Sudden reversal in the pressure dependence of $T_c$ in the iron-based superconductor CsFe$_2$As$_2$: A possible link between inelastic scattering and pairing symmetry

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We report a sudden reversal in the pressure dependence of $T_c$ in the iron-based superconductor CsFe$_2$As$_2$, similar to that discovered recently in KFe$_2$As$_2$ [Tafti et al., Nat. Phys. 9, 349 (2013)]. As in KFe$_2$As$_2$, we observe no change in the Hall coefficient at $T \to 0$, again ruling out a Lifshitz transition across the critical pressure $P_c$. We interpret the $T_c$ reversal in the two materials as a phase transition from one pairing state to another, tuned by pressure, and we investigate what parameters control this transition. Comparing samples of different residual resistivity $\rho_0$, we find that a 6-fold increase in impurity scattering does not shift $P_c$. From a study of X-ray diffraction on KFe$_2$As$_2$ under pressure, we report the pressure dependence of lattice constants and As-Fe-As bond angle. The pressure dependence of the various lattice parameters suggests that $P_c$ should be significantly higher in CsFe$_2$As$_2$ than in KFe$_2$As$_2$, but we find on the contrary that $P_c$ is lower in CsFe$_2$As$_2$, indicating that other factors control $T_c$. Resistivity measurements under pressure reveal a change of regime across $P_c$, suggesting a possible link between inelastic scattering and pairing symmetry.

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I. INTRODUCTION

To understand what controls $T_c$ in high temperature superconductors remains a major challenge. Several studies suggest that in contrast to cuprates where chemical substitution controls electron concentration, the dominant effect of chemical substitution in iron-based superconductors is to tune the structural parameters – such as the As-Fe-As bond angle – which in turn control $T_c$. This idea is supported by the parallel tuning of $T_c$ and the structural parameters of the 122 parent compounds BaFe$_2$As$_2$ and SrFe$_2$As$_2$. In the case of Ba$_{1-x}$K$_x$Fe$_2$As$_2$, at optimal doping ($x = 0.4$, $T_c = 38$ K) the As-Fe-As bond angle is $\alpha = 109.5^\circ$, the ideal angle of a non-distorted FeAs$_4$ tetrahedral coordination. Underdoping, overdoping, or pressure would tune the bond angle away from this ideal value and reduce $T_c$ by changing the electronic bandwidth and the nesting conditions.

CsFe$_2$As$_2$ is an iron-based superconductor with $T_c = 1.8$ K and $H_{c2} = 1.4$ T. Based on the available X-ray data, the As-Fe-As bond angle in CsFe$_2$As$_2$ is 109.58°, close to the ideal bond angle that yields $T_c = 38$ K in optimally-doped Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. If the bond angle were the key tuning factor for $T_c$, CsFe$_2$As$_2$ should have a much higher transition temperature than 1.8 K.

In this article, we show evidence that $T_c$ in (K,Cs)Fe$_2$As$_2$ may be controlled by details of the inelastic scattering processes that are not directly related to structural parameters, but are encoded in the electrical resistivity $\rho(T)$. The importance of inter- and intra-band inelastic scattering processes in determining $T_c$ and the pairing symmetry of iron pnictides has been emphasized in several theoretical works. Recently, it was shown that a change of pairing symmetry can be induced by tuning the relative strength of different competing inelastic scattering processes, i.e. different magnetic fluctuation wavevectors.

In a previous report, we examined the pressure dependence of $T_c$ in KFe$_2$As$_2$, the fully hole-doped member of the Ba$_{1-x}$K$_x$Fe$_2$As$_2$ series. No sudden change was observed in the Hall coefficient or resistivity across the critical pressure $P_c = 17.5$ kbar, indicating that the transition is not triggered by a change in the Fermi surface. Recent dHvA experiments under pressure confirm that the Fermi surface is the same on both sides of $P_c$, ruling out a Lifshitz transition and strengthening the case for a change of pairing state. We interpret the sharp $T_c$ reversal as a phase transition from $d$-wave to $s$-wave symmetry. Bulk measurements such as thermal conductivity and penetration depth favor $d$-wave symmetry at zero pressure. Because the high-pressure phase is very sensitive to disorder, a likely $s$-wave state is one that changes sign around the Fermi surface, as in the $s_\pm$ state that changes sign between the $\Gamma$-centered hole pockets, as proposed by Maiti et al. It appears that in KFe$_2$As$_2$ $s$-wave and $d$-wave states are nearly degenerate, and a small pressure is
enough the push the system from one state to the other.

