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Pressure-temperature phase diagrams of $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$ superconductors

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The pressure dependence of the magnetic and superconducting transitions, as well as that of the superconducting upper critical field is reported for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, the first example of an Fe-based superconductor with spin-vortex-crystal-type magnetic ordering. Resistance measurements were performed on single crystals with two substitution levels ($x = 0.033, 0.050$) under hydrostatic pressures up to 5.12 GPa and in magnetic fields up to 9 T. Our results show that, for both compositions, magnetic transition temperatures, T_N , are suppressed upon applying pressure, the superconducting transition temperatures T_c are suppressed by pressure as well, except for $x = 0.050$ in the pressure region where T_N and T_c cross. Furthermore, the pressure associated with the crossing of the T_N and T_c lines also coincides with a minimum in the normalized slope of the superconducting upper critical field, consistent with a likely Fermi-surface reconstruction associated with the loss of magnetic ordering. Finally, at $p \sim 4$ GPa, both Ni-substituted $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$ samples likely go through a half-collapsed-tetragonal (hcT) phase transition, similar to the parent compound $\text{CaKFe}_4\text{As}_4$.

I. INTRODUCTION

Since the discovery of Fe-based superconductors (FeSC)^{1–4}, many studies have been done on them and they have expanded into a large family. Among them the AeFe_2As_2 compounds ($\text{Ae}=\text{Ca}, \text{Sr}, \text{Ba}, \text{Eu}$) have received significant attention because large, high-quality single crystals can be obtained with a variety of chemical substitution^{5,6}. Studies have revealed that members of this family share a global phase diagram upon tuning by substitution or pressure^{5,7}. At ambient pressure, the parent compounds undergo a structural/magnetic transition upon cooling; substitution or pressure induce superconductivity after sufficiently suppressing the structural/magnetic transitions^{5,6,8–11}. This suggests a competition between the magnetism and superconductivity, and that magnetic fluctuations play an important role in forming superconductivity in this system^{7,12–16}.

Recently, a new FeSC $\text{AeAFe}_4\text{As}_4$ ($\text{A}=\text{K}, \text{Rb}, \text{Cs}$) structural type ($P4/mmm$) was discovered by Iyo *et al*¹⁷. This is not a homogeneous substitution as in $(\text{Ae}_{0.5}\text{A}_{0.5})\text{Fe}_2\text{As}_2$ where Ae/A share the same crystallographic site. Each Ae and A in the $\text{AeAFe}_4\text{As}_4$ structure has a unique, well-defined, crystallographic site, forming alternating Ae and A planes along the c -axis^{17,18}. Among them, single crystals of $\text{CaKFe}_4\text{As}_4$ were synthesized and found to be superconducting at ~ 35 K and no other phase transition from 1.8 K to 300 K at ambient pressure^{18,19}. A pressure study up to 6 GPa shows that the superconducting transition temperature, T_c , is suppressed to about 28.5 K before it undergoes half-collapsed-tetragonal (hcT) phase transition at ~ 4 GPa and loses bulk superconductivity²⁰. The hcT phase transition occurs due to the As-As bonding across the Ca-layer under pressure, like the collapsed-tetragonal transition in CaFe_2As_2 at ~ 0.35 GPa^{21–23}.

From the perspective of electron count, $\text{CaKFe}_4\text{As}_4$ is analogous to $(\text{Ba}_{0.5}\text{K}_{0.5})\text{Fe}_2\text{As}_2$ and many of its prop-

erties are consistent with this¹⁸. In the later compound, the stripe-type spin density wave associated with BaFe_2As_2 is suppressed by hole doping⁷ (substitution K for Ba). A recent study revealed that adding electrons to $\text{CaKFe}_4\text{As}_4$ via Ni or Co substitution drives the system back towards a magnetic phase. In contrast to the stripe-type antiferromagnetism in the "122" systems, the order in the Ni- or Co-substituted $\text{CaKFe}_4\text{As}_4$ is experimentally identified as a new hedgehog spin-vortex-crystal (SVC) magnetism that has no structural phase transition associated with it²⁴. This type of magnetic order had been theoretically predicted but until the discovery of Ni- or Co-substituted $\text{CaKFe}_4\text{As}_4$, was considered to be a "missing link"^{25–27}. Increasing the substitution level of Ni or Co in $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$ leads to the suppression of the superconducting transition temperature T_c and stabilizing the SVC magnetism and increasing T_N ²⁴.

The application of pressure to $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ suppresses AFM (T_N falls) and increases T_c ²⁸. This has been taken as an indication that pressure, like doping, can tune T_N and the associated AFM fluctuations to favor the superconducting state when $T_N > T_c$. Therefore, it is natural to study how the SVC magnetic order behaves under pressure, specifically, how the magnetism and superconductivity interact in this system and whether this interaction is similar to $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$.

In this work, we present the first pressure study on Ni-substituted $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$ ($x = 0.033$ and 0.050) up to 5.12 GPa. The pressure-temperature ($p - T$) phase diagrams inferred from resistance measurements allow comparison of $T_N(p)$ and $T_c(p)$. Specifically, $p - T$ phase diagrams reveal that T_N is suppressed with pressure for both substitution levels. In contrast to $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, T_c is suppressed as well, although more slowly. For $x = 0.050$, it exhibits an anomaly at the pressure where T_c and T_N cross. At ~ 4 GPa both compositions appear to undergo the hcT transition as was observed in the undoped $\text{CaKFe}_4\text{As}_4$. Furthermore, su-

perconducting upper critical fields studied up to 9 T suggests a Fermi-surface reconstruction when $T_N(p)$ crosses $T_c(p)$.

