Anomalous high-magnetic field electronic state of the nematic superconductors FeSe$_{1-x}$S$_x$

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Understanding superconductivity requires detailed knowledge of the normal electronic state from which it emerges. A nematic electronic state that breaks the rotational symmetry of the lattice can potentially promote unique scattering relevant for superconductivity. Here, we investigate the normal transport of superconducting FeSe$_{1-x}$S$_x$ across a nematic phase transition using high-magnetic fields up to 69 T to establish the temperature and field dependencies. We find that the nematic state is dominated by a linear resistivity at low temperatures that evolves towards Fermi-liquid behavior, depending on the composition $x$ and the impurity level. Near the nematic end point, we find an extended temperature regime with $\sim T^{1.5}$ resistivity, different from the behavior found near an antiferromagnetic critical point. The variation of the resistivity exponent with temperature reflects the importance of the nematoelastic coupling that can also suppress divergent critical fluctuations at the nematic end point. The transverse magneto-resistance inside the nematic phase has a $\sim H^{1.55}$ dependence over a large magnetic field range and it displays an unusual peak at low temperatures inside the nematic phase. Our study reveals anomalous transport inside the nematic phase, influenced by both changes in the electronic structure and the scattering with the lattice and spin fluctuations.

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Magnetic field is a unique tuning parameter that can suppress superconductivity to reveal the normal low-temperature electronic behavior of many unconventional superconductors [1,2]. High-magnetic fields can also induce new phases of matter, probe Fermi surfaces, and determine the quasiparticle masses from quantum oscillations in the proximity of quantum critical points [1,3]. In unconventional superconductors, close to antiferromagnetic critical regions, an unusual scaling between a linear resistivity in temperature and magnetic fields was found [4,5]. Magnetic fields can also induce metal-to-insulator transitions, as in hole-doped cuprates, where superconductivity emerges from an exotic electronic ground state [2].

FeSe is a unique bulk superconductor with $T_c \sim 9$ K, which displays a variety of complex and competing electronic phases [6]. FeSe is a bad metal at room temperature and it enters a nematic electronic state below $T_T \sim 87$ K. This nematic phase is characterized by multiband shifts driven by orbital ordering that lead to Fermi-surface distortions [6,7]. Furthermore, the electronic ground state is that of a strongly correlated system and the quasiparticle masses display orbital-dependent enhancements [7,8]. FeSe shows no long-range magnetic order at ambient pressure, but complex magnetic fluctuations are present at high energies over a large temperature range [9]. Below $T_T$, the spin-lattice relaxation rate from nuclear magnetic resonance (NMR) experiments is enhanced as it captures the low-energy tail of the stripe spin fluctuations [10,11]. Furthermore, recent $\mu$SR studies invoke the close proximity of FeSe to a magnetic quantum critical point as the muon relaxation rate shows unusual temperature dependence inside the nematic state [12].

The changes in the electronic structure and magnetic fluctuations of FeSe can have a profound implication on its transport and superconducting properties. Scanning tunneling microscopy (STM) reveals a highly anisotropic superconducting gap driven by orbital-selective Cooper pairing [13]. Due to the the presence of the small inner bands, whose Fermi energies are comparable to the superconducting gap, FeSe was placed inside the BCS-BEC crossover regime [14]. In large magnetic fields, when the Zeeman energy is comparable to the gap and Fermi energies, a peculiar, highly polarized superconducting state may occur [14].

To establish the role played by different competing interactions on nematicity and superconductivity, an ideal route is provided by the isoelectronic substitution of selenium by sulfur ions in FeSe$_{1-x}$S$_x$ [15]. This tuning parameter suppresses
nematicity and leads to changes in the electronic structure, similar to the temperature effects, with the Fermi surface becoming isotropic in the tetragonal phase and the electronic correlations becoming weaker [3,6,15,16]. As nematicity is suppressed, it creates ideal conditions to explore a potential nematic critical point [17] in the absence of magnetism. The superconducting dome extends outside the nematic state, but anisotropic pairing remains robust [18], and a different superconducting state was suggested to be stabilized in the tetragonal phase [19].

In this paper, we study the normal electronic state across the nematic transition in FeSe$_{1-x}$S$_x$ using magnetotransport studies in high-magnetic fields up to 69 T. We find that the nematic state shows unusual transport behavior with temperature and transverse magnetoresistance ($\sim H^{1.55}$), reflecting an unconventional scattering mechanism. Just outside the nematic phase, resistivity is dominated by a $\sim T^{1.5}$ dependence, similar to studies under pressure [20]. The transverse magnetoresistance is significant inside the nematic phase and it shows an unusual change in slope at low temperatures. Inside the nematic phase at low temperatures, we find linear resistivity followed by Fermi-liquid behavior for certain $c$ and impurity levels. We identify a qualitative link between spin fluctuations and the linear resistivity regime. The variation of the resistivity exponent with temperature suggests that the nematoelastic coupling plays an important role in these systems [21,22].

