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Superconductivity induced by Ni doping in BaFe₂As₂ single crystals

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Abstract. A series of 122 phase BaFe_{2-x}Ni_xAs₂ ($x = 0, 0.055, 0.096, 0.18$ and 0.23) single crystals was grown by the self-flux method and a dome-like Ni doping dependence of the superconducting transition temperature was discovered. The transition temperature T_c^{on} reaches a maximum of 20.5 K at $x = 0.096$, and it drops to below 4 K as $x \geq 0.23$. The negative thermopower in the normal state indicates that the electron-like charge carrier indeed dominates in this system. This Ni-doped system provides another example of superconductivity induced by electron doping in the 122 phase.

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1. Introduction

Following the discovery of superconductivity at a T_c of 26 K in F-doped LaFeAsO [1], the so-called 1111 phase family of high- T_c superconductors with the ZrCuSiAs-type structure, i.e. LnFeAsO $_{1-x}$ F $_x$ [2]–[8], LnFeAsO $_{1-\delta}$ [9] and Ln $_{1-x}$ Th $_x$ FeAsO [10, 11] (where Ln = La, Ce, Pr, Nd, Sm, Gd and Tb), has been reported. Similar to the cuprate high-temperature superconductors, the (Fe $_2$ As $_2$) layers are conducting layers and essential to the occurrence of superconductivity, whereas the (Ln $_2$ O $_2$) layers inject charge carriers into the former via chemical doping and also retain the structural integrity of the (Fe $_2$ As $_2$) layers. Furthermore, a 122 phase family of high- T_c superconductors AFe $_2$ As $_2$ (A = Ca, Sr and Ba) [12]–[16] with ThCr $_2$ Si $_2$ -type structure (space group $I4/mmm$), which possess the same (Fe $_2$ As $_2$) layers as LnFeAsO but separated by simple A-layers, has been discovered. Superconductivity in the so-called 111 phase compound LiFeAs with a similar structure has also been reported [17]. In the 1111 phase family, superconductivity is mostly induced by electron doping, i.e. partial substitution of O by F, Ln by Th, or O-vacancy although superconductivity with a T_c of 25 K has indeed been reported in hole-doped La $_{1-x}$ Sr $_x$ FeAsO [18]. In contrast, superconductivity is mostly induced by hole doping in the 122 phase, i.e. partial substitution of Ca, Ba or Sr by K or Na, etc. No superconductivity has been observed by means of A-site electron doping, such as the partial substitution of divalent ions (Ba $^{2+}$ or Sr $^{2+}$) by trivalent ions (La $^{3+}$) [15].

In contrast to high- T_c cuprates, superconductivity can also be induced by partial substitution of Fe in the conducting layers by other transition metal elements like Co [19, 20] and Ni [21] in the 1111 phase, and measurements of the Hall effect and thermopower indicate that the electron-type charge carriers dominate in the Co-doped case. For the 122 phase, superconductivity with T_c as high as 25 K has also been observed in Co-doped BaFe $_{2-x}$ Co $_x$ As $_2$ [22] and SrFe $_{2-x}$ Co $_x$ As $_2$ [23] systems. As implied by the negative thermopower value in the normal state [15, 20, 24], Co doping appears to donate the extra electrons into Fe layers as itinerant charge carriers, and thus induces superconductivity for either the 1111 phase or the 122 phase. In this paper, we report that superconductivity is induced in the Ni-doped BaFe $_{2-x}$ Ni $_x$ As $_2$ single crystals. A dome-like Ni doping dependence of T_c is established and the highest T_c^{on} of 20.5 K is realized at the optimal doping level $x = 0.1$. Compared with the Co-doped 122 phase, the superconducting window is narrower and the optimal Ni doping concentration is only about half. This Ni-doped system provides another example of superconductivity induced by electron doping in the 122 phase.