II. EXPERIMENTAL DETAILS

Single crystals of $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$ ($x = 0.033$ and 0.050) with sharp superconducting transitions at ambient pressure [See Figs 1(b)-3(b)] were grown using high-temperature solution growth^{18,19}. The substitution level, x , was determined by performing wavelength-dispersive x-ray spectroscopy (WDS) as described in Ref. 24.

The in-plane ab resistance was measured using standard four-probe configuration. The $25\ \mu\text{m}$ Pt wires were soldered to the samples using a Sn:Pb-60:40 alloy. For $x = 0.033$, two samples, #1 and #2, were cut from one single crystal. They were then measured in a piston-cylinder cell (PCC)²⁹ and a modified Bridgman Anvil Cell (mBAC)³⁰ respectively. For $x = 0.050$, a single sample was prepared and measured in the mBAC. Pressure values for both cells, at low temperature, were inferred from the $T_c(p)$ of lead³¹. For the PCC, a 4:6 mixture of light mineral oil:n-pentane was used as the pressure medium, which solidifies, at room temperature, in the range of 3-4 GPa. For the mBAC, a 1:1 mixture of iso-pentane:n-pentane was used as the pressure medium, which solidifies, at room temperature, in the range of 6-7 GPa. Both of the solidification pressures are well above the maximum pressures achieved in the pressure cells, which suggests good hydrostatic conditions^{29,32,33}.

The ac resistance measurements were performed in a Quantum Design Physical Property Measurement System using $I = 1\ \text{mA}$; $f = 17\ \text{Hz}$ excitation, on cooling with the rate of $0.25\ \text{K/min}$ and the magnetic field was applied along the c axis.

III. RESULTS AND DISCUSSIONS

Figs. 1(a) and 2(a) show the pressure dependence of the temperature dependent resistance for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.033$. Sample #1 was measured in the PCC for pressures up to 1.83 GPa. Sample #2 was measured in the mBAC for pressures up to 5.12 GPa. For both samples, the 0 GPa resistance was corrected for geometric changes to the sample via normalization. (Details of the normalization are described in the Appendix.) Fig. 3(a) shows the pressure dependence of the temperature dependent resistance for the $x = 0.050$ sample that was measured in the mBAC for pressures up to 5.12 GPa. In general, for all samples, the resistance decreases under applied pressure.

For both compositions, the magnetic phase transition T_N appears as a kink-like anomaly in the lower temperature data and is more pronounced in the $x = 0.050$ compound. This feature is more clearly revealed as a step-like anomaly in the temperature derivative dR/dT

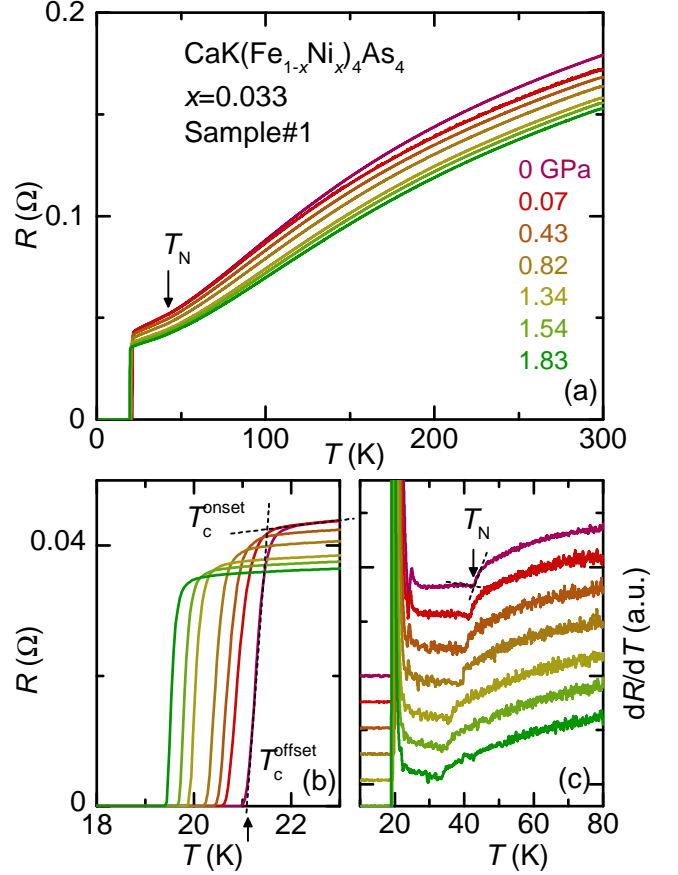


FIG. 1. (a) Evolution of the in-plane resistance with hydrostatic pressures up to 1.83 GPa measured in a PCC for the $\text{CaK}(\text{Fe}_{0.967}\text{Ni}_{0.033})_4\text{As}_4$, sample #1. (b) Blowup of the low temperature region. Criteria for T_c^{onset} and T_c^{offset} are indicated in the figure. (c) Temperature derivative, dR/dT , showing the evolution of the magnetic transition T_N with offset criteria as shown in the figure.

[Figs. 1(c), 2(c) and 3(c)]. These plots demonstrate that T_N is suppressed by increasing pressure before it disappears at higher pressures.

The blowups of the low temperature resistance [Figs. 1(b), 2(b) and 3(b)] show how T_c changes under increasing pressure. For $x = 0.033$, T_c monotonically decreases in the studied pressure range. In contrast, for $x = 0.050$, after 2.41 GPa there is a slight enhancement of the T_c before it is suppressed again at higher pressures.

