Non-metallic, non-Fermi-liquid resistivity of FeCrAs from 0 to 17 GPa

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Abstract
An unusual, non-metallic resistivity of the 111 iron–pnictide compound FeCrAs is shown to be relatively unchanged under pressures of up to 17 GPa. Combined with our previous finding that this non-metallic behaviour persists from at least 80 mK to 800 K, this shows that the non-metallic phase is exceptionally robust. Antiferromagnetic order, with a Néel temperature $T_N \sim 125$ K at ambient pressure, is suppressed by pressure at a rate of $7.0 \pm 0.4$ K GPa$^{-1}$, falling to $\sim 50$ K at 10 GPa. We conclude that the formation of a spin-density-wave gap at $T_N$ does not play an important role in the non-metallic resistivity of FeCrAs at low temperatures.

1. Introduction

According to band theory, the resistivity of an insulator rises exponentially with decreasing temperature because thermally activated carriers are being frozen out. The resistivity of a metal, in contrast, falls with decreasing temperature, because the density of carriers is fixed while thermally excited scattering is frozen out. However, there are exceptions—‘non-metallic metals’—where the resistivity rises at low temperature as the temperature falls, but without a gap opening in the electronic excitation spectrum. Some of these exceptions are well understood, for example Kondo systems or disordered metals showing weak or strong localization according to whether they have high or low carrier densities. There are also exceptions that are not understood, such as the underdoped cuprates [1, 2], and some disordered heavy fermion compounds [3]. These systems may have high carrier density yet display strongly non-metallic resistivity as the temperature is varied between room temperature and $T \to 0$ K. Moreover, in the $T \to 0$ K limit the non-metallic heavy fermion systems may show non-Fermi-liquid properties in their resistivity, linear specific heat coefficient, and magnetic susceptibility [3]: $\rho \propto \rho_0 - AT$, $C/T \propto -\ln(T)$, $\chi(T) \propto 1 - c\sqrt{T}$.

The distinction between ‘non-metallic’ and ‘non-Fermi-liquid’ behaviour is important in these materials. Nozières [4] argued in the context of the single-impurity Kondo problem that a metal with internal degrees of freedom such as fluctuating spins should be a Fermi liquid, with $T$-linear specific heat, Pauli-like susceptibility, and a $T^2$ but non-metallic resistivity $\rho(T) = \rho_0 - AT^2$, provided that the temperature is low enough that internal degrees of freedom are locked (corresponding to $T \ll T_K$ in the Kondo problem).

Recently, the high temperature resistivity of some iron–pnictides has also been found to be flat, or slowly rising, with decreasing temperature, even in undoped parent compounds where disorder levels are low [5]. In these systems, however, the resistivity generally begins to fall, and becomes metallic, when magnetic order sets in, typically around 100–200 K. It has been suggested that many-body effects, arising from orbital degeneracy [6] or spin-fluctuations [7], may be responsible for the anomalous temperature dependence of their high temperature resistivity.

We recently showed that the hexagonal iron–pnictide, FeCrAs, is an extreme example of such behaviour [8]: over four decades of temperature from 800 K to below 80 mK, single crystals show a resistivity in the hexagonal plane that rises monotonically with decreasing temperature, but...
without showing the presence of a gap (see figure 1). The c-axis resistivity is similar, except for a local maximum at a Néel ordering transition near 125 K. In the limit as \( T \to 0 \) K, the resistivity shows a non-metallic rise with a non-Fermi-liquid power law dependence on temperature, \( \rho(T) = \rho_0 - A T^{0.65 \pm 0.1} \). In the context of the Nozières argument cited above, the non-\( T^2 \) temperature dependence of the resistivity of FeCrAs is significant, particularly because the specific heat, \( C(T) \), and the ac-susceptibility, \( \chi(T) \), obey Fermi liquid power laws as \( T \to 0 \) K. This combination of non-Fermi-liquid transport with Fermi liquid thermodynamic properties is very unusual. Moreover, the linear coefficient of specific heat is large, \( C(T)/T \sim 30 \text{ mJ (mol K}^2) \), as is the Pauli susceptibility, showing that FeCrAs has a large density of states at the Fermi energy. If the carrier density were small, as in some non-metallic metals, this value of \( C(T)/T \) would require extremely massive quasiparticles, which would itself be unusual for a 3d-electron system. Band-structure calculations, however, predict a large carrier density [9], with three large, three-dimensional Fermi surfaces [10]. According to neutron diffraction measurements, levels of disorder in our single crystals are low [11].

