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Superconducting gap and critical behavior in the Iron-Pnictides

M.M.E. Barakat [a,b], T.A. Abdel-Baset [a,c], M. Belhaj [a], D. El-Said Bakeer [d,e], A.N. Vasiliev [f,g], M. Abdel-Hafiez [h,i,*]

[a] Department of Physics, Faculty of Science, Taibah University, Yanbu, Saudi Arabia
[b] Department of Physics, Faculty of Science, Alexandria University, Alexandria, Egypt
[c] Department of Physics, Faculty of Science, Fayoum University, Fayoum, Egypt
[d] Department of Physics, Faculty of Science and Arts, Taibah University, Al-Ula, Saudi Arabia
[e] Department of Physics, Faculty of Science, Damanhour University, Damnhour, Egypt
[f] Lomonosov Moscow State University, Moscow 119991, Russia
[g] National University of Science and Technology "MISiS", Moscow 119049, Russia
[h] Department of Physics and Astronomy, Uppsala University, Box 516, Uppsala SE-75120, Sweden
[i] Department of Applied Physics and Astronomy, University of Sharjah, P. O. Box 27272, Sharjah, United Arab Emirates

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ABSTRACT

In the phase diagram of iron pnictides, superconductivity arises at the border of antiferromagnetism, which raises the question of the role of symmetry of the gap and quantum criticality. Although more than 15-years of extensive research, the microscopic origin of the pairing symmetry inside the superconducting (SC) dome and its link to quantum criticality still remains elusive. Here, we report two new findings on $BaFe_{2-x}Ni_xAs_2$: (1) A sharp peak in the x-dependence of the lower and upper critical fields, the SC critical current density J_c, the size of the jump in the specific heat $\Delta C_{el}/T$ and the Sommerfeld coefficient (γ) at the optimum composition $x = 0.10$, where the SC transition temperature T_c reaches a maximum. Our obtained reliable values as a function of doping of the normal-state Sommerfeld coefficient increase with doping, illustrating the strong competition between magnetism and superconductivity and attributed to closing of spin density wave gap with Ni doping. (2) We show that doping induced a sudden change of the gap structure from nodeless to nodal. Our results imply that the superconductivity in $BaFe_{2-x}Ni_xAs_2$ is closely linked to the quantum criticality and is characterized by a complex order parameter.

Introduction

In many correlated electron materials, several ground states are often in competition and it is usually possible to tune from one ground state to another by adjusting some control parameter[1,2]. Typical control parameters include magnetic field, pressure, electron density, and chemical composition. Such a transition from one zero temperature phase to another is called a quantum phase transition, when the transition is continuous, it defines a quantum critical point (QCP)[3,4]. Superconductivity in heavy fermions and in cuprates is believed by many to be mediated by fluctuations associated with a quantum critical point[5–8], whose relationship to the occurrence of superconductivity has been one of the central issues in condensed-matter physics in the last decades. In cuprate across a QCP near optimal doping, an evidence of a pairing symmetry transition from under doped compositions to overdoped compositions is reported[9]. Chubukov and Hirschfeld[10] have shown that Fe-based superconductors, with their multiple Fermi surface (FS) pockets, are the most likely candidates to show a change in the pairing symmetry upon doping.

$BaFe_{2-x}Ni_xAs_2$ is an electron-doped system where every Ni donates two electrons in contrast to Co-doping that contributes only one electron [11,12]. Interestingly, substitution of Fe by Co(Ni) in iron arsenides was argued to keep the carrier number unchanged, instead the extra electrons are localized around the impurity atoms and such substitution should effectively be isoelectronic[13]. On the other hand, the upper critical field $H_{c2}(T)$, is one of the fundamental parameters in type-II superconductors, which provides valuable information on the microscopic origin of pair breaking and reflects the electronic structure responsible for superconductivity[14]. Fe-based superconductors usually possess an extremely large upper critical fields, $H_{c2}(T)$, limiting its

* Corresponding author.
E-mail address: mahmoud.hafiez@physics.uu.se (M. Abdel-Hafiez).