Upon increasing pressures above $\sim 4\ \text{GPa}$, the sharp superconducting transition at lower pressures becomes broadened at higher pressures. A similar behavior was also observed in the parent compound $\text{CaKFe}_4\text{As}_4$ and has been associated with the $h\text{cT}$ phase transition at $p \gtrsim 4\ \text{GPa}$ ²⁰. In order to understand the nature of the broadening in the substituted system, analysis similar to that in Ref. 20 was carried out.

Fig. 4 presents the temperature dependence of the resistance under magnetic field up to 9 T for selected pressures. The superconducting transition width, $\Delta T =$

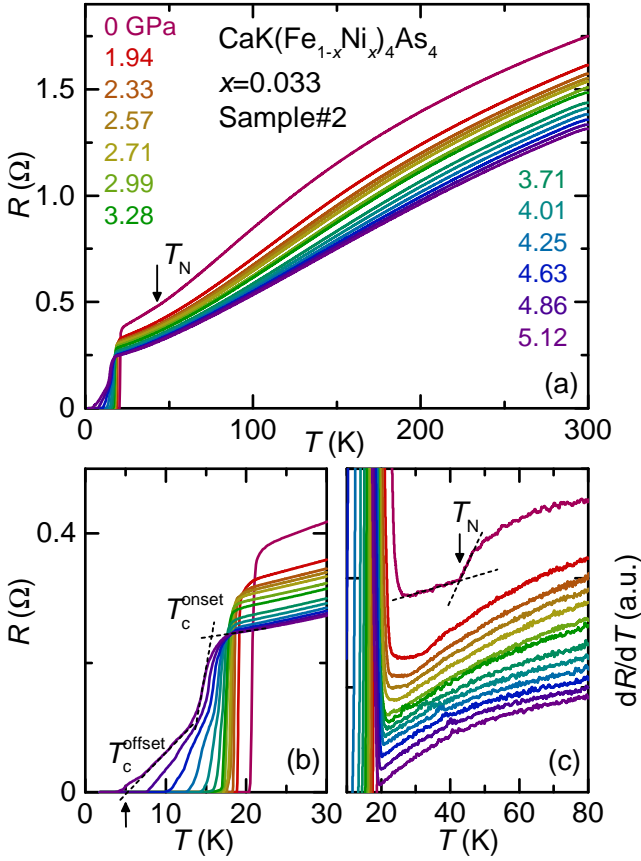


FIG. 2. (a) Evolution of the in-plane resistance with hydrostatic pressures up to 5.12 GPa measured in a mBAC for $\text{CaK}(\text{Fe}_{0.967}\text{Ni}_{0.033})_4\text{As}_4$, sample#2. (b) Blowup of the low temperature region. (c) Temperature derivative, dR/dT , showing the evolution of magnetic transition T_N .

$T_c^{\text{onset}} - T_c^{\text{offset}}$, is broadened with increasing pressure, with the criteria for T_c^{onset} and T_c^{offset} shown in Figs. 1(b), 2(b) and 3(b). In order to determine whether the broadening is associated with any sort of phase transition, or is simply due to pressure inhomogeneities in the pressure medium when larger loads are applied, the field dependence of the superconducting transition width $\Delta T(H)$ was studied²⁰. Specifically, the transition width at magnetic fields 0 T and at 3 T (indicated by thicker lines in Figs. 4) were determined, and then the difference between them, $\Delta T(3T) - \Delta T(0)$, was calculated. Any broadening due to the pressure inhomogeneities are expected to be equally present in the $H = 0$ T and 3 T data. Figs. 5(a) and (c) present the pressure dependence of the transition width difference. As it is clearly shown, for both compositions, $\Delta T(3T) - \Delta T(0)$ increases dramatically as pressure goes above $p^* \sim 4$ GPa (indicated by arrows in Figs. 5(a), (c)). Note that for $x = 0.050$, at 5.12 GPa, the transition width difference was taken between $H = 0$ T and 1 T, because T_c^{offset} is not clearly defined at $H = 3$ T. But we would expect the transition width difference between $H = 0$ T and 3 T to be even

