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Direct observation of a Fermi liquid-like normal state in an iron-pnictide superconductor

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There are two prerequisites for understanding high-temperature (high-\(T_c\)) superconductivity: identifying the pairing interaction and obtaining a correct description of the normal state from which superconductivity emerges. The nature of the normal state of iron-pnictide superconductors, and the role played by correlations arising from partially screened interactions, are still under debate. Here we show that the normal state of carefully annealed electron-doped BaFe\(_{2-x}\)Co\(_x\)As\(_2\) at low temperatures has all the hallmark properties of a local Fermi liquid, with a more incoherent state emerging at elevated temperatures, an identification made possible using bulk-sensitive optical spectroscopy with high frequency and temperature resolution. The frequency dependent scattering rate extracted from the optical conductivity deviates from the expected scaling \(M_\omega(\omega, T) \propto (\omega^2 + (p\pi k_BT)^2\) with \(p \approx 1.47\) rather than \(p = 2\), indicative of the presence of residual elastic resonant scattering. Excellent agreement between the experimental results and theoretical modeling allows us to extract the characteristic Fermi liquid scale \(T_0 \approx 1700\) K. Our results show that the electron-doped iron-pnictides should be regarded as weakly correlated Fermi liquids with a weak mass enhancement resulting from residual electron-electron scattering from thermally excited quasi-particles.

Strong electronic correlations and Mott physics have played an important role in shaping our understanding of high-\(T_c\) superconductivity (HTSC)\(^1\). With the discovery of the iron-pnictide family of HTSCs a new playground to study correlation effects has emerged\(^2\). Unlike the cuprate HTSC, the pnictides are properly classified as moderately correlated semi-metals\(^3\). By studying their normal state properties a new picture has started to emerge\(^4\) where intra-atomic exchange processes (Hund’s coupling) govern the degree of correlation effects. In the resulting “Hund’s metal” state\(^5\), Hund’s coupling reduces the propensity towards a strongly correlated Mott insulating state, while simultaneously reducing the coherence temperature below which Fermi liquid (FL) properties emerge. A strong dependence of the nature of this Hund’s metal state on orbital filling has been found, providing a natural explanation for the differences between hole- and electron-doped pnictides\(^6,7\). Recently, Werner et al. showed\(^8\) that the combined effect of dynamic screening (manifested through a single particle self-energy, \(\Sigma(\omega, T)\)) and orbital occupancy results in a Fermi-liquid-like state in electron-doped pnictides, while a spin-freezing transition separates an incoherent metal regime from the FL regime in hole-doped materials (for a more extensive review of the role of Hund’s coupling in the iron-pnictides, see ref. 2). A clear experimental identification of both these regimes is currently lacking. Here, we provide direct experimental confirmation of the Fermi liquid state in the electron-doped case.

Optical spectroscopy is a powerful tool to probe self-energy effects\(^9\) as a function of frequency and temperature simultaneously. The complex-valued free charge optical conductivity\(^10\) can be written as
\[ \sigma(\omega, T) = \sigma_1(\omega, T) + i \sigma_2(\omega, T) = \frac{i \omega_p^2}{4 \pi \omega + M(\omega, T)}, \]

where \( \omega_p^2 = \frac{m^2}{e^2} \) is the plasma frequency and \( M(\omega, T) = M_1(\omega, T) + i M_2(\omega, T) \) is the complex memory function. For a simple Drude metal \( M(\omega, T) = i \Gamma \omega \) is frequency independent, while interactions beyond simple impurity scattering introduce a frequency and temperature dependence. In the latter case Eq. (1) is referred to as the "Extended Drude model". The single particle self-energy \( \Sigma(\omega, T) \) thus manifests itself in the free charge carrier response, appearing as a deviation from a classical Drude response. For a local FL with a momentum independent interaction between electrons, \( \Sigma(\omega, T) \) and consequently \( M(\omega, T) \) (see Methods) follow a universal quadratic dependence on both energy and temperature

\[ M_2(\omega, T) = \frac{2}{3 \pi k_B T_0} [ (h \omega)^2 + (p \pi k_B T)^2], \]

where \( k_B T_0 \) is an overall energy scale characterising the correlation strength and \( p \) is a non-universal constant. For a local FL one expects \( p = 2 \); however, deviations arise in the presence of additional elastic resonant scattering channels\(^{12}\). To date the only known example with \( p = 2 \) is Sr$_2$RuO$_4$,\(^{14}\), while \( p \neq 2 \) has been reported for several correlated materials\(^{11,13-18}\). When applied to the iron-pnictide superconductors, the accurate determination of \( M(\omega, T) \) is hampered by the presence of low-lying interband transitions. In the following we first show that \( M(\omega, T) \) extracted for carefully annealed BaFe$_{1.8}$Co$_{0.2}$As$_2$ single crystals indeed displays the characteristic \( \omega, T \) scaling predicted by Eq. (2). We then introduce an analysis of the complex optical conductivity that represents a direct confirmation of the Fermi liquid normal state of these electron doped iron-pnictides. This is made possible by the 2 K temperature resolution in our experiments, which allows us to compare the detailed frequency and temperature dependence with similar resolution.