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determination to temperatures near T_c giving rise to less reliable extrapolations of $H_{c2}(0)$. Therefore, large magnetic fields are required for studying the $H_{c2}(T)$. Although there is a general consensus that spin fluctuations play an important role in the formation of Cooper pairs in pnictides, many aspects such as the role of magnetism, evidence for a QCP in the temperature-doping phase diagram, the nature transition; when the transition is continuous, it defines a QCP[3,4].

In this work, bulk thermodynamic experiments were measured for 13 samples as marked by the arrows in Fig. 3. Here, in order to reveal how the superconducting gap evolves and critical behaviors in the Iron-Pnictides, $BaFe_{2-x}Ni_xAs_2$, we report on high quality complementary measurements of specific heat, transport, magnetization, and upper and critical fields studies for 13 samples as marked by the arrows in Fig. 3. We measured the temperature dependence of the in-plane electrical resistivity for various doping along with the isothermal magnetic field sweep resistance data up to 60T taken at different temperatures in magnetic fields aligned along the c-axis and along the direction of conducting plane [see Supplementary Figs. S1-S3 for details]. We find a critical point at the optimum composition $x = 0.10$, where the superconducting transition temperature T_c reaches a maximum of 21 K. We noticed sharp peak in the x-dependence of the lower and upper critical fields, the superconducting critical current density J_c, $\Delta C_{el}/T$ and the Sommerfeld coefficient (γ) in clean samples of $BaFe_{2-x}Ni_xAs_2$ at the optimum composition $x = 0.10^{13}$. This sharp peak around $x = 0.10$ [11] may arise from electronic spin fluctuations associated with a quantum critical point. Our obtained reliable values as a function of doping of the normal-state Sommerfeld coefficient (Fig. 2c and d), increase with introducing Ni to the undoped $BaFe_2As_2$, illustrating the strong competition between magnetism and superconductivity. Additionally, it can be attributed to closing of spin density wave gap with Ni doping. We show, also, that electron doping drives a crossover of the pairing symmetry from the isotropic $s-$type to anisotropic d-wave through specific heat and resistance measurements. Thus, our observation that such distinct systems display remarkably similar unexpected anomalies unveils a puzzling and seemingly universal manifestation of quantum fluctuations in the iron pnictides.

Experimental details

$BaFe_{2-x}Ni_xAs_2$ ($x = 0$, 0.03, 0.065, 0.085, 0.092, 0.096, 0.10, 0.12, 0.15, 0.18, 0.20, 0.22, and 0.30) single crystals were grown by the FeAs self-flux method, details for the growth process and sample characterization were published elsewhere[11,12]. The actual Ni level was determined to be 80% of the nominal level × through the inductively coupled plasma analysis of the as-grown single crystals. Magnetization measurements were performed by using a Quantum Design superconducting quantum interference magnetometer. The low-T specific heat down to 0.4 K was measured in its Physical Property Measurement System (PPMS) with the adiabatic thermal relaxation technique. Specific heat measurements were performed down to 70mK by using a heat-pulse technique within a dilution refrigerator along H || c up to H = 9T. The upper critical field H_{c2} at low temperatures was measured in a 60T pulsed magnet at Wuhan National High Magnetic Field Center of China. The resistance was measured with a standard four-probe method with an excitation current of 5 mA at frequency of ≈70 kHz. A high-speed data acquision card recorded the reference voltage of current and the voltage from sample at a rate of 3 MHz. The data then was extracted by a software with a phase-locked method.