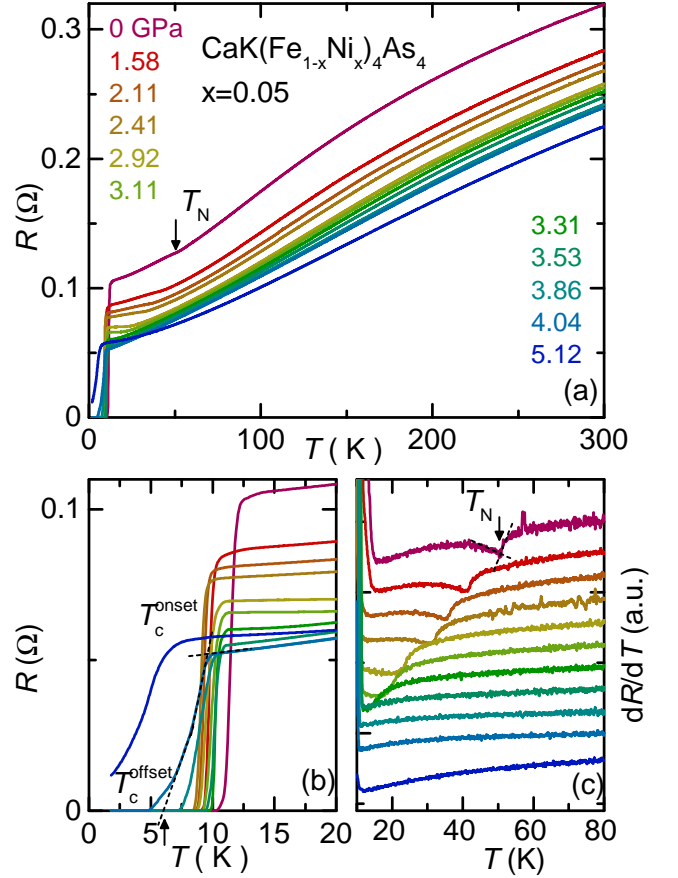


FIG. 3. (a) Evolution of the in-plane resistance with hydrostatic pressures up to 5.12 GPa measured in a mBAC for $\text{CaK}(\text{Fe}_{0.95}\text{Ni}_{0.05})_4\text{As}_4$. (b) Blowup of the low temperature region. (c) Temperature derivative, dR/dT , showing the evolution of magnetic transition T_N .

larger at this pressure. Furthermore, the pressure dependence of the resistance $R(p)$ at fixed temperatures for both compositions (Figs. 5(b), (d)) shows anomaly at the same pressure at 40 K (indicated by arrows in the figure), though subtle for $x = 0.033$. Based on the analogy with the parent compound $\text{CaKFe}_4\text{As}_4$ ²⁰, we identify this anomaly as an indication of the hcT phase transition that exists from base temperature up to at least 40 K. As was the case for pure $\text{CaKFe}_4\text{As}_4$, we believe that superconductivity is not bulk for $p \gtrsim 4$ GPa (i.e. in the hcT phase).

The upper superconducting critical field H_{c2} can be evaluated from Fig. 4 at pressures lower than p^* , where superconductivity is considered bulk, using the offset criteria defined in Figs. 1-3. The temperature dependence of H_{c2} at various pressures is presented in Figs. 6 and 7 for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.033$ and 0.050 respectively. For $x = 0.033$, both Sample#1 and Sample#2 were analyzed and plotted in Fig 6. Note that at ambient pressure, T_c^{offset} values for two samples differ by ~ 0.5 K, possibly due to a small difference of the substitution level at different positions of the crystal they were

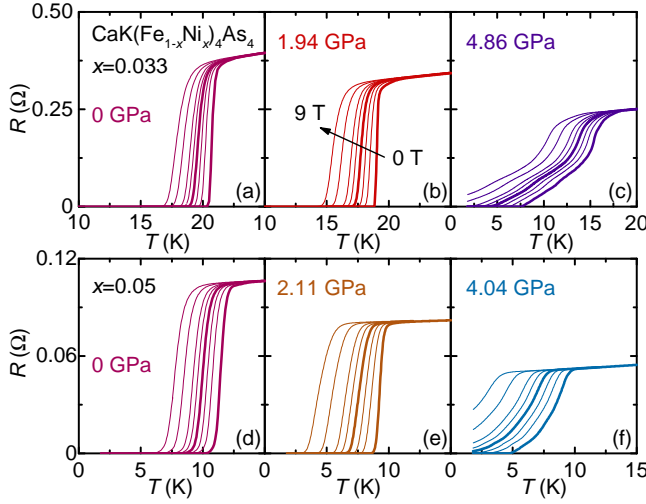


FIG. 4. Temperature dependence of resistance under magnetic field up to 9 T for selective pressures for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.033$ ((a)-(c)), $x = 0.050$ ((d)-(f)). Superconducting transition becomes broader as pressure is increased for both compounds, to explore the nature of the broadening, transition width at 0 T and 3 T (indicated by thick lines in the figures) were analyzed and described in details in the text.

cut from. As is shown in Figs. 6 and 7, for $x = 0.033$, H_{c2} is systematically suppressed by increasing pressure, whereas, for $x = 0.050$, the evolution of the temperature dependent H_{c2} is nonmonotonic. For both compositions, H_{c2} is linear in temperature except for magnetic fields below 1 T. The curvature at low fields has been observed in other FeSC and can be explained by nature of superconductivity^{34–36}, which is also the case for the parent compound $\text{CaKFe}_4\text{As}_4$ ³⁷.

Figs. 8(a) and 9(a) present the p - T phase diagrams for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.033$ and 0.050 respectively, with T_c^{offset} and T_N values obtained using the criteria shown in Figs. 1-3 and the indication of non-bulk superconductivity above p^* . For both compositions, T_N is suppressed by pressure, specifically, T_N is suppressed from 43 K to 25 K at 2.71 GPa for $x = 0.033$ and suppressed from 51 K to 13.8 K at 3.31 GPa for $x = 0.050$.

In terms of superconductivity, for $x=0.033$, T_c^{offset} is monotonically suppressed with increasing pressure. It drops from 20.5 K to 15.1 K at 4.01 GPa before superconductivity becomes non-bulk. A closer examination reveals that T_c^{offset} is initially linearly suppressed by pressure up to 2.71 GPa, then a small, but clear deviation from the linear suppression was observed above 2.99 GPa. An extrapolation of T_N shows that the deviation happens near the crossing of T_N and T_c^{offset} lines. For $x = 0.050$, the behavior of $T_c^{\text{offset}}(p)$ is distinctly non-monotonic. T_c^{offset} is initially linearly suppressed from 11 K to a local minimum of 8.7 K at 2.41 GPa. Then it rises to a maximum of 10 K at 3.31 GPa, exhibiting a dome shape. This dome of enhanced T_c^{offset} coincides

