Heat capacity jump at T_c and pressure derivatives of superconducting transition temperature in the Ba_{1-x}Na_xFe₂As₂ (0.1 $\leq x \leq 0.9$) series

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We present the evolution of the initial (up to ~10 kbar) hydrostatic pressure dependencies of T_c and of the ambient pressure, and the jump in the heat capacity associated with the superconducting transition as a function of Na doping in the Ba_{1-x}Na_xFe₂As₂ family of iron-based superconductors. For Na concentrations $0.15 \le x \le 0.9$, the jump in specific heat at T_c , $\Delta C_p |_{T_c}$, follows the $\Delta C_p \propto T^3$ (the so-called BNC scaling) found for most BaFe₂As₂-based superconductors. This finding suggests that, unlike the related Ba_{1-x}K_xFe₂As₂ series, there is no significant modification of the superconducting state (e.g., change in superconducting gap symmetry) in the Ba_{1-x}Na_xFe₂As₂ series over the whole studied Na concentration range. Pressure dependencies are nonmonotonic for x = 0.2 and 0.24. For other Na concentrations, T_c decreases under pressure in an almost linear fashion. The anomalous behavior of the x = 0.2 and 0.24 samples under pressure is possibly due to the crossing of the phase boundaries of the narrow antiferromagnetic tetragonal phase, unique for the Ba_{1-x}Na_xFe₂As₂ series, with the application of pressure. The negative sign of the pressure derivatives of T_c across the whole superconducting dome (except for x = 0.2) is a clear indication of the nonequivalence of substitution and pressure for the Ba_{1-x}Na_xFe₂As₂ series.

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I. INTRODUCTION

Of many recently discovered Fe-based superconductors and related materials [1–3], the so called 122 family ($AEFe_2As_2$, with AE = alkaline earth and Eu) is the most studied [4–6]. This family allows for substitution on all three cystallographic sites, and, as a result, an intricate combination of carrier doping and anisotropic steric effects can be studied, while maintaining the same, tetragonal, ThCr₂Si₂-type crystal structure.

The main body of the published work on the 122 family focused on the $AE(Fe_{1-x}TM_x)_2As_2$ series with transition metals, TM, being substituted for Fe [4,5]. This is due to the relative ease of growing homogeneous, high-quality single crystals. Substitutions for AE, as in the $Ba_{1-x}K_xFe_2As_2$ series [7–10], or As, as in the $BaFe_2(As_{1-x}P_x)_2$ series [11,12] have been explored, but both series require significant efforts to achieve homogeneity and/or reasonably sized crystals.

At first glance, substitution for Ba $(Ba_{1-x}K_xFe_2As_2)$, Fe $[Ba(Fe_{1-x}Co_x)_2As_2]$, or As $[BaFe_2(As_{1-x}P_x)_2]$ in BaFe₂As₂, as well as application of pressure [13] to this parent compound, results in similar phase diagrams [14]. First, the temperature of structural and magnetic transitions decreases, and then superconductivity emerges with a region of coexistence of superconductivity and antiferromagnetism. Upon further substitution (or under higher pressure), the magnetic and structural transitions are suppressed, and the superconducting transition temperature passes through the maximum and gradually goes to zero or to a small finite value as in the case of complete substitution of K for Ba in KFe₂As₂. A closer examination of the globally similar phase diagrams, however, points to clear differences in the details, enabling us to gain insight into the complex physics of these materials [2,4–6].

Na substitution for AE appears to be one of the less explored branches in the 122 family tree. A possibility to induce superconductivity by Na substitution in CaFe₂As₂ was realized fairly early [15,16], whereas a tentative *x*-*T* phase diagram for the Ca_{1-x}Na_xFe₂As₂ series was published a few years later [17]. A Sr_{1-x}Na_xFe₂As₂ sample with a *T_c* value as high as \approx 35 K was studied in Ref. [18], and later an evolution of the physical properties in Sr_{1-x}Na_xFe₂As₂ (for a rather limited range of substitution, *x* \leq 0.4) was presented [19].