The physics behind the non-metallic resistivity of FeCrAs is not known. Its general behaviour is somewhat reminiscent of systems that are on the border of Anderson localization, in which the Fermi energy is close to the mobility edge separating extended states from localized states in disordered semiconductors [12]. Such materials can show a non-metallic resistivity combined at low temperature with a linear \( C(T) \), however the carrier density in FeCrAs seems much too high, and the level of disorder much too low, for it to be in this regime. Thus, for example, in well-known cases with a non-metallic resistivity such as phosphorus-doped silicon or non-stoichiometric Ce\(_{3-x}\)S\(_4\) [13, 14] the resistivity on the border of Anderson localization is over 100 times larger than we see in FeCrAs, while in Si:P which has a linear specific heat at low temperature, \( C(T)/T \) is 300 times smaller than we see in FeCrAs [15].

An obstacle to understanding the physics of FeCrAs is the antiferromagnetic transition near 125 K. As samples are cooled through this transition, the c-axis resistivity falls abruptly in the highest quality crystals, before continuing to rise again at lower temperature, as shown in figure 1. This behaviour suggests that there is some spin-fluctuation scattering, but it might also be compatible with an orbital mechanism. The \( ab \)-plane resistivity does not fall upon cooling below \( T_N \); indeed if anything the slope of \( d\rho_{ab}/dT \) is steepest just below \( T_N \) (see figure 1), producing a weak ‘bump’, or concave downwards region, below \( T_N \).

It would be interesting to know the low temperature limiting behaviour of the resistivity in the absence of antiferromagnetic order. A possible scenario is that the resistivity would saturate, or even begin to fall, as \( T \to 0 \) K in the absence of magnetic order that opens a spin-density-wave gap over part or all of the Fermi surface. Or, at the other extreme, perhaps a full gap would open over the entire Fermi surface, leading to diverging resistivity as \( T \to 0 \) K, but antiferromagnetism prevents the opening of this gap and leaves a semi-metallic state with a small Fermi surface.

The antiferromagnetic order in FeCrAs is itself unusual, and indicative of some level of frustration due to the \( P62m \) crystal structure, which can be viewed as a triangular lattice of iron ‘trimers’, plus a distorted Kagome sublattice of Cr ions. Magnetic order is found only on the Cr sublattice, in the form of a commensurate spin-density-wave that triples the unit cell in the hexagonal plane, while along the c-axis the moments are parallel [8, 16]. At low temperature in the commensurate spin-density-wave state, neutron scattering measurements find that the ordered moments on the Cr sublattice vary between 2.2 and 0.6 \( \mu_B \) [8], while Mössbauer spectroscopy places an upper limit of 0.1 ± 0.03 \( \mu_B \) on the ordered moment on the Fe sites [17]. The Néel temperature is low compared with the comparable tetragonal systems Fe\(_2\)As and Cr\(_2\)As, which have \( T_N \sim 350 \) K [18] and \( T_N \sim 393 \) K [19] respectively. In FeCrAs, the iron site is tetrahedrally coordinated by As, as in the iron–pnictide superconductors, and the absence of a measurable ordered moment below \( T_N \) is in agreement with the predictions of band-structure calculations [9] that found that the partial density of states on the iron sites is too low to meet the Stoner criterion for magnetic moment formation, and indeed in a recent paper the iron K\( \beta \) x-ray emission spectrum from FeCrAs was used to provide a non-magnetic reference compound [20]. It should be noted, however, that in the related tetrahedral compound Fe\(_2\)As the iron moment on the tetrahedrally coordinated site is 1.28 \( \mu_B \) [18], suggesting that this site is close to the magnetic/non-magnetic moment-formation boundary, and that frustration may also play a role in moment suppression on the iron site.