Methods. Single crystals of FeSe$_{1-x}$S$_x$ were grown by the KCl/AlCl$_3$ chemical vapor transport method [23]. The composition for samples from the same batch was checked using energy-dispersive x-ray (EDX) spectroscopy, as reported previously in Ref. [3]. Note that in Refs. [24,25], the nominal $x_{\text{nom}}$ is used to identify the composition (which is the value used during the growth process), which typically corresponds to about 80% of the real $x$ (see also Refs. [3,17,26]). The structural transition at $T_S$ also provides useful information about the expected $x$ value, as shown in Fig. S1 in the Supplemental Material (SM) [27]. More than 30 samples were screened for high-magnetic field studies to test their physical properties. The residual resistivity ratio varies between 15 and 25 in the tetragonal phase above $T_S$, and its magnitude across the phase diagram, as shown in Figs. 1(f) and 1(g). The resistivity slope $d\rho_{xx}/dT$ in 34 T of FeSe changes sign around a crossover temperature, $T^* \sim 14$ K, as shown in Fig. 1(f) [also in the color plot of the slope in Fig. 3(d)]. With increasing sulfur substitution from FeSe towards $x \sim 0.07$ (defined as the nematic A region), the position of $T^*$ shifts to a slightly higher temperature of $\sim 20$ K, and the peak in magnetoresistance is much smaller than for FeSe. For higher concentrations, approaching the nematic phase boundary ($x \sim 0.11$–0.17 defined as the nematic B region), there is a small peak at $T^*$ but the negative slope $d\rho_{xx}/dT$ in 34 T is strongly enhanced at low temperatures, which is different from the nematic A phase [see Figs. 1(h), 1(i) and 3(d)]. Lastly, in the tetragonal phase, the magnetoresistance shows a conventional behavior and increases quadratically in magnetic fields [Figs. 1(e) and 1(j)].

The unusual downturn in resistivity in high-magnetic fields below $T^*$ inside the nematic A phase was previously assigned to large superconducting fluctuations in FeSe in magnetic fields up to 16 T [10,11]. We find that this behavior remains robust in magnetic fields that are at least a factor of 2 higher than the upper critical field of $\sim 16$ T for $H/\mu_0$ [10]. Furthermore, it also manifests in $x \sim 0.07$ inside the nematic A phase, but it disappears for higher $x \geq 0.1$. As $T_S$ and the upper critical field inside the nematic phase for different $x$ remain close to that of FeSe [3,28], the changes in the resistivity slope in high magnetic fields are likely driven by field-induced effects that influence scattering and/or the electronic structure.

The Hall coefficient, $R_H = \rho_{xy}/\mu_0 H$, extrapolated in the low-field limit (below 1 T) for FeSe$_{1-x}$S$_x$ has an unusual temperature dependence, as shown in Fig. 2(b). For a compensated metal, the sign of the Hall coefficient depends on the difference between the hole and electron mobilities [29]. In the tetragonal phase above $T_S$ and for $x \geq 0.18$, $R_H$ is close to zero [Fig. 2(b)], as expected for a two-band compensated metal. On the other hand, in the low-temperature nematic A phase, the sign of $R_H$ is negative, suggesting that transport is dominated by a highly mobile electron band [15,30]. It becomes positive inside the nematic B phase, dominated by a hololead-like band [Fig. 2(a)]. It is worth mentioning that inside the nematic B phase, the quantum oscillations are dominated by a low-frequency pocket with light mass that disappears at the nematic end point [3]. Thus, the behavior of $R_H$ is...
Deep inside the nematic phase, the inner hole band and inner electron bands are brought in the vicinity of the Fermi level. A magnetic field can induce changes in scattering and in multiband systems that have pockets with small Fermi energy is close to the Zeeman energy, in particular field-induced Fermi-surface effects in the limit when the symmetry points at the top of the Brillouin zone, \(x\) in FeSe below \(T_s\) and for different \(x\) (based on ARPES data reported in Refs. [6,7,15,16]). The horizontal lines represent the location of distinct regions in the magnetotransport behavior called nematic A \((x = 0.07)\), nematic B \((x = 0.11, 0.17)\), and the tetragonal phase for \(x \gtrsim 0.18\). In the tetragonal phase, the compensated semimetal is formed of two electron and two holelike bands. Deep inside the nematic phase, the inner hole band and inner electron bands are brought in the vicinity of the Fermi level.

