Spin-liquid polymorphism in a correlated electron system on the threshold of superconductivity

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We report neutron scattering measurements which reveal spin-liquid polymorphism in an “11” iron chalcogenide superconductor. It occurs when a poorly metallic magnetic state of FeTe is tuned toward superconductivity by substitution of a small amount of tellurium with isoelectronic sulfur. We observe a liquid-like magnetic response, which is described by the coexistence of two disordered magnetic phases with different local structures whose relative abundance depends on temperature. One is the ferromagnetic (FM) plaquette phase observed in undoped, nonsuperconducting FeTe, which preserves the C_4 symmetry of the underlying square lattice and is favored at high temperatures, whereas the other is the antiferromagnetic plaquette phase with broken C_4 symmetry, which emerges with doping and is predominant at low temperatures. These findings suggest the coexistence of and competition between two distinct liquid states, and a liquid–liquid phase transformation between these states, in the electronic spin system of $\text{FeTe}_{1-x}(\text{S,Se})_x$. We have thus discovered the remarkable physics of competing spin-liquid polymorphs in a correlated electron system approaching superconductivity. Our results facilitate an understanding of large swaths of recent experimental data in unconventional superconductors. In particular, the phase with lower C_2 local symmetry, whose emergence precedes superconductivity, naturally accounts for a propensity for forming electronic nematic states which have been observed experimentally, in cuprate and iron-based superconductors alike.

spin liquid | magnetism | superconductivity | neutron scattering | polymorphism

Ideas about liquid polymorphism date as far back as Mendeleev’s work on water–alcohol mixtures and Röntgen’s proposals that many of the anomalous properties of water, such as its density maximum at 4 °C, can be understood if water is viewed as a mixture of two liquid “species,” different in local structure and density (1–3). The possible existence of different liquid polymorphs and liquid–liquid phase transformations (LLPT) between such polymorphs in simple molecular fluids continues to receive considerable attention, but the issue has not been settled yet (4–10). This is mainly because competition between different liquid phases is governed by the energy balance of intermolecular interactions and is thus expected only at low temperatures, where it outweighs thermal entropy. For most simple liquids this is expected only in a deeply supercooled region, well below the freezing temperature (4–10). On the other hand, such competition arises quite naturally in a system of atomic magnetic moments in a crystal, where the development of magnetic order is hindered by competing interactions (frustration) (11, 12), reduced dimensionality (13, 14), or both (15). In these cases, the system remains disordered even at temperatures much lower than the energy of magnetic interactions, thus realizing a spin-liquid state. Hence, many exotic quantum properties of liquids have been discovered by studying spin liquids (11–15).

Liquid-like states play a prominent role in theories of unconventional superconductivity in cupric oxides and iron pnictides and chalcogenides, which feature 2D square nets of magnetic atoms with a strongly reduced tendency to order magnetically (16–18).

There is strong experimental evidence that superconductivity in these materials is coupled with antiferromagnetic correlations, which could be responsible for electron pairing (18). A simple, pedestrian way to visualize such coupling is to imagine that an electron moving in a spin-liquid background, by virtue of the hybridization of its wave function with those of magnetic electrons, is “dressed” by a magnetic polaron, and that this magnetic polarization cloud plays a similar role to the polarization of atomic displacements in the phonon mechanism of Cooper pairing in conventional superconductors. The presence of delocalized electrons interacting with the system of atomic spins also affects the nature of the spin-liquid state: The emerging patterns of short-range magnetic correlation not only have to optimize the orbital overlap energy of localized valence electrons (spin superexchange) but also the hybridization energy with the delocalized electron.

Motivated to uncover magnetic signatures of electronic pairing interactions, we carried out neutron scattering studies of dynamical magnetic correlations in $\text{FeTe}_{0.87}\text{S}_{0.13}$, an iron chalcogenide of “11” superconductor family, where substitution of only 13% of Te with S, far below any percolation phenomena, leads to the onset of superconductivity (19). Through neutron scattering measurements, we have discovered not only the hallmark of spin-liquid behavior, but also clear evidence of an LLPT between two spin-liquid polymorphs, one featuring ferromagnetic (FM) plaquettes present in the parent Fe_{1+y}Te compound (20), and the new one, which can be associated with emergent superconductivity in $\text{FeTe}_{1-x}(\text{S,Se})_x$ materials (21–31) (see also the *Supporting Information*).