The physics of frustrated metallic magnets still has many open questions [21, 22]. Based on the frustrated magnetic sublattices and the absence of a magnetic moment on the iron...
sites, Rau et al have put forward a theory that the anomalous behaviour of FeCrAs arises from a 'hidden spin liquid' on the iron sublattice [23]. In this picture, the conduction electrons fractionalize, and anomalous transport is due to scattering of bosonic charge degrees of freedom off strong gauge fluctuations.

In this paper we try to determine whether the antiferromagnetism is playing an important role, particularly in the $T \to 0$ K limit, by using pressure to adjust $T_N$. We find that, despite suppressing $T_N$ by more than a factor of two, and possibly all the way to 0 K in our highest pressure measurements, the general behaviour of the anomalous resistivity is not dramatically modified, suggesting that the opening of a spin-density-wave gap does not play an important role in the non-metallic resistivity of FeCrAs.

2. Experiment

We have carried out four-terminal resistivity measurements on single crystal samples of FeCrAs at high pressure. Crystals were grown from a stoichiometric melt in an alumina crucible within a sealed quartz tube. The material was melted twice and then annealed at 900 °C for 150 h. The sample quality in FeCrAs is revealed by the sharpness of the resistive transition at $T_N$, the value of $T_N$, and the temperature at which glassy behaviour in the magnetic susceptibility sets in. The crystals used in these measurements were from our highest quality batch [8], in which $T_N \sim 125$ K in susceptibility measurements, $T_N \sim 133$ K according to the cusp in the $c$-axis resistivity, and in which glassy behaviour is very weak and only sets in below 10 K. Details of crystal growth and characterization can be found in [11].

Electrical contacts to the samples were made with Dupont 6838 epoxy. These had high resistivities at ambient pressure, but under pressure they fell to the range of a few ohms.

We pressurized two single crystals, one with $I \parallel c$ which measures $\rho_c$, and the other with $I \perp c$, measuring $\rho_{ab}$. The $\rho_c$ sample had dimensions $250 \times 200 \times 30 \mu$m$^3$. It was pressurized in a Moissanite anvil cell with 800 $\mu$m culets. The gasket was beryllium–copper, with a 400 $\mu$m hole, insulated with a mixture of alumina-powder and stycast 1266 epoxy. The $\rho_{ab}$ sample had dimensions $250 \mu$m $\times 100 \mu$m $\times 25 \mu$m and was pressurized in a diamond anvil cell with 600 $\mu$m culets, using a fully hardened T301 stainless-steel gasket.

Daphne oil 7373 was used as the pressure medium. The pressure was determined at room temperature using ruby fluorescence; the pressure may shift by up to $\sim 0.4$ GPa while the cell is cooled. Moreover, Daphne oil 7373 is known to become non-ideal with increasing pressure; Tateiwa and Haga [24] using ruby fluorescence in a diamond anvil cell found that non-hydrostaticity/uniaxial deviations versus pressure at $77$ K grow quite rapidly above $2.0$ GPa, from $\pm 0.05$ GPa at $2.0$ GPa to around $\pm 1.0$ GPa at $10$ GPa.

The $\rho_c$ sample survived up to 10 GPa before an anvil broke, while the $\rho_{ab}$ sample survived up to 17 GPa.

In order to track the pressure dependence of $T_N$ we made use of the peak in $\rho_c$ at $T_N$. Unfortunately, $\rho_{ab}$ does not have a well-defined anomaly at $T_N$, as can be seen in figure 1, so we could only follow $T_N$ versus pressure with confidence up to 10 GPa.

