

SUPPLEMENTARY INFORMATION

The isotopically substituted polycrystalline samples with nominal compositions $\text{SmFeAsO}_{1-x}\text{F}_x$ ($x=0, 0.15$) were synthesized by conventional solid state reaction using SmAs , SmF_3 , As , Fe and $\text{Fe}_2^{16}\text{O}_3/\text{Fe}_2^{18}\text{O}_3$ as starting materials for oxygen isotope exchange, while using SmAs , SmF_3 , As , $^{54}\text{Fe}/^{56}\text{Fe}$ and $^{54}\text{Fe}_2\text{O}_3/^{56}\text{Fe}_2\text{O}_3$ as starting materials for iron isotope exchange. $\text{Fe}_2^{16}\text{O}_3$ and $\text{Fe}_2^{18}\text{O}_3$ were obtained by sintering Fe powder in natural abundance oxygen $^{16}\text{O}_2$ and $^{18}\text{O}_2$ with an 80% enrichment, respectively. The $^{54}\text{Fe}_2\text{O}_3$ and $^{56}\text{Fe}_2\text{O}_3$ were prepared by sintering ^{54}Fe and ^{56}Fe powder in flowing natural abundance oxygen atmosphere, respectively. The iron isotope polycrystalline samples of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ($x=0, 0.4$) were synthesized using BaAs , KAs , and $^{56}\text{Fe}_2\text{As}$ and $^{54}\text{Fe}_2\text{As}$ as starting materials, respectively. $^{56}\text{Fe}_2\text{As}$ and $^{54}\text{Fe}_2\text{As}$ were synthesized by reacting ^{56}Fe and ^{54}Fe powder with As powder in evacuated quartz tubes at 650°C for 24 h, respectively. To keep the same condition, the isotope exchange samples with the same composition were sealed in the same evacuated quartz tube for annealing. To mitigate the difference of F content in isotope exchange samples $\text{SmFeAsO}_{0.85}\text{F}_{0.15}$, SmAs , SmF_3 and As were first mixed according to stoichiometric ratio and grounded, then mixture of SmAs , SmF_3 and As was equally separated into two parts. Finally, $^{54}\text{Fe}/^{56}\text{Fe}$ and $^{54}\text{Fe}_2\text{O}_3/^{56}\text{Fe}_2\text{O}_3$ as starting materials were weighed and put into the separated mixture of SmAs , SmF_3 and As for iron isotope exchange. It guarantees the same F content for isotope exchange in the beginning of sample process. They were loaded into the same quartz tube for annealing.

Natural abundance iron was used for the oxygen isotope exchange experiments and obtained from

Alfa Aesar, while the ^{56}Fe and ^{54}Fe enriched isotopes from ISOFLEX (San Francisco, USA) were used for the iron isotope experiments. The purity of the natural abundance iron $^{\text{n}}\text{Fe}$ is 99.9%, while the purity of the ^{56}Fe (enrichment: 97%+) is 99.78%, and the purity of the ^{54}Fe (enrichment: 96%+) is 99.86%. Natural abundance oxygen was used for the iron isotope exchange experiments for the system $\text{SmFeAsO}_{1-x}\text{F}_x$. The purity of the natural abundance oxygen $^{\text{n}}\text{O}$ is 99.99%. The purity of the ^{18}O (enrichment: 80%) is 99.99%. Raman spectra were obtained on a LABRAM-HR Confocal Laser MicroRaman Spectrometer using the 514.5 nm line from an argon-ion laser with in-plane light polarization. X-ray diffraction (XRD) was performed by MAC MXPAPHF X-Ray diffractometer with graphite monochromated $\text{CuK}_{\alpha 1}$ radiation ($\lambda=0.15406$ nm) at room temperature. The superconducting transition temperatures and the SDW transition temperatures were determined by standard four-probe resistance and susceptibility measurements. The resistance was measured by an AC resistance bridge (LR-700, Linear Research). Magnetic susceptibility measurements were performed with a superconducting quantum interference device magnetometer (Quantum Design MPMS-7).

