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Coexistence of orbital and quantum critical magnetoresistance in FeSe$_{1-x}$S$_x$

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The recent discovery of a nonmagnetic nematic quantum critical point (QCP) in the iron chalcogenide family FeSe$_{1-x}$S$_x$ has raised the prospect of investigating, in isolation, the role of nematicity on the electronic properties of correlated metals. Here we report a detailed study of the normal state transverse magnetoresistance (MR) in FeSe$_{1-x}$S$_x$, for a series of S concentrations spanning the nematic QCP. For all temperatures and x values studied, the MR can be decomposed into two distinct components: one that varies quadratically in magnetic field strength $\mu_0H$ and one that follows precisely the quadrature scaling form recently reported in metals at or close to a QCP and characterized by a $H$-linear MR over an extended field range. The two components evolve systematically with both temperature and S substitution in a manner that is determined by their proximity to the nematic QCP. This study thus reveals the coexistence of two independent charge sectors in a quantum critical system. Moreover, the quantum critical component of the MR is found to be less sensitive to disorder than the quadratic (orbital) MR, suggesting that detection of the latter in previous MR studies of metals near a QCP may have been obscured.

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I. INTRODUCTION

Many strongly interacting electron systems lie in close proximity to a quantum critical point (QCP), realized by suppressing a finite temperature ordering transition to zero temperature via some nonthermal tuning parameter [1]. Metallic quantum critical systems exhibit anomalous transport and thermodynamic properties, including (but not restricted to) a $T$-linear resistivity at low temperatures [2–4] and a logarithmic divergence of the electronic specific heat [5]. Recently, a new feature of metallic quantum criticality was discovered in the transverse magnetoresistance (whereby the magnetic field is applied perpendicular to the current) in the iron pnictide compound BaFe$_2$(As$_{1-x}$P$_x$)$_2$ (Ba122) near its antiferromagnetic QCP [6]. In particular, the magnetoresistivity, when expressed as $\Delta \rho / T$ (where $\Delta \rho = \rho[H, T] - \rho[0, 0]$) was found to exhibit an unusual quadrature scaling form $\sqrt{T} + \gamma(\mu_B \mu_0 H/k_B T)^2$ where $0.5 \leq \gamma \leq 1$ is a dimensional parameter, $k_B$ is Boltzmann’s constant, and $\mu_B$ is the Bohr magneton [6,7]. Thus, in addition to a $T$-linear resistivity at zero field, $\Delta \rho$ is found to vary linearly with magnetic field strength over a wide field range. A similar scaling of the transverse MR was also reported recently in the electron-doped cuprate La$_{2-x}$Ce$_x$CuO$_4$ (LCCO), again near its antiferromagnetic QCP [8].

In ordinary metals, the low-field orbital MR $\delta \rho / \rho[0, T] = (\rho[H, T] - \rho[0, T]) / \rho[0, T] \propto (\omega_c \tau)^2$ where $\omega_c = e \mu_0 H / m^*$ is the cyclotron frequency, $m^*$ is the effective mass of the charge carriers, $e$ is the electric charge, and $\tau$ the scattering time [9]. In the limit where $\omega_c \tau < 1$, $\delta \rho / \rho[0, T]$ thus varies quadratically with field and given that $\rho[0, T] \propto 1 / \tau$, the transverse MR has a strong temperature dependence that often obeys another form of scaling, known as Kohler’s scaling, in which plots of $\delta \rho / \rho[0, T]$ versus $(H / \rho[0, T])^2$ at different temperatures collapse onto a single curve [10]. In certain correlated metals, such as the hole-doped cuprates [11] and the heavy fermion CeCoIn$_5$ [12], a modified Kohler’s scaling is observed, whereby plots of $\delta \rho / \rho[0, T]$ versus $(H / \tan \Theta_H)^2$ collapse onto a single curve, where $\tan \Theta_H$ is the tangent of the Hall angle. By contrast, the MR curves in Ba122 and LCCO display no intrinsic temperature dependence—they simply present a set of parallel curves (at high field) offset by the change in $\rho[0, T]$ [6–8,13].