A possible concern with all high pressure measurements is pressure-induced structural phase transitions. Among the 111 pnictides, however, FeCrAs should be relatively immune to such transitions. The 111 pnictides come in three main crystal structures: tetrahedral, hexagonal, and orthorhombic, in order of decreasing unit cell volume [25]. Both Fe$_2$As and Cr$_2$As have the tetrahedral structure, and there is only a narrow range of stability of the hexagonal phase around the FeCrAs stoichiometry, thus FeCrAs must be just barely below the volume criterion of stability for the hexagonal phase. We thus expect it to be able to withstand quite a lot of compression before it transforms to the orthorhombic phase, and indeed in our measurements we did not see any abrupt changes in resistivity that would indicate a change of structure.

Resistivity measurements were carried out at many pressures, as shown in figures 2 and 5. At each pressure the temperature was varied between room temperature and 2 K, using a dipping probe to control the temperature.

3. Results

Figure 2 shows $\rho_c$ versus $T$ at pressures between 0.8 and 9.7 GPa. The main features of this plot are: (1) the curves shift downwards with increasing pressure, showing that the sample becomes more conducting with increasing pressure, however the overall effect is small; (2) $T_N$ is suppressed by pressure, as shown by the shift to lower temperature of the peak in $\rho_c$, which is known from our previous measurements to coincide with $T_N$ [8]; and (3) the overall shape of $\rho_c$ versus $T$ does not change markedly.

Point (3) is our key result. Despite the fact that $T_N$ is suppressed by as much as a factor of two, for $T > T_N$ the resistivity remains non-metallic with little change in slope. The $T \to 0$ K slope is also roughly independent of pressure,
Figure 3. The first (a) and second (b) derivatives of $\rho_c(T)$ versus $T$. The second-derivative curves have been shifted vertically for clarity. The lowest pressure curves show zero-crossings of $d\rho/dT$ and well-defined minima of $d^2\rho/dT^2$ that accurately locate $T_N$, plotted in figure 4. At the highest pressures $T_N$ was estimated from the changes in slope of $d\rho/dT$ and, especially, $d^2\rho/dT^2$.

Figure 4. $T_N$, extracted as described in the text from the data in figure 2, versus pressure. A linear extrapolation (dashed line) would place the quantum critical point at $P_c \approx 16.5 \pm 1$ GPa. The inset shows the pressure dependence of the slope $d\rho/dT$ versus $T$ for $T > T_N$, obtained by fitting a linear function to the data between 150 and 300 K. No significant pressure dependence is observed, showing the robustness of the non-metallic resistivity behaviour against pressure.

remaining non-Fermi-liquid like at all pressures, with the same power law behaviour within the error, $\rho_c \sim \rho_{c,0} - AP^{0.7 \pm 0.1}$, as was observed at ambient pressure.

We have constructed a phase diagram of $T_N$ versus $P$, extracting $T_N$ with the help of plots of the first and second derivatives of $\rho_c(T)$ with respect to $T$, which are shown in figure 3. The derivatives at each temperature $T$ were obtained by fitting a fifth-order polynomial to $\rho_c(T)$ in a region 40 K wide surrounding $T$, and then taking the derivatives of the polynomial. Up to 6.3 GPa, $d\rho_c/dT$ has a clear zero-crossing (corresponding to a local maximum of $\rho_c(T)$), while $d^2\rho/dT^2$ has a well-defined minimum, and using these as criteria for $T_N$ gives results that agree well with each other, as indicated by the comparatively small error bars on $T_N$ for these pressures in the main panel of figure 4. For the highest pressure curves, however, neither of these criteria is well defined, perhaps due to the increased non-hydrostaticity of the Daphne oil pressure medium. At these pressures the only robust feature is a sharp upturn in $d^2\rho/dT^2$ with decreasing $T$. At low pressure this upturn is located just below the $T_N$ minimum, so for the 8.8 and 9.7 GPa data we have estimated $T_N$ to be located at the first shoulder above the upturn, but with large error bars indicating the uncertainty of this assignment. The resulting estimated values of $T_N$ are consistent with the maxima in $d\rho/dT$ at these pressures.