The Ba_{1-x}Na_xFe₂As₂ series, where Na is substituted for Ba, offers an almost complete range of substitution [20,21]. One of the complications for this series is that its end member, NaFe₂As₂ ($T_c \sim 11-12$ K), was reported to be metastable, and as such it cannot be formed by a solid-state reaction technique, but can only be obtained by the mild oxidation of NaFeAs [22–24]. Additionally, deviations from stoichiometry (Na_{1-y}Fe_{2-x}As₂, with $y \approx 0.1$ and $x \approx 0.3$) for the obtained material were suggested [24].

In generic terms, for the overlapping x values, the x-T phase diagram of the $Ba_{1-x}Na_xFe_2As_2$ series [21] bears a close similarity to that of the $Ba_{1-x}K_xFe_2As_2$ series [8,10], but a comprehensive study of the Na-doped series and a direct comparison with the results for its K-doped counterpart would help to address several issues of relevance for the physics of Fe-based superconductors. First, in the $Ba_{1-x}K_xFe_2As_2$ series, several experimental observations point to a significant modification of the superconducting state (possibly a change in the superconducting pairing symmetry) for K concentration, x > 0.7 [25–29]. It would be of interest to examine the overdoped part of the phase diagram of the $Ba_{1-x}Na_xFe_2As_2$ series for similar features. Second, in addition to the lowtemperature antiferromagnetic/orthorhombic phase, which is ubiquitous in Fe-based superconductors, an antiferromagnetic tetragonal, C4, phase was reported in the $Ba_{1-x}Na_xFe_2As_2$ series over a narrow Na-concentration region around $x \sim$ 0.24 [21,30]. The tip of the narrow C4 dome was suggested to be at \sim 50 K; at lower temperatures, this new magnetic phase was suggested to coexist with superconductivity. The bulk physical properties of the members of the series close to and in the C4 phase as well as the effect of this magnetic phase on superconductivity are interesting to study in detail.

In this work, we utilize the same approach as we did in the recent studies of the $Ba_{1-x}K_xFe_2As_2$ series [27]; we present two sets of data for the $Ba_{1-x}Na_xFe_2As_2$ series, with Na concentrations covering underdoped, optimally doped, and overdoped regions of the x-T phase diagram. The first set consists of the data on the evolution of the jump in heat capacity at the superconducting transition. Many Fe-based, 122 superconductors follow the empirical trend suggested in Ref. [31] and expanded in Refs. [32] and [33], the so-called BNC scaling, $\Delta C_p|_{T_c} \propto T_c^3$. This set is to be compared with the data on the $Ba_{1-x}K_xFe_2As_2$ series [27], which show a clear deviation from the BNC scaling for K concentration x > 0.7. The second set contains the initial ($P \lesssim 10$ kbar) pressure dependencies of the superconducting transition temperatures, $T_c(P)$, similar in scope to the data reported for $Ba(Fe_{1-x}Co_x)_2As_2$ [34] and $Ba_{1-x}K_xFe_2As_2$ [27]. Such data evaluate the possible equivalence of pressure and doping that was suggested for several 122 series [35-38]. Moreover, under favorable circumstances, such a dataset can shed light on the details of the mechanism of superconductivity [39–41].