At present, there is no consensus as to the origin of the quadrature form for the transverse MR in quantum critical (QC) metals nor for the violation of Kohler scaling in other highly correlated metals. Moreover, it is not known how these very distinct MR responses are related, if at all. In particular, there is, as yet, no system in which signatures of the different MR behavior have been shown to coexist, suggesting that they
are forms associated with different limits (e.g., the low- and high-field limits or the behavior of systems located near or far from a QCP).

In this contribution, we report the observation of two additive components in the transverse MR of a series of FeSe$_{1-x}$S$_x$ single crystals that collectively span a QCP—in this case a nematic QCP. One component has a quadratic-in-field MR response up to the highest fields studied (in all S-doped samples), suggesting that this $H^2$ MR is not the limiting low-field form of the quadrature component, but something distinct, presumably reflecting the (near-)perfect compensation of the electron and hole carriers in this family of semimetals. The second component, obtained by subtracting the $H^2$ term, exhibits the quadrature scaling form to a very high degree of precision, unambiguously demonstrating its coexistence with the conventional, orbital contribution. By studying the evolution of the MR over such a wide range of dopings and temperatures, we are able to rule out the second component originating from Dirac cone states, as reported, for example, in the iron pnictides [14–16]. Rather, the two components are found to evolve systematically with both temperature and S substitution in a manner that is determined by the proximity to the QCP. This study thus reveals the coexistence of two charge sectors in a quantum critical system. Finally, comparison of the MR response of two samples with very different residual resistivities reveals a marked difference in the sensitivity of the two components to disorder.

II. NEMATIC QUANTUM CRITICALITY IN THE IRON CHALCOGENIDES

The iron chalcogenide family FeSe$_{1-x}$S$_x$ [crystal structure displayed in Fig. 2(a)] represents a class of quantum critical metals in which the QCP is due to electronic nematicity rather than antiferromagnetism [17–20]. Recently, the evolution of the (in-plane) resistivity across the nematic QCP was studied in high magnetic fields applied in the longitudinal field configuration ($H//I//ab$) in order to suppress superconductivity while at the same time, minimizing the normal state MR [21]. To orientate the subsequent analysis and discussion, we reproduce in Fig. 1 a schematic of the low-temperature phase diagram of FeSe$_{1-x}$S$_x$ as deduced from the temperature-dependent exponent $\alpha$ of the in-plane resistivity across the doping series at temperatures below 30 K [21]. The top color scale in Fig. 1 denotes the magnitude of $\alpha$ at different $T$ and $x$. At $x = x_c = 0.16$, $\rho[T]$ is $T$ linear down to 1.5 K while on either side of the QCP, $\rho[T]$ is found to cross over to a $T^2$ dependence characteristic of a correlated Fermi liquid. $A^*$—the coefficient of the $T^2$ resistivity (once corrected for the growth in total carrier density with S doping)—was found to become strongly enhanced on approach to $x_c$ (from either side), as indicated by the lower color scale. All these observations are consistent with those found in other quantum critical metals and suggest a strong coupling of the charge carriers to quantum fluctuations of the relevant order parameter.

It should be acknowledged here that there is currently no recognized theory for a $T$-linear resistivity down to $T = 0$ at a nematic QCP in a clean system [22]. While FeSe exhibits only nematic order below $T_c$, a spin-density-wave (SDW) state is found to be stabilized under applied pressure [23].

Moreover, enhanced spin fluctuations (at ambient pressure) and critical behavior have been reported below $T_c$ [24,25], in the same range over which $\rho_{ab}[T]$ is quasi-$T$-linear, suggesting a possible link between the $T$-linear resistivity and antiferromagnetic, rather than nematic fluctuations. With increasing $S$ substitution, however, the nematic and SDW states become decoupled [26], and as the pressure range of nematic order shrinks, eventually vanishing at $x_c$, the dome of SDW order shifts to progressively higher pressures. Thus, at $x = x_c$, the SDW phase is located far from the ambient pressure axis at which our experiments are performed. At the same time, NMR experiments have shown that spin fluctuations, although present in FeSe$_{1-x}$S$_x$ at low $x$ values, are strongly suppressed with S substitution [27]. These combined results suggest that the critical behavior at $x = x_c$ is not associated with proximity to a magnetic phase.