The phase diagram, figure 4, allows us to elaborate on points (2) and (3) above. The main figure shows a plot of $T_N$ versus $P$. We see that $T_N$ falls roughly linearly with pressure. Fitting a straight line to the points in figure 3 gives $dT_N/dP = -7.0 \pm 0.4 \text{ K GPa}^{-1}$. If we extrapolate this linear behaviour to estimate the pressure at which the quantum critical point, $T_N = 0 \text{ K}$, would be reached, then we find $P_c \sim 16.5 \pm 1$ GPa. It should be noted, however, that such extrapolations are not always reliable: $T_N$ versus $P$ curves can turn downwards [26].
or saturate [27], so that $P_c$ may be significantly smaller or larger than this estimate.

The inset of figure 4 shows that the high temperature slope of $\rho_c$ versus $T$ is unaffected by pressure, within the error, emphasizing that pressure has little effect on the non-metallic resistivity at $T > T_N$.

Figure 5 shows $\rho_{ab}$ versus $T$ at four pressures between 4.3 and 17.3 GPa. Unfortunately, $\rho_{ab}$ does not have a sharp feature at $T_N$ (see figure 1), so we cannot continue our $T_N$ versus $P$ curve using these data. As with $\rho_c$, $\rho_{ab}$ decreases with increasing pressure, the overall effect is small, and the non-metallic temperature dependence is relatively unaffected by pressure up to 17.3 GPa. The high temperature slope, shown in the inset, is unchanged within the error.

The $T \to 0$ K non-metallic behaviour of $\rho_{ab}$ versus $T$ also persists to high pressure, however the slope is smaller in the two highest pressure curves, and the unusual sublinear temperature dependence crosses over to become more linear in $T$. At intermediate temperatures the concave region, seen at ambient pressure below $T_N$, seems to have been suppressed in these highest pressure curves. This would be consistent with the extrapolation of $T_N$ in figure 4, so antiferromagnetism may indeed have been suppressed by 17.3 GPa. As a result of the suppression of this concave downward section, $\rho_{ab}$ at 17.3 GPa looks quasilinear over the whole temperature range.

We have seen no indication of superconductivity in any of our data, although we have measured only down to 2 K.

4. Discussion

In these measurements we have suppressed $T_N$ by at least a factor of two, and if the extrapolation of $T_N$ versus $P$ beyond 10 GPa can be trusted, then in our measurements up to 17.3 GPa on the $\rho_{ab}$ sample $T_N$ may have been suppressed to 0 K. Despite this suppression of antiferromagnetic order, the overall shape of the resistivity curves is not dramatically affected: the high temperature resistivity remains non-metallic with minimal change of its large, negative slope, while the $T \to 0$ K resistivity also remains non-metallic, with non-Fermi-liquid power laws persisting to all but the two highest pressures, and with remarkably little change of slope. The most notable changes that we do observe are the gradual disappearance of the concave downward regions in $\rho$ versus $T$ that are associated with antiferromagnetic order, and an indication in the $\rho_{ab}$ sample that for $P > 10$ GPa the $T \to 0$ K resistivity crosses over from sublinear to linear dependence on $T$. This latter change may have to do with the antiferromagnetic quantum critical point being approached. Linear resistivities are typical of antiferromagnetic quantum critical points, although in all of the cases that we know of, the slope of $\rho$ versus $T$ is positive, not negative as in FeCrAs.