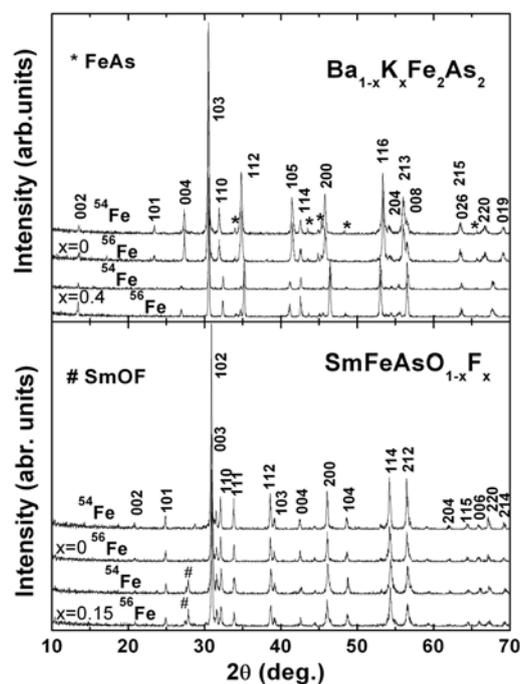


Figure S1: X-ray diffraction patterns at room temperature for the samples $\text{SmFeAsO}_{1-x}\text{F}_x$ and $\text{Ba}_{1-x}\text{K}_x\text{Ba}_2\text{As}_2$ with iron isotope exchange. As shown in the Figure, XRD patterns look like the same for isotope exchange. The XRD patterns are fitted by Rietveld analysis. The obtained lattice constants are: $a=0.3961(1)$ nm, $c=1.3015(4)$ nm for BaFe_2As_2 with ^{54}Fe , $a=0.3962(1)$ nm, $c=1.3017(4)$ nm for BaFe_2As_2 with ^{56}Fe ; $a=0.3918(2)$ nm, $c=1.3289(7)$ nm for $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ with ^{54}Fe , $a=0.3917(2)$ nm, $c=1.3288(7)$ nm for $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ with ^{56}Fe ; $a=0.39415(6)$ nm, $c=0.8502(3)$ nm for SmFeAsO with ^{54}Fe , $a=0.39410(6)$ nm, $c=0.8501(3)$ nm for SmFeAsO with ^{56}Fe ; $a=0.39320(5)$ nm, $c=0.8490(2)$ nm for $\text{SmFeAsO}_{0.85}\text{F}_{0.15}$ with ^{54}Fe , $a=0.39326(5)$ nm, $c=0.8491(2)$ nm for $\text{SmFeAsO}_{0.85}\text{F}_{0.15}$ with ^{56}Fe , respectively. The lattice constants are the same within experimental error.