II. EXPERIMENTAL DETAILS

Homogeneous, single-phase $Ba_{1-x}Na_xFe_2As_2$ polycrystalline powder with nominal x = 0.1, 0.15, 0.2, 0.24, 0.3, 0.35, 0.4, 0.5, 0.6, 0.7, 0.8, and 0.9 were synthesized by a variation of a previously reported procedure [21]. Handling of all starting materials was performed in an Ar-filled glove box. Mixtures of Ba, Na, and FeAs were loaded in alumina crucibles, which were sealed in Nb tubes under argon, and then sealed again in fused silica tubes under vacuum. The mixtures were heated twice at 800 °C for 1 and 3 days, respectively, and at 850 °C for 1 day, with quenching in air. Between each annealing cycle, the mixtures were ground to a fine, homogeneous powder. After the third annealing step, the powder was pressed into a pellet, reloaded into the alumina crucible, sealed first inside the Nb tube, and then into a fused silica tube. The pellet was heated at 850 °C for approximately 1 day before quenching in air. The superconducting transition temperatures of the samples, as determined by magnetization measurements, were compared with those in the previously reported x-T phase diagram [21] in order to confirm the x values of the compositions.

samples synthesis of homogeneous The of $Ba_{1-x}Na_xFe_2As_2$ is challenging due to the volatility of Na upon heating. For this reason, sealed Nb tubes are utilized to suppress and contain Na vaporization at high temperature. This is particularly crucial in the highly underdoped region (x < 0.3) where small changes in x can lead to large changes in T_c . Because of the sensitivity of T_c in the highly underdoped samples with respect to Na content, the pellets of these samples needed to be annealed multiple times for shorter durations in order to achieve sharp superconducting transitions. Additionally, for the overdoped samples, a small amount of Na was added to the pelletized mixture before the final annealing in order to finely tune the x parameter. The most highly overdoped sample, Ba_{0.1}Na_{0.9}Fe₂As₂, is also difficult to prepare due to its tendency to phase-segregate into an underdoped composition with higher thermal stability and a stable ternary compound NaFeAs (with a different structure type) [42]. It should be noted here that the end-member NaFe₂As₂ (x = 1) cannot be formed by this solid-state technique; instead, it can be obtained by the mild oxidation of NaFeAs [22–24].

Low-field dc magnetization under pressure was measured in a Quantum Design Magnetic Property Measurement System (MPMS-5) SQUID magnetometer using a commercial, HMD, Be-Cu piston-cylinder pressure cell [43]. Daphne oil 7373 was used as a pressure medium and superconducting Pb or Sn (to have its superconducting transition well separated from that of the sample) as a low-temperature pressure gauge [44]. The heat capacity was measured using a hybrid adiabatic relaxation technique of the heat capacity option in a Quantum Design Physical Property Measurement System (PPMS-14) instrument.

III. RESULTS

A. Jump in specific heat and BNC scaling

Of the samples studied in this work, Ba_{0.9}Na_{0.1}Fe₂As₂ is not superconducting whereas Ba_{0.15}Na_{0.85}Fe₂As₂ is, but low-field, zero-field-cooled, magnetization measurements display two transitions. The x = 0.1 sample is not included in the analysis of the jump in specific heat at T_c . For x = 0.15, the feature at ~ 5 K is the only feature present in the specific heat data. For this sample, the difference between the data taken in zero field and 140 kOe applied field was analyzed, and the error bars in the obtained ΔC_p at T_c value are expected to be rather large. The other samples studied in this work show a distinct feature in specific heat at T_c (see Fig. 1 as an example). The T_c and $\Delta C_p|_{T_c}$ values were determined by a procedure consistent with that used in Refs. [27,31]. The specific heat jump data for the $Ba_{1-r}Na_rFe_2As_2$ series obtained in this work were added in Fig. 2 to the BNC plot taken from Ref. [27] and updated by including several more recent data points taken



FIG. 1. (Color online) Temperature-dependent heat capacity of Ba_{0.2}Na_{0.8}Fe₂As₂ near the superconducting transition plotted as C_p/T vs T. Criteria for T_c and $\Delta C_p|_{T_c}$ (isoentropic construct) are shown. Inset: $C_p(T)$ in a wider temperature range.



