

From mean-field localized magnetism to itinerant spin fluctuations in the “nonmetallic metal” FeCrAs

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FeCrAs displays an unusual electrical response that is neither metallic in character nor divergent at low temperatures, as expected for an insulating response, and therefore it has been termed a “nonmetal metal.” The anomalous resistivity occurs for temperatures below ~ 900 K. We have carried out neutron scattering experiments on powder and single crystal samples to study the magnetic dynamics and critical fluctuations in FeCrAs. Magnetic neutron diffraction measurements find Cr^{3+} magnetic order setting in at $T_N = 115$ K ~ 10 meV with a mean-field critical exponent. Using neutron spectroscopy we observe gapless, high velocity, magnetic fluctuations emanating from magnetic positions with propagation wave vector $\vec{q}_0 = (\frac{1}{3}, \frac{1}{3})$, which persists up to at least 80 meV ~ 927 K, an energy scale much larger than T_N . Despite the mean-field magnetic order at low temperatures, the magnetism in FeCrAs therefore displays a response which resembles that of itinerant magnets at high energy transfers. We suggest that the presence of stiff high-energy spin fluctuations extending up to a temperature scale of ~ 900 K is the origin of the unusual temperature dependence of the resistivity.

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I. INTRODUCTION

There is a growing list of materials which behave as neither a metal nor an insulator [1]. Recent examples of interest include underdoped and high temperature superconducting cuprates, heavy fermions, iron based chalcogenides [2–4], and oxyselenides [5–7]. The underlying cause of this unconventional behavior is not understood on a general level and, as found in at least the cuprates [8] and iron based superconductors [9], is often complicated by several competing structural and magnetic orders. Here, we report neutron scattering measurements studying the magnetic fluctuations in an extreme example of this unusual electronic behavior found in FeCrAs.

FeCrAs displays very unusual properties which have led to it being termed a “nonmetallic metal” [10,11]. Thermodynamic measurements reveal a highly enhanced Fermi liquid: The linear coefficient of specific heat is $\gamma \sim 30$ mJ/mole K^2 , while the susceptibility is Pauli like and quite large, leading to a Wilson ratio of approximately 4. On the other hand, not only does the electrical resistivity $\rho(T)$ show a strong departure from Fermi liquid T^2 behavior—as $T \rightarrow 0$ K it

has a *sublinear* power law $\rho(T) \simeq \rho_0 + AT^{0.6}$ —but it is also “nonmetallic” in the sense that the A coefficient is *negative*. That is, the resistivity rises with decreasing temperature, but without any evidence of a gap in the density of states. In contrast to the Kondo effect, where such behavior is seen only at low temperature, in FeCrAs the resistivity has a negative slope over a huge temperature range. The *ab*-plane resistivity rises monotonically with decreasing temperature from near 900 K down to the lowest measured temperatures of 80 mK, while the *c*-axis resistivity has a similar rising form interrupted only by a sharp fall just below the antiferromagnetic ordering temperature $T_N \simeq 125$ K. The magnitude of the resistivity is in the range of a few hundred $\mu\Omega$ cm, which is very large for a metal. First principles calculations predict a carrier density of approximately $2 \times 10^{28} \text{ m}^{-3}$, and for this density the measured resistivity would suggest an extremely short mean-free-path, well below one lattice spacing.

FeCrAs has a hexagonal crystal structure (space group $P\bar{6}2m$ with lattice constants $a = 6.068$ Å and $c = 3.657$ Å). The Fe sublattice forms a triangular lattice of trimers, while the Cr ions form a highly distorted Kagome framework within the basal plane (see Fig. 1). However, the interlayer Cr-Cr distance is relatively short (3.657 Å), suggesting that the interlayer hopping is substantial in this material. This is consistent with the small resistivity anisotropy ($\rho_c/\rho_{ab} < 2$). The Cr magnetic

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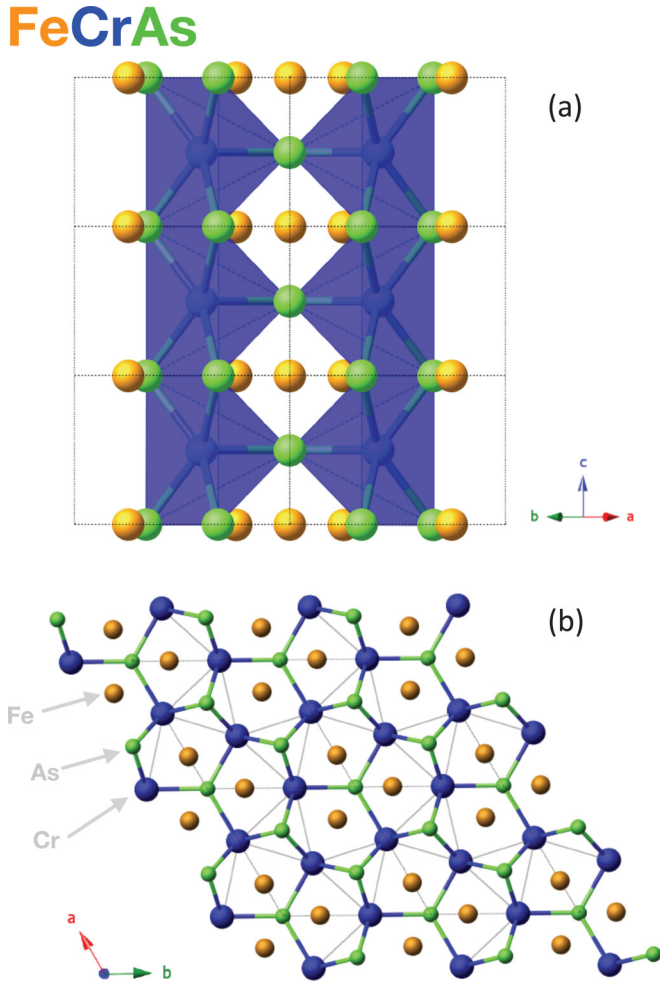