III. METHODS

The single crystals of FeSe$_{1-x}$S$_x$ used in this study were grown at two different locations. The bulk of the samples were grown in Kyoto by the chemical vapor transport technique [17]. The actual sulfur composition $x$ was determined by energy dispersive x-ray (EDX) spectroscopy, and was found to be around 80% of the nominal S content. The Berkeley sample discussed exclusively in Sec. V was grown using the KCl flux technique [28] with a nominal concentration of 18% selenium replaced by sulfur, whose composition was also confirmed by EDX. To be consistent with the data presented in Ref. [21] (carried out on the same Kyoto crystals), all $x$ values quoted here are the nominal values. The crystals were cut into regularly shaped platelets and electrical contacts applied to each sample in a Hall bar geometry. The magnetotransport
At the lowest fields, $d \rho \, d(\mu_0 H)$ is linear in the field with a zero intercept, implying that the low-field MR is strictly quadratic. The slope of the derivative is labeled $2 \beta_\mu \mu_0 H$ and is indicated in each case by a green dotted line. For $2T < \mu_0 H < 7$ T, the slope of $d \rho / d(\mu_0 H)$ gradually decreases until above 7 T, it becomes linear once more, albeit with a finite intercept. The presence of this finite intercept implies that for $\mu_0 H > 7$ T, the MR has two components, one linear in field, the other quadratic. A similar field-dependent MR was reported by Sun et al. for $x = 0$ and $x = 0.14$ albeit over a more limited field and temperature range [29]. In our study, both components are found to persist up to the highest field measured (35 T). Note that such a field dependence cannot be captured by a simple two-carrier model involving electrons and holes [30]. A three-carrier model [31] can produce an MR with a field dependence that resembles those displayed in Fig. 3, but only over a narrow range of field values and parameters. We will return to this point in the following section.

The slope of $d \rho / d(\mu_0 H)$ at high field is defined here as $2 \beta_\mu \mu_0 H$, where $\beta_\mu$ is the magnetoresistance coefficient of the individual MR curves. As indicated by the dotted black lines in panels (d) and (f), the contribution of the MR to the field dependence of the derivative is revealed. As indicated by the dotted black lines in panels (d) and (f), the contribution to the MR is found to follow the same quadratic form, i.e., $\rho [\mu_0 H] - \beta_\mu \mu_0 H^2 = a H^2 / (1 + b H^2)$ that was first reported in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ by Sun et al. for $x = 0.14$ albeit over a more limited field and temperature range [29]. In our study, both components are found to persist up to the highest field measured (35 T). Note that such a field dependence cannot be captured by a simple two-carrier model involving electrons and holes [30]. A three-carrier model [31] can produce an MR with a field dependence that resembles those displayed in Fig. 3, but only over a narrow range of field values and parameters. We will return to this point in the following section.

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MR contributions persist over the entire range of temperatures and $S$ concentrations studied. Only in stoichiometric FeSe, where the orbital MR is extremely large, is this term found to deviate from $H^2$ at high fields and low $T$, though even here, the form of the MR (having subtracted off the quadrature component) is found to be consistent with the usual Drude expression for two-carrier (i.e., electron and hole) magnetotransport (see Fig. S2 in the Supplemental Material [32] for more details).

**B. Two-component magnetoresistance**

The data presented in Fig. 3 thus reveal the presence of two contributions to the MR response of FeSe$_{1-x}$S$_x$ which individually extend over a wide field, temperature, and doping range. A similar form of the MR has also been observed in the iron pnictide family Ba(Fe$_{1-x}$T$_x$)$_2$As$_2$ ($T =$ Co, Ni, and Cu) [14–16]. There, as in FeSe$_{1-x}$S$_x$ [29], the $H$-linear component was attributed [14,15] to the MR response of Dirac-like states beyond the quantum limit [33]. A few reports have claimed evidence for the presence of Dirac cones inside the nematic phase of FeSe$_{1-x}$S$_x$ [29,34,35]. The fact that the $H$-linear MR component in FeSe$_{1-x}$S$_x$ persists beyond the nematic phase, however, suggests that it is unlikely that such Dirac-like states are responsible for the anomalous MR component found in FeSe$_{1-x}$S$_x$. There is also no reason a priori why the MR response of Dirac states should exhibit both the quadrature form and $H/T$ scaling found in FeSe$_{1-x}$S$_x$ across the entire phase diagram (discussed in more detail in the following section).