FIG. 2. (Color online) ΔC_p at the superconducting transition vs T_c for the Ba_{1-x}Na_xFe₂As₂ series, plotted together with literature data for various FeAs-based superconducting materials. The plot from [27] was updated to include published data for K_{1-x}Na_xFe₂As₂ ($0 \le x \le 0.31$), Ca_{1-x}Na_xFe₂As₂, Ba_{1-x}Na_xFe₂As₂ (x = 0.35, 0.4), and LaFeAs_{0.9}F_{0.1} [17,45–48]. The line corresponds to $\Delta C_p \propto T_c^3$. Numbers near the symbols are Na concentrations x.

from the evolving literature. There appears to be a clear trend: all data points $(0.15 \le x \le 0.9)$ from this study follow the BNC scaling, in agreement with the literature data for two close Na concentrations [45,46]. This behavior is clearly different from that reported in the Ba_{1-x}K_xFe₂As₂ series, where the data for K concentrations of $0.8 \le x \le 1.0$ clearly deviate from the BNC scaling, and it is similar to the previously studied Ba(Fe_{1-x}TM_x)₂As₂ (TM = transition metal) series, for which the BNC scaling was observed for the samples covering the full extent of the superconducting dome. For the Ba_{1-x}Na_xFe₂As₂ series, ΔC_p at T_c increases and decreases as T_c rises and falls to form the superconducting dome.

B. Pressure dependence of T_c

An example of M(T) data taken at different pressures is shown in Fig. 3. An onset criterion was used to determine T_c . Alternative criteria (e.g., maximum in dM/dT) yield similar pressure dependencies. The Ba_{0.9}Na_{0.1}Fe₂As₂ sample was measured at ambient pressure and at 11.1 kbar. No traces of superconductivity down to 1.8 K were observed. The pressure dependencies of the superconducting transition temperatures of the samples with Na concentration in the range of $0.15 \le x \le 0.9$ are shown in Fig. 4. For the $Ba_{1-x}Na_xFe_2As_2$ sample with Na concentration of x =0.15, the pressure dependence associated with the lowertemperature feature, which is consistent with the ambient pressure phase diagram, is presented [21]. It is noteworthy that (i) $T_c(P)$ are nonmonotonic (even in a limited pressure range of this work) for two Na concentrations, x = 0.2 and 0.24; (ii) for all other concentrations studied in this work, both in underdoped and overdoped regimes, T_c decreases under pressure. The data for $Ba_{1-x}Na_xFe_2As_2$ samples with x = 0.2 and 0.24 are shown separately in Figs. 5(a) and 5(b).



FIG. 3. (Color online) Example of temperature-dependent magnetization (zero-field-cooled, taken in 25 Oe applied magnetic field) of Ba_{0.7}Na_{0.3}Fe₂As₂ measured at 1.1, 2.9, 5.9, and 10.7 kbar. The arrow indicates increasing pressure. The onset criterion for T_c used in this work is shown for P = 10.7 kbar curve as an example. Superconducting transitions in Pb used as a pressure gauge are seen near 7 K.

Two pressure runs on two different samples from the same batch were performed for $Ba_{0.8}Na_{0.2}Fe_2As_2$. The results are fairly consistent. For both the x = 0.2 and 0.24 samples, the measurements were performed upon pressure increase and pressure decrease [Figs. 5(a) and 5(b)]. For both samples, the observed nonmonotonic behavior is robust and not affected by pressure cycling. One can join these two data sets by shifting the data for $Ba_{0.76}Na_{0.24}Fe_2As_2$ by +8 kbar along the X axis and by -4 K along the Y axis. We can understand the grounds for such a two-axis shift if we assume that both the steric effect and hole doping cause changes in the superconducting transition temperature.