**Results**

The in-plane optical conductivity of as-grown and annealed BaFe$_{1.8}$Co$_{0.2}$As$_2$ (see Methods) are shown in Fig. 1a,b, respectively. After annealing we observe a decrease in the depth of the minimum around 70 meV separating the free-charge and interband optical conductivity. This arises from a reduction of a broad incoherent response associated with high energy interband processes, rather than from changes in the free-charge response (see Supplementary Online Material, SOM). A spectral weight analysis (see SOM) indeed displays the characteristic \( \omega, T \) state of these electron doped iron-pnictides. This is made possible by the 2 K temperature resolution in our experiments, which allows us to compare the detailed frequency and temperature dependence with similar resolution.

**Experimental signatures of the Fermi liquid state.** Subtle changes in the free charge carrier response are more easily analysed in terms of equations (1) and (2), but the extended Drude model analysis assumes that interband transitions do not contribute to the optical conductivity in the energy range of interest. The multi-band nature of the pnictides complicates the extraction of \( M(\omega, T) \) since inter-band processes have a significant contribution to the optical conductivity\(^{19-22}\). In the SOM we describe the procedure used to extract the memory functions and its range of validity. We find that even though the determination of the memory functions comes with uncertainty at higher energies, at low energies \((h \omega \leq 50 \text{ meV})\) interband transitions only weakly affect the frequency dependence. In the following we subtract the full frequency dependence of the interband response as outlined in the SOM; however, we note that our conclusions remain the same when alternative methods for accounting for the interband transitions are applied.

The frequency and temperature dependence of the imaginary part of the memory function \( M_2(\omega, T) \), shown in Fig. 1c,d for the as-grown and annealed crystal, respectively, indicates the presence of residual interactions beyond a classical Drude response. We fit both datasets with a power-law form \( M_2(\omega, T) = \frac{1}{\tau(0, T) + B(T) \omega^{\eta(T)}} \), where \( \frac{1}{\tau(0, T)} \) is the zero-frequency scattering rate and \( \eta(T) \approx 2 \) is expected for a FL. These parameters are determined independently at each temperature. The temperature dependence of \( \frac{1}{\tau(0, T)} \) and \( \eta(T) \) are displayed in Fig. 1e,f for both the as-grown and annealed crystal. We find that the annealed crystal displays characteristic FL behaviour with \( \frac{1}{\tau(T)} \sim T^2 \) (Fig. 1e) and \( \eta(T) \sim T^2 \) (Fig. 1f) over a large range of energy \((10 \text{ meV} \leq h \omega \leq 50 \text{ meV})\) and temperature \((8 \text{ K} \leq T \leq 100 \text{ K})\). We further find that the prefactor \( B(T) \) is temperature independent in the same temperature range as is expected from Eq. (2) (see SOM). The as-grown crystal on the other hand does not display FL behaviour. Instead, the zero frequency scattering rate follows a more linear temperature dependence, while the frequency exponent \( \eta(T) < 2 \). Given the approximate \( T^2 \) and \( \omega^2 \) dependence of the memory function apparent in Fig. 1e,f, we test whether the scaling form of Eq. (2) applies to the annealed crystal. Figure 1g demonstrates that \( M_2(\omega, T) \) indeed follows a universal FL scaling as function of the scaling variable \( \xi^2 = (h \omega)^2 + (p \pi k_B T)^2 \), with \( p \approx 1.47 \) (see SOM).

We highlight three deviations from universal FL behaviour that can be discerned in Fig. 1g. First, universal FL behaviour disappears above 100 K. Second, for \( \xi^2 \gtrsim 2500 \text{ meV}^2 \), \( M_2(\xi) \) changes slope, as...
indicated by the dashed pink line, signalling a crossover to a nearly energy independent $M_2(\omega, T)$ for $\omega \geq 50$ meV (Fig. 1d). Third, $p = 1.47$ rather than 2, indicating that an additional elastic contribution is present beyond residual electron-electron scattering. We note that the precise value of $p$ determined by collapsing the data on a universal curve comes with some uncertainty as it depends on the assumed strength and frequency dependence of the interband contribution (see SOM).