The remarkable agreement between the “residual” MR (upon subtraction of the $H^2$ component) and the quadrature form of the MR in BaFe$_2$As$_2$ has also been modeled using a three-carrier Drude model [16]. While the three-carrier model is able to generate a MR curve that approximates a $H + H^2$ form (over a limited field range at least), it can only do so for a limited range of parameters. Moreover, for the same set of parameters, the corresponding Hall response is found to be highly nonlinear, whereas for $x \gg x_c$, the Hall resistivity of our crystals is found to be either linear or show only small deviations from linearity [36]. Finally, it is not possible to simulate the very different ratios of the $H$ and $H^2$ terms observed in FeSe$_{1-x}$S$_x$ and over such a wide field range. Therefore, we do not believe it is appropriate to model the MR response in FeSe$_{1-x}$S$_x$ using the three-carrier model, at least one in which the carrier densities and mobilities are assumed to be independent of field strength. Of course, one could always add further complexity in the model (e.g., by allowing parameters to vary with field strength or introduce $k$ dependence in a Boltzmann-type analysis), but in our opinion, this would not constitute a robust analysis.

The remarkable agreement between the “residual” MR (upon subtraction of the $H^2$ component) and the quadrature form of the MR, including the $H/T$ scaling, leads us to conclude that the charge dynamics of FeSe$_{1-x}$S$_x$ must contain two distinct sectors: one that generates a conventional orbital MR, presumably involving quasiparticle transport, and one akin to the quantum critical sector found in Ba122 and LCCO (that exhibits scale invariance). In such a scenario, the total (zero-field) conductivity $\sigma_{tot}$ should be expressed as a sum of the individual contributions, i.e., $\sigma_{tot}[T] = \sigma_{QC}[T] + \sigma_{FL}[T]$, where the subscripts refer to the quantum critical and
quasiparticle (Fermi-liquid) sectors, respectively. The transverse magnetoconductance is then given by a weighted sum [37]:

$$\frac{\Delta \sigma_{\text{tot}}}{\sigma_{\text{tot}}} = \frac{\sigma_{\text{QC}}}{\sigma_{\text{tot}}} \frac{\Delta \sigma_{\text{QC}}}{\sigma_{\text{tot}}} + \frac{\sigma_{\text{FL}}}{\sigma_{\text{tot}}} \frac{\Delta \sigma_{\text{FL}}}{\sigma_{\text{tot}}}. \quad (1)$$

In reality, of course, it is the magnetoresistance, rather than the magnetoconductance that is measured, the former being related to the latter via inversion of the (in-plane) conductivity tensor.

$$\frac{\delta \rho}{\rho[0,T]} = \frac{\Delta \sigma_{\text{tot}}}{\sigma_{\text{tot}}} - \left( \frac{\sigma_{\text{xy}}}{\sigma_{\text{tot}}} \right)^2 \quad (2)$$

where $\rho[0,T] = 1/\sigma_{\text{tot}}$ is the zero-field resistivity at the temperature at which an individual field sweep is taken, $\sigma_{\text{xy}}$ is the Hall conductivity, and $\sigma_{\text{xy}}/\sigma_{\text{tot}}$ the corresponding Hall angle. In order to proceed, it is necessary to estimate first the magnitude of the Hall angle ($\sigma_{\text{xy}}/\sigma_{\text{tot}}$) relative to $\delta \rho/\rho[0,T]$ (as measured). In the temperature range $20 \text{K} < T < 50 \text{K}$ over which we currently have data overlap, the square of the Hall angle (for $x = 0.16$) varies between 5% and 20% of the as-measured MR [36]. Thus, we can conclude that the MR is dominated by the magnetoconductance term and rewrite Eq. (1) as

$$\frac{\delta \rho}{\rho[0,T]} = \frac{\sigma_{\text{QC}}}{\sigma_{\text{tot}}} \frac{\delta \rho_{\text{QC}}}{\rho[0,T]} + \frac{\sigma_{\text{FL}}}{\sigma_{\text{tot}}} \frac{\delta \rho_{\text{FL}}}{\rho[0,T]}, \quad (3)$$