In this article, we report the discovery of a similar $T_c$ reversal in CsFe$_2$As$_2$. The two systems have the same tetragonal structure, but their lattice parameters are notably different. Our high-pressure X-ray data reveal that at least 30 kbar of pressure is required for the lattice parameters of CsFe$_2$As$_2$ to match those of KFe$_2$As$_2$. Yet, surprisingly, we find that $P_c$ is smaller in CsFe$_2$As$_2$ than in KFe$_2$As$_2$. This observation clearly shows that structural parameters alone are not the controlling factors for $P_c$ in (K,Cs)Fe$_2$As$_2$. Instead, we posit that competing inelastic scattering processes are responsible for tipping the balance between pairing symmetries.

II. EXPERIMENTS

Single crystals of CsFe$_2$As$_2$ were grown using a self-flux method. Resistivity and Hall measurements were performed in an adiabatic demagnetization refrigerator, on samples placed inside a clamp cell, using a six-contact configuration. Hall voltage is measured at plus and minus 10 T from $T = 20$ to 0.2 K and antisymmetrized to calculate the Hall coefficient $R_H$. Pressures up to 20 kbar were applied and measured with a precision of ±0.1 kbar by monitoring the superconducting transition temperature of a lead gauge placed beside the samples inside the clamp cell. A pentane mixture was used as the pressure medium. Two samples of CsFe$_2$As$_2$, labelled “sample 1” and “sample 2”, were measured and excellent reproducibility was observed.

High pressure X-ray experiments were performed on polycrystalline powder specimens of KFe$_2$As$_2$ up to 60 kbar with the HXMA beam line at the Canadian Light Source, using a diamond anvil cell with silicon oil as the pressure medium. Pressure was tuned blue with a precision of ±2 kbar using the R$_1$ fluorescent line of a ruby chip placed inside the sample space. XRD data were collected using angle-dispersive techniques, employing high energy X-rays ($E_i = 24.35$ keV) and a Mar345 image plate detector. Structural parameters were extracted from full profile Rietveld refinement using the GSAS software. Representative refinements of the X-ray data are presented in appendix A.

III. RESULTS

Fig. 1a shows our discovery of a sudden reversal in the pressure dependence of $T_c$ in CsFe$_2$As$_2$ at a critical pressure $P_c = 14 ± 1$ kbar. The shift of $T_c$ as a function of pressure clearly changes direction from decreasing (Fig. 1b) to increasing (Fig. 1c) across the critical pressure $P_c$. $T_c$ varies linearly near $P_c$, resulting in a $V$-shaped phase diagram similar to that of KFe$_2$As$_2$.

Measurements of the Hall coefficient $R_H$ allow us to rule out the possibility of a Lifshitz transition, i.e., a sudden change in the Fermi surface topology. Fig. 2 shows the temperature dependence of $R_H$ at five different pressures. In the zero-temperature limit, $R_H(T → 0)$ is seen to remain unchanged across $P_c$ (Fig. 2 inset). If the Fermi surface underwent a change, such as the disappearance of one sheet, this would affect $R_H(T → 0)$, which is a weighted average of the Hall response of the various sheets. Similar Hall measurements were also used to rule out a Lifshitz transition in KFe$_2$As$_2$ in agreement with the lack of any change in dHvA frequencies.

Several studies on the Ba$_{1-x}$K$_x$Fe$_2$As$_2$ series suggest that lattice parameters, in particular the As-Fe-As bond angle, control $T_c$. To explore this hypothesis, we measured the lattice parameters of KFe$_2$As$_2$ as a function of pressure, up to 60 kbar, in order to find out how much pressure is required to tune the lattice parame-
show the pressure variation of $R_H$ in CsFe$_2$As$_2$ (sample 2), at five selected pressures, as indicated. The low-temperature data converge to the same value for all pressures, whether below or above $P_c$. Inset: The value of $R_H$ extrapolated to $T = 0$ is plotted at different pressures. Horizontal and vertical error bars are smaller than symbol dimensions. $R_H(T = 0)$ is seen to remain unchanged across $P_c$.