FIG. 1. (a) Crystal structure of FeCrAs illustrating the CrAs₅ pyramids and connectivity along the *c* axis. (b) The structure projected onto the *ab* plane.

moments order at $T_N \sim 125$ K forming a spin-density wave with the ordered moments varying from 0.6 to $2.2 \mu_B$ [12]. Given that the Cr magnetic moment measured with neutrons is proportional to gS (g is electron gyromagnetic ratio and S is spin quantum number), it is likely that Cr has a valence of $3+$ (hence $S = \frac{3}{2}$) and therefore lacks an orbital degeneracy in pyramidal crystal field environment. In contrast to iron based pnictides, earlier studies report that the Fe site in FeCrAs does not carry an observable moment at any temperature. The neutron diffraction results found no ordered moment at the Fe site [12]. A fluctuating Fe moment should result in some induced polarization when the Cr sublattice orders, but this is not observed in Mössbauer spectroscopy [13]. Linear spin density approximation calculation suggests significant covalency between Fe and As, so that moment formation is negligible (i.e., it is below the Stoner criterion) [14]. All of these studies are also consistent with the suppressed Fe $K\beta'$ fluorescence line observed by x-ray emission spectroscopy, which is sensitive to any fluctuating moment down to the x-ray time scale [15]. These combined observations provide compelling evidence that any static and dynamic Fe moment is negligibly small in FeCrAs.

A number of theoretical studies have been devoted to understanding the strange metallic properties of FeCrAs. The magnetic phase diagram of the coupled Fe trimer lattice and the distorted Kagome lattice of Cr has been mapped out predicting magnetic order consistent with experiment [16]. Given the lack of observable static magnetic order on the Fe sublattice, a hidden spin-liquid phase has been proposed arising from the close proximity to a metal-insulator transition. The strong charge fluctuations associated with this nearby critical point have been implicated as the origin of the unusual transport properties [17]. An alternate explanation has been proposed in the context of “Hund’s metals” where large localized moments are coupled to more itinerant electrons [18,19]. There have been only a limited number of spectroscopic studies to put these theories to the test. Charge excitations have been investigated using optical spectroscopy which revealed that the anomalous temperature dependence of resistivity was dominated by the temperature dependence of scattering rate, rather than carrier concentration [11]. In addition, they found that two Drude components with drastically different energy scales contribute to the low energy charge dynamics. On the other hand, the spin dynamics in FeCrAs have not been investigated to date.

In this study, we apply neutron scattering to investigate the magnetic properties of FeCrAs with emphasis on the static order and fluctuations originating from the Cr³⁺ sites. We first present diffraction work showing the magnetic order associated with the propagation wave vector of $\vec{q}_0 = (\frac{1}{3}, \frac{1}{3})$ is described with a mean-field critical exponent. We then measured the powder averaged fluctuations showing stiff magnetic fluctuations extending up to at least ~ 80 meV, while the low energy excitations seem to be well described with gapless spin waves emanating from the ordering wave vector. These results illustrate spin excitations in FeCrAs resemble those in itinerant magnets. We further discuss the magnetic excitation spectrum in the context of the unusual transport properties. Our finding of a high energy scale for magnetic fluctuations suggests that magnetic fluctuations could be responsible for the anomalous scattering that is observed up to high temperatures, despite the Néel temperature occurring at much lower temperature. Although our observations do not directly speak to the mechanism of nonmetallic and non-Fermi-liquid resistivity in the $T \rightarrow 0$ K limit, it seems natural to hypothesize that anomalous magnetic correlations begin to form at very high temperature in FeCrAs and continue to evolve down to very low temperature, somehow producing the nonmetallic metal state.

II. EXPERIMENT DETAILS

The powder samples of FeCrAs were prepared by melting high purity Fe, Cr, and As in stoichiometric ratios following Ref. [20]. A small single crystal (with mass 25 mg) was also produced by slow cooling from the stoichiometric melt. The single crystal used here was from the same batch as those used in earlier transport and thermodynamic studies discussed in Ref. [10].