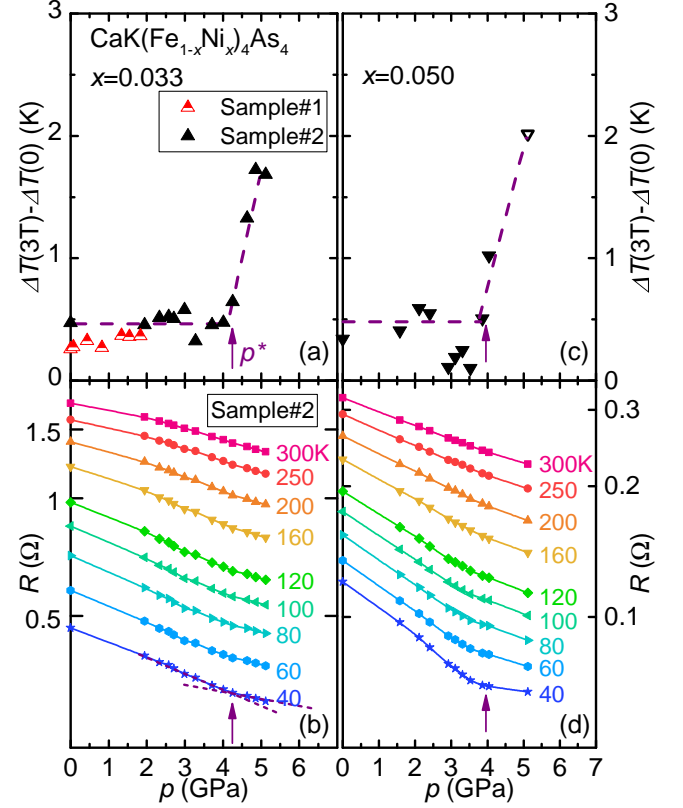


FIG. 5. (a),(c) Pressure dependence of the superconducting transition widths difference for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.033$ and 0.050 respectively. The superconducting transition widths is $\Delta T = T_c^{\text{onset}} - T_c^{\text{offset}}$ and the widths difference is taken between 0 field and 3 T. Open symbol in panel (c) is the widths difference taken between 0 field and 1 T because of no clear definition of T_c^{offset} at 3 T for 5.12 GPa. (b), (d) Pressure dependence of resistance at $R(p)$ fixed temperatures for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.033$ and 0.050 respectively. The critical pressure p^* (Arrows in the figure) which is associated with the hcT phase is described in details in the text.

with the disappearance of T_N . After the local maximum in T_c^{offset} there is a much more rapid suppression of T_c^{offset} with increasing p until the hcT transition at p^* . For both compositions, a change in $T_c^{\text{offset}}(p)$ happens at the pressure where T_N and T_c^{offset} lines cross.

Both compositions show signatures of non-bulk superconductivity above $p^* \sim 4$ GPa (blue symbols in Figs. 8(a), 9(a)) similar to the parent compound $\text{CaKFe}_4\text{As}_4$ ²⁰, suggesting the same hcT phase transition. Pressure dependent resistance data in Fig. 5 demonstrates that the hcT phase transition is discernable up to at least 40 K for the substituted compounds. The transition pressure does not appear to change with Ni-substitution. This is not too surprising given the fact that the hcT transition does not involve the Fe-plane but is, instead As-As bonding across the Ca plane.

To better understand the superconducting properties of $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, the superconducting upper crit-

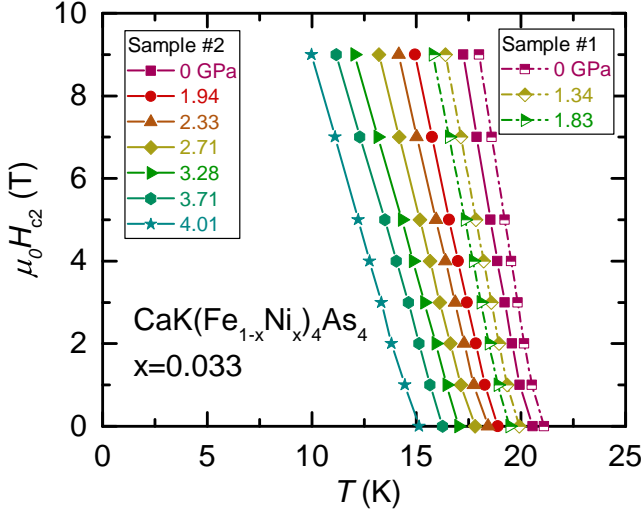


FIG. 6. Temperature dependence of the upper superconducting critical field, $H_{c2}(T)$, under selected pressures for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.033$. T_c^{offset} is used for the figure. Half filled and solid symbols are two samples measured in PCC and mBAC respectively.

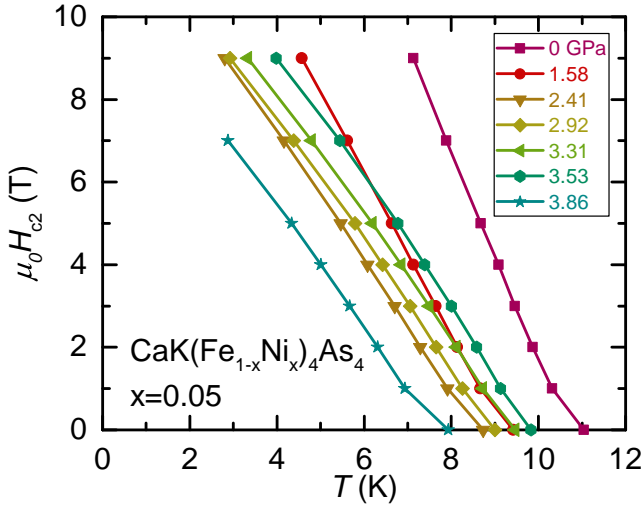


FIG. 7. Temperature dependence of the upper superconducting critical field, $H_{c2}(T)$, under selected pressures for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.050$. T_c^{offset} is used for the figure.