The experimental signatures of a liquid arise from well-defined microscopic local structures, fluctuating in time, that balance the energy of intermolecular interactions and the entropy of thermal motion, which are at the origin of liquid state (4–10). In a glass, another common amorphous state of condensed matter which is solid, the local order is static and survives averaging over practically

Significance

Liquid polymorphism and liquid–liquid phase transformations are remarkable phenomena of great fundamental and practical interest, as is unconventional superconductivity. Yet, despite being intensely studied, they still remain enigmatic. Our results not only provide a rare experimental example of liquid polymorphism, which occurs in the spin system of an iron chalcogenide superconductor, but also establish its unexpected connection with the onset of unconventional superconductivity.

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infinite time, giving rise to elastic scattering. In a liquid it is dynamic, so a well-defined local structure is only present as an instantaneous snapshot. The corresponding equal-time correlation function is obtained by energy-integrating the inelastic scattering signal measured in a neutron scattering experiment, where energy- and wave-vector-dependent patterns in Fourier space arising from time- and space-dependent real-space correlations are detected (32). For a system of electronic spins in a crystal, it is the correlation of magnetization fluctuations, rather than of atomic positions, that determines the character of the equilibrium state. The hallmark of a spin liquid is the presence in magnetic neutron scattering intensity, in a broad range of energies, of a well-defined diffuse pattern in the wave-vector space, which is characteristic of a particular short-range order. Indeed, if such a pattern persists in the energy range containing the dominant portion of scattering spectral weight, then this same pattern will dominate the single-time correlation describing the instantaneous local spin structure of the liquid state.

A liquid-like, diffuse scattering pattern persisting in a wide energy range appears as a broad vertical column of intensity in the (Q, E) coordinate plane cutting the four-dimensional (Q, E) phase space along a particular wave-vector trajectory (15). This is precisely how the measured intensity is shaped in $\text{FeTe}_{0.87}\text{S}_{0.13}$ (Fig. 1). In Fig. 1 *A–C*, the (Q, E) slice along the $(h, 1-h)$ line of the 2D Brillouin zone corresponding to the $a-b$ plane of Fe square lattice is shown, as it evolves upon cooling from 300 to 3 K. At $h = 0.5$ this trajectory passes through $Q = (\pi, \pi)$, the point where spin resonance at $E \approx 7$ meV develops in the superconducting $\text{FeTe}_{1-x}(\text{S,Se})_x$ samples (21, 22). We observe no resonance even at 3 K, which is below the superconducting

transition temperature $T_c \approx 8$ K of our material (19). This is consistent with the filamentary character of superconductivity (19), which leaves the bulk of the sample probed in our neutron experiment near critical doping and at the verge of being superconducting. The columns of magnetic scattering appear at $h \approx 0.3$ and ≈ 0.7 in the units of P4/nmm ($a = b \approx 3.8$ Å) reciprocal lattice (Fig. 1 *D–F*). This pattern is strikingly ubiquitous in $\text{FeTe}_{1-x}\text{Se}_x$ samples where superconductivity is partially or completely suppressed by imperfect composition, Fe substitution with Cu or Ni, or temperature (21–31).

The scattering we observe in $\text{FeTe}_{0.87}\text{S}_{0.13}$ does bear some similarities with the parent Fe_{1+y}Te material: it is consistent with a similarly large Fe local moment, which is also obtained from Curie–Weiss analysis of the magnetic susceptibility (19), is stronger near $Q = (\pi, 0)$ (Fig. 1 *G* and *H*) and increases in intensity upon warming, which indicates thermally enhanced electronic localization. However, there is also a marked distinction, which renders the four-spin up-up-up-up (UUUU) FM plaquette local structure of Fig. 24 inapplicable. In Fe_{1+y}Te , where the overwhelming majority of magnetic spectral weight is also inelastic and spin-liquid-like, it can be very accurately described by the short-range antiferromagnetic (AFM) correlation of UUUU plaquettes illustrated in Fig. 2 *A* and *D* (20). The scattering intensity in this model is governed by the UUUU plaquette structure factor, which is exactly zero along the $(h, 1-h)$ line and its square-symmetric replica, giving rise to zero-intensity square in Fig. 2*D*. Hence, whereas the practical absence of scattering along these lines in Fe_{1+y}Te provides a unique fingerprint of the FM plaquette liquid, this model fundamentally cannot account for the intensity observed in $\text{FeTe}_{0.87}\text{S}_{0.13}$ (Fig. 1 *A–C*) (20).

