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SUPPLEMENTARY INFORMATION

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Sudden reversal in the pressure dependence of T_c in the iron-based superconductor KFe_2As_2

METHODS

Single crystals of KFe_2As_2 were obtained by KAs flux growth at USTC [13]. Three crystals of KFe_2As_2 were measured: two pure and one doped with 3.4% of Co, labeled A, B, and C, respectively. The two pure samples (A and B) have T_c ~ 3.8 K and a residual resistivity ratio (RRR = ρ(300 K) / $\rho(T \rightarrow 0)$) ~ 1200, at ambient pressure. Sample C has T_c = 1.7 K and RRR = 66.

Resistivity (ρ_{xx}) and Hall resistance (ρ_{xy}) were measured under pressure in a six-probe AC configuration. Six contacts were soldered on each sample; one longitudinal pair to pass the electric current, another longitudinal pair to measure ρ_{xx}, and one transverse pair to measure ρ_{xy}. The samples were mounted so that the magnetic field H was parallel to the tetragonal c axis. Hall measurements were done by reversing the field and anti-symmetrizing the ρ_{xy} data. At each pressure, the Hall signal was measured by sweeping temperature from 20 K to 2 K at H = + 13 T and H = - 13 T.

Samples were pressurized hydrostatically in a piston-cylinder clamp cell made of Be-Cu alloy 25 with the inner jacket made of alloy MP35N. The pressure inside the cell was determined via the T_c of a lead wire; we estimate its uncertainty to be ± 0.2 kbar. Measurements on the two pure samples (A and B) were repeated in two different pressure cells, with 4-mm and 6-mm inner jacket diameters, respectively. They were also performed in two different pressure media: Daphne oil 7373 and a 1/1 mixture of pentane and 3-methyl-1-butanol. As shown in Fig. S1, the values of T_c vs P obtained are all in excellent agreement, demonstrating that our results are highly reproducible and independent of the choice of sample, pressure medium or pressure cell. (They are also independent of the choice of T_c criterion – see below.)

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The resistivity under pressure was measured in the temperature range from 300 K to 0.3 K using a Cambridge Magnetic Refrigerator. A calibrated Cernox attached directly on the cell was used for thermometry. For measurements below 2 K, a calibrated ruthenium oxide thermometer mounted on the low-temperature stage was used. Slow cooling rates of 100 mK / min (between 300 K and 2 K) and 20 mK / min (between 2 K and 0.3 K) were used to maintain a good thermal equilibrium between the cell and the stage. Resistivity curves below 2 K were obtained in both cooling and heating cycles with no detectable thermal hysteresis.

DETERMINATION OF T_c

In this paper, the superconducting transition temperature T_c is defined as the temperature below which the resistance is zero. We emphasize that in both samples A and B the width of the superconducting transition was narrow and, importantly, it remained constant as a function of pressure, as can be seen in Figs. 2b and 2c. This implies that the dependence of T_c on pressure reported in Fig. 2a (from sample A) and Fig. 4 (from sample B) does not depend on our choice of criterion (*e.g.* whether it is R = 0 or 10%, 50%, 90% of the resistive drop), modulo a rigid offset. The best way to demonstrate this is to plot (normalized) ρ vs T / T_c for several isobars, as done in Fig. S2. This scaling analysis avoids having to choose a particular criterion for defining T_c and affords greater accuracy, with an uncertainty of less than ± 2% on the relative value of T_c (as it changes with pressure). In Fig. S2a, we see that T_c varies strictly linearly with P on both sides of P_c, within this very small error bar. This confirms that T_c has a kink at P_c, the signature of a transition.

Note that for the sample with 3.4% Co impurities we use a 50% criterion for T_c (see Fig. S3), because these preliminary data were not taken down to the lowest temperatures. This difference in criterion has no impact on the qualitative conclusions we draw from these data.