High energy inelastic neutron scattering measurements on powder samples were performed using the MARI direct geometry chopper spectrometer (ISIS, Didcot). Measurements were performed with incident energies of $E_i = 150$ meV and 300 meV that were selected using the “relaxed” Fermi chopper

spinning at $f = 300$ Hz and 450 Hz, respectively, with the data being collected in a time of flight mode. Details of the background subtraction are provided below. Single crystal spectroscopy measurements were not successful owing to the small sample size.

Further higher resolution neutron spectroscopy measurements were performed on the MACS cold triple-axis spectrometer (NIST, Gaithersburg). Instrument and design concepts can be found elsewhere [21,22]. Data was collected by measuring momentum space cuts at constant energy transfers by fixing the final energy at $E_f = 2.4$ meV using the 20 double-bounce PG(002) analyzing crystals and detectors and varying the incident energy defined by a double-focused PG(002) monochromator. Each detector channel was collimated using 90' Soller slits before the analyzing crystal and a cooled Be filter was placed before the analyzing crystals. Maps of the spin excitations as a function of energy transfer were then constructed from a series of constant energy scans at different energy transfers. All of the data has been corrected for the $\lambda/2$ contamination of the incident-beam monitor and an empty cryostat measurement was used to estimate the background.

Single crystal magnetic neutron diffraction measurements were performed on the 1T1 thermal triple axis spectrometer (LLB, Saclay) utilizing an open collimation sequence, double focusing monochromator, and vertically focusing analyzer. The crystal was aligned in the (HK0) scattering plane of the hexagonal unit cell for the duration of the experiment.

III. RESULTS

A. Magnetic order from neutron diffraction

Neutron diffraction characterizing the magnetic order is presented in Fig. 2. The resolution limited magnetic Bragg peaks in Fig. 2(b) confirm the presence of long-range magnetic order with a $(\frac{1}{3}, \frac{1}{3})$ propagation vector as observed in previous powder diffraction measurements [12]. The integrated neutron scattering intensity which is proportional to the squared magnetic order parameter is plotted in Fig. 2(a). We observe the onset of magnetic Bragg intensity at $T_N = 115.5(5)$ K, a temperature significantly lower than the $T_N = 125$ K Néel temperature extracted from resistivity and magnetic susceptibility measurements on the same sample (Ref. [10]). The value of T_N in FeCrAs is known to vary across different samples between 100 and 125 K depending on the synthesis conditions and sample quality. Those samples with a higher T_N are observed to have a splitting of field cooled and zero field cooled magnetic susceptibility at lower temperatures and the highest quality samples are associated with the highest T_N [23]. However, neutron diffraction and magnetic susceptibility measurements were performed on the same sample so the origin of this discrepancy is presently not clear.

In a mean field approximation, for localized magnetism, the critical temperature is related to the magnetic exchange interaction via the relation,

$$k_B T_N \sim k_B \Theta_{CW} = \frac{2}{3} S(S+1) z J,$$

where S is the spin value, presumably $\frac{3}{2}$ for Cr^{3+} , and J is the average exchange constant with z representing the number of nearest neighbors. The FeCrAs magnetic structure is highly

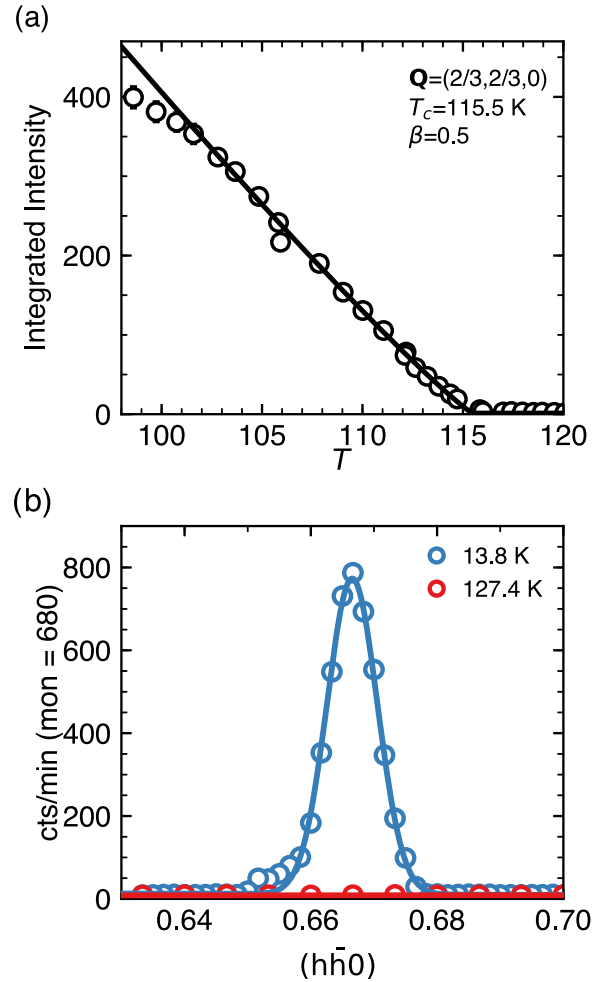


FIG. 2. (a) Integrated intensity of the $(2/3, 2/3, 0)$ magnetic Bragg peak measured on 1T-1 (LLB); solid line is a fit to $(1 - T/T_N)^{2\beta}$ with $T_N = 115.5 \pm 0.5$ K and $\beta = 0.54 \pm 0.05$. (b) Transverse scans through the magnetic Bragg peak at $(2/3, 2/3, 0)$.