2. Experimental

Single crystals with nominal formula BaFe $_{2-x}$ Ni $_x$ As $_2$ ($x = 0, 0.05, 0.1, 0.16$ and 0.2) were prepared by the self-flux method [22]. All the starting materials, Ba rods, Fe powder, Ni powder and As pieces, are of high purity ($\geq 99.99\%$). First, FeAs and NiAs binaries were prepared by reacting Fe/Ni powder and As powder in an evacuated silicon tube. They were heated slowly to 873 K, kept for 10 h, and then cooled to room temperature. A ratio of Ba : FeAs/NiAs = 1 : 5 was used and the extra FeAs material acted as the flux. The mixtures of Ba and FeAs/NiAs were loaded in finely designed thin corundum crucibles. All these processes were carried out in a glove box filled with high-purity argon. Then the crucibles were sealed in evacuated quartz tubes. Finally, the quartz tubes filled with the mixtures were slowly heated to 973 K, held for 5 h and then heated to 1453 K, and held for 10 h. The temperature was decreased to

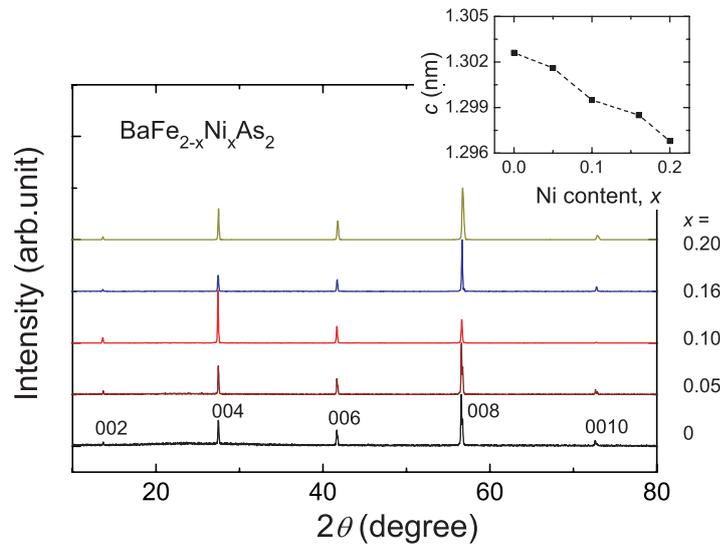


Figure 1. X-ray diffraction pattern at room temperature for $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ single crystals. The inset shows the variation of the c -axis lattice constant with x , where the x values are only the nominal Ni contents according to starting materials.

1363 K at a rate of -2 K h^{-1} , followed by decanting the quartz tube at 1363 K, and finally cooled down to room temperature slowly. Single crystals with a diameter of about 5 mm were obtained.

X-ray diffraction (XRD) was performed at room temperature using a D/Max-rA diffractometer with $\text{Cu-K}\alpha$ radiation and a graphite monochromator. Lattice parameters were refined by a least-squares fit. Chemical analysis by energy-dispersive x-ray (EDX) microanalysis was performed on an EDAX GENESIS 4000 x-ray analysis system affiliated to a scanning electron microscope (SEM, model SIRION). For each doped single crystal, EDX microanalysis was performed at more than three different spots to check the distribution of Ni content. The electrical resistivity was measured using a standard four-probe method. The temperature dependence of dc magnetization was measured on a Quantum Design magnetic property measurement system (MPMS-5). The thermopower was measured by a steady-state technique.

3. Results and discussion

The XRD patterns of $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ crystals are shown in figure 1. Only $(00l)$ reflections appear, indicating that the c -axis is perpendicular to the cleaved surface. The $(00l)$ reflections with only even l were observed because the system belongs to the body-centered space group. The c -axis lattice constant was calculated as 1.303 nm for the undoped parent compound BaFe_2As_2 , which is consistent with previous reports [25]. The inset of figure 1 shows the variation of the c -axis lattice constant with nominal Ni content x . The c -axis lattice constant decreases monotonically with increasing x . The EDX measurements show that the actual average Ni content (x) is slightly different from the nominal composition, but the variation of

