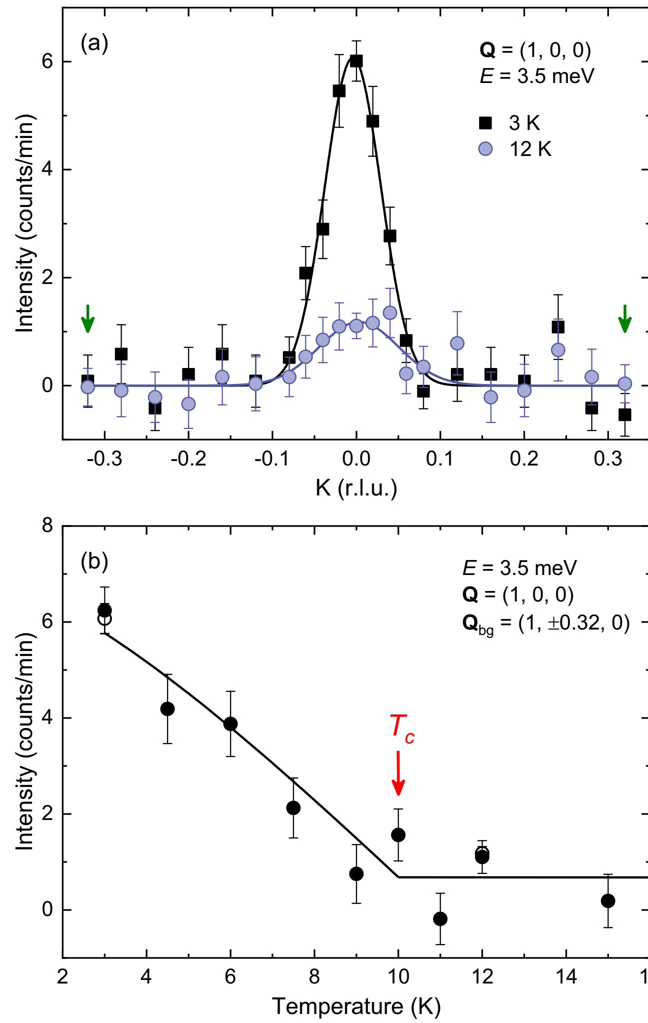

FIG. 2. (a) Momentum K scans through (1,0,0) at $E = 3.5$ meV below (black squares) and above (blue circles) T_c. The solid lines are Gaussian fits to the data. Linear backgrounds have been subtracted. (b) Temperature dependence of the magnetic scattering intensity at $E = 3.5$ meV. Background intensities are obtained at $\mathbf{Q}_{bg} = (1, \pm 0.32, 0)$—indicated by the green arrows in (a)—and subtracted. Open circles represent the peak amplitudes extracted from the fits in (a).

at the stripe antiferromagnetic wave vector $\mathbf{Q} = (1, 0, 0)$. Upon entering the superconducting state, a sharp resonance mode appears at $E_r = 3.5$ meV, below the superconducting gap [46,49], suggesting a sign-reversed pairing. Stripe spin fluctuations are strongly coupled to the nematic order. This is demonstrated by the drastic enhancement of low-energy spin fluctuations when cooling into the nematic state. The persistent direct coupling between stripe spin fluctuations and nematicity as the underlying interactions are simultaneously tuned by the chemical pressure is consistent with the spin-driven scenario of nematicity.

II. EXPERIMENTAL DETAILS

High-quality $FeSe_{0.95}S_{0.05}$ single crystals were grown using $KCl/AlCl_3$ flux under a permanent temperature gradient, as described in Ref. [50]. The temperature dependence of resistivity in the ab plane shows metallic characteristics, with a small but sharp kink at ∼80 K, indicating a nematic ordering phase transition. When the temperature is further lowered, a sharp superconducting transition at $T_c = 10$ K is observed. Superconducting and nematic transitions are further confirmed by susceptibility measurements [Fig. 1(b)]. More sample characterizations are provided in the Supplemental Material [51].

Our inelastic neutron scattering experiments were conducted using the PANDA cold triple-axis spectrometer at FRMII, Heinz Maier-Leibnitz Zentrum, Münich, Germany. A pyrolytic graphite [PG (002)] was used as the monochromator and analyzer. With the final neutron energy fixed at $E_f = 5.1$ meV, an instrumental energy resolution of 0.16 meV was obtained. Approximately 2 g of $FeSe_{0.95}S_{0.05}$ single crystals were coaligned in the $(H, K, 0)$ scattering plane. The wave vector $\mathbf{Q}$ at (q_x, q_y, q_z) was defined as $(H, K, L) =$