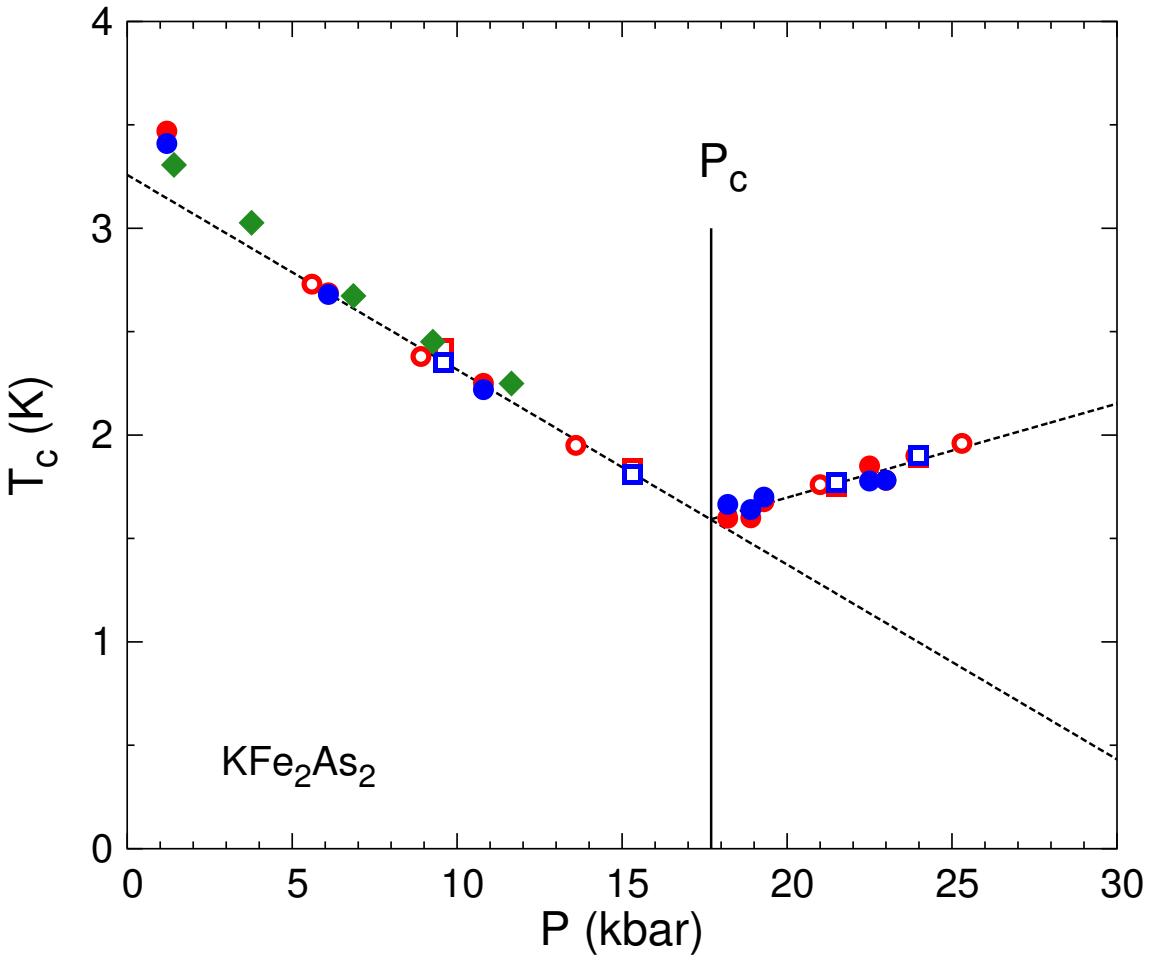

Figure S1 | Comparing different measurements of T_c vs pressure in KFe_2As_2.

Superconducting critical temperature T_c of KFe_2As_2, defined as the point of zero resistance, measured on our two pure samples (A, blue symbols; B, red symbols). Data obtained in two pressure cells (6-mm cell, circles; 4-mm cell, squares) and in two pressure media (Daphne oil, full symbols; pentane mixture, open symbols) are compared. Excellent reproducibility is observed, across different samples, cells and media. Our data are also in good agreement with those of a prior study of KFe_2As_2 under pressure (green diamonds, from ref. 28).