Hence,

$$\delta \rho[H] = \left( \frac{\sigma_{\text{QC}}}{\sigma_{\text{tot}}} \right)^2 \delta \rho_{\text{QC}}[H] + \left( \frac{\sigma_{\text{FL}}}{\sigma_{\text{tot}}} \right)^2 \delta \rho_{\text{FL}}[H] \quad (4)$$

where at high fields $\rho[0,T]$ can be expressed as

$$\delta \rho[H] = \beta_{\text{QC}} \mu_0 H + \beta_{\text{FL}} (\mu_0 H)^2. \quad (5)$$

Here $\beta_{\text{QC}}$ and $\beta_{\text{FL}}$ are, respectively, the (as-measured) magnitudes of the $H$-linear and $H^2$ MR terms, which according to Eqs. (4) and (5) represent the quantum critical $\delta \rho_{\text{QC}}[H]$ and quasiparticle $\delta \rho_{\text{FL}}[H]$ contributions to the total MR, weighted by the square of the contribution of the two sectors to the total (zero-field) conductivity.

C. Evolution across the phase diagram

The ratio $\beta_{\text{QC}}/\beta_{\text{FL}}$ for all samples, determined at a temperature (15 K) at which there are no discernible superconducting fluctuation conductivities, is plotted in Fig. 4 (as open red circles). The ratio is found to peak around $x_c = 0.16$, in a manner that is strikingly similar to the enhancement of the quasiparticle effective mass as expressed through $\Lambda^*$, the renormalized coefficient of the $T^2$ resistivity (and plotted as empty black squares in Fig. 4) [21]. It is important to realize that these two quantities are determined in very different ways yet together, they appear to reveal a consistent picture in which the (magneto)transport properties of FeSe1-xSx are heavily influenced by the presence of quantum critical fluctuations.

As described above, plots of $\Delta \rho / T$ versus $H/T$ in P-doped Ba122 at the critical doping collapse onto a single curve of the quadrature form [6]. Such scaling can only be realized if the high-field ($H$-linear) slopes of the individual MR curves are the same, i.e., $\Delta \rho_{\text{QC}} = X_1 \mu_0 H$ independent of temperature. Since there are two contributions to the MR in FeSe1-xSx, whose relative strengths are weighted by their respective contributions to the total conductivity, the same $\Delta \rho / T$ scaling cannot be gleaned directly from our data by simply subtracting off the orbital MR term. Nevertheless, further analysis outlined below and presented in Secs. III and IV of the Supplemental Material [32] provides strong evidence that $H/T$ scaling is also realized in FeSe1-xSx.

Firstly, according to the scaling ansatz of Hayes et al. [6], the residual MR (obtained by subtracting the $H^2$ term from the total MR) should have the same dependence with field for all samples when measured at the same temperature, irrespective of its absolute magnitude. As shown in Fig. S3 of the Supplemental Material [32], the (normalized) residual MR at $T = 15 \text{ K}$ is indeed found to follow the same form right across the phase diagram. Secondly, when the residual MR for one sample is plotted versus $H/T$ for a range of temperatures inside the QC fan (see Fig. S4 of the Supplemental Material [32]), the data are found to collapse onto a single curve. Finally, as described in the Discussion section, a second sample with a doping close to $x_c$ but with a larger residual resistivity (that effectively quenches the orbital component to the MR), is found to exhibit precisely the same MR scaling as seen in Ba122 and LCCO. Thus, we can conclude that the QC component to the MR in FeSe1-xSx follows the exact same scaling relation, and since $d(\Delta \rho_{\text{QC}})/dH = d(\delta \rho_{\text{QC}})/dH$ (only the intercepts differ), we obtain from Eqs. (4) and (5) the following relation between $\beta_{\text{QC}}$ and $X_1$:

$$\beta_{\text{QC}} = \left( \frac{\sigma_{\text{QC}}}{\sigma_{\text{tot}}} \right)^2 X_1. \quad (6)$$