Results and discussion

(a) Upper critical fields, $H_{c2}(T)$

Fig. 1(a) summarizes the doping dependence of $H_{c2}(0)$ for the whole superconducting dome from × = 0.065 to 0.22. The overall doping-dependent features in $H_{c2}(0)$ presents a sharp peak at × = 0.10, while

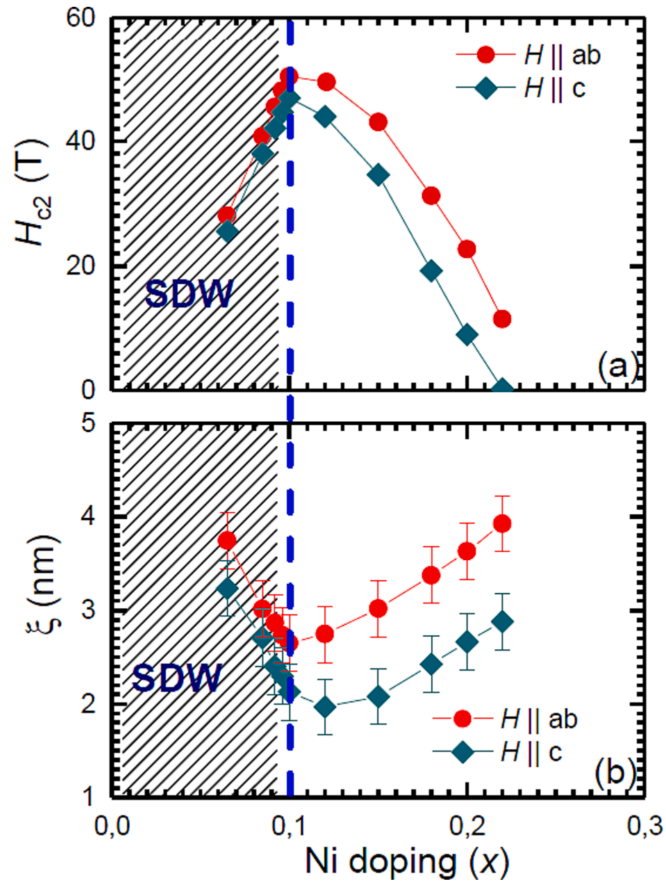

Fig. 1. Upper critical field and coherence length as a function of concentration x. (a) Hc2(0) in BaFe2-xNixAs2 estimated from the pulsed high magnetic field measurements up to 60 T on the × -doped sample. Hc2(0) forms a sharp peak correlated with the SC transition temperature. (b) Doping dependence of the coherence length derived from the Hc2 data. The results indicate a growing coherence length from optimal doping point towards either underdoping or overdoping Tc.

their difference quickly increases, especially on the overdoped side, resulting in an abrupt increase of the anisotropy. The doping dependence of the coherence length ξ is obtained form $H_{c2} = \varphi_0/(2\pi\xi^2)$ is shown in Fig. 1(b). Clearly, it is shown that from × = 0.10 the ξ grows towards underdoping or overdoping. The nearly isotropic $H_{c2}(0)$ and $\xi(0)$ for most of the optimally doped iron pnictides may originate either from Pauli-limiting or band-warping effects along the k_z direction [15,16]. The doping-dependent anisotropy of the superconductivity in $BaFe_{2-x}Ni_xAs_2$ can be understood by the dual effects from the Fermi-surface (FS) topology and impurity scattering[17]. Thus, the optimum doping with the most isotropic superconductivity correlates with an isotropic scattering from different FSs with a fine-tuned scattering rate and FS topology. A comparison with the behaviour observed in $BaFe_2(As_{1-x}P_x)_2$ is also interesting[18]. Here one sharp peak in $H_{c2}(0)$ as a function of doping (x) has been reported. It shows that the proximity of the QCP yields unexpected anomalies in the superconducting critical fields. Both the lower and upper critical fields in $BaFe_2(As_{1-x}P_x)_2$ do not follow the behaviour, predicted by conventional theory, resulting from the observed mass enhancement near the QCP. Additionally, two peaks in $H_{c2}(0)$ as a function of doping in $YBa_2Cu_3O_{7-\delta}$ have been reported [19]. These approximately coincide with critical points where other evidence suggests that the FS reconstructs. The upper critical field data presented here exhibit intriguing anisotropic and optimal doping effects. The maximum H_{c2} at the optimal Ni doping level with an in-plane magnetic field suggests enhanced superconducting properties due to a delicate balance of electronic interactions. On the other hand, the