The spatial structure of short-range spin-liquid correlations is revealed by constant-energy maps of the magnetic scattering in Figs. 3 and 4. The first important observation here is that all scattering features, including the characteristic ridges, which extend diagonally from $(\pm 0.5, 0)$ and $(0, \pm 0.5)$ positions and correspond to diffuse columns in Fig. 1 *A–C*, are extremely broad. Resolution-corrected Lorentzian fits to the data shown in Fig. 1 *D–H* suggest a correlation length of only about one lattice repeat, which is the distance between the diagonal next-nearest-neighbor Fe atoms. Hence, the elementary local plaquette describing such correlations cannot consist of more than four Fe sites; the calculated structure factor for an ordered cluster of eight spins already has features that are sharper than those seen in Figs. 3 and 4. Among all four-spin plaquettes, the best fit to our data is provided by the slanted AFM up-down-up-down (UDUD) plaquette shown in Fig. 2 *B* and *E*. In hindsight, the presence of this particular local order is quite natural: Its long-range wave-like propagation with wave vector $(\pi, 0)$ describes bicollinear AFM partial order observed in Fe_{1+y}Te , suggesting that the corresponding spin-liquid state (Fig. 2*B*) is close in energy.

Surprisingly, the ridges characteristic of the AFM UDUD plaquettes in $\text{FeTe}_{0.87}\text{S}_{0.13}$ become less pronounced upon warming (Fig. 3 *B* and *C*), whereas the scattering intensity shifts to smaller Q , acquiring a shape reminiscent of the UUUU plaquette liquid in Fe_{1+y}Te (Fig. 2*D*). Similar temperature evolution is seen even more clearly in the pattern of (quasi-)elastic scattering (Fig. 4), which exhibits the characteristic square pattern already at 100 K. We therefore introduce a model where the two contributions coexist, and refine the relative composition of the two liquid components by fitting the data at different temperatures. Although this model is not very sensitive to small admixtures of one of the components, it clearly shows that UUUU plaquettes form a minority at 3 K, but a majority at 300 K. That such a simple model, with only few parameters, provides such an accurate description of all experimental data (see also the [Supporting Information](#)) is strong evidence that it correctly captures the physics of the observed spin-liquid state, which is a mixture of two components with different local structures.

Fig. 1. Dynamical magnetic correlations in $\text{FeTe}_{0.87}\text{S}_{0.13}$ measured by inelastic neutron scattering. (*A–C*) (Q, E) intensity maps along $(h, 1-h, 0)$ direction in the $a-b$ plane, (c1), measured at 298, 100, and 3.1 K, respectively. (*D–F*) Corresponding panels (*Right*) show representative constant- E linear scans with resolution-corrected Lorentzian fits; the intensity is offset by adding $(E-3) \times 30$ counts to avoid overlaps. Dashed lines show the average peak positions. (*G*) (Q, E) intensity map, and (*H*) representative scans along $(0.5, k, 0)$ direction, (g1), at $T = 5$ K. Ellipses show the FWHM of the instrument resolution.

Fig. 2. Electronic density and magnetization correlation in plaquette spin-liquid models. (A–C) Height shows correlated electronic density corresponding to the isotropic atomic magnetic form factor of Fe^{2+} and enlarged by the covalence factor of 2, which best fits the data in Fig. 3. Color represents the amplitude, in arbitrary units, of magnetic correlation for the interplaquette correlation length equal to two nearest-neighbor Fe–Fe spacings. A damped-wave correlation of four-iron FM UUUU plaquettes, propagating with the wave vector $(\pi, 0)$, gives rise to short-range checkerboard in FeTe (A). For AFM UDUD plaquettes emergent in $\text{FeTe}_{1-x}(\text{S,Se})_x$, a correlation with wave vector $(\pi, 0)$ gives short-range bistrispe (B), whereas for wave vector (π, π) it gives collinear antiferromagnetism observed in iron pnictides (C). (D–F) show pattern of scattering intensity corresponding to the model on the left, averaged over possible plaquette orientations to restore the macroscopic C_4 symmetry.