ical field H_{c2} was analyzed following Refs. 35, 36, and 38. Generally speaking, the slope of the upper critical field normalized by T_c , is related to the Fermi velocity and superconducting gap of the system³⁴. In the clean limit, for a single-band,

$$-(1/T_c)(d\mu_o H_{c2}/dT)|_{T_c} \propto 1/v_F^2, \quad (1)$$

where v_F is the Fermi velocity. Even though the superconductivity in $\text{CaKFe}_4\text{As}_4$ compounds is multiband, Eq. 1 can give qualitative insight into changes induced by pressure.

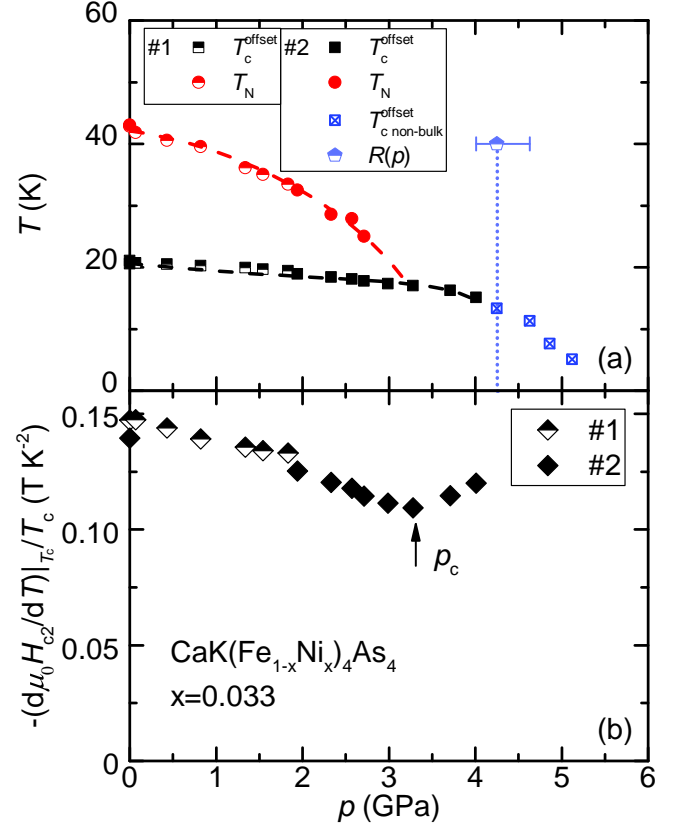


FIG. 8. (a) Temperature-pressure phase diagram of $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.033$, as determined from resistance measurement. The squares and circles represent the superconducting T_c^{offset} and magnetic T_N phase transition. Half filled and solid symbols are two samples measured in the PCC and the mBAC respectively. Blue symbols represent T_c^{offset} for filamentary superconductivity. Dashed lines are guides to the eye. Blue dotted line indicates the half-collapsed-tetragonal phase transition up to 40 K, inferred from the pressure dependent resistance $R(p)$ data in Fig. 5. (b) Pressure dependence of the normalized upper critical field slope $-(1/T_c)(d\mu_o H_{c2}/dT)|_{T_c}$. A local minimum in the slope at p_c (indicated by arrow) is observed near the pressure where T_c^{offset} and T_N lines cross.

As is shown in Figs. 8(b) and 9(b), the normalized slope of the upper critical field $-(1/T_c)(d\mu_o H_{c2}/dT)|_{T_c}$ (the slope $d\mu_o H_{c2}/dT|_{T_c}$ is calculated by linear fitting the data from 1-5 T in Figs. 6 and 7) exhibits a similar pressure dependence for $x = 0.033$ and 0.050. It initially decreases upon increasing pressure and then begins to increase above pressure p_c , resulting in a minimum of $-(1/T_c)(d\mu_o H_{c2}/dT)|_{T_c}$ in the studied pressure range. In both compositions, p_c coincides with the crossing of T_N and T_c^{offset} lines, suggesting a common origin of this feature.

In Fe-based superconductors, especially the "122" system, Fermi-surface nesting can lead to a partial opening of a gap at the Fermi-surface below T_N . By tuning with doping or applying pressure, a Fermi-surface