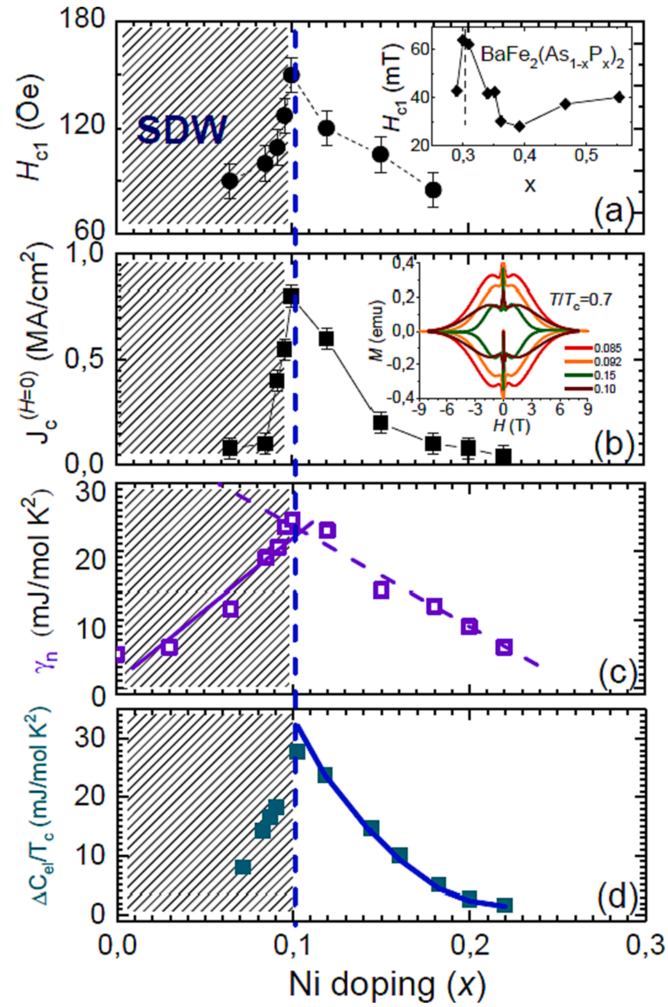

Fig. 2. Concentration × dependence of lower critical field, critical current density and specific heat jump, and the Sommerfeld coefficient. (a) Hc1(0) in BaFe2-xNixAs2 estimated from the magnetization data of each x-doped sample. Error bars on Hc1 represent the combination of uncertainties in extrapolating Hc1 to T = 0 and in the demagnetizing factor. The inset shows the lower critical field data taken from BaFe2(As1-xPx)2 system. (b) Doping evolution of the zero-field critical current in BaFe2-xNixAs2. The critical current was evaluated from the magnetic measurements, see Ref. [27]. (c) summarizes The Sommerfeld coefficient as a function of doping. Our data show the increase of the Sommerfield coefficient (γ) from 5.6 mJ/mol K2 for × = 0 to about 24 mJ/mol K2 close to the optimal concentration. This can be attributed to closing of spin density wave gap with Ni doping. The finite value of r in the underdoped regime indicates a finite electronic density of states at low energy, even in zero applied field. (d) The size of the jump in the specific heat Cel/T as a function of *x*. The line shows a fit to the logarithmic behavior expected near a QCP.

maximum H_{c2} beyond the optimal Ni doping with an out-of-plane magnetic field indicates the influence of competing phases and disorder on the unconventional superconductivity. Understanding these trends is essential for gaining insights into the underlying physics of Ni-doped Fe-based superconductors and paves the way for further exploration and potential applications of these fascinating materialsOur data, also, show that the upper critical fields for both crystallographic orientations are higher than the weak-limit paramagnetic limiting $H_p \approx$ 40T. These high values may come from the strong coupling nature of superconductivity in iron pnictides, or indeed reflect paramagnetic limiting at low temperatures, as was suggested in several studies [20,21].