In the Fe_{1+y}Te parent material, the dominant UUUU plaquette correlation persists for temperatures up to at least $T_r = 300$ K, and only increases in intensity upon warming, as a result of thermally induced localization of weakly itinerant electrons (the effective spin localized on each Fe atom changes from $S = 1$ at $T = 10$ K to $S = 3/2$ at 300 K). That this correlation is so insensitive to thermal disorder suggests that formation of the UUUU plaquette spin liquid in Fe_{1+y}Te is governed by the hybridization energy notably exceeding $k_B T_r \approx 30$ meV. These very strong interactions define the energy–time domain of liquid behavior. The slow, dissipative fluid dynamics which preserves the local plaquette structure in this domain is governed by the residual balance of interplaquette interactions. It remained a puzzle, why partial bicollinear magnetic order observed in Fe_{1+y}Te below $T_N \approx 60$ K, despite only affecting a minority of the spectral weight, does not accommodate UUUU plaquettes, thus corresponding to a different liquid state. One solution to this puzzle is offered by the (co)existence of two competing spin-liquid polymorphs, only one of which crystallizes into an ordered structure on cooling. Our present results indeed provide strong experimental support for this idea. A fraction of magnetic spectral weight in $\text{FeTe}_{0.87}\text{S}_{0.13}$ also freezes in a short-range glassy state below ≈ 50 K, corroborating that UDUD

plaquettes are favored by energy, rather than by entropy, which favors higher-symmetry UUUU plaquettes. Thus, the parent, higher-temperature phase can be thought of as a classical spin liquid, as both the electronic decoherence, and the higher, C_4 local symmetry of the FM plaquette, are favored by thermal rather than quantum fluctuations.

A closer inspection of the scattering pattern in the UDUD plaquette model as a function of the interplaquette correlation length ξ leads to another surprising discovery. Recently, a variety of seemingly distinct, rather mysterious, and ill-understood neutron patterns, including the spin resonance, was observed by different groups in various $\text{FeTe}_{1-x}\text{Se}_x$ samples, both with suppressed and well-developed superconductivity (21–31). In the absence of even a qualitative description, these results were often compared with ad hoc functions, such as a Sato–Maki function, leading to data parameterizations void of much physical meaning (22). Remarkably, it appears that all of the variety of the observed neutron patterns can be qualitatively well-described by the UDUD plaquette model with only slightly varying interplaquette correlation length, $\xi \sim 1 - 2$ lattice units. Hence, they all manifest very similar local correlation, revealing amazing intrinsic universality uncovered by our real-space model (Supporting Information).

That the observed correlation length is very short is not surprising. Long-range phases are suppressed in layered, quasi-2D systems with disorder (33), and strong quenched disorder is invariably present in doped samples. However, the low-symmetry phase can still be manifest as the “vestigial nematicity,” which survives to finite disorder strength (34). The local C_4 symmetry breaking can also be detected as a localized electronic nematic order in scanning tunneling spectroscopy (35). In principle, the correlation length can be increased by reducing disorder, or by applying an order-favoring field such as the macroscopic strain,

Fig. 3. Neutron scattering pattern of dynamical magnetic correlations in the $a - b$ plane at constant energy $E = 6$ meV. (A–C) data at $T = 3.1$, 100, and 298 K, respectively, as a function of the in-plane wave vector $(h, k, 0)$. (D–F) Fits of the above data to the model of a mixture of two types of plaquette spin liquids shown in Fig. 2. The calculated intensity is orientation-averaged to restore the macroscopic C_4 symmetry observed in experiment. In addition to the background and the intensity scale factor, the interplaquette correlation length, ξ , covalent compression of the magnetic form factor, and the percentage fraction of the FM UUUU plaquettes were varied in these fits. Ellipses show the FWHM of the instrument resolution; dotted lines show locations of cuts presented in Fig. 1.