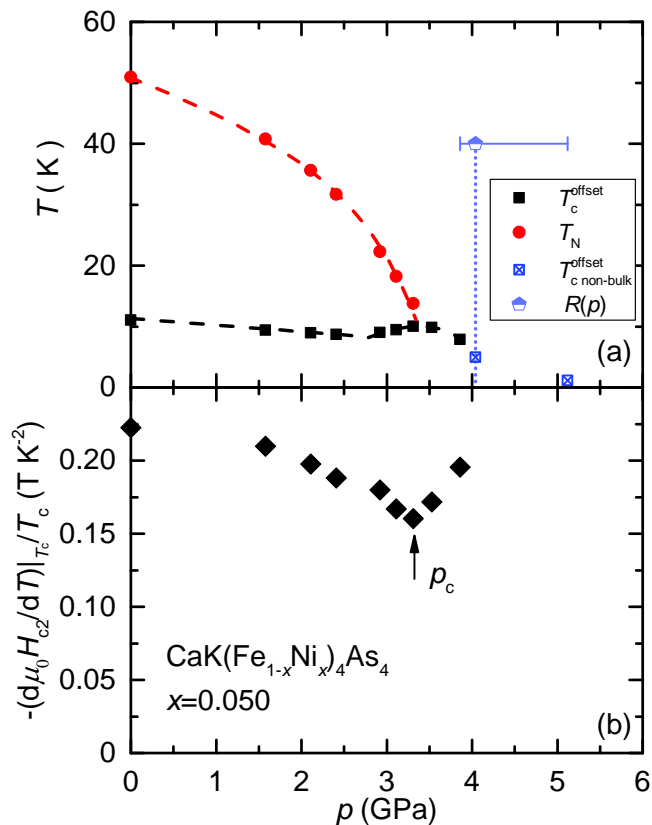


FIG. 9. (a) Temperature-pressure phase diagram of $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.050$, as determined from resistance measurement. The squares and circles represent the superconducting T_c^{offset} and magnetic T_N phase transition. Blue symbols represent T_c^{offset} for filamentary superconductivity. Dashed lines are guides to the eye. Blue dotted line indicates the half-collapsed-tetragonal phase transition up to 40 K, inferred from the pressure dependent resistance $R(p)$ data in Fig. 5. (b) Pressure dependence of the normalized upper critical field slope $-(1/T_c)(d\mu_o H_{c2}/dT)|_{T_c}$. A local minimum in the slope at p_c (indicated by arrow) is observed near the pressure where T_c^{offset} and T_N lines cross.

reconstruction could happen due to the disappearance of magnetism^{39–47}. For $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$ ($x = 0.033$ and 0.050), a clear change of the pressure dependence of the normalized slope $-(1/T_c)(d\mu_o H_{c2}/dT)|_{T_c}$ is observed at p_c , indicating a possible Fermi-surface reconstruction near p_c . Note that for $x = 0.050$, there appears to be a discontinuous change in the normalized slope $-(1/T_c)(d\mu_o H_{c2}/dT)|_{T_c}$ and a subtle anomaly in $T_c(p)$ from 2.41 GPa to 2.92 GPa, suggesting there may be a Lifshitz transition near this pressure. Such features are not observed for $x = 0.033$.

Figs. 8 and 9, then, combine surprising and not unexpected features. The hcT phase transition pressure appears insensitive to Ni substitution. This is reasonable because this transition involves bonding of As atoms across the Ca-plane. The clear feature at p_c in $-(1/T_c)(d\mu_o H_{c2}/dT)|_{T_c}$, as well as the more subtle features

in $T_c(p)$, are again not too surprising and can be associated with the change (with increasing p) from $T_N > T_c$ to $T_N < T_c$, i.e. T_c occurring in an AFM ordered state to T_c occurring in a state lacking the AFM order and associated additional periodicities. The surprising feature shown in Figs. 8 and 9 is the weak suppression of T_c concurrent with the strong suppression of T_N . This is contrary to what is seen in Co substitution and pressure study on BaFe_2As_2 (where T_c increases, as T_N is suppressed)^{5,6,11,28} and brings into question the exact effects suppression of T_N has on the magnetic fluctuations that the superconducting state is nominally built out of.

IV. CONCLUSION

In conclusion, the resistance of Ni-substituted iron-based superconductor $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$ ($x = 0.033$ and 0.050) has been studied under pressures up to 5.12 GPa and in magnetic fields up to 9 T. For both substitution levels, hedgehog spin-vortex-crystal magnetic transition temperature, T_N , is suppressed with increasing pressure. In both compositions, T_c is initially suppressed as well and exhibits a weak anomaly near the crossing of T_N and T_c lines. As pressure exceeds ~ 4 GPa, both compositions likely go through the half-collapsed-tetragonal phase transition, similar to the one observed in the parent compound. This demonstrates the insensitivity of the hcT transition pressure to Ni-substitution. The minimum observed in the normalized slope of the upper critical field, $-(1/T_c)(d\mu_o H_{c2}/dT)|_{T_c}$, at the pressure where T_N and T_c lines cross indicate a possible Fermi-surface reconstruction associated with the disappearance of antiferromagnetism.

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V. APPENDIX

Fig. 10 presents the evolution of the in-plane resistance with hydrostatic pressure for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.033$, solid lines in the figure are the actual measured resistance data, dashed lines are the resistance after normalization. Sample#1 was measured in a PCC for pressures up to 1.83 GPa and Sample#2 was measured in a mBAC for pressures up to 5.12 GPa. Note that the 0 GPa resistance data was measured on a PPMS puck