A near-linear magnetoresistance is detected for \(x \sim 0.07\) in Fig. 1(b) and for a “dirtier” sample with lower residual resistivity ratio of 8.5 (that can result from defect concentrations like dislocations and vacancies) in Fig. S9 in the SM [27]. The quasilinear field magnetoresistance at low temperature can arise from squeezed trajectories of carriers in semiclassically large magnetic fields in the case of small Fermi surfaces \((\omega \tau \gg 1)\) [32,33]. Another explanation for an almost linear magnetoresistance is the presence of mobility fluctuations caused by spatial inhomogeneities, as found in low carrier density systems [33–35].

Classical magnetoresistance in systems with a single dominant scattering time is expected to follow a \(H^2\) dependence [32]. This results in Kohler’s rule, which is violated in FeSe\(_{1-x}\)S\(_x\), suggesting that the magnetoresistance is not dominated by a single scattering time, as shown in Figs. S2(a)–S2(c) in the SM [27]. Magnetoresistance is quadratic in magnetic fields up to 69 T in the tetragonal phase \((x \gtrsim 0.19)\) (see Fig. 1(e) and Figs. S4(e) and S4(f) in the SM [27]), but not inside the nematic phase. FeSe\(_{1-x}\)S\(_x\) are compensated multiband systems [6] where the high-field magnetoresistance is expected to be very large and dependent on the scattering times of the electron and hole bands [29]. As these systems are very clean and quantum oscillations have been observed, the overall estimated value for the \(\omega \tau\) is 1.4 in 5 T, which places these systems in the high-field limit where the details...
of the Fermi surface and unusual type of scattering may become important [29,32]. Magnetoresistance has a complex form and instead simpler scaling have been sought to reveal its importance, in particular in the vicinity of critical points [4,5]. For example, in BaFe$_2$(As$_{1−x}$S$_x$)$_2$ for $x \sim 0.33$ at the antiferromagnetic critical point, a universal $H−T$ scaling was empirically found between the linear resistivity in temperature and magnetic field [4]. For FeSe$_{1−x}$S$_x$ near the nematic end point at $x \sim 0.17$, we find that a $H−T$ dependence collapses onto a single curve, as shown in Fig. 2(e) in the SM [27]. Despite this, the energy scaling of magnetoresistance used to described the antiferromagnetic critical point in Ref. [4] is not obeyed in the vicinity of the nematic end point in FeSe$_{1−x}$S$_x$, as detailed in Figs. 2(g)−2(i) in the SM [27]. This could be due to additional constraints to be included either to account for the nematoleastic coupling [21] and/or the effect of additional effects induced by sulphur substitution. For example, a very dirty sample of FeSe$_{1−x}$S$_x$ close to $x_{nom} \sim 0.18$ was recently suggested to obey $H−T$ scaling [25].

For reasons described above, we propose a different approach to model the magnetoresistance data in the nematic state of FeSe$_{1−x}$S$_x$, using a power law in magnetic fields given by $\rho_{xx}(H) = \rho_{H=0} + bH^\delta$ for each temperature. Strikingly, we find that all the magnetoresistance data inside the nematic phase can be described by a unique exponent $\delta \sim 1.55(5)$ over a large field region, as shown by the color plot in Fig. 3(c) as well as in Figs. 2(c) and Figs. S4(a)−S4(d) in the SM [27]. A detailed method of the extraction of $\delta$ and its stability over a large temperature and field window is shown in Fig. S3 in the SM [27]. Furthermore, this gives $\delta \sim 2$ for samples in the tetragonal phase [see Fig. 3(e)]. Inside the nematic phase, the Fermi surface of FeSe$_{1−x}$S$_x$ distorts anisotropically [6,7] and an unusual type of scattering could become operational due to the presence of hot and cold spots along certain directions [36].