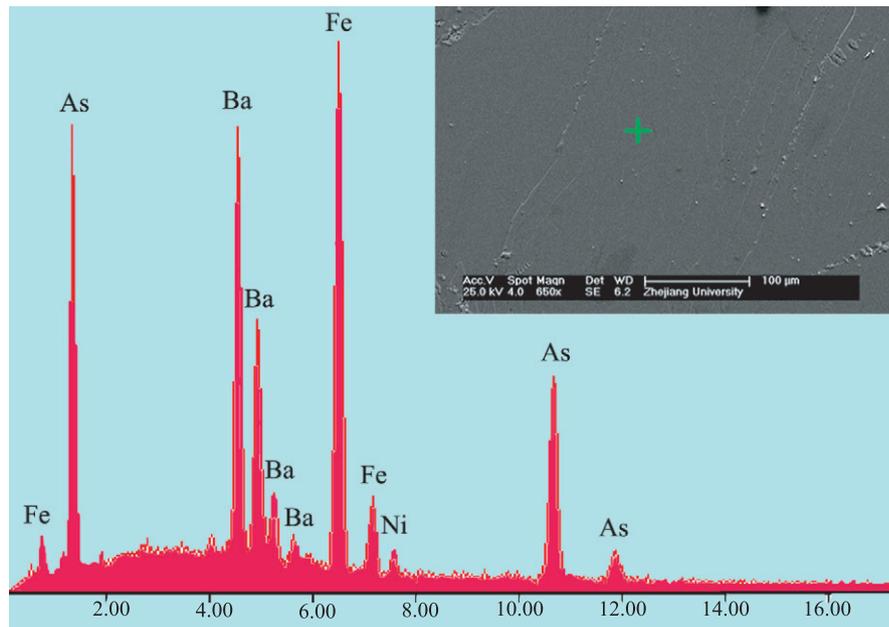


Figure 2. A representative EDX spectrum for a single crystal with a nominal x of 0.1. The inset shows the SEM photo of this sample. The scale at the bottom of the photo is $100\ \mu\text{m}$. The actual Ni content in this sample is between 0.092–0.099 according to the EDX spectra.

Ni content is less than 5% in each sample except for the sample with a nominal x of 0.05, which indicates that the distribution of Ni doping is quite uniform in these samples. A representative EDX spectrum for a single crystal with a nominal x of 0.1 is shown in figure 2 and the inset shows a SEM photo of this sample. According to the EDX reports, the average x value is 0.055, 0.096, 0.18 and 0.23 for the samples with the nominal x of 0.05, 0.1, 0.16 and 0.2, respectively.

Figure 3 shows the temperature dependence of in-plane resistivity. Similar to the previous reports [15, 25], there is an anomalous decrease in the resistivity of the parent compound around a T_{an} of 140 K, which is associated with a structural phase transition. Neutron studies have confirmed that the spin-density wave order occurs simultaneously with the structural phase transition in the 122 phase [26]–[28]. The resistivity anomaly is shifted to a lower temperature, about 90 K, for the $x = 0.055$ sample. However, the resistivity exhibits a sharp increase instead of a decrease around T_{an} . Such an anomalous increase in the resistivity has also been observed in other doped 122 phases [23]. With further decreasing temperature, a superconducting transition occurs below T_{c}^{on} (defined as the onset point in the resistive transition) of 12.5 K for the same $x = 0.055$ sample, but zero resistivity cannot be reached even for T as low as 4 K. It is hard to distinguish whether there is a microscopic phase separation (inhomogeneous distribution of Ni content) or a co-existence of SDW order and superconducting order according to current measurements. With increasing Ni content, the resistivity anomaly disappears. At the optimal doping level $x = 0.096$, T_{c}^{on} reaches a maximum of 20.5 K, $T_{\text{c}}^{\text{mid}}$ is 20.2 K and the transition width ΔT_{c} is less than 1.0 K, suggesting a sharp superconducting transition. The temperature dependence of susceptibility as shown in figure 3(b) also indicates very sharp superconducting transitions for $x = 0.096$ and 0.18. However, the superconducting transition