frustrated [24] potentially making the sum over neighbors quite complicated. However, this expression does allow us to obtain an estimate of the mean-field spin-wave velocity of $zSJa \sim 3k_B T_N a / 2(S+1) \sim 20$ meV \AA if we assume local spin moments. We compare this energy scale to the measured spin fluctuations below.

A fit of the temperature dependent integrated neutron intensity to a power law near T_c finds the mean-field critical exponent $\beta = 0.54 \pm 0.05$. This differs from the critical exponent of $\beta \sim 0.25$ found in iron based langasite [25] and other two-dimensional triangular magnets [26]. The fluctuations critical to magnetic order in FeCrAs also differ from iron based pnictides and chalcogenides which broadly display Ising universality class behavior [7,27–29]. However, the mean-field critical exponent is expected for an itinerant ferromagnetic transition. For example, Moriya's spin-fluctuation theory predicts the temperature dependence of $M \sim (1 - T/T_c)^{1/2}$ [30].

B. Magnetic dynamics from inelastic neutron scattering

We now discuss the magnetic dynamics as measured by inelastic neutron scattering. Figure 3 illustrates the high-energy

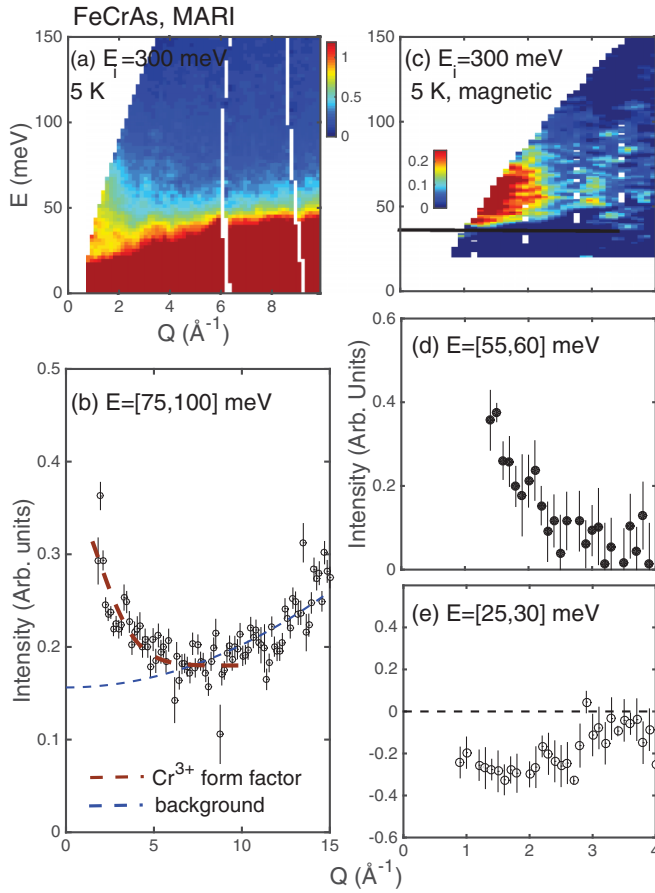


FIG. 3. (a) Powder-averaged inelastic neutron spectrum in FeCrAs taken on MARI. The intensity between 75 meV and 100 meV is integrated and plotted as a momentum cut in panel (b). The blue dashed line is an estimate of the background from extrapolating from large momentum transfers as described in the text and the dark red curve is the scale magnetic Cr^{3+} form factor. (c) Illustrates the same data as in panel (a), but with the background removed. The solid line at $E = 40$ meV shows where the background subtraction fails due to strong and highly structured in momentum phonon scattering. Constant energy cuts from this panel are plotted in (d) for the energy interval $E = [55, 60]$ meV and (e) for the energy interval $E = [25, 30]$ meV.

spectroscopy measurements performed on the MARI chopper spectrometer. Panel (a) displays a powder-averaged energy-momentum map at 5 K showing the presence of scattering at low momentum transfers above ~ 50 meV which decays rapidly with Q . The white region corresponds to where no data could be taken due to kinematic constraints of neutron scattering imposed by a minimum scattering angle of $2\theta \sim 3^\circ$. Panel (b) shows a constant energy cut illustrating the presence of two components to the scattering: one rapidly decaying with momentum, indicative of magnetic fluctuations and well described by the Cr^{3+} form factor, and the other slowly increasing at large momentum transfers, characteristic of a phonon contribution. To extract magnetic fluctuations at high energy transfers, we relied on the fact that the magnetic scattering is confined to small momentum transfers and decays with increasing Q while the phonon background increases with Q^2 .