Fig. 4. Quasi-elastic magnetic correlations measured at $E = 0$, showing most clearly change in the nature of dominant magnetic plaquette with temperature. (A and B) Elastic intensity at 4 and 100 K, respectively. Data in B were measured for $k < 0$ and then symmetrized with respect to $k = 0$. (C and D) Fits of the corresponding data on the left to the model of a mixture of UDUD plaquettes (E), prevailing at low temperature, and the UUUU (FM) plaquettes (F), emergent on warming.

which could lead to a long-range order amenable to detection in macroscopic measurements (36).

Thus, we find that the UDUD plaquette spin liquid emerges with doping in $\text{FeTe}_{1-x}(\text{S,Se})_x$, as it begins to develop superconductivity. It breaks the C_4 symmetry of the square lattice, and therefore can be related to nematicity observed in many unconventional superconductors (18, 35). It is favored energetically, and in FeTe is stabilized by bicollinear static antiferromagnetism. Symmetric UUUU plaquettes, on the other hand, are favored by entropy and are preferred at high temperature, as we observe. Our findings suggest that a liquid–liquid phase transformation occurs in the system of electronic spins in FeTe when it is doped, with Se or S, to become a superconductor. The newly emerging slanted UDUD plaquettes in all probability reflect a new electronic hybridization pattern, which is likely favored by the shift of atomic energy levels that results from doping, and which facilitates electron

pairing. The time scales of the local plaquette–liquid fluctuations are controlled by strong electronic Coulomb interactions and hybridization, thus suggesting an antiadiabatic pairing mechanism. The structure of a plaquette is reflective of particular electronic Wannier functions, which define the orbital content of electronic bands in the tight-binding view. That this change of the hybridization pattern belongs to the low-energy domain such that it is affected not only by small amounts of dopant atoms, but also by temperature, is a surprising discovery that calls for a profound revision of the tight-binding paradigm. So is the remarkable competition of two different electronic spin-liquid polymorphs in a material on the threshold of unconventional superconductivity, which suggests new understanding of electronic nematicity and non-Fermi-liquid behavior (18, 34–36).

Materials and Methods

The $\text{FeTe}_{1-x}\text{S}_x$ [$x = 0.13(1)$, $m = 2.6$ g] crystal used in our present measurements was grown from Te–S self-flux using a high-temperature flux method (19) and had a mosaic of $<1^\circ$ full width at half maximum (FWHM). Elemental composition, structure, and bulk properties, including the superconducting transition, of a small piece of the same crystal were characterized in ref. 19. The sample was mounted on an aluminum holder attached to the cold head of a closed-cycle refrigerator on the HB3 triple-axis spectrometer at the High Flux Isotope Reactor at Oak Ridge National Laboratory. Measurements in Figs. 1–4 were performed in the $(h, k, 0)$ zone with the crystal c -axis vertical; measurements in the $(h, 0, l)$ zone are presented in the [Supporting Information](#). Constant-energy scans were acquired by varying the wave-vector transfer for fixed monitor counts in a low-efficiency detector between the pyrolytic graphite [PG (002)] monochromator and the sample and with fixed scattered neutron final energy $E_f = 14.7$ meV selected by a PG(002) analyzer. Beam angular collimation was set to $48^\circ\text{--}60^\circ\text{--}80^\circ\text{--}240^\circ$, from monochromator to detector, and a polycrystalline PG filter after the sample was used to minimize intensity at harmonics of the desired wavelength. The resultant energy resolution measured by the FWHM of the incoherent elastic scattering signal was 1.35 meV. The wave-vector resolution calculated using standard methods ([Supporting Information](#)) is shown by FWHM ellipses in Figs. 1–4. The ambient neutron background (BG) of 9 cpm was measured with Cd-blocked analyzer entrance and subtracted from all data. Q -independent BG arising from incoherent elastic scattering in the sample subtracted in Fig. 4 was estimated by averaging $<1\%$ of lowest-intensity points.

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