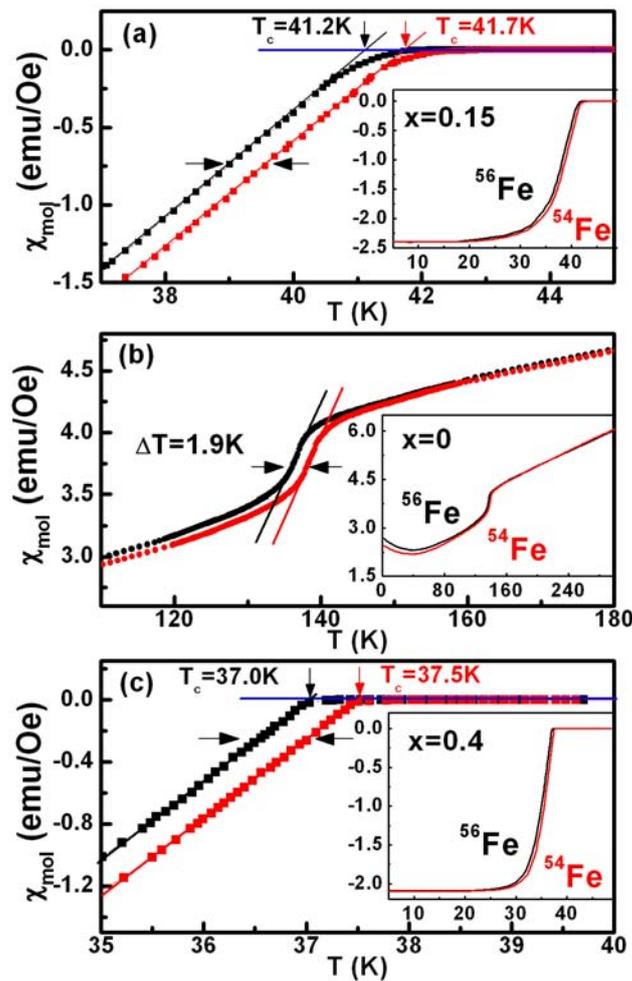


Figure S2: Temperature dependence of susceptibility for the typical samples of $\text{SmFeAsO}_{0.85}\text{F}_{0.15}$ and $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ isotopically substituted with ^{56}Fe and ^{54}Fe . (a): $\text{SmFeAsO}_{0.85}\text{F}_{0.15}$; (b): $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ with $x=0$; (c): $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ with $x=0.4$. To confirm the results from resistivity measurements, the susceptibility of the same samples used in Figure 3 is measured. The shift of T_C and T_{SDW} determined from susceptibility measurements is consistent with that obtained from the resistivity measurements as shown in Figure 3. The susceptibility of superconducting samples $\text{SmFeAsO}_{0.85}\text{F}_{0.15}$ and $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ is measured in the zero-field cooling process under the magnetic field of 10 Oe. The susceptibility of parent compound BaFe_2As_2 is measured in the field-cooled process under the magnetic field of 50000 Oe.