A more compact way to look at the pressure dependence of T_c in the Ba_{1-x}Na_xFe₂As₂ series is presented in Fig. 6. The superconducting transition temperature values obtained from specific heat and magnetization measurements are very similar, and these data sets are consistent with the literature [21]. The initial low-pressure value of dT_c/dP [and $d(\ln T_c)/dP$]) for x = 0.2 is positive and relatively high. For other Na concentrations studied, the pressure derivatives of T_c are negative. Whereas for the optimally doped and overdoped regions the absolute values of dT_c/dP and $d(\ln T_c)/dP$ are rather small and change smoothly with concentration (Fig. 6), there appears to be a break in the trend in the underdoped region.

Figure 7 presents a comparison of the relative changes in the superconducting transition temperature under pressure and with Na doping. For $0.4 \le x \le 0.9$, both sets of data can be scaled reasonably well, illustrating the apparent equivalence of the effect of pressure and doping on T_c , suggested for other members of the 122 family [35–38] and also observed in the limited range of K concentrations for a closely related Ba_{1-x}K_xFe₂As₂ [27]. This scaling, however, fails for Na concentrations $0.15 < x \le 0.35$. Not only can the values of $d(\ln T_c)/dP$ and $d(\ln T_c)/dx$ not be scaled in this region of



FIG. 4. (Color online) Summary plot of the pressure dependence of T_c for the Ba_{1-x}Na_xFe₂As₂ (0.15 $\leq x \leq 0.9$) series studied in this work. Dashed lines are linear fits to the data except for x = 0.2 and 0.24, where they are a guide for the eye. The linear fits are extended to P = 0.

concentrations, but (except for x = 0.2) the signs of these derivatives are different. In the underdoped region, an increase in x causes an increase in T_c , and pressure causes a decrease in T_c ; however, in the optimally doped and overdoped regions, both an increase in x and pressure cause a decrease in T_c (Fig. 7, inset).

IV. DISCUSSION AND SUMMARY

Both K and Na substitutions in BaFe₂As₂ provide hole doping and induce superconductivity with comparable maximum values of T_c of 34–38 K at similar K or Na concentrations of $x \approx 0.4$. Whereas in the Ba_{1-x}K_xFe₂As₂ series a clear deviation from the BNC scaling is observed for $0.8 \le x \le 1$ [27], in the Ba_{1-x}Na_xFe₂As₂ series the data for $0.15 \le x \le$ 0.9 follow the BNC scaling, $\Delta C_p|_{T_c} \propto T_c^3$, fairly well. This probably means that either there is no significant modification of the superconducting state (e.g., a change in superconducting gap symmetry) in the Ba_{1-x}Na_xFe₂As₂ series over the whole studied Na concentration range, or, if such modification exists, it is very subtle in its implications for the BNC scaling. The fact that the Ba_{1-x}Na_xFe₂As₂ series does not extend, in single-phase form, to x = 1.0 prevents us from carrying this study to pure NaFe₂As₂, as we were able to do for KFe₂As₂.

The negative sign of the pressure derivatives of T_c for the underdoped samples (except for x = 0.2) is a clear indication of the nonequivalence of substitution and pressure for the Ba_{1-x}Na_xFe₂As₂ series in this range, which is



FIG. 5. (Color online) Pressure dependence of the superconducting transition temperature for (a) $Ba_{0.8}Na_{0.2}Fe_2As_2$ and (b) $Ba_{0.76}Na_{0.24}Fe_2As_2$. Different symbols in panel (a) correspond to two different pressure runs. Numbers near the symbols in panels (a) and (b) correspond to the order in which the pressure was changed. Panel (c) shows both data sets with the one for x = 0.24 shifted by +8 kbar along the X axis and by -4 K along the Y axis.

different from the gross overall equivalence suggested for other 122 series [27,35–38]. It has to be noted that for the Ba(Fe_{1-x}TM_x)₂As₂ (TM = transition metal) (at least for TM concentrations that cover the superconducting dome) [49–51]