We have separated the two components by fitting the high angle detector intensity (where magnetic scattering is expected to be negligibly weak) to $I_{BG} = B_0 + B_1 Q^2$ and extrapolating to small momentum transfers. An example of this analysis is illustrated by the dashed curves in Fig. 3(b) which show a cut integrated over energies between 75 and 100 meV. The dashed lines in (b) show an estimate of the background based on a fit to the high angle detectors and also the Cr^{3+} form factor scaled by a constant factor to agree with the low- Q momentum dependence. The result of applying this analysis to each energy transfer and subtracting the high- Q background is shown by the false color image in panel (c). Individual cuts integrating over $E = [55, 60]$ meV and $E = [25, 30]$ meV are plotted in panels (d) and (e). The analysis successfully extracts magnetic intensity for energy transfers above ~ 45 meV but failed to separate out the magnetic and phonon contribution at lower energy transfers resulting in an over subtraction of intensity. This is seen in the false color image in panel (c) and further displayed through constant energy cuts in panels (d) and (e). While the background subtraction works at large energy transfers as shown in panel (d), the assumptions behind this background correction break down for low-energy transfers, where the phonon scattering becomes intense and highly structured in momentum as shown in panel (e). Therefore, we have removed the region below 20 meV from the plots. We note that this technique for background subtraction has been successfully applied previously to studying high energy $d-d$ transitions in NiO and CoO [31,32]. It was also applied to extract the magnetic fluctuations in $\alpha\text{-NaMnO}_2$ [33]. In all of these cases the analysis was only applied to a region in momentum energy where the powder averaged phonon contribution was small and unstructured in Q .

Given the failure to extract reliable magnetic scattering below ~ 40 meV using the MARI direct geometry spectrometer we have used the MACS cold triple-axis spectrometer with a low fixed $E_f = 2.4$ meV to investigate the magnetic response at low energy transfers. This configuration kinematically affords access to low momentum transfers where phonon scattering is expected to be negligible. The background corrected data from MACS is compared against the high-energy magnetic response extracted used in MARI in Figs. 4(a) and 4(b). Steeply dispersing magnetic fluctuations are observable at low energies below ~ 6 meV, emanating from Q positions which correspond to the propagation vector of $\vec{q}_0 = (\frac{1}{3}, \frac{1}{3})$. Further magnetic fluctuations are observable above ~ 40 meV using MARI. Data between these two energy ranges, bridging the MACS and MARI data sets, could not be reliably extracted, as discussed above, due to both kinematic constraints of neutron scattering and also the substantial phonon background over this energy range highlighted in Fig. 3 panel (c).

The MACS data in Fig. 4(b) reveal additional weak magnetic scattering near 3 meV suggestive of a second low-frequency magnetic mode. It is possible that this mode is the second transverse mode (magnon) with a gap of about 3 meV resulting from a weak easy plane anisotropy. Another possibility is that this intensity arises from a longitudinal mode, similar to what has been found in other metallic magnets [34]. Experiments using single crystal samples are necessary to address the nature of these low energy modes. The high velocity, or stiff, spin excitations extend up to 6 meV beyond

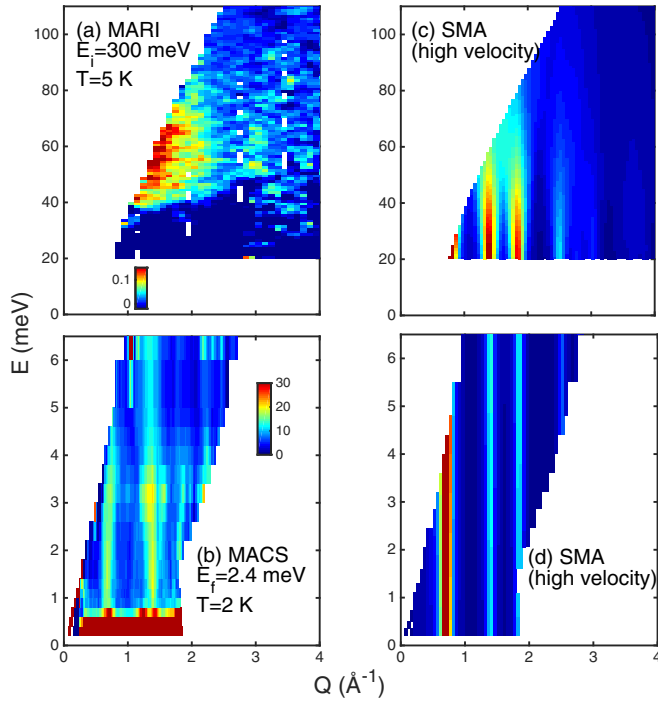


FIG. 4. The powder averaged magnetic response at 5 K in FeCrAs measured with (a) $E_i = 300$ meV (MARI, ISIS) and (b) $E_f = 2.4$ meV (MACS, NIST). The variation in pixel size as a function of energy transfer in the MACS data, panel (b), is due to the difference in the way the data is collected. MARI data was collected in a time of flight configuration while MACS is a triple-axis which each energy transfer corresponding to a different constant energy scan. (c),(d) The powder averaged heuristic parametrization based on the single mode approximation (SMA) discussed in the text. The calculation was done assuming two-dimensional linear spin waves with a velocity of 200 meV \AA .