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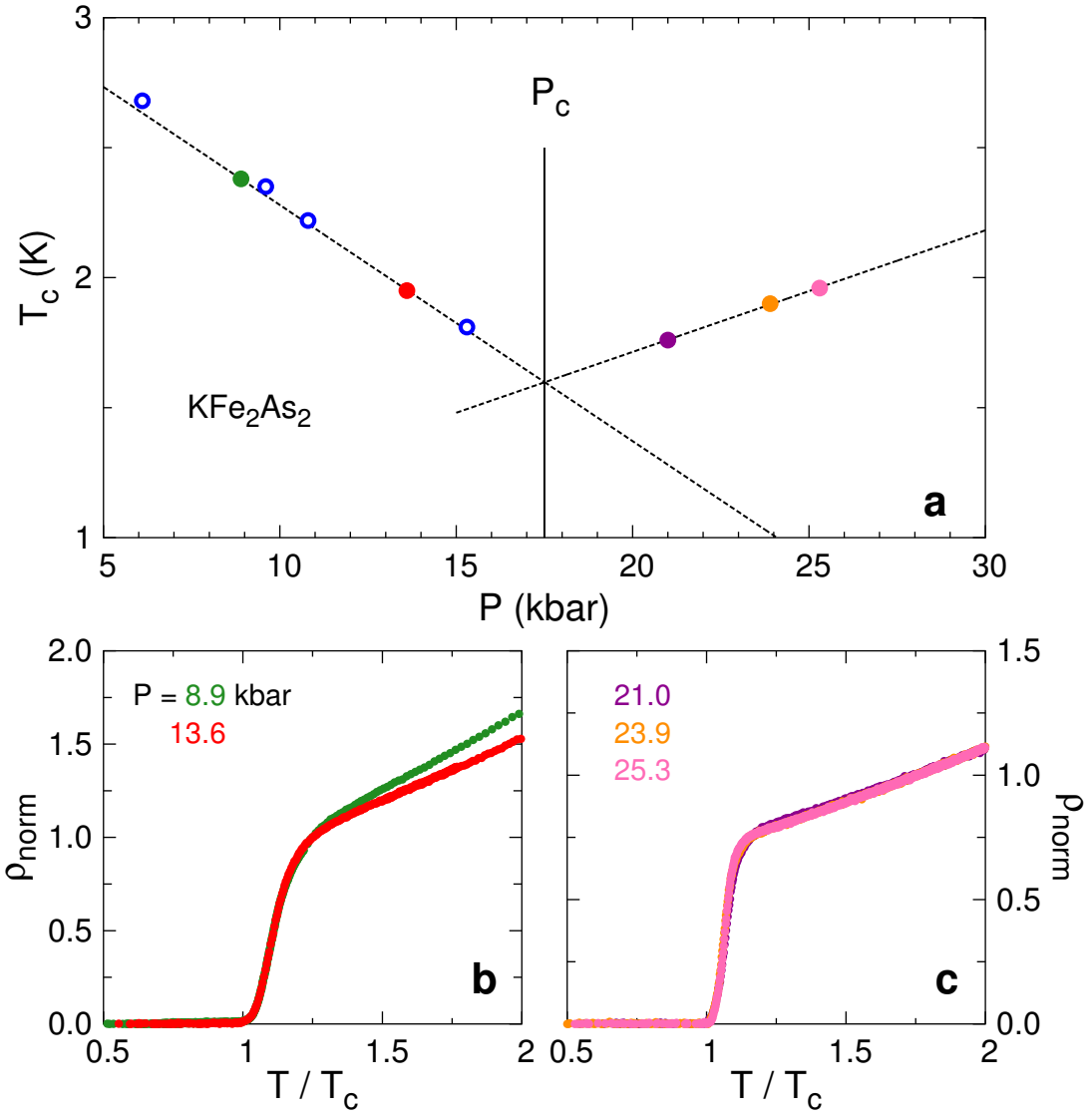

Figure S2 | Determination of T_c in KFe_2As_2 .

a) Pressure dependence of T_c in sample A (full circles; from data in Fig. 2) and sample B (open circles; Fig. 4) obtained from isobars as in panel b (for $P < P_c$) and panel c (for $P > P_c$). The colour of full circles matches that of the corresponding isobars in panels b and c. The two dashed lines are a linear fit through the full circles on either side of P_c. **b)**, **c)** Isobars of $\rho(T)$ in sample A at different pressures as indicated, plotted as $\rho(T)$ vs T / T_c, where ρ is normalized and T_c is adjusted so that all curves collapse onto a single curve. The resulting T_c values, accurate to better than ± 2%, are plotted in panel a and in Fig. 2a (as colour-coded full circles).