FIG. 6. (Color online) $d(\ln T_c)/dP$, dT_c/dP , and T_{c0} (top to bottom) as a function of Na concentration in Ba_{1-x}Na_xFe₂As₂. Stars: T_c values from heat capacity measurements. For Ba_{0.8}Na_{0.2}Fe₂As₂ and Ba_{0.76}Na_{0.24}Fe₂As₂, initial, low-pressure, pressure derivative values are used (half-filled hexagons).

and for the $Ba_{1-x}K_xFe_2As_2$ series [8,10], the concentration dependence of the lattice parameters is monotonic and close to linear. For the $Ba_{1-x}Na_xFe_2As_2$ series, the *a* lattice parameter decreases with an increase of *x* in an almost linear fashion, but the *c* lattice parameter initially increases and then



FIG. 7. (Color online) Na-concentration dependence of the normalized concentration derivatives, $d(\ln T_c)/dx = \frac{1}{T_{c0}}dT_c/dx$ (left axis, circles), and the normalized pressure derivatives, $d(\ln T_c)/dP = \frac{1}{T_{c0}}dT_c/dP$ (right axis, triangles), of the superconducting transition temperatures. For Ba_{0.8}Na_{0.2}Fe₂As₂ and Ba_{0.76}Na_{0.24}Fe₂As₂, initial, low-pressure, normalized pressure derivative values are used (halffilled hexagons). Inset: schematic exhibiting *different* signs of the change in T_c with substitution and with pressure for the underdoped samples, and *similar* effects of substitution and pressure for the overdoped samples.

decreases, with a maximum at $x \sim 0.4$ in its dependence of Na concentration [21]. Although we do not know which particular structural parameter in the Ba_{1-x}Na_xFe₂As₂ series has the dominant contribution to the pressure dependence of T_c , this nonmonotonic behavior of c(x) might be responsible for the negative sign of the dT_c/dP for underdoped samples. Detailed structural studies under pressure would be useful for a deeper understanding of this problem.

On the one hand, the unusual nonmonotonic behavior of the superconducting transition temperature under pressure for the $Ba_{1-x}Na_xFe_2As_2$ samples with x = 0.2 and 0.24 (Fig. 5) could be considered to be consistent with what one would expect for a Lifshitz transition [52]. At this moment, the data on the Fermi surface of $Ba_{1-x}Na_xFe_2As_2$ [46] are regarded as very similar to those for $Ba_{1-x}K_xFe_2As_2$ [53,54], and no change of the Fermi surface topology, from the angle-resolved photoemission spectroscopy measurements, has been reported between the parent, $BaFe_2As_2$, compound and $Ba_{1-x}Na_xFe_2As_2$ with x values up to 0.4, so the Lifshitz transition hypothesis seems unlikely.

On the other hand, an important feature, unique to the x-T ambient pressure phase diagram of the Ba_{1-x}Na_xFe₂As₂ series, is the existence of a distinct, narrow antiferromagnetic and tetragonal C4 phase [21]. Both of the samples with nonmonotonic pressure dependences of T_c are located, at ambient pressure, close to the phase boundaries of this C4 phase. It is thus possible that the observed anomalies in $T_c(P)$ behavior for the samples with x = 0.2 and 0.24 are associated with the crossing of these phase boundaries under pressure. If this supposition is correct, the effect of the antiferromagnetic orthorhombic phase that is omnipresent in Fe-As materials, since crossing of the antiferromagnetic orthorhombic phase line under pressure either has no effect on T_c or this effect has previously been missed.

In summary, it appears that the nonmonotonic behavior of the *c* crystallographic lattice parameter and the narrow antiferromagnetic tetragonal *C*4 phase both affect the pressure dependencies of the superconducting transition temperature in the Ba_{1-x}Na_xFe₂As₂ series. Synthesis of homogeneous single crystals with a finely controlled Na concentration around 20–25 % and further comprehensive measurements of superconducting and magnetic properties (and their interplay) in this range of concentrations would be desirable to understand the distinct properties of the Ba_{1-x}Na_xFe₂As₂ series.

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