The four panels of Fig. 3 show the pressure variation of the lattice constants $a$ and $c$, the unit cell volume ($V = a^2c$), and the intra-planar As-Fe-As bond angle ($\alpha$) in KFe$_2$As$_2$. The red horizontal line in each panel marks the value of the corresponding lattice parameter in CsFe$_2$As$_2$. In order to tune $a$, $c$, $V$, and $\alpha$ in KFe$_2$As$_2$ to match the corresponding values in CsFe$_2$As$_2$, a negative pressure of approximately $-10$, $-75$, $-30$, and $-30$ kbar is required, respectively. Adding these numbers to the critical pressure for KFe$_2$As$_2$ ($P_c = 17.5$ kbar), we would naively estimate that the critical pressure in CsFe$_2$As$_2$ should be $P_c \approx 30$ kbar or higher. We find instead that $P_c = 14$ kbar, showing that other factors are involved in controlling $P_c$.

It is possible that the lower $P_c$ in CsFe$_2$As$_2$ could be due to the fact that $T_c$ itself is lower than in KFe$_2$As$_2$ at zero pressure, i.e., that the low-pressure phase is weaker in CsFe$_2$As$_2$. One hypothesis for the lower $T_c$ in CsFe$_2$As$_2$ is a higher level of disorder. To test this idea, we studied the pressure dependence of $T_c$ in a less pure KFe$_2$As$_2$ sample. Fig. 3 compares the $T$-$P$ phase diagram in three samples: 1) a high-purity KFe$_2$As$_2$ sample, with $\rho_0 = 0.2 \mu \Omega$ cm (from ref. 12); 2) a less pure KFe$_2$As$_2$ sample, with $\rho_0 = 1.3 \mu \Omega$ cm, measured here; 3) a CsFe$_2$As$_2$ sample (sample 2), with $\rho_0 = 1.5 \mu \Omega$ cm. Different disorder levels in our samples are due to growth conditions, not to deliberate chemical substitution or impurity inclusions. First, we observe that a 6-fold increase of $\rho_0$ has negligible impact on $P_c$ in KFe$_2$As$_2$. Secondly, we observe that $P_c$ is 4 kbar smaller in CsFe$_2$As$_2$ than in KFe$_2$As$_2$, for samples of comparable $\rho_0$. These observations rule out the idea that disorder could be responsible for the lower value of $P_c$ in CsFe$_2$As$_2$ compared to KFe$_2$As$_2$.

IV. DISCUSSION

We have established a common trait in CsFe$_2$As$_2$ and KFe$_2$As$_2$: both systems have a sudden reversal in the pressure dependence of $T_c$, with no change in the underlying Fermi surface. The question is: what controls that transition? Why does the low-pressure superconducting state become unstable against the high-pressure state?

In a recent theoretical work by Fernandes and Millis,
it is demonstrated that different pairing interactions in 122 systems can favour different pairing symmetries.\(^\text{11}\) In their model, SDW-type magnetic fluctuations, with wavevector \((\pi, 0)\), favour \(s_\pm\) pairing, whereas Néel-type fluctuations, with wavevector \((\pi, \pi)\), strongly suppress the \(s_\pm\) state and favour \(d\)-wave pairing. A gradual increase in the \((\pi, \pi)\) fluctuations eventually causes a phase transition from an \(s_\pm\) superconducting state to a \(d\)-wave state, producing a V-shaped \(T_c\) vs \(P\) curve.\(^\text{11}\)

In KFe\(_2\)As\(_2\) and CsFe\(_2\)As\(_2\), it is conceivable that two such competing interactions are at play, with pressure tilting the balance in favor of one versus the other. We explore such a scenario by looking at how the inelastic scattering evolves with pressure, measured via the inelastic resistivity, defined as \(\rho(T) - \rho_0\), where \(\rho_0\) is the residual resistivity. Fig. 4(a) shows raw resistivity data from the KFe\(_2\)As\(_2\) sample with \(\rho_0 = 1.3\) \(\mu\Omega\) cm below 30 K. To extract \(\rho(T) - \rho_0\) at each pressure, we make a cut through each curve at \(T = 20\) K and subtract from it the residual resistivity \(\rho_0\) that comes from a power-law fit \(\rho = \rho_0 + A T^n\) to each curve. \(\rho_0\) is determined by disorder level and does not change as a function of pressure. The resulting \(\rho(T = 20\) K\) - \(\rho_0\) values for this sample are then plotted as a function of normalized pressure \(P/P_c\) in Fig. 4(b). Through a similar process we extract the pressure dependence of \(\rho(20\) K\) - \(\rho_0\) in CsFe\(_2\)As\(_2\) and the purer KFe\(_2\)As\(_2\) sample with \(\rho_0 = 0.2\) \(\mu\Omega\) cm in Fig. 4(c) and (d). In all three samples, at \(P/P_c > 1\), the inelastic resistivity varies linearly with pressure. As \(P\) drops below \(P_c\), the inelastic resistivity in (K,Cs)Fe\(_2\)As\(_2\) shows a clear rise below their respective \(P_c\) over and above the linear regime. Fig. 4 therefore suggests a connection between the transition in the pressure dependence of \(T_c\) and the appearance of an additional inelastic scattering process. Note that our choice of \(T = 20\) K for the inelastic resistivity is arbitrary. Resistivity cuts at any finite temperature above \(T_c\) give qualitatively similar results.