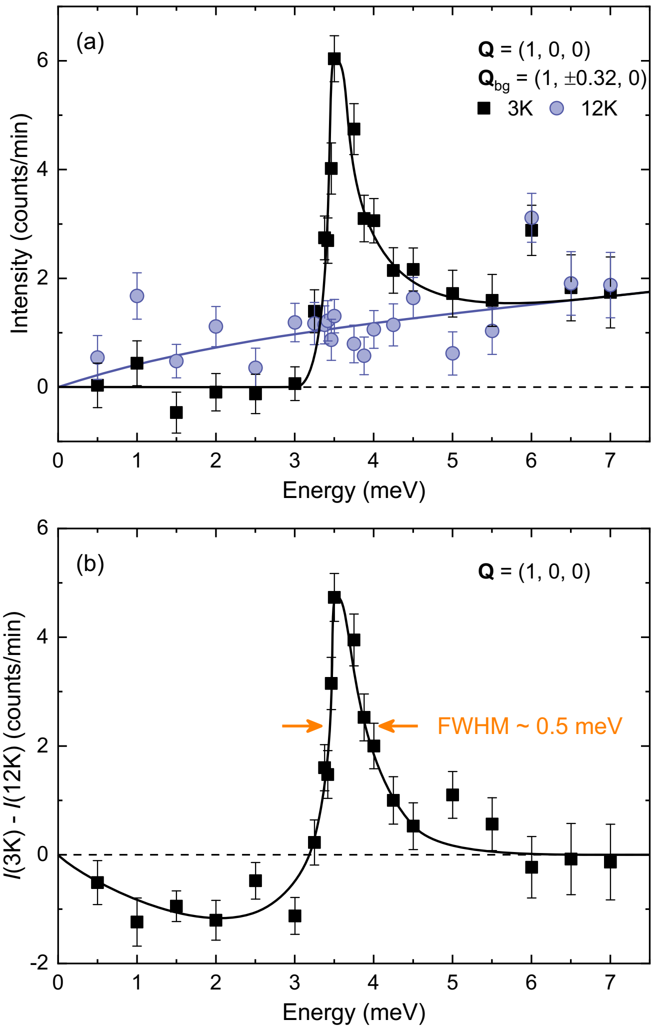

FIG. 3. (a) Low-energy stripe spin-fluctuation spectra of $FeSe_{0.95}S_{0.05}$ in the superconducting (black squares) and normal (blue circles) states. Backgrounds determined at $(1, \pm0.32, 0)$ are subtracted. (b) Intensity difference between the 3- and 12-K spectra at (1,0,0). Solid curves are guides to the eye.

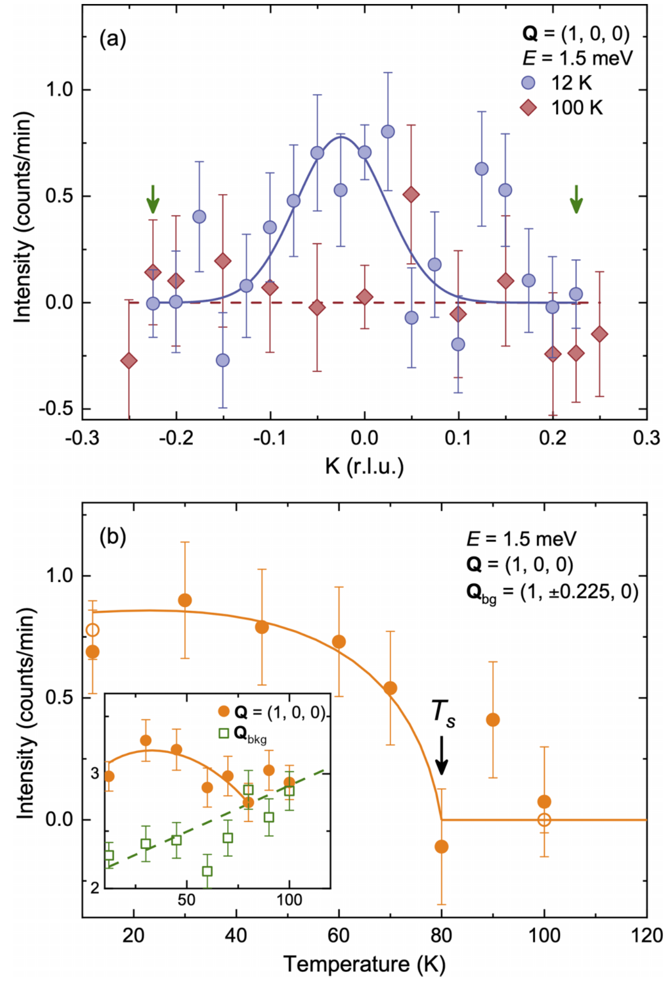

FIG. 4. (a) Momentum scans through $\mathbf{Q} = (1, 0, 0)$ at $E = 1.5$ meV along the K direction at 12 K (blue circles) and 100 K (red diamonds). The solid line is a fit using a Gaussian function. Linear backgrounds have been subtracted. (b) Temperature dependence of the spin-fluctuation intensity at $E = 1.5$ meV. Open circles represent the peak intensities extracted from the fits in (a). Background scatterings measured at $\mathbf{Q} = (1, \pm0.225, 0)$—indicated by the green arrows in (a)—are subtracted. Raw data are displayed in the inset. The solid circles and open squares denote the scattering intensities at the signal and background positions, respectively. Solid and dashed lines are guides to the eye.