**Fermi liquid signatures in the optical conductivity.** Our analysis provides compelling evidence that the normal state of BaFe$_{1.8}$Co$_{0.2}$As$_2$ below 100 K is properly classified as a FL. We emphasise that the specific method of accounting for interband processes does not alter the conclusion that the low frequency and temperature dependence of $M_2(\omega, T)$ follows $\omega^2$ and $T^2$ scaling (see SOM). In contrast, the same analysis applied to the as-grown crystal does not show such clear signatures of FL behaviour, despite its similar plasma frequency and high-energy optical properties. Nevertheless, the determination of the parameters characterising the Fermi liquid state using the extended Drude analysis remains sensitive to the choice for the interband contribution. To fortify our conclusions, and to determine the characteristic properties of the Fermi liquid state more accurately, we now turn our attention to an analysis of the complex optical conductivity, which provides a more direct comparison between theory and experiment and does not require a model specific choice for the interband processes.

Berthod et al. showed\textsuperscript{11} that in a local FL a dome is defined by the locus of points where $\sigma_1(\omega, T) = \sigma_2(\omega, T)$, which bounds a 'thermal' regime in which FL behaviour emerges. Zero crossings signalling the presence of a dome have been clearly observed\textsuperscript{14} in Sr$_2$RuO$_4$ at low temperatures. Despite the clean Fermi liquid
The deviation from scaling in Fig. 1e–g around 100 K signals a crossover temperature where $\hbar \omega \approx 55 \text{ meV}$ and $D \approx 1 \text{ eV}$ are motivated below. In addition to the free charge response, we also include the frequency dependent interband response from Supplementary Table S1. The only remaining free parameter, $T_c \approx 1700 \text{ K}$, is determined by two criteria: (i) the maximum of the dome of zero crossings (at $\hbar \omega \approx 55 \text{ meV}$) and (ii) the low temperature zero-crossing at $\hbar \omega \approx 100 \text{ meV}$. To facilitate the estimation of $T_c$, we derive an approximate analytical expression, $\Delta \sigma (\omega, T)$ (see Methods and SOM), for these zero-crossings taking an energy independent interband response (e.g. $\varepsilon_c$) into account. The consistency between $T_c (\omega)$, the data, and the calculation (which includes the full frequency dependence of the interband response) shows that the details of the interband response are unimportant for obtaining this level of agreement.

**Characteristics of Fermi liquid properties of Co-doped $\text{BaFe}_2\text{As}_2$.** The deviation from scaling in Fig. 1e–g around 100 K signals a crossover temperature where $\hbar \omega \leq \pi \hbar q T$, above which an incoherent regime emerges. This suggests a natural cutoff $\hbar \omega_c \approx 1.47 \pi \hbar q T$ with $T \approx 100 \text{ K}$, resulting in $\omega_c \approx 41 \text{ meV}$. The deviation from scaling in Fig. 1e–g around 100 K signals a crossover temperature where $\hbar \omega \leq \pi \hbar q T$, above which an incoherent regime emerges. This suggests a natural cutoff $\hbar \omega_c \approx 1.47 \pi \hbar q T$ with $T \approx 100 \text{ K}$, resulting in $\omega_c \approx 41 \text{ meV}. \mu$m
The cutoff $D \approx 1$ eV is less critical but is motivated by the value of $T_0$. Dynamical Mean Field Theory (DMFT) calculations for a single band Hubbard model\(^\text{11}\) indicate that $k_B T_0 \approx 0.57 \delta W$ where $W$ is half the bandwidth and $\delta$ is the carrier density. This yields $W \approx 1.3$ eV in our case, which is reasonable compared to combined density functional theory and DMFT (e.g LDA + DMFT) estimates\(^\text{8}\).

Apart from the saturation in $\Sigma_2 (\omega, T)$, $\omega_c$ also introduces a frequency dependence in $\Sigma_1 (\omega, T)$ (see Fig. 3a), which should be manifest as a frequency dependent mass enhancement $m'/m (\omega \rightarrow 0) \approx 1.2$. Note that the data and fits are offset from their actual value with increments of 0.1 at successive temperatures above the 8 K curves. (c) $\omega (T)$ dependence of $\Delta \sigma (\omega, T)$ for the as-grown crystal. The dome of zero-crossings is smaller compared to the annealed crystal. This difference is highlighted by the dashed (as-grown) and dashed-dotted (annealed) semi-circles calculated from $T_\infty (\omega)$. The dashed semi circle is calculated using the same parameters as in Fig. 2 except for $\Gamma_0 \approx 8$ meV and $p \approx 1.34$.

