Supplementary Information to: Collapse of Metallicity and High-T_c Superconductivity in the High-Pressure phase of $\mathrm{FeSe}_{0.89}\mathrm{S}_{0.11}$

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In this document, we provide additional analyses which support the central conclusions drawn in the main manuscript.

SUPPLEMENTARY NOTE

In the main manuscript, we extract a resistivity exponent k within a narrow pressure region located between the structural transition at p_s and the loss of metallic behaviour around $p \approx 5\,\mathrm{GPa}$. Here, we provide a refined analysis based on effective medium theory to demonstrate the continuous validity of the resistivity exponent in this regime. Exemplary, we can separate the metallic and non-metallic components of the resistivity at a pressure of $p = 5.2\,\mathrm{GPa}$, measured on sample B, and then carry out the same resistivity exponent analysis as in the main manuscript, but solely on the metallic component.

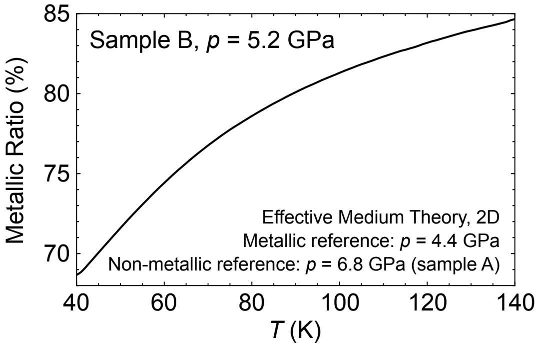
Within effective medium theory in two dimensions, the effective (measured) conductivity σ_e is given through the implicit equation:

$$\frac{\sigma_1 - \sigma_e}{\sigma_1 + \sigma_e} \cdot r = \frac{\sigma_2 - \sigma_e}{\sigma_2 + \sigma_e} \cdot (r - 1). \qquad (1)$$

In our case, σ_1 shall denote the metallic conductivity within the tetragonal phase, σ_2 the non-metallic conductivity of the high-pressure phase, and r is the fraction of the metallic phase. In this case, σ_e will describe a mixed conductivity assuming a random spatial distribution of metallic and non-metallic domains.

To estimate the non-metallic component, we observe that the resistivity at $p = 6.8\,\mathrm{GPa}$ for sample A (Fig. 1 in the main manuscript) appears very similar to both the $p = 13.5\,\mathrm{GPa}$ and $p = 15\,\mathrm{GPa}$ curves in FeSe as previously reported by Sun *et al.* [1]. This indicates that at this pressure, the phase transition is complete and the resistivity has saturated to its non-metallic value. We therefore take $\rho(T) = 1/\sigma_2$ at 6.8 GPa as a reference value for the resistivity of the non-metallic phase.

Next, we estimate the ratio between metallic and non-metallic phases for sample B at $p = 5.2\,\mathrm{GPa}$, by using its $p = 4.4\,\mathrm{GPa}$ data as the metallic reference σ_1. In this way, we obtain a metallic ratio of $r = 70 - 85\,\%$ between the onset of superconductivity and 140 K, as shown in Supplementary Figure 1 below. This estimate demonstrates the large majority of metallic conductivity at this pressure, as expected. As an interesting and reassuring side note, we add that carrying out the same analysis for different samples and pressures for which a small tail starts to appear at the bottom of the superconducting transition, we obtain consistently $r \approx 60\,\%$, i.e. very close to the percolation threshold of many 2D lattices. This appears fully consistent with the assumed spatial phase separation and the formation of superconducting islands.



Supplementary Figure 1. Extracted metallic ratio within effective medium theory, under a pressure located within the phase-separated region close to p_s.

As a next step, we assume an average and temperature independent $r = 79\,\%$ metallic ratio for the temperature range $T = 40 - 140\,\mathrm{K}$ in order to extract the expected metallic resistivity from the original $p = 5.2\,\mathrm{GPa}$ dataset (if we were to use the full temperature dependence of r, we would simply recover the metallic reference data $1/\sigma_1$). The thus obtained temperature dependence for $1/\sigma_1$ is shown below in Supplementary Figure 2 on the left, compared against the originally measured dataset. The corresponding fits to power law behaviour as well as the obtained resistivity exponents k using

$$\rho(T) = \rho_0 + aT^k \qquad (2)$$

between 40-100K (consistent with Fig. 4(c) in the main manuscript) are given as well. This shows that an attempt to extract the purely metallic component of the

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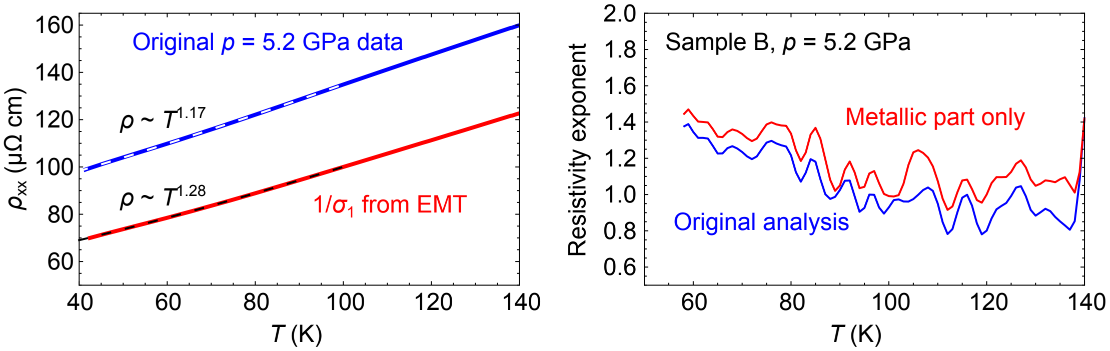

Supplementary Figure 2. Left: Extracted metallic ratio within effective medium theory, within the phase separated phase close to p_s. Right: Extracted resistivity exponents before and after extracting the metallic component.

resistivity only yields a marginally larger resistivity exponent on the right-hand side of the quantum critical fan. We also note that the exponents differ substantially from the metallic reference data, for which we have $k = 0.74$.

Finally, we extract the temperature dependence of the resistivity exponent in the same way as in the manuscript, i.e. by using the double-logarithmic derivative. The result is shown in Supplementary Figure 2 on the right, compared against the data shown in the main manuscript. Clearly, we recover the same temperature dependence, i.e. the exponent drops towards higher temperatures, which is fully consistent with the fact that this dataset is located on the right hand side of the quantum critical fan. The only difference is the marginal and nearly constant offset, which however does not influence any conclusions drawn.

In summary, accounting for a non-metallic conductivity within a spatially inhomogeneous sample still recovers a quantum critical fan, even though the fan would appear somewhat sharper. This suggests that the remaining metallic regions preserve the manifestation of the quantum critical fan over the narrow pressure range $p_s < p < 5.2\,\mathrm{GPa}$.

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[1] J. P. Sun, K. Matsuura, G. Z. Ye, Y. Mizukami, M. Shimozawa, K. Matsubayashi, M. Yamashita, T. Watashige, S. Kasahara, Y. Matsuda, J.-Q. Yan, B. C. Sales, Y. Uwatoko, J.-G. Cheng, and T. Shibauchi, Dome-shaped magnetic order competing with high-temperature superconductivity at high pressures in FeSe, Nature Communications **7**, 12146 (2016), arXiv:1512.06951.