IMPURITY SCATTERING IN KFe_2As_2

One way to differentiate a *d*-wave superconductor from a standard *s*-wave superconductor (s_{++}) is to study the effect of non-magnetic impurities on T_c. In a *d*-wave superconductor, where the sign change in the order parameter around the Fermi surface is imposed by symmetry, impurity scattering rapidly suppresses T_c because it mixes the phase of initial and final states. For a single-band system, the critical scattering rate Γ_c for suppressing T_c to zero is on the order of the clean limit T_{c0} [31] :

$$\hbar \Gamma_c = 0.88 \, k_B \, T_{c0}$$

This is indeed what is found in KFe_2As_2 [11, 12, 13], and this is one of several arguments in favour of a *d*-wave state in this iron arsenide at ambient pressure [11, 12]. The situation is very different in the iron-based superconductors $CaFe_2As_2$, $BaFe_2As_2$, and $SrFe_2As_2$ at optimal doping, where $\hbar \Gamma_c \sim 45 \, k_B \, T_{c0}$ [29]. This robustness is consistent with an s_{++} state. It can also be consistent with a type-I $s_{\pm}$ state (with sign change between hole and electron pockets; Fig. 1b) if inter-band impurity scattering is weak [3].

The type-II $s_{\pm}$ state proposed by Maiti *et al*. [27] (with sign change between h_1 and h_2 hole pockets; Fig. 1d) is likely to be more sensitive to disorder than the type-I $s_{\pm}$ state. This is because for those two rather similar hole pockets, inter-band and intra-band impurity scattering are very likely to be comparable.

To determine the sensitivity of T_c to impurity scattering in KFe_2As_2 as a function of pressure, the resistivity and Hall effect were measured in a sample of KFe_2As_2 with 3.4% Co impurities (sample C). At ambient pressure, T_c = 1.7 K [13]. As seen in Fig. S3, pressure rapidly reduces T_c to values below 0.3 K, and superconductivity does not reappear at high pressure. This therefore rules out an s_{++} state at $P > P_c$. As in the pure sample, the Hall coefficient $R_H(0)$ of the Co-doped sample does not show any change across P_c , confirming here as well that there is no Lifshitz transition.

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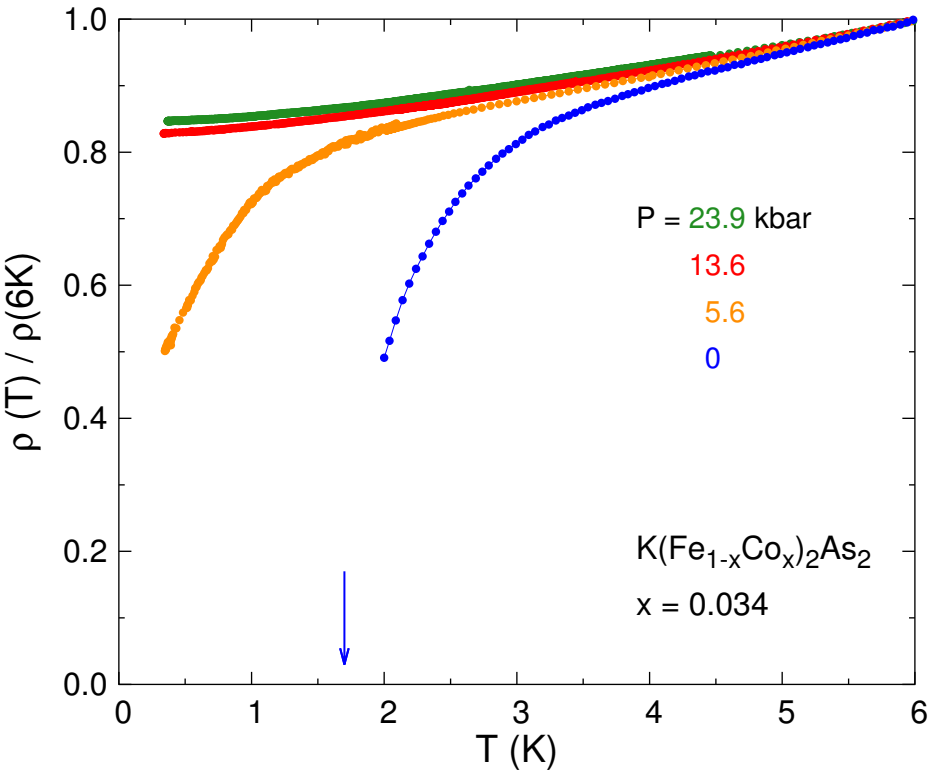

Figure S3 | Resistivity of a KFe_2As_2 sample with Co impurities.

Electrical resistivity of a sample of KFe_2As_2 with 3.4% Co impurities, normalized to its value at T = 6 K, for different pressures as indicated. At ambient temperature, T_c = 1.7 K [13] (arrow). At higher pressure, T_c drops below 0.3 K and remains undetected up to the highest pressures. This shows that the superconducting states below and above T_c are both easily destroyed by a low level of impurity scattering. The T_c values displayed in Fig. 4 (as full green squares) are obtained using the criterion that ρ has fallen to 50% of its value at T = 6 K.