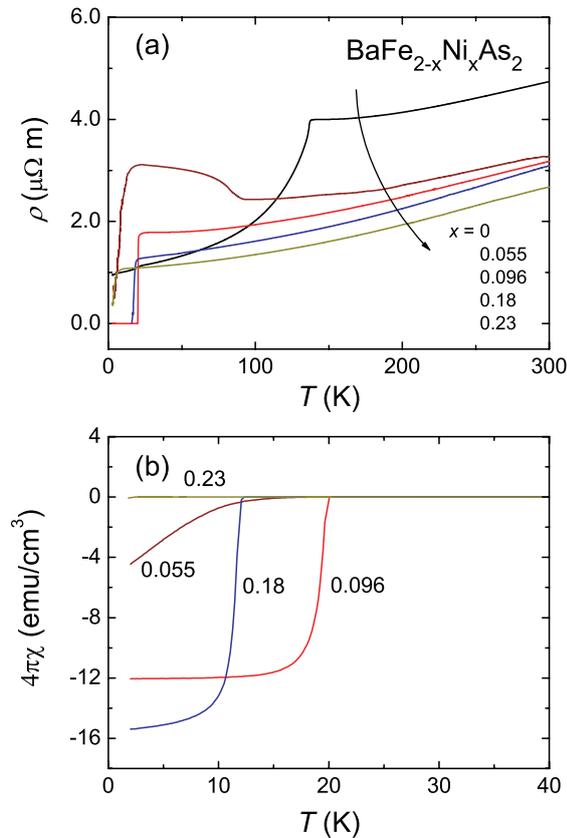


Figure 3. (a) Temperature dependence of in-plane resistivity and (b) temperature dependence of magnetic susceptibility for $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ single crystals. The susceptibility was measured under zero-field cooling (ZFC) condition and the applied magnetic field H is 10 Oe along the c -axis direction. Because of the extremely large demagnetization factor of the samples, the observed superconducting diamagnetic volume fraction is obviously larger than 100%.

in magnetic susceptibility of the $x = 0.055$ sample is rather broad, which means that the distribution of Ni dopants could be very inhomogeneous in this sample.

Figure 4 shows the variation of T_c^{on} and T_c^{mid} with Ni content x . In the superconducting window ($0.055 \leq x \leq 0.23$), a dome-like $T_c(x)$ curve is roughly established, similar to that of the Ni-doped 1111 phase. It should be noted that the optimal doping level $x = 0.096$ corresponds to about 5% of Fe substituted by Ni in the 122 phase, which is very close to the optimal Ni doping level in the 1111 phase (4% of Fe substituted by Ni) [21]. However, compared to the Co-doped 1111 and 122 phases, the superconducting window in Ni-doped system is much narrower, and the optimal Ni doping concentration (about 5% Ni content) is only about half of that of Co-doped systems [20, 24]. Compared to Fe^{2+} ion, Co^{2+} ($3d^7$) has one more 3d electron, but Ni^{2+} ($3d^8$) has two more 3d electrons. Thus it is expected that each Ni dopant induces two extra itinerant electrons, whereas each Co dopant induces only one extra itinerant electron. Thus, the fact that the optimal doping content of Ni is only about half of that of Co can be understood. Actually, both Ni-doped and Co-doped systems show maximum T_c at the same effective doping level. A universal doping (charge carrier concentration) dependence of T_c has

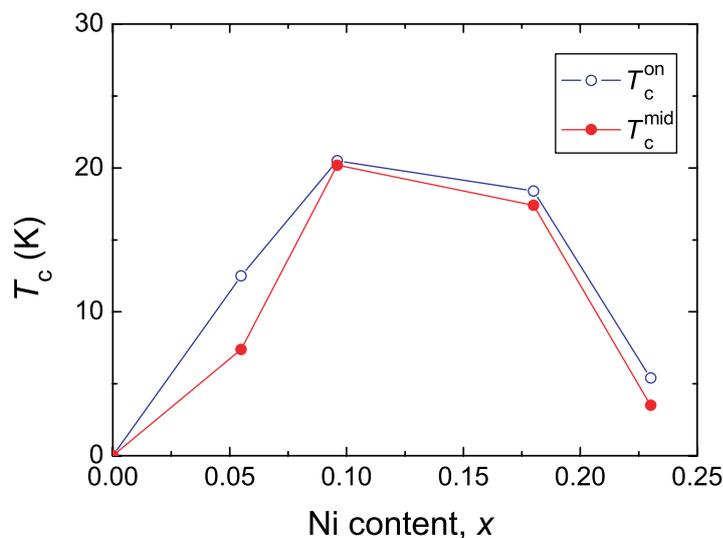


Figure 4. Variations of T_c^{on} and T_c^{mid} with Ni content x for $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ single crystals.