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**Discussion**

To conclude we discuss the deviation of $p$ from the FL value $p = 2$. The most likely origin appears to be scattering of quasi-particles on weak, localised magnetic moments\(^\text{12}\). Such localised moments could be associated with the presence of Co impurities in the Fe lattice, although no local moment has been detected for Co impurities in BaFe$_2$As$_2$\(^\text{23}\). Regardless the origin, this resonant elastic term has a strong influence on the normal state properties. Figure 3c displays $\Delta \sigma (\omega, T)$ for the as-grown crystal, displaying a suppressed dome of zero-crossings compared to the annealed crystal. The dashed semi-circle is calculated using exactly the same parameters as for the annealed case, except for a slightly higher $\Gamma_0 \approx 8$.
and $p = 1.34$. This smaller value of $p$ corresponds to a two-fold stronger elastic term in the single particle self-energy $\Sigma(\omega, T)$, indicating that annealing strongly reduces the influence of this scattering channel. Given the concomitant change in superconducting critical temperature, we suggest that this scattering channel could be pair-breaking, possibly providing an interesting direction for future work.

### Methods

**Method subsection.** A large $4 \times 5 \times 0.1 \text{ mm}^3$ single crystal of BaFe$_{1.8}$Co$_{0.2}$As$_2$, grown from self-flux, was cut into two pieces and one piece was subsequently annealed for 75 hours at 800°C. The dc resistivity and dc susceptibility show an increase of the critical temperature $\Delta T_c/T_c \approx 0.3$ upon annealing, while the overall value of the resistivity decreases. Further details of the experiments are presented in the SOM. The theoretical formalism is based on the Allen-Kubo formula for the optical conductivity$^{24}$,

$$\sigma(\omega, T) = \frac{\omega^2}{i 4 \pi \omega} \int_{-\infty}^{+\infty} \frac{n_p(\omega + x, T) - n_p(x, T)}{\omega - \Sigma(x + \omega, T) + \Sigma^*(x, T) + i \Gamma_{\text{imp}}} dx$$

(3)

which we evaluated numerically. The imaginary part of the single particle self-energy appearing in the denominator is given by$^{12}$,

$$\Sigma_2(\omega, T) = -\frac{i}{\pi k_B T_0} [(1 + a)(\hbar \omega)^2 + (\pi k_B T)^2]$$

(4)

for a local Fermi liquid with an additional elastic resonant scattering contribution (note $a = (p^2 - 4)/(1 - p)^{12}$). Such an energy and temperature dependent $\Sigma_2(\omega, T)$ results at low temperature in an imaginary memory function$^{12,11}$,

$$M_2(\omega, T) = \frac{2}{3\pi k_B T_0} [(1 + a)(\hbar \omega)^2 (2\pi k_B T)^2]$$

(5)

Together with equation (1) for the optical conductivity and an interband contribution characterised by an energy independent value $\varepsilon_{\infty, fB}$, equation (5) leads to the following expression for the dashed semi-circle displayed in Fig. 2:

$$T_{\text{imp}}(\omega) = \frac{1}{4} \sqrt{\frac{3}{\varepsilon_{\infty, fB} \pi \omega}} \sqrt{T_0^2 - \frac{3}{4} \varepsilon_{\infty, fB} \omega^4 - 4\varepsilon_{\infty, fB} \omega^2 - 4 \varepsilon_{\infty, fB}^2 \omega^4} - \frac{3 T \omega^2}{\varepsilon_{\infty, fB} \pi \omega} - \frac{6 T_0^2}{\pi} - \frac{\varepsilon_{\infty, fB} \omega^2}{\pi}.$$

(6)

For a derivation and further details see the SOM. The parameters used to calculate the dashed semi-circles in Fig. 2a,b are the same as for the full calculation except for $\varepsilon_{\infty, fB} \approx 100$. For the full calculation of equation (3), we introduce two cutoffs, $\omega_c$ and $D$ in equation (4) such that the imaginary part of the self-energy is given by $\Sigma_2(\omega) \propto \omega^2$ for $|\omega| < \omega_c$ and $\Sigma_2(\omega) \propto \omega_c^2$ for $|\omega| > \omega_c$, and $\Sigma_2(\omega) = 0$ otherwise. In the SOM we derive analytical expressions for the real part of the self-energy obtained from Kramers-Kronig transformation.

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Author Contributions
A.T. carried out and analysed experiments. As-grown and annealed single crystals were provided by Y.K.H. A.T. and A. de V. carried out and analysed transport and magnetisation experiments on the crystals used in this study. S.J. performed theoretical calculations, provided theoretical support and wrote the manuscript. E.v.H designed the experiment, analysed data, performed theoretical calculations and modelling, and wrote the manuscript.

Additional Information
Supplementary information accompanies this paper at http://www.nature.com/srep

Competing financial interests: The authors declare no competing financial interests.


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