In the absence of magnetic field, the transport behavior can also be described by a power law, $\rho(T) = \rho_0 + AT^\gamma$. Figure 3(a) shows a color plot of the exponent $\gamma$, which is close to unity at low temperatures inside the nematic phase and becomes sublinear close to the nematic phase boundary, indicating a significant deviation from Fermi-liquid behavior (a value of $\gamma = 1.1(2)$ was previously reported for FeSe [37]). Outside the nematic phase, a $T^{1.5}$ dependence of resistivity describes the data well over a large temperature range up to 120 K [see Figs. 2(a) and 3(a)], in agreement with previous studies of FeSe$_{1−x}$S$_x$ under pressure [20]. Using the high-magnetic field data below $T_n$, we extract the low-temperature normal resistivity in the absence of superconductivity, $\rho(T→0)$. Figures 2(d)–2(f) show resistivity against temperature for different values of $x$, together with the extrapolated high-field points, using longitudinal magnetoresistance when $H||\langle ab\rangle$ plane, shown in Figs. S5 and S6 in the SM [27]. We also use transverse magnetoresistance data (in the regime where quantum oscillations were not dominating the response) to extract the zero-field resistivity, using the established power law $H^{1.55}$, as shown in detail in Figs. S4 and S7 in the SM [27]. From both measurements, we find evidence for a linear resistivity in the low-temperature regime, below $T^*$, inside the nematic phase. At low temperatures, we observe that Fermi-liquid behavior recovers in the tetragonal phase (see, also, Refs. [24,38]) and inside the nematic phase, below $T_{FL}$ [see Figs. 2(d)−2(f) and 3(b)]. This is strongly dependent on composition and impurity level, even in the vicinity of the nematic end point (see Figs. S8 and S9 in the SM [27]).

We find that $T_{FL}$ is highest for the samples with the largest residual resistivity ratio (above $≈16$) (see Figs. S1(c) and S6 in the SM [27]).

A related study of FeSe$_{1−x}$S$_x$ detected linear resistivity from the 35 T temperature dependence of the longitudinal magnetoresistance in Ref. [24], assumed to occur near the nematic critical point. In this study, the sulfur-doping level is given as the nominal concentration, which is an overestimate of $x$. For example, the $x_{nom} \sim 0.16$ suggested to be at the nematic critical point shows a structural transition at $T_s \sim 51$ K, which corresponds to $x \sim 0.13$ in our phase diagrams in Fig. 3 and in Fig. S1(b) in the SM [27] (see, also, the resistivity derivative in Ref. [24]). However, the linear resistivity in Ref. [24] agrees with our findings inside the nematic state, once corrected for the doping shift [Fig. 3(b)].

Theoretical models suggest that the temperature exponent $\gamma$ in the vicinity of nematic critical points is highly dependent on the presence of cold spots on different Fermi surfaces, due
to the symmetry of the nematic order parameter [36,39,40]. Furthermore, resistivity near a nematic critical point can have a variation of $\gamma$ with temperature due to the scattering from acoustic phonons [22]. This is a potential cause for the variation of $\gamma$, as we observe experimentally in FeSe$_{1-x}$S$_x$ near the nematic end point. Furthermore, the scale at which the crossover to Fermi-liquid behavior occurs at $T_{FL}$ also depends on the strength of the coupling to the lattice [21]. To assess the critical behavior in FeSe$_{1-x}$S$_x$, it is worth emphasizing that the effective masses associated to the outer hole bands do not show any divergence close to the nematic end point $x \sim 0.18$ [3]. This agrees with the variation of the $A^{1/2}$ coefficient (see Fig. S11 in the SM [27]) and previous studies under pressure [20], suggesting that the critical nematic fluctuations are quenched by the coupling to the lattice along certain directions in FeSe$_{1-x}$S$_x$. This effect would lead to the resistivity exponent varying with temperature, as we find experimentally and predicted theoretically [22].

An overall representation of the resistivity slope in high fields $d\rho_{xx}/(34 T) dT$ for FeSe$_{1-x}$S$_x$ as a function of temperature is shown in the phase diagram in Fig. 3(d). The low-temperature manifestation of the nematic A and B phases is clearly different below $T^*$. In order to identify possible sources of scattering responsible for these changes, we consider the role of spin fluctuations. Recent NMR data found that antiferromagnetic spin fluctuations are present inside the nematic phase of FeSe$_{1-x}$S$_x$, being strongest around $x \sim 0.1$ [26]. In FeSe, spin fluctuations are rather anisotropic [26,41] and strongly field dependent below 15 K [11]. Interestingly, the spin-fluctuation relaxation rate is enhanced below $T^*$ [Fig. 3(d)], suggesting a correlation between spin-dependent scattering, the high-field magnetoresistance, and...
the low-temperature transport inside the nematic state. Highmagnetic fields are expected to align magnetic spins and could affect the energy dispersion of low-energy spin excitations and spin-dependent scattering in magnetic fields. In FeSe, the spin-relaxation rate in different magnetic fields up to 19 T deviates at \( T^* \) [11], but it remains relatively constant in 19 T at the lowest temperatures. This may suggest that the variation in magnetoresistance in high-magnetic fields at low temperatures in FeSe\(_{1-x}\)\( S_x \) is more sensitive to the changes in the electronic behavior, rather than to the spin fluctuations across the nematic phase.