which they are outside of the observation window on MACS. The fact that these excitations form steep rods in Q seen in the MACS data allows us to speculate that they link to the high-energy response observed on MARI. We discuss this point below by applying a parametrization, illustrated in Figs. 4(c) and 4(d), based on the first moment sum rule.

Above, we have relied on the momentum dependence to extract the magnetic intensity. To further confirm the magnetic origin of the low-angle response, we have measured the magnetic fluctuations at higher temperature shown in Fig. 5 which plots the extracted magnetic scattering with $E_i = 150$ meV at 5 K and 150 K, below and above T_N , respectively. Background corrected false color maps at these two temperatures are shown in panels (a) and (b) with constant energy cuts shown in panels (c) and (d). While the low- Q excitations are still present at high temperatures, indicative of a large underlying energy scale, a decrease in the scattering confirms the magnetic origin of this scattering present at small momentum transfers.

C. Parameterization in terms of high velocity damped spin waves

The two data sets from time of flight and triple-axis spectroscopy show magnetic excitations at high and low energy

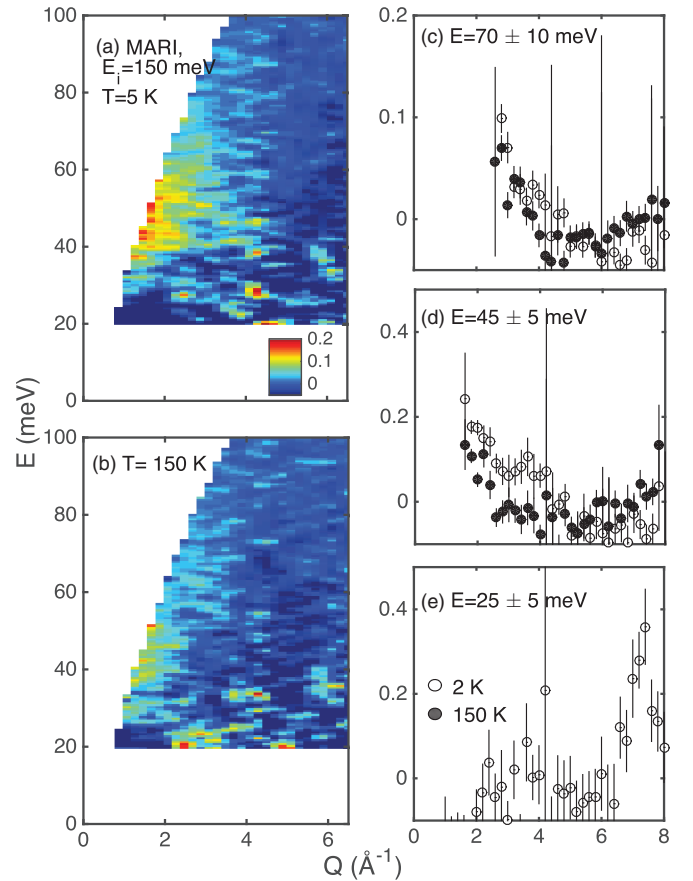


FIG. 5. The powder averaged magnetic response measured with $E_i = 150$ meV at (a) $T = 5$ K and (b) 150 K. (c),(d) Constant energy cuts illustrating the decay of magnetic intensity with momentum transfer. Panel (e) illustrates an energy cut at 25 meV where phonon scattering prevents a reliable subtraction of the background.

regime quite clearly; however, we note that it is difficult to measure magnetic excitations in the intermediate energy regime connecting these two data sets because of strong phonon scattering. To illustrate a consistent link between the low and high energy data sets, we have parameterized the spin fluctuations by high velocity damped spin waves from the magnetic $(\frac{1}{3}, \frac{1}{3})$ positions. We have simulated the scattering using the following form motivated by the Hohenberg-Brinkmann first moment sum rule applied in the case of a dominant single mode, known as the single mode approximation [35]. This approach has been applied to low-dimensional organic magnets (Refs. [36,37]) and the form reflects that used to describe magnetic excitations in powder samples of triangular magnets (Refs. [33,38]).