The Fermi surface of KFe\(_2\)As\(_2\) includes three \(\Gamma\)-centered hole-like cylinders. A possible pairing state is an \(s_\pm\) state where the change of sign occurs between the inner cylinder and the middle cylinder, favored by a small-\(Q\) interaction.\(^\text{14}\) By contrast, the intraband inelastic scattering wavevectors that favor \(d\)-wave pairing are large-\(Q\) processes.\(^\text{20}\) Therefore, one scenario in which to understand the evolution in the inelastic resistivity with pressure (Fig. 5), and its link to the \(T_c\) reversal, is the following. At low pressure, the large-\(Q\) scattering processes that favor \(d\)-wave pairing make a substantial contribu-

**FIG. 4.** Pressure dependence of \(T_c\) in three samples: pure KFe\(_2\)As\(_2\) (black circles), less pure KFe\(_2\)As\(_2\) (grey circles), and CsFe\(_2\)As\(_2\) (sample 2, red circles). Even though the \(T_c\) values for the two KFe\(_2\)As\(_2\) samples are different due to different disorder levels, measured by their different residual resistivity \(\rho_0\), the critical pressure is the same \((P_c = 17.5\) kbar\). This shows that the effect of disorder on \(P_c\) in KFe\(_2\)As\(_2\) is negligible. For comparable \(\rho_0\), the critical pressure in CsFe\(_2\)As\(_2\), \(P_c = 14\) kbar, is clearly smaller than in KFe\(_2\)As\(_2\).

**FIG. 5.** a) Resistivity data for the KFe\(_2\)As\(_2\) sample with \(\rho_0 = 1.3\) \(\mu\Omega\) cm at five selected pressures. The black vertical arrow shows a cut through each curve at \(T = 20\) K and the dashed line is a power law fit to the curve at \(P = 23.8\) kbar from 5 to 15 K that is used to extract the residual resistivity \(\rho_0\). Inelastic resistivity, defined as \(\rho(T = 20\) K\) - \(\rho_0\) is plotted vs \(P/P_c\) in b) the less pure KFe\(_2\)As\(_2\) sample, c) the purer KFe\(_2\)As\(_2\) sample, and d) CsFe\(_2\)As\(_2\) (sample 2) where \(P_c = 17.5\) kbar for KFe\(_2\)As\(_2\) and \(P_c = 14\) kbar for CsFe\(_2\)As\(_2\). In panel (b), (c), and (d) the dashed black line is a linear fit to the data above \(P/P_c = 1\).
tion to the resistivity, as they produce a large change in momentum. These weaken with pressure, causing a decrease in both $T_c$ and the resistivity. This decrease persists until the low-$Q$ processes that favor $s_\pm$ pairing, less visible in the resistivity, come to dominate, above $P_c$.