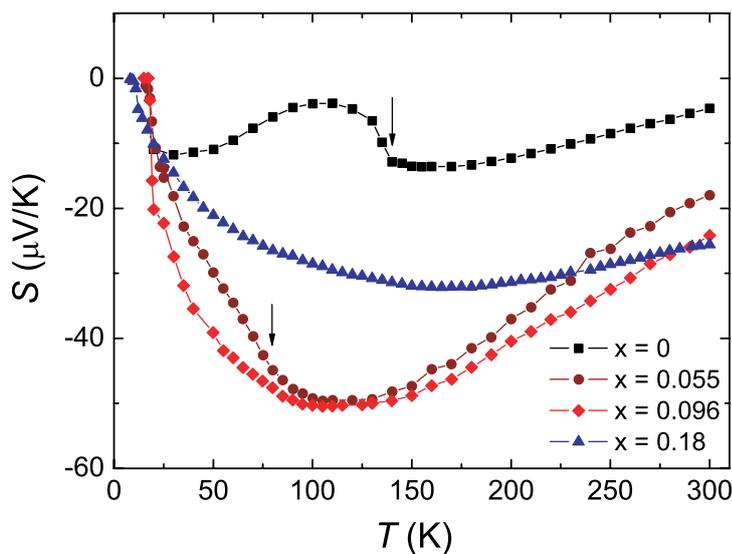


Figure 5. Temperature dependence of thermopower (S) for $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ single crystals. The arrows indicate the anomaly in thermopower associated with structural phase transition and/or SDW ordering.

been established in high- T_c superconducting cuprates. Our result implies that there might exist a universal doping dependence of T_c in iron-based arsenide superconductors. For the higher Ni-doping levels ($x \geq 0.23$), superconductivity is no longer observed for $T > 4$ K, but the resistivity becomes even more metallic. It should be noted that the other end member, BaNi_2As_2 , is a superconductor with a T_c of only about 0.7 K [29]. However, the first-principles calculations suggested that BaNi_2As_2 could be a conventional phonon-mediated superconductor [30].

Figure 5 plots the thermopower (S) as a function of temperature. All the samples show negative thermopower, which means that electron-type charge carriers dominate. For the

undoped parent compound ($x = 0$), thermopower exhibits an anomalous enhancement just below the structural phase transition temperature T^{on} of about 140 K. Similar behavior has been observed in undoped parent compounds such as LaFeAsO [31], TbFeAsO [11] and SmFeAsO [24]. With Ni doping, this anomaly is suppressed quickly. For $x = 0.055$, only a slight enhancement can be observed below about 80 K (indicated by the arrow in figure 5). Meanwhile, the absolute value of normal state thermopower ($|S|$) increases remarkably with increasing Ni content, reaches a maximum at the optimal doping level $x = 0.096$, and then decreases with further Ni doping. It has been suggested that there is a correlation between the normal state thermopower and T_c according to the studies on the thermopower of the Co-doped SmFeAsO system [24]. In the Ni-doped 122 phase, such a correlation should also exist, although the absolute value of thermopower is much smaller than that of F-doped 1111 phase and Co-doped 1111 and 122 phases.

4. Conclusion

In conclusion, a series of Ni-doped 122 phase single crystals $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ ($x = 0, 0.055, 0.096, 0.18$ and 0.23) was successfully synthesized by the self-flux method, and superconductivity with T_c values as high as 20 K was observed. The thermopower is negative in the normal state, suggesting that the electron-like charge carrier dominates. This Ni-doped system provides another example of superconductivity induced by electron doping in the 122 phase.

Acknowledgments

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