Thus, under the inference that the QC component to the MR in FeSe1-xSx exhibits scale invariance, $\beta_{\text{QC}}$ provides a direct measure of the contribution of $\sigma_{\text{QC}}$, the QC component, to the total conductivity. This quantity is plotted in Fig. 5 for

\[\text{FIG. 4. Open circles: Variation of } \beta_{\text{QC}}/\beta_{\text{FL}}, \text{ the ratio of the } H\text{-linear transverse MR to the (high-field) } H^2 \text{ component as a function of } x. \text{ All } \beta_{\text{QC}}/\beta_{\text{FL}} \text{ values were obtained at } T = 15 \text{ K. Open squares: Corresponding values of } A^*, \text{ the coefficient of the } T^2 \text{ resistivity, normalized to a fixed carrier density [21].}\]
all the S concentrations studied (bar $x = 0.00$ for which $\beta_{QC}$ is hard to extract due to its exceptional high-field behavior). What is most striking here is the evolution in the behavior of $\delta\rho_{QC}[T]$ across the phase diagram. For samples with $x < x_c$, $\beta_{QC}[T]$ follows the same $T$ dependence, reaching a maximum at or around the temperature below which $\rho(T)$ is no longer $T$ linear, i.e., below the QC fan, implying that the QC component is reduced as one approaches the FL ($\rho \sim T^2$) regime (see Fig. 1) and may even vanish in the zero-temperature limit.

By contrast, for $x_c = 0.16$, $\beta_{QC}$ increases monotonically with decreasing temperature, consistent with the observation that the $T$-linear resistivity extends down to the lowest $T$ accessed to date and indicating that as the temperature is lowered, the QC component emerges as the dominant contribution. This nonmonotonic behavior of the $H$-linear component for $x < x_c$ and its correlation with the evolution of the zero-field resistivity is further evidence that it is not related to a contribution from Dirac-like states for which one would expect a monotonic increase in its magnitude as $T$ decreases. Finally, beyond $x_c$, the magnitude of $\beta_{QC}$ gradually softens with further S doping, though crucially, even for $x = 0.25$, $\beta_{QC}$ remains finite.

Although $\beta_{QC}$ is claimed to be proportional to $(\sigma_{QC}/\sigma_{tot})^2$, we cannot determine $\sigma_{QC}/\sigma_{tot}$ directly as we have no way of obtaining $X_1$ independently. However, one can gain an estimate for $\sigma_{QC}/\sigma_{tot}$ by simulating the zero-field $\rho(T)$ assuming parallel conduction, a point we shall return to later. Nevertheless, Fig. 5 reveals a very systematic evolution in the fraction of the total conductivity that can be attributed to the QC component.

For completeness, we now turn to consider the second component $\delta\rho_{FL} = \rho_{FL}(\mu_0 H)^2$. The large field range over which this MR component remains perfectly quadratic suggests that the electron and hole pockets in our FeSe$_{1-x}$S$_x$ crystals are close to being fully compensated. Moreover, as shown in Fig. 6(b) for the $x = 0.25$ sample, the $T$ dependence of $\delta\rho_{FL}/\rho_{FL}[0]$ (where $\rho_{FL}[0] = \rho_{FL}[0, T]$) is estimated from the zero-field resistivity shown in Fig. 7 and discussed below) is found to have a Fermi-liquid (FL) form: $\delta\rho_{FL}/\rho_{FL}[0] = (\omega_c \tau)^2 = 1/(A + BT^2)^2$ between 1 and 30 K (since for a FL, $1/\tau \propto T^2$). At the QCP, the $T$ dependence of $\delta\rho_{FL}/\rho_{FL}[0]$ cannot be captured by the same expression [Fig. 6(a)].

Nevertheless, the very strong $T$ dependence observed in both cases supports the notion that this contribution is controlled by orbital effects (i.e., by $\omega_c \tau$).

### D. Two-component conductivity

The presence of two distinct components in the transverse MR of FeSe$_{1-x}$S$_x$ implies that there must also be two contributions to the zero-field conductivity, i.e., $\sigma_{tot} = \sigma_{FL} + \sigma_{QC}$; the first term giving rise to the conventional, orbital MR and the second to the QC quadrature term. While the QC component $\sigma_{QC}$ is linked directly to $\beta_{QC}$ through Eqs. (4) and (5), it cannot be determined uniquely since $X_1$ itself is not known. We can, however, allow the magnitude of $X_1$ to vary (recall that $X_1$ will have a unique value for each sample but its magnitude is independent of $T$) and inspect the resultant $T$ dependence of $\rho_{QC} = 1/\sigma_{QC}$ and $\rho_{FL} = 1/\sigma_{FL} = 1/(\sigma_{tot} - \sigma_{QC})$ where $\sigma_{tot} = 1/\rho[0, T]$, in order to see whether or not a self-consistent picture for both the zero-field resistivity and the transverse MR emerges from the data.