(b) Lower critical fields, $H_{c1}(T)$

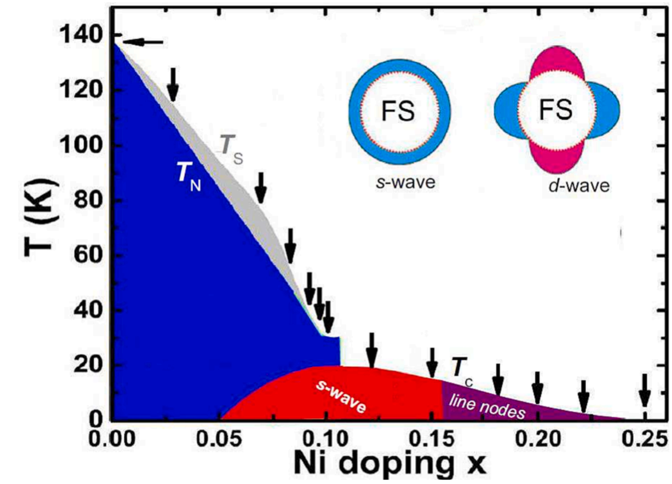

Fig. 3. Gap symmetry crossover in electronic phase diagram of BaFe2-xNixAs2. The phase diagram obtained from the magnetic, resistance, and specific heat data, showing the suppression of the magnetic (TN) and structural (TS) phase transitions with an increasing Ni concentration and the appearance of the SC transitions. The arrows indicate the various doping levels studied in this work. Superconductivity exists in a dome-like region below the transition temperature Tc. With increasing ×, Tc rises to reach a maximal value of 20 K at optimal doping. We show that the symmetry of the SC state changes from s-wave to d-wave (line nodes). The lower inset illustrates the sketch of the SC gap of s-wave vs. d-wave gap. The s-wave has isotropic gap and it does not change sign. The d-wave gap changes sign four times as it goes around the Fermi surface and as a result the gap necessarily goes to zero at four nodes.

We measured the lower critical field, H_{c1}, in $BaFe_{2-x}Ni_xAs_2$ samples using two different methods as described in Refs.[22,23]. Care must be taken in identifying H_{c1} because, in some cases, surface pinning and geometrical barriers should be not considered. However, in our measurements[11,12], several different checks, such as the equality of the investigated samples rule this out. The temperature dependence of H_{c1} is found to be linear in T at low temperature for all × [see Supplementary Fig. S4 for details], which again is indicative of a lack of surface barriers that tend to become stronger at low temperature causing an upturn in H_{c1} (T)[12]. Extrapolating this linear behaviour to zero temperature gives us H_{c1} (0), which is plotted versus doping in Fig. 2a. Most importantly, a distinct peak-shaped anomaly at × = 0.10 is observed. As being theoretically reported[24], a rapid increase is observed in H_{c1} of the underdoped region. This is due to the coexisting antiferromagnetic (AFM) and superconducting orders. On the other hand, over doped samples show a decrease in H_{c1}, due to an increasing pair breaking scattering with larger amount of substitutional disorder and larger superconducting gap anisotropy[25].