$$S(\vec{Q}, E) \propto \frac{1}{\epsilon(\vec{Q})} \gamma(\vec{Q}) f^2(Q) \delta(E - \epsilon(\vec{Q})),$$

where $\gamma(\vec{Q})$ is a geometric term chosen to peak at the Bragg positions with propagation vector $(\frac{1}{3}, \frac{1}{3})$, $f^2(Q)$ is the magnetic form factor for Cr^{3+} , $\delta(E - \epsilon(\vec{Q}))$ is an energy conserving delta function, and $\epsilon(\vec{Q})$ is the dispersion relation for the spin excitations. We only consider Cr^{3+} moments here because the iron moment is negligibly small as discussed above. Given

that the scattering is concentrated at low momentum transfers and a large portion is kinematically inaccessible, we are not able to derive an accurate measure of the total integrated intensity for comparison to sum rules of neutron scattering. For the calculations shown here, we have taken the spin wave dispersion to be two dimensional (within the a - b plane) and also be linear given that no upper band is observed. Powder averaging was done using a finite grid of 10^4 points and summed at each momentum and energy transfer. We note due to powder averaging it is difficult to make any conclusions from the data regarding any continuum scattering that may exist owing to longitudinal spin fluctuations as observed in other itinerant systems [39]. As displayed in Fig. 4 panel (d), the combination of powder averaging results in scattering over an extended range in momentum transfer. Given kinematics associated with the $(\frac{1}{3}, \frac{1}{3})$ type order, we are not able to draw any conclusions about possible ferromagnetic fluctuations that may exist near $Q = 0$.

The results of this calculation using a linear and three-dimensional spin-wave velocity of $\hbar c = 200$ meV Å and performing the powder average are shown in Figs. 4(c) and 4(d). The calculation confirms that the two experimental data sets presented in Figs. 4(a) and 4(b) can be consistently understood in terms of high velocity spin waves emanating from the $(\frac{1}{3}, \frac{1}{3})$ positions. As seen in Fig. 4(c), the magnetic form factor ensures that the magnetic scattering is suppressed at large momentum transfers. The value used in this calculation, $\hbar c = 200$ meV Å should be considered as a lower bound of the spin wave velocity. The steep velocity ensures magnetic scattering is confined to low scattering angles as observed experimentally which are eventually completely masked at high energy transfers by kinematic constraints of neutron scattering. One thing that is not clear in this analysis is the highest energy scale of the steeply dispersing magnetic excitations. Our measurements do not reveal a high energy peak in the powder averaged spectra that would result from an enhanced density of states for zone boundary spin waves and instead we observe an apparent high energy continuum. This may be attributed to either a combination of kinematic constraints and the magnetic form factor, or possibly to strong damping of the highest energy magnetic excitations that results from coupling to conduction electrons. The latter case occurs in classic itinerant magnets [40]. We note that our model of three-dimensional spin waves emanating from magnetic $(\frac{1}{3}, \frac{1}{3})$ does not capture the momentum dependent intensity of the spin excitations at low energy transfers measured on MACS [Fig. 4(b)]. We speculate that such modulation with momentum originates from a more complex momentum dependence not captured in our analysis originating from unusual magnetic structure. To refine a model to capture this, single crystal data is required.

Our parametrization of the data in terms of three-dimensional and high velocity spin waves emanating from $\vec{q}_0 = (1/3, 1/3)$ positions is arguably the simplest model that is consistent with the three-dimensional nature of the resistivity and also the structure discussed above. However, it should be emphasized that powder averaging does mask features that would become clear in single crystals. It is possible that the three-dimensional character of the spin excitations is only present at low energies crossing over to two-dimensional

excitations at higher energies. Indeed, our heuristic model does not capture the additional scattering at ~ 3 meV measured on MACS which could be suggestive of such a scenario. As an example, we point to powder averaged spin excitations in BaFe_2As_2 [41] which did give clear ridges of scattering up to high energies while later single crystal work confirmed the two-dimensional character. Our data and parametrization does show that high velocity spin excitations are present up to unusually high energies in FeCrAs with the exact nature of the dimensionality made ambiguous from the powder averaging.

IV. DISCUSSION

Our neutron diffraction measurements (with resolution ~ 2 meV) show that the magnetic order sets in around $T_N = 115$ K in FeCrAs, and the sublattice magnetization is well described with a mean-field critical exponent of $\beta = 1/2$. The inelastic neutron scattering measurements show that the low-energy spin excitations of FeCrAs are well-defined gapless spin waves extending up to ~ 6 meV. The spin excitations are observed up to high energy transfers of at least 80 meV. Such a large energy scale of these spin excitations indicates an underlying magnetic energy scale that is significantly larger than that estimated from local moment molecular field model (see Sec. III A).