Examples of this procedure are shown in Figs. 7(a) and 7(b) for the $x_c = 0.16$ and $x = 0.25$ samples with $X_1 = 2.5$ and 1.7, respectively. Here, we have ensured that the two components add in parallel to give the total, as-measured resistivity. $\rho_{QC}(T)$ is found to be $T$ linear in both cases, at least up to 25 K. For $x = 0.25$, $\rho_{FL}(T)$ retains its $T^2$ character up to 30 K, even though the raw resistivity curve is quadratic only up to 12 K. For $x = 0.16$, $\rho_{FL}(T)$ shows an approximately quadratic (FL-like) $T$ dependence (indicated by a dotted line) only below 15 K. Above 15 K, the form of $\rho_{FL}(T)$ deviates from its low-$T$ form and tends towards a constant value. The presence of the quasiparticle component in both the zero-field resistivity and the transverse MR may indicate that this sample is located close to, though not necessarily at the QCP. Further measurements down to lower temperatures (in higher fields) would be helpful in confirming the form of $\beta_{QC}/\beta_{FL}$ at $x = x_c$ below 1.5 K.

### V. DISCUSSION

The observation of two distinct components in the transverse MR of FeSe$_{1-x}$S$_x$ raises the question why previous
studies of correlated metals (both in the vicinity of or far from a QCP) found only an orbital MR response (that may or may not have violated Kohler’s rule) or the quadrature scaling form, but never the combination [11,12]. A comparative study of two crystals with different levels of disorder, presented below, provides one possible explanation for these distinct behaviors.

Figure 8(a) shows the low-T resistivity of the two crystals in question (both with nominal composition \( x = 0.18 \)). The crystal with the lower residual resistivity (S018a) was synthesized in Kyoto using identical starting constituents and growth conditions as the other crystals described in the preceding section. As with the other crystals from this source, it exhibits both components in the transverse MR that evolve with temperature as summarized in Figs. 4 and 5. The second crystal (S018b) was prepared in Berkeley using a different technique and found to have a residual resistivity that is approximately five times higher. A series of MR curves obtained on this crystal over a wide temperature range \( 1.5 \text{K} < T < 80 \text{K} \) is shown in Fig. 8(b). In contrast to the multiple crossing points realized in the other crystals [an example of which is shown in Fig. 2(b)], the MR curves for S018b (beyond the field-induced superconductor-to-metal transition) are parallel to one another and become \( H \)-linear at high fields. Moreover, when the MR curves are replotted as \( \delta \rho_T / \rho_0 \) versus \( H / T \), as shown in Fig. 8(c), they are found to collapse onto a single curve that fits the same quadrature form \( \delta \rho_T / \rho_0 = \sqrt{1 + \gamma (\mu_B \mu_0 H / k_B T)^2} \) (with \( \gamma \approx 0.5 \)) that was observed in the cleaner crystal (though now unfettered by the presence of the orbital MR term).

The observation of QC scaling in the MR response of the second crystal reveals that while the orbital component is effectively quenched with increasing impurity scattering (a 5-fold increase in the residual resistivity would correspond to a 25-fold decrease in the orbital MR at low \( T \)), the QC component remarkably survives.
It has been argued previously that the $H$-linear transverse MR and $H/T$ scaling found in pnictides [6] and cuprates [8] may arise due to a variation in the carrier composition within a given sample [38], as postulated previously for two-dimensional electron gases [39] and even elemental metals [40]. The current study, however, suggests that this is not necessarily the case in QC systems. As shown in Fig. 4, the ratio of the $H$-linear ($\beta_{QC}$) to $H^2$ ($\beta_{FL}$) components shows a very systematic evolution with $S$ substitution and peaks strongly at the QCP, even though the residual resistivities are comparable across the entire series of S-doped crystals [21]. Moreover, $\beta_{QC}/\beta_{FL}$ in FeSe is the same as in FeSe$_{0.75}S_{0.25}$, despite the fact that the former’s residual resistivity is one order of magnitude smaller than the latter and clear quantum oscillations are observed in the former. Finally, the sharpness of the kinks in $d\rho/dT$ [21] at $T = T_c$ (for $x < x_c$) imply homogeneous doping for all these samples. Thus it appears unlikely that the $H$-linear component to the transverse MR is due to an extrinsic longitudinal contribution arising from a variation in carrier density along each crystal.