There are few materials to which we can compare these results. The robustness of the non-metallic resistivity of FeCrAs under pressure is in sharp contrast to that of CeCuAs$_2$ [28], whose strongly non-metallic resistivity between room temperature and 1.8 K is completely suppressed at 10 GPa to produce a metallic resistivity over the entire temperature range. LaFeAsO is the iron-pnictide superconductor whose resistivity most closely resembles FeCrAs: for $T > 200$ K its ambient pressure resistivity is nearly flat, although unlike FeCrAs its resistivity becomes metallic for $T < 200$ K. As pressure is increased up to 12 GPa, the magnitude of the high temperature resistivity falls, but the non-metallic slope remains flat [29]. Thus, the behaviour is like FeCrAs in that the resistivity versus temperature curves are displaced downwards by pressure but their slope is not changed. Another 1111 pnictide, CaFeAsF, has a quasilinear, weakly metallic, slope at high temperature, and again this slope is nearly independent of pressure while the resistivity curves displace downwards [29]. The 122 pnictides are even more metallic at high temperature, and in these systems pressure increases the metallic slope somewhat, so pressure makes these materials more metallic, e.g. [30]. One major difference between FeCrAs and all of these systems, however, is that pressure suppresses the total resistivity much more slowly in FeCrAs. For example, in LaFeAsO [29], the non-metallic resistivity at room temperature falls by 55% between 0 and 12 GPa—from 3.8 to 1.7 m$\Omega$ cm—while in FeCrAs $\rho_{ab}$ at room temperature falls by only 10% between 4 and 17 GPa—from 380 to 340 $\mu$Ω cm.

While our results rule out magnetic long-range order as an important factor in the non-metallic resistivity of FeCrAs at low temperature, they do not necessarily exclude spin-fluctuations as playing a role. The iron-pnictide superconductors are believed to be incipient Mott insulators.

In a theoretical study based on this picture, Dai et al decomposed the electronic excitations into a coherent part near the Fermi energy and an incoherent part further away [7]. The latter comprises incipient lower and upper Hubbard bands, which accommodate localized Fe moments. This model supports a magnetic quantum critical point as a result of the competition between magnetic ordering of the local moments and the mixing of the local moments with the coherent electrons. The spectral weight of the coherent quasiparticle peak changes as a result of mixing and once it exceeds a critical value, magnetism disappears. In analogy with this model, the non-metallic behaviour of FeCrAs could be a manifestation of incipient Mott insulating behaviour.

However, there are good reasons for thinking that the physics of FeCrAs may be different. In FeCrAs, local moments reside on the Cr and not the Fe sites, so the incoherent carriers would have to be released from Cr d-orbitals while the coherent part of the spectrum most likely would stem from Fe d-orbitals hybridized with As p-orbitals. Non-Fermi-liquid behaviour could then arise due to a coupling between the two species of carriers. Within this theoretical framework, we naively expect that, because pressure increases the mixing between the coherent and incoherent parts of the spectrum, at some critical pressure the magnetic order must vanish. This model has not been worked out for FeCrAs, and as far as we know it would not explain the Fermi liquid specific heat and magnetic susceptibility that are seen at ambient pressure.

Moreover, it is far from clear that FeCrAs is close to a Mott transition. As noted in the introduction, band-structure calculations predict three large Fermi surfaces, and would
seem to place the system far from a Mott state. Alternative models, for example involving orbital effects [6], a hidden spin liquid [23], microscopic phase-separation [2], or even some exotic form of Kondo effect may more accurately capture the physics.

It would be of interest to carry out optical conductivity and NMR measurements to see if a pseudogap is forming as the resistivity rises with decreasing temperature, and to investigate the spin dynamics. In terms of possible spin liquid states [23], thermal conductivity measurements at low temperature may be enlightening.

5. Conclusions

We have found that suppression of antiferromagnetic long-range order does not strongly affect the non-metallic resistivity seen across several decades of temperature in FeCrAs. Moreover, the non-Fermi-liquid power law behaviour of the resistivity as \( T \to 0 \) K at most crosses over from sublinear below 10 GPa to linear above 10 GPa. These results rule out the formation of spin-density-wave gaps on the Fermi surface as playing an important role in the anomalous non-metallic resistivity of this material.

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