These observations are quite reminiscent of the spin excitations observed in chromium metal. The incommensurate spin-density wave order in Cr is considered as a textbook example of magnetic order driven by the Fermi surface nesting [42]. Its spin excitation spectrum has been the subject of intense investigation both theoretically and experimentally [34,43–47]. Experimentally, spin-wave-like excitations with very steep dispersion have been observed by inelastic neutron scattering measurements [34,46,47]. Theoretical studies showed that the transverse spin fluctuations in the long wavelength limit can be described by spin-wave modes even for this type of itinerant systems [43–45]. That is, $\omega = c|q|$, but the spin-wave velocity is given by $c = v_F/\sqrt{3}$, where v_F is the Fermi velocity which originates from charge physics and therefore is much larger than typical spin-wave velocity observed in a localized spin model. In addition to the transverse spin waves, a longitudinal mode is allowed and, in fact, has been observed to be quite strong [47]. The observed spin wave velocity is a weighted combination of transverse and longitudinal modes and so can differ significantly from $c = v_F/\sqrt{3}$. In Cr, the longitudinal fluctuation renormalizes the apparent spin-wave velocity down [34] and the apparent spin-wave velocity $\hbar c(\text{Cr})$ is given by $\hbar c(\text{Cr}) \sim \hbar\sqrt{c_L c_T} \sim 1000$ meV Å where c_L and c_T denote longitudinal and transverse velocity of Cr.

Since the carrier density in FeCrAs is known from the first principles calculation ($n = 2 \times 10^{28}$ m $^{-3}$), we can estimate $v_F/\sqrt{3} \sim 4000$ meV Å. Although this value is much larger than the spin wave velocity used in Fig. 4, we do not consider this a significant numerical discrepancy. First, the spin-wave velocity used in Fig. 4 is just a lower bound, and the data will be still adequately described with a larger value of c . Second, the Sommerfeld coefficient of 30 mJ/mol K 2 suggests a large renormalization of the bare Fermi velocity. Finally, longitudinal magnetic excitations are expected to reduce the apparent spin-wave velocity. Of course a calculation based on the real

band structure would be necessary to obtain a more quantitative comparison between itinerant theory and experiment.

We would like to point out that there is a growing list of materials which display low-energy localized excitations but itinerant fluctuations at higher energy transfers. Fe_{1+x}Te [48–52] has been found to have localized transverse fluctuations at low energies which cross over to high energy fluctuations resembling more itinerant fluctuations [53,54]. CeRhIn_5 shows well defined localized spin waves which break-down into a multiparticle continuum [39]. $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ similarly displays localized low-energy fluctuations but itinerant fluctuations at high energies [55,56]. However, unlike above materials, FeCrAs is far from the quasi-two-dimensional limit. The observed weak resistivity anisotropy is a strong indicator of this, with an additional support provided by our observation of mean-field critical exponent. In the parent compounds of iron or copper based superconductors, the critical behavior is usually governed by strong 2D fluctuations, giving rise to critical exponents in the range of $\beta \sim 0.2\text{--}0.3$, much smaller than the observed mean-field exponent [7,27–29,57].

We now discuss the relation between the spin excitations and the unusual response measured in resistivity. The data shows fast spin excitations at low momentum transfers. The MACS data illustrate that the excitations are originating from finite Q but extend up to high energy transfers. A central question in FeCrAs is the origin of the unusual metallic properties with the resistivity increasing in a power-law fashion from 600 K. The resistivity from spin fluctuations has been suggested to have the following form in the context of work done on the cuprate superconductors [58,59]

$$\rho(T) \propto T \int_{-\infty}^{\infty} \frac{E}{T} d\left(\frac{E}{T}\right) \frac{e^{E/T}}{(e^{E/T} - 1)^2} \int d^3q \chi''(\vec{q}, E).$$

Given that the neutron scattering cross section $I(Q, E) \propto S(Q, E) = \frac{1}{\pi}[n(E) + 1]\chi''(Q, E)$, an energy independent $\int d^3q \chi''(\vec{q}, E)$ would result in a resistivity which has linear temperature dependence. If, however, this local susceptibility integral term decreased slowly with increasing temperature, then a temperature independent resistivity may be explained.

While the kinematic constraints of our experiment preclude measurement of the temperature dependent local susceptibility, we do observe only a weak decrease of high energy magnetic intensity with increasing temperature implying that the associated change in local susceptibility is small. The large energy scale of the fluctuations inevitably will affect the resistivity over a very broad temperature scale.

The measurements above find two results in the context of the dynamics: first that the magnetic excitations are gapless down to the energy scale set by the ~ 0.5 meV resolution of MACS, and second, the high energy scale fluctuations are present at high temperatures above T_N . The large energy scale and gapless nature of the spin fluctuations may provide an explanation for the unusual transport response. A similar coupling between spin fluctuations and the electron response was suggested in Fe_{1+x}Te which also display little change in the resistivity over a broad range in temperature [60]. Indeed, only when the magnetic fluctuations become gapped in Fe_{1+x}Te does the resistivity drop and the two can be correlated using the relation above [4]. FeCrAs may represent an extreme example with gapless spin excitations that extend up to at least 80 meV (~ 926 K).

In summary, we studied critical behavior of the magnetic order parameter near the Neel transition in FeCrAs and observed that the temperature dependence of the magnetic order parameter is described with the mean-field critical exponent. Our neutron spectroscopy measurements reveal high velocity gapless spin wave excitations which extend up to at least ~ 80 meV, which resembles spin excitations in itinerant magnets. We suggest that coupling between this broad-band spin fluctuations is the origin of the unusual resistivity measured in this “nonmetal metal.”

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