$(q_x a/2\pi, q_y b/2\pi, q_z c/2\pi)$ reciprocal lattice units (r.l.u.) using an orthorhombic unit cell, with $a = b = 5.385$ Å, $c = 5.52$ Å.

III. RESULTS AND DISCUSSION

Figure 2(a) shows transverse (along the K direction) momentum scans near the stripe wave vector $\mathbf{Q} = (1, 0, 0)$ for $E = 3.5$ meV. A commensurate magnetic peak is observed at $T = 12$ K in the normal state, which is significantly enhanced when cooling below T_c. As shown in Fig. 3, the normal state spin-excitation spectrum at $\mathbf{Q} = (1, 0, 0)$ is gapless and the intensity gradually increases with energy up to 7 meV. A sharp spin resonance mode appears in the superconducting state around $E_r = 3.5$ meV below the superconducting gap [46,49]. The energy width of the resonance mode is approximately 0.5 meV [full width at half maximum (FWHM)]. These results suggest that the resonance mode is likely a bound spin exciton below the superconducting gap rather than an enhanced paramagnon excitation. We note that the energy width of the resonance mode is still slightly larger than the instrumental resolution, which could have resulted from the reduced lifetime due to doping-induced disorder. Alternatively, the anisotropy of the superconducting gap may also have broadened the mode. The temperature dependence of the spin-fluctuation intensity at $E = 3.5$ meV reveals an order-parameter-like behavior with an abrupt increase occurring at T_c [Fig. 2(b)], corroborating the intimate coupling between the stripe spin fluctuations and superconductivity. The presence of the resonance mode suggests that the Cooper pairing in this system has a sign-reversed symmetry. Along with the experimentally observed high anisotropy of the superconducting gap [46,52], this puts a strong constraint on the pairing function.

To address the relation between spin fluctuations and nematicity, we measured the evolution of stripe spin fluctuations across the nematic transition. A comparison of stripe spin fluctuations at 1.5 meV below and above $T_s = 80$ K is shown in Fig. 4(a). While a peak is observed at the stripe wave vector $\mathbf{Q} = (1, 0, 0)$ at 12 K, it vanishes at 100 K. The detailed temperature evolution reveals that low-energy stripe spin fluctuations onset at the nematic transition [Fig. 4(b)], resembling the observations in undoped FeSe [41]. We thus find a persistent coupling between spin fluctuations and nematicity when the latter is tuned by chemical pressure.

We then discuss the discrepancy between NMR/μSR and neutron scattering studies. Our previous neutron scattering measurements have revealed the coexistence of the stripe and Néel spin fluctuations in FeSe [50], suggesting a frustrated magnetic ground state. A considerable amount of spectral weight at low energy is transferred from the Néel to the stripe wave vector upon crossing into the nematic phase. This is consistent with the model that the nematic order is driven by frustrated magnetic interactions [53]. If similar magnetic frustration persists in sulfur-doped FeSe, the spectral weight change across T_s may not be resolvable by NMR and μSR measurements, because these measurements are only sensitive to momentum-integrated spin fluctuations and the increased stripe spin fluctuations below T_s may be compensated for by the spectral weight loss at the Néel wave vector. The persistent coupling between low-energy spin fluctuations and nematicity in $FeSe_{1-x}S_x$ lends strong support to the spin-driven scenario of nematicity.

Notably, the resonance energy in $FeSe_{0.95}S_{0.05}$ is slightly lower than that of undoped FeSe, even though sulfur doping enhances T_c. This suggests that the superconducting gap in $FeSe_{0.95}S_{0.05}$ may be lower than that of FeSe, assuming that the spin exciton has similar binding energies in these two compounds. A recent angle-resolved photoemission spectroscopy (ARPES) study has revealed two superconducting gaps in the hole bands in $FeSe_{1-x}S_x$ [46]. The magnitude of the larger gap in the outer hole pocket decreases with sulfur doping whereas the small gap in the inner hole pocket is essentially doping independent. This is consistent with our observation that the resonance energy does not increase with sulfur doping, which further confirms the intimate relationship between the resonance mode and superconducting gap.

To summarize, we use inelastic neutron scattering to study low-energy spin fluctuations in sulfur-doped $FeSe_{0.95}S_{0.05}$. Our data reveal a spin gap of ∼3 meV and a sharp resonance mode at $E_r = 3.5$ meV at the stripe wave vector. The evolution of the resonance mode with sulfur doping is consistent with the doping dependence of the superconducting gap determined by ARPES measurements, implying an intimate coupling between the resonance mode and superconducting gap. The stripe spin fluctuations are strongly coupled with the nematic order, evidenced by an abrupt enhancement of low-energy spin fluctuations in the nematic state. These observations are in favor of a spin-driven pairing and nematicity mechanism.

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