In the low-temperature regime of FeSe\(_{1-x}\)\( S_x \), below \( T^* \) we find a temperature regime with a linear resistivity across the whole nematic phase. Linear resistivity is usually found near an antiferromagnetic critical point, such as in BaFe\(_2\)(As\(_{1-x}\)P\(_x\)) [37], and this behavior is a potential manifestation of scattering induced by critical spin fluctuations in clean systems [42].\( \mu \)SR studies place FeSe near an itinerant antiferromagnetic quantum critical point at very low temperatures [12]. Spin fluctuations are present inside the nematic state in FeSe\(_{1-x}\)\( S_x \) [11,26], being suppressed at the nematic end point where a Lifshitz transition was detected in quantum oscillations [3]. Thus, in FeSe\(_{1-x}\)\( S_x \), we find a qualitative link between the linear resistivity and spin fluctuations below \( T^* \) only inside the nematic phase.

The striking difference in magnetotransport behavior between the nematic and tetragonal phases in FeSe\(_{1-x}\)\( S_x \) can have significant implications on what kind of superconductivity is stabilized inside and outside the nematic phase as different pairing channels may be dominant in different regions, as found experimentally [18,19]. Linear resistivity found at low temperatures inside the nematic state is present in the region where spin fluctuations exist. Furthermore, the absence of superconductivity enhancement at the nematic end point in FeSe\(_{1-x}\)\( S_x \) is supported by the lack of divergent critical fluctuations, found both with chemical pressure [3] and applied pressure [20]. It is expected that the coupling to the relevant lattice strain restricts criticality in nematic systems only to certain high-symmetry directions [21,43]. Future theoretical work needs to be dedicated to understanding multiband transport phenomena of FeSe\(_{1-x}\)\( S_x \) and address the role played by both small and large pockets in relation to the BEC-BCS crossover, the effect on the Zeeman energy on different bands, as well as the possible field-induced effects in scattering.

In conclusion, we have studied the evolution of the low-temperature magnetotransport behavior in FeSe\(_{1-x}\)\( S_x \) in high-

magnetic fields up to 69 T. We find that the nematic state displays unconventional power laws in magnetic field, reflecting the dominant anomalous scattering inside the nematic phase. The temperature variation of the resistivity exponent near the nematic end point reflects the nematic elastic coupling with the lattice that also suppresses the divergent nematic critical fluctuations in FeSe\(_{1-x}\)\( S_x \). In high-magnetic fields, well above the upper critical fields, the transverse magnetoresistance shows a change in slope that reflects the changes in the spin fluctuations and/or the electronic structure. In the low-temperature limit, we find an extended linear resistivity in temperature where spin fluctuations are present. Fermi-liquid behavior recovers at low temperatures depending on the composition, impurity level, and strength of the nematic. In conclusion, we have studied the evolution of the low-temperature magnetotransport behavior in FeSe\(_{1-x}\)\( S_x \) in high-magnetic fields up to 69 T. We find that the nematic state displays unconventional power laws in magnetic field, reflecting the dominant anomalous scattering inside the nematic phase. The temperature variation of the resistivity exponent near the nematic end point reflects the nematic elastic coupling with the lattice that also suppresses the divergent nematic critical fluctuations in FeSe\(_{1-x}\)\( S_x \). In high-magnetic fields, well above the upper critical fields, the transverse magnetoresistance shows a change in slope that reflects the changes in the spin fluctuations and/or the electronic structure. In the low-temperature limit, we find an extended linear resistivity in temperature where spin fluctuations are present. Fermi-liquid behavior recovers at low temperatures depending on the composition, impurity level, and strength of the nematic. We thank Lara Befatto, Dmitrii Maslov, Rafael Fernandez, Erez Berg, Shigeru Kasahara, Steve Simon, Siddharth Parameswaran, and Stephen Blundell for useful comments and discussions. We thank and acknowledge previous contributions from Matthew Watson, Mara Bruma, Samuel Blake, Abhinav Naga, and Nathaniel Davies. This work was mainly supported by EPSRC (Grants No. EP/L001772/1, No. EP/1004475/1, and No. EP/1017836/1). A.A.H. acknowledges the financial support of the Oxford Quantum Materials Platform Grant (Grant No. EP/M020517/1). A portion of this work was performed at the National High Magnetic Field Laboratory, which is supported by National Science Foundation Cooperative Agreement No. DMR-1157490 and the State of Florida. Part of this work was supported by HFML-RO/1004475/1). A.A.H. acknowledges financial support through an EPSRC Career Acceleration Fellowship (Grant No. EP/1004475/1).

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