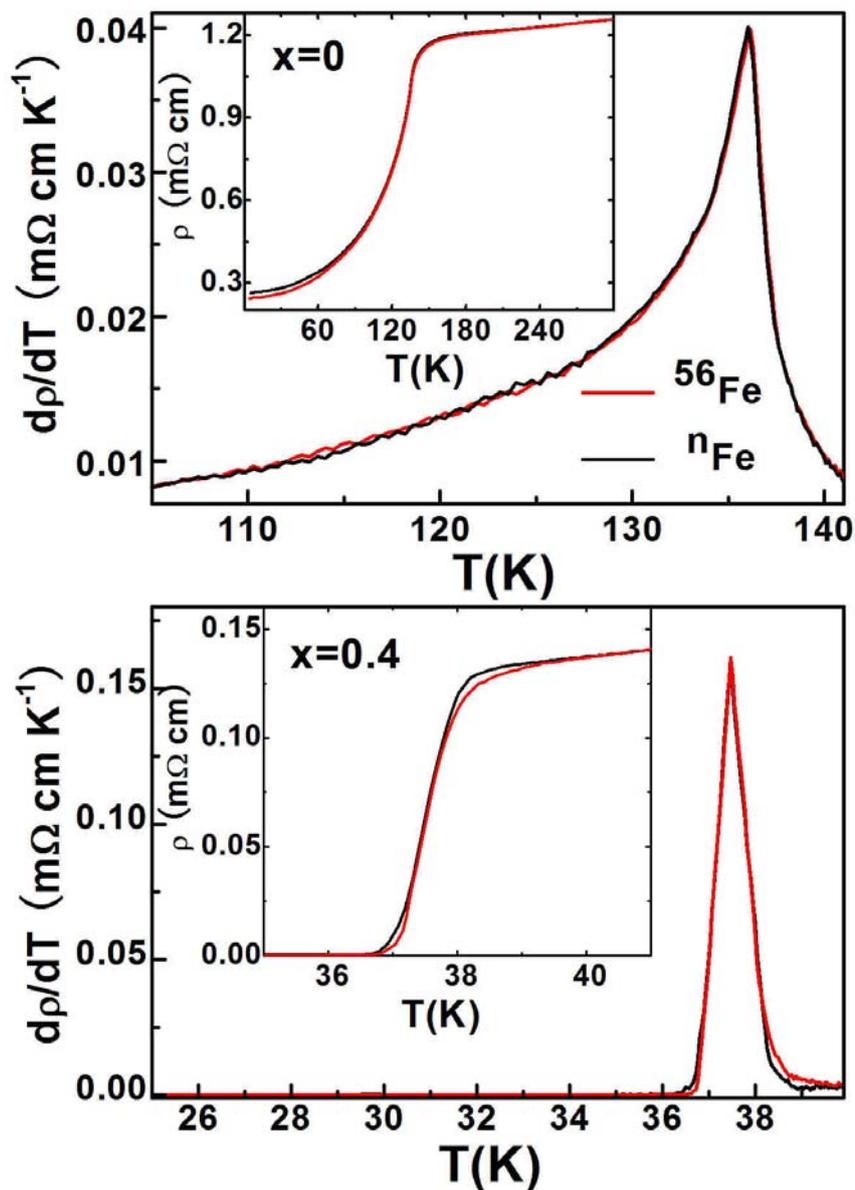


Figure S3: Temperature dependence of resistivity (ρ) and its derivative ($d\rho/dT$) for the samples $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ($x=0$ and 0.4) with nature abundance iron $^{\text{n}}\text{Fe}$ and ^{56}Fe . The natural abundance iron $^{\text{n}}\text{Fe}$ with a 91.8% enrichment of ^{56}Fe and purity of 99.9%; the enrich iron ^{56}Fe with a 97%+ enrichment and purity of 99.78%. It clearly shows that no apparent change in T_{SDW} and T_{C} is observed in the samples with nature abundance iron $^{\text{n}}\text{Fe}$ and enriched iron ^{56}Fe .

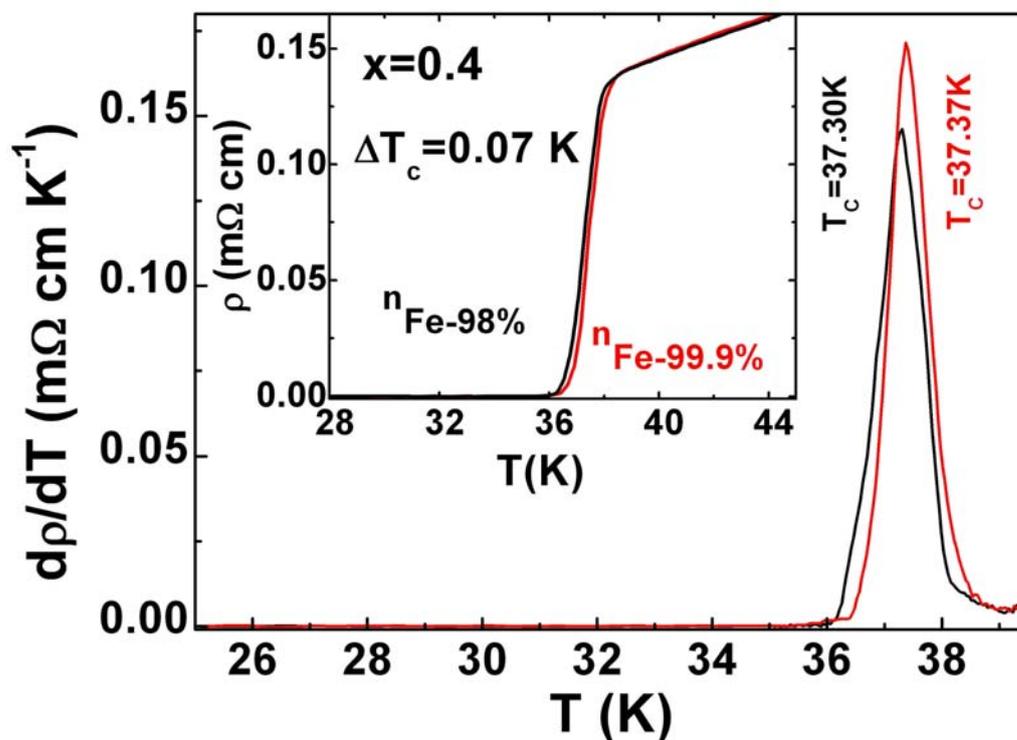


Figure S4: Temperature dependence of resistivity (ρ) and its derivative ($d\rho/dT$) for the samples $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ with nature abundance irons ${}^{\text{n}}\text{Fe}$ with purity of 98% and 99.9%, respectively. To check the effect of impurity on T_c , the samples with different purity of iron ${}^{\text{n}}\text{Fe}$ are studied. It clearly shows that the shift of T_c for the two samples with different purity of iron is about 0.07 K. It indicates that the effect of impurity on T_c is very little. In addition, the purity of isotope iron used in our work is 99.78% for ${}^{56}\text{Fe}$ and 99.86% for ${}^{54}\text{Fe}$, so that the change of T_c from impurity effect should be much less than 0.07 K due to smaller purity difference. Therefore, the effect of impurity on T_c can be ignored, and it further indicates that the isotope effect reported here is intrinsic.