(c) Critical current density, J_c, and specific heat

Critical current density (J_c) experiments in unconventional superconductors serve as a powerful tool for detecting hidden quantum critical points (QCPs) due to their extreme sensitivity to changes in the superconducting state. Quantum critical points are associated with continuous phase transitions driven by quantum fluctuations, leading to exotic electronic properties. In unconventional superconductors, J_c measurements can provide valuable insights into the interplay between superconductivity and other electronic phases. As a QCP is approached, the superconducting gap structure and coherence length may undergo significant modifications, affecting the ability of Cooper pairs to flow coherently, thus altering J_c. Consequently, observing unexpected behavior or sharp changes in J_c as a function of some parameter, such as pressure or doping level, can indicate the presence of a hidden quantum critical point. Such experiments not only aid in identifying and characterizing quantum criticality but also offer a deeper understanding of the complex interactions that govern unconventional superconductivity, paving the way for the design and discovery of novel quantum materials with enhanced functionalities. To further support our results, we measured the critical current density (J_c) for all samples. J_c, a fundamental superconducting parameter, is a powerful tool for investigating the presence of a hidden QCP inside the superconducting dome without destroying the superconducting phase[26]. The zero-field J_c of the hole-doped cuprate superconductor has a sharp peak that is centered on a critical hole-doping where the pseudo gap boundary line projects to zero temperature, and that is attributed in model calculations to changes in the superfluid density[26]. These results indicate that J_c measurements may provide an opportunity to explore the relationship between unconventional SC and any QCP that is hidden beneath the SC dome [27]. From the magnetization hysteresis loops in M(H), we have extracted the magnetic field dependence of the J_c for the investigated single crystals [see Ref. [27] for details].

Additionally, it is evident from the data in Fig. 2c and d that the size of the anomaly $\Delta C/T_c$ and the Sommerfeld coefficient (γ) depends very strongly on × and T_c. As the strong increase in ΔC with × is accompanied by a relatively small increase in T_c close to the optimum composition × = 0.10. we suggest that this increase in $\Delta C/T_c$ reflects the increase in the normal state density of states and hence the γ. We find that our data, for $0.1 \geq x \geq 0.22$, are well described by the logarithmic critical behavior $\Delta C/T_c = c_0 + c_1 \ln(x - x_c)$ expected close to a QCP[4]. Furthermore, specific heat jumps at T_c obtained for these materials scale relatively well with its T_c in light of the careful results for the pnictide superconductors [28]. These materials with a wide range of T_c could be described by the scaling law $\Delta C \propto T^3$. This has been interpreted as either originating from quantum critically or from strong impurity pair breaking.

In correlated materials there are obvious reasons for changing the character of interactions with doping, e.g., the electron-hole asymmetry: the effective masses of the hole-like bands at the Γ point are larger than those of the electron-like bands at the *M* point. The importance of the FS proximity to nesting and spin fluctuations in the pairing mechanism therefore change as relative sizes of the electron and hole bands vary with doping. This is quite different from that of cuprates in which almost all have a nodal pairing state[7]. Therefore, the microscopic origin of the pairing symmetry inside the superconducting dome and its link to quantum criticality still remains elusive. Therefore, we turn, then, to one the main results on the pairing symmetry inside the SC dome in $BaFe_2(As_{1-x}P_x)_2$. Such complex and variable data on the pairing gap poses a question, whether or not the symmetry type in pnictides changes with doping and, if"yes", than how does it evolve?. For clarifying this situation, we chose bulk thermodynamic quantities, which are a sensitive probe of low-energy excitations of a complex quantum system. Low-energy excitations contain useful information about the nature of the ground state and can help to evaluate the presence or absence of nodes in the SC order parameter.