In summary, we discovered a pressure-induced reversal in the dependence of the transition temperature $T_c$ on pressure in the iron-based superconductor CsFe$_2$As$_2$, similar to our previous finding in KFe$_2$As$_2$. We interpret the $T_c$ reversal at the critical pressure $P_c$ as a transition from one pairing state to another. The fact that $P_c$ in CsFe$_2$As$_2$ is smaller than in KFe$_2$As$_2$, even though all lattice parameters would suggest otherwise, shows that structural parameters alone do not control $P_c$. We also demonstrate that disorder has negligible effect on $P_c$. Our study of the pressure dependence of resistivity in CsFe$_2$As$_2$ and KFe$_2$As$_2$ reveals a possible link between $T_c$ and inelastic scattering. Our proposal is that the high-pressure phase in both materials is an $s_\pm$ state that changes sign between $\Gamma$-centered pockets. As the pressure is lowered, the large-$Q$ inelastic scattering processes that favor $d$-wave pairing in pure KFe$_2$As$_2$ and CsFe$_2$As$_2$ grow until at a critical pressure $P_c$ they cause a transition from one superconducting state to another, with a change of pairing symmetry from $s$-wave to $d$-wave. The experimental evidence for this is the fact that below $P_c$ the inelastic resistivity, measured as the difference $\rho(20 \text{ K}) - \rho_0$, deviates upwards from its linear pressure dependence at high pressure.

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Appendix A: X-ray data

All our X-ray measurements are performed at room temperature using angle-dispersive technique with the HXMA beam line at CLS. Figure 6 includes two representative structural refinements of the X-ray diffraction data at $P = 1.6$ kbar and $P = 51$ kbar. 2D diffraction data from the image plate detector were reduced to 1D using the FIT2D program and plotted as intensity versus $\Theta$. The structural refinements were performed using the GSAS software package. The experimental data points are illustrated by red crosses, the best fit to the diffraction pattern is illustrated by the solid black line, and the difference between the two curves is denoted by the solid blue line. The Bragg reflections corresponding to the tetragonal $I4/mmm$ structure of KFe$_2$As$_2$ are indicated by the black tick marks below the data.

Appendix B: Bond angles

Within the tetragonal structure of KFe$_2$As$_2$, there are two bond angles in each FeAs$_4$ tetrahedron as indicated in the inset of Fig. 7. The intra-planar bond angle ($\alpha$) that spans the bond from one As plane to an Fe atom and back to an As atom in the original plane and the inter-planar bond angle ($\beta$) that spans the bond from one As plane through an Fe atom to the next As plane. In the case of an ideal undistorted tetrahe-
In Fig. 7, we present only the intra-planar bond angle $\alpha$ to show that about $\sim$30 kbar is required to tune $\alpha$ from its value in KFe$_2$As$_2$ to CsFe$_2$As$_2$. For completeness, here we plot the pressure evolution of both bond angles in Fig. 7. $\alpha$ decreases as a function of pressure while $\beta$ increases, hence, the size of the tetragonal distortion in KFe$_2$As$_2$ grows progressively larger as the pressure increases. Interestingly, the form of this tetragonal distortion is opposite to that observed in Ca$_{0.67}$Sr$_{0.33}$Fe$_2$As$_2$ where applied pressure causes intra-layer bond angles to increase and inter-layer bond angles to decrease.

**Appendix C: Anisotropic compressibility in KFe$_2$As$_2$**

In Fig. 3 we fit our data to the Murnaghan equation of state,

$$P(V) = \frac{K}{K'} \left[ \left( \frac{V}{V_0} \right)^{-K'} - 1 \right]$$

and extend it smoothly to negative pressures to find how much pressure is required to tune the lattice parameters of KFe$_2$As$_2$ to those of CsFe$_2$As$_2$. Note that the compressibility of KFe$_2$As$_2$ appears to be anisotropic. The fits also allow us to extract the bulk modulus $K$ and its pressure derivative $K' = \partial K/\partial P$ in KFe$_2$As$_2$. Table 1 summarizes the values of the bulk modulus $K$ as well as the moduli of elasticity along the $a$- and $c$-axes. The modulus of elasticity appears to be almost identical along the $a$- and the $c$-axes, but the first derivative of the modulus is over an order of magnitude larger along the $a$-axis. This accounts for the roughly 40% smaller compression observed for the in-plane lattice constant.

| $K_a$ (GPa) | $K_c$ (GPa) | $K'$ (GPa) | $K''$ (GPa) |
|-------------|-------------|------------|-------------|
| 105 $\pm$ 5 | 115 $\pm$ 3 | 40 $\pm$ 1 | 400 $\pm$ 2 |
| $3.3 \pm 0.8$ | $6.1 \pm 0.4$ |

**TABLE I.** The moduli of elasticity along $a$-axis $K_a$ and $c$-axis $K_c$ as well as the bulk modulus $K$ are extracted by fitting our data to the Murnaghan equation of state. The pressure derivatives of $K_a$, $K_c$, and $K'$ are also reported.

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