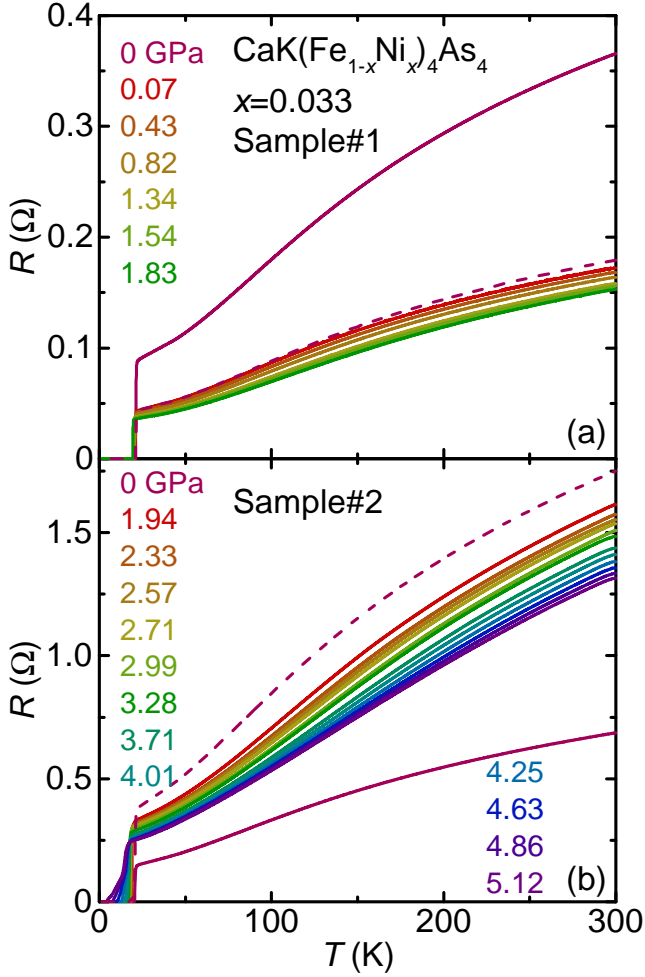


FIG. 10. Evolution of the in-plane resistance with hydrostatic pressure of Sample#1 measured in a PCC (a) and Sample#2 measured in a mBAC (b) for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.033$. Solid lines are the actual resistance data measured, dashed lines are the normalized resistance for 0 GPa. Notice that the 0 GPa resistance is measured on PPMS puck outside of either pressure cell (i.e. ambient pressure); in both cases there is a sudden change between the resistance measured at ambient pressure and inside pressure cell. Possible reasons for the sudden change and details of normalization are explained in details in the text.

outside of either pressure cell (i.e. ambient pressure), a sudden change of resistance between ambient pressure and inside pressure cell was observed in both samples. For Sample#1, when the sample was moved from PPMS puck and mounted onto the PCC, one contact of the voltage channel became detached from the sample and that contact had to be re-attached. As a result, the changed position of the contact led to changes in the resistance before and after. For Sample#2, nothing was intentionally done to the sample before and after it was put into the mBAC, the sudden change of the resistance is most likely due to the exfoliation or cracking of the sample when pressure was first applied as the pressure cell was closed.

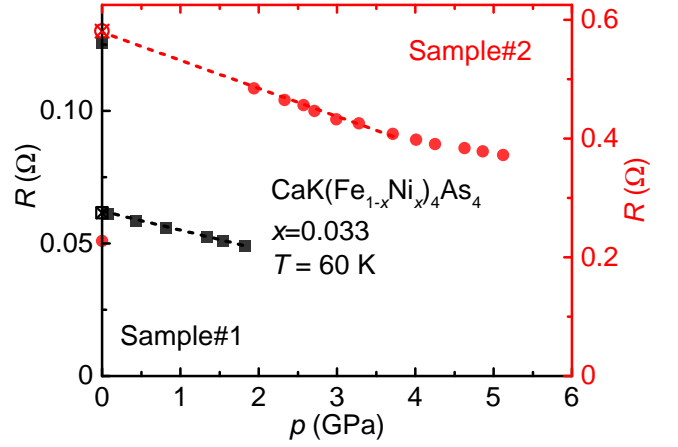


FIG. 11. Pressure dependence of resistance at 60 K for $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$, $x = 0.033$, black solid squares are data from Sample#1 measured in PCC, red solid circles are data from Sample#2 measured in mBAC. Dashed lines are linear fitting of the data before 4 GPa (not including 0 GPa), notice the clear deviation from the linear fitting for the 0 GPa data. Open symbols are the corresponding normalized 0 GPa resistance for Sample#1 and Sample#2 at 60 K.

Despite the abrupt change of resistance from ambient pressure to the first finite pressures inside the pressure cell, the resistance of $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$ ($x = 0.033$ and 0.05) continuously and systematically decreases upon increasing pressure, consistent with the behavior that is observed in parent compound $\text{CaKFe}_4\text{As}_4$ ²⁴ and many "122" systems^{38,48,49}.

To better evaluate the resistance evolution with pressure, especially the pressure dependence of resistance at various temperatures (Fig. 5 (b)(d)), the ambient pressure resistance is shifted via normalization (assuming in each case that the shift was due to geometric changes). Fig. 11 presents the pressure dependence of the resistance at $T = 60$ K for Sample#1 and Sample#2 (solid symbols). Note $T = 60$ K was chosen because the pressure values are determined from the $T_c(p)$ of lead³¹ at ~ 7 K, and the pressure cells are known to have pressure changes with temperature. With the pressure cells and liquid medium we used in this study, the pressure change from room temperature to 7 K can be $0.2 \sim 0.3$ GPa^{30,50}. 60 K was chosen based on the idea that at this temperature, the pressure medium has already solidified³³, the temperature dependence of the thermal expansion of cell materials flattens at low temperature, and the pressure difference between 60 K and 7 K should be small⁵⁰. The fact that 60 K is still above the magnetic transition temperature T_N guarantees that pressure dependence of resistance at this temperature gives no feature related to magnetism. As shown in Fig. 11, except the ambient pressure data, the 60 K resistance for both samples are linearly suppressed by pressure before 4 GPa, so it is assumed that the ambient pressure resistance should also follow this pressure dependence (open symbols in Fig.

11). To do that, the ambient pressure resistance curves for the two samples are multiplied by two corresponding factors and moved to the dashed lines as shown in Fig. 10.

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