HALL EFFECT

The temperature dependence of the Hall coefficient R_H is displayed in Fig. S4 up to 20 K. $R_H(T)$ is seen to go from one value at $T = 0$ to another value at high temperature. This reveals a difference in the ***k*** dependence (or anisotropy) of elastic and inelastic scattering. The growth of inelastic scattering with temperature reduces the mobility of certain regions of the Fermi surface more than others and hence causes a shift in the relative balance of contributions to R_H from the three Fermi surfaces. This is consistent with ***k***-dependent antiferromagnetic fluctuations being the cause of inelastic scattering.

PRIOR WORK

Our T_c values obtained from resistivity at different pressures are in good agreement with those of ref. 28 (Fig. S1), where T_c in KFe_2As_2 was defined as the onset of the magnetization drop. Note that because the maximal pressure in that study was 12 kbar, the transition at P_c was not observed.

REFERENCE

[31] Alloul, H. *et al*. Defects in correlated metals and superconductors. *Rev. Mod. Phys.* **81**, 45 (2009).

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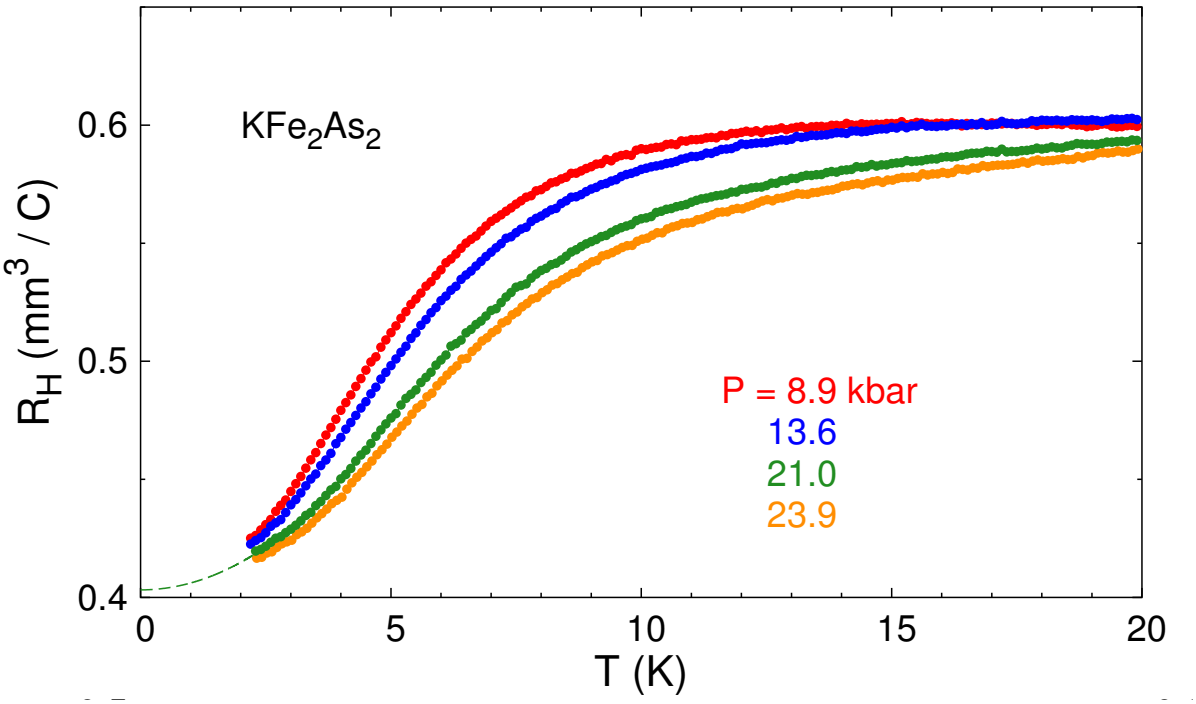

Figure S4 | Hall coefficient of KFe_2As_2 vs temperature.

In-plane Hall coefficient R_H of KFe_2As_2 as a function of temperature, measured in a magnetic field of 13 T along the *c* axis of the tetragonal lattice, for different values of the applied pressure *P*, as indicated. The green dashed line is a T^2 extrapolation of the data at P = 21.0 kbar.