Recent models of strange metals, invoking either holographic methods [41] or based on the Sachdev-Ye-Kitaev picture of itinerant, nonquasiparticle transport [42], have succeeded in obtaining certain aspects of the MR scaling, but as of yet, not in tandem with a more conventional, orbital MR. The key task now therefore is to understand how these two components can coexist.

We consider here first the possibility that the two components arise from excitations that occupy different regions of the Brillouin zone. FeSe and its derivatives are known to contain (equal) numbers of electron- and holelike carriers and correlated metals often display an electron-hole dichotomy, most evident in the respective phase diagrams of electron- and hole-doped cuprates, for example [43]. In such a scenario, the electron and hole pockets found in FeSe$_{1-x}$S$_x$ would harbor different types of excitations that contribute respectively to the orbital and QC MR responses. Alternatively, the two excitations may reside within both pockets, albeit at different points on the Fermi surface; for example, the QC component may arise from excitations near hot spots—strong scattering sinks that destroy the quasiparticle character of excitations there—leading to strong momentum-dependent scattering as realized, for example, in the cuprates [44]. Indeed, the superconducting gap in FeSe has been shown to be strongly anisotropic in both the electron and hole pockets, indicating anisotropic (and possibly orbitally selective) pairing interactions [45]. Both scenarios, however, appear inconsistent with the observation of quantum oscillations on both the electron and hole pockets (at least for S concentrations located away from the QCP) that indicate the presence of coherent quasiparticle states around the Fermi surface of both pockets [46,47].

The lack of oscillations at $x = x_c$ itself, however, is consistent with the notion that the quasiparticles are much heavier close to the QCP where the QC component of the MR is also dominant. The gradual crossover from quantum critical to quasiparticle contributions to the MR away from the QCP suggests in fact that the low-lying excitations near the Fermi level have dual character, i.e., the quasiparticle and the quantum critical sectors are two “flip sides” of the same electronic states, whose weighting depends on their proximity to the QCP. Whatever the origin, these findings clearly call for further theoretical studies in order to understand the interplay of the two sectors across the phase diagram, and more experimental studies to determine quantitatively the role of disorder in the realization of the $H/T$ scaling in the transverse MR not only in more disordered FeSe$_{1-x}$S$_x$, but also in other candidate QC systems. The latter comparison is important to establish whether it is merely a question of disorder or length scales (e.g., between electron-electron collisions and electron-impurity collisions), or whether it is the nematic character of the quantum fluctuations in FeSe$_{1-x}$S$_x$ that allows both the quantum critical and quasiparticle sectors to reveal themselves, even at the QCP itself.

VI. CONCLUSIONS

In summary, we have carried out a systematic study of the transverse MR in a series of FeSe$_{1-x}$S$_x$ single crystals in high magnetic fields up to 38 T for S concentrations that span the nematic QCP. The field derivatives of the MR curves reveal the ubiquitous presence of two distinct (and additive) components to the MR in FeSe$_{1-x}$S$_x$: the normal orbital $H^2$ MR and an anomalous component that follows precisely the quadrature scaling first observed in the iron pnictide P-doped BaFe$_2$As$_2$ near the spin-density-wave QCP.
The ratio of the two MR components follows a very similar evolution with doping as the (renormalized) $T^2$ resistivity, suggesting that the component with the quadrature form is associated with scale-invariant quantum critical fluctuations that are also responsible for the quasi-particle mass enhancement on approaching the QCP. The quantum critical contribution is found to be increased with decreasing $T$ at the QCP, but is suppressed inside the FL regime away from the QCP. With increased disorder content, the orbital MR is quenched, leading to the appearance of strict quantum critical scaling at or near the QCP.

These collective findings provide evidence for the coexistence of two charge sectors in a quantum critical system whose quantum critical scaling at or near the QCP. The task now is to identify how these two sectors coexist and to establish whether this is a universal behavior.

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