Fig. S5a, in supplementary materials, illustrates the electronic specific-heat (C_{el}) contribution of an underdoped sample with $x = 0.096$, obtained by subtracting the lattice specific heat from the raw data following the procedure given previously[12]. A precise description of the experimental data for the two-gap *s*-wave model is obtained by using $\Delta_1(0) = 1.8 k_B T_c$ and $\Delta_2(0) = 0.74 k_B T_c$. Additionally, the two-gap model also reproduces the C_{el} (see SI Data Fig. S5a). Meanwhile, C_p/T varies linearly with magnetic field (Fig. S5c). The observations of the temperature and magnetic field dependencies of the specific heat show that the SC gap is nodeless in the underdoped and optimally doped compounds. This is also supported by penetration depth data[12]. On the other hand, the observed T^2 term (Fig. S5b) together with the *H* behavior (Fig. S5d) of the specific heat in the SC state for the overdoped samples evidences *d*-wave superconductivity in almost all FS sheets. In this case the gap would necessarily have four-line nodes that run vertically along the *c* axis. In such a nodal structure, zero energy nodal quasiparticle will conduct heat not only in the plane but also along the *c* axis by an amount proportional to the *c*-axis dispersion of the FS. For overdoped sample ($x = 0.18$), the *d*-wave scenario without Pauli term agrees well with the experimental $H_{c2}(T)$ data shown in Fig. S5d. In good agreement with the specific heat data, the upper critical field data further supports the existence of *d*-wave superconductivity for overdoped $BaFe_{2-x}Ni_xAs_2$. The model fitting parameters are summarized in The Methods, Extended Data Table.

Conclusion

We summarize in Fig. 3 the doping dependence of structural, magnetic, and SC properties of $BaFe_{2-x}Ni_xAs_2$ single crystals as a function of Ni content[11,12]. The undoped parent ($x = 0$) undergoes a structural transition from a high-temperature tetragonal phase to a low-temperature orthorhombic one at 136(3)K, accompanied by a paramagnetic to antiferromagnetic transition at about the same temperature [see Supplementary Fig. S6 for details]. Upon the Ni substitution, the transitions gradually broaden and shift to lower temperatures[12]. Upon further increasing the Ni concentration, these transitions are no longer visible, while superconductivity maintains at slightly lower temperatures. Compared with the phase diagram shown in Fig. 3, these anomalies obtained from $H_{c2}(0)$, $H_{c1}(0)$, $J_c(0)$, $\Delta C_{el}/T$ and $\gamma(0)$ are precisely at the point where the AFM order disappears (by extrapolation to $T = 0$). These combined data for the specific heat, transport, magnetization, and upper and critical fields show compelling signatures for the existence of a quantum critical point close to $x = 0.10$ in the system. Our study demonstrates that the $H_{c2}(0)$, $H_{c1}(0)$, and $J_c(0)$, a fundamental superconducting parameters, are a powerful tool for investigating the presence of a hidden QCP inside the superconducting dome without destroying the superconducting phase. The anomalous peak in the lower critical field data observed at the optimal doping level in quantum critical superconductors can be attributed to the strongly anisotropic superconducting gap structure and the microscopic mixing of antiferromagnetism and superconductivity. This unique feature is expected to be a general characteristic of quantum critical superconductors, highlighting the intricate nature of their electronic properties. Understanding these phenomena is essential for advancing our knowledge of superconductivity and could have implications for the design and application of novel quantum materials. Further research and experimental investigations are required to unveil the full extent of these intriguing behaviors in quantum critical superconductors. Our results present a sharp peak in the Ni doped of the lower and upper critical fields. We also present the size of the jump in the specific heat C_{el}/T as a function of *Ni*. Furthermore, we show that doping induced a sudden change of the gap structure from nodeless to nodal. This observation of a change of pairing symmetry in the same system upon doping would be unprecedented and is another reason why researchers are so excited about Fe-based superconductors.

CRediT authorship contribution statement

M.M.E. Barakat: Conceptualization, Methodology, Software, Investigation, Writing – review & editing. **T.A. Abdel-Baset:** Data curation, Conceptualization, Methodology, Software, Investigation, Writing – original draft. **M. Belhaj:** Software, Investigation, Writing – review & editing. **D. El-Said Bakeer:** Methodology, Investigation, Funding acquisition. **A.N. Vasiliev:** Methodology, Investigation, Funding acquisition. **M. Abdel-Hafiez:** Investigation, Validation, Writing – review & editing, Methodology, Funding acquisition, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rinp.2023.106832.

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