Phase-Sensitive Evidence for the Sign-Reversal $s_{\pm}$ Symmetry of the Order Parameter in an Iron-Pnictide Superconductor Using $Nb/Ba_{1-x}Na_xFe_2As_2$ Josephson Junctions

A. A. Kalenyuk,[1,2] A. Pagliero,[1] E. A. Borodianskyi,[1] A. A. Kordyuk,[2,3] and V. M. Krasnov[1,*]
[1]*Department of Physics, Stockholm University, AlbaNova University Center, SE-10691 Stockholm, Sweden*
[2]*Institute of Metal Physics of National Academy of Sciences of Ukraine, 03142 Kyiv, Ukraine*
[3]*Kyiv Academic University, 03142 Kyiv, Ukraine*

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Josephson current provides a phase-sensitive tool for probing the pairing symmetry. Here we present an experimental study of high-quality Josephson junctions between a conventional s-wave superconductor Nb and a multiband iron-pnictide $Ba_{1-x}Na_xFe_2As_2$. Junctions exhibit a large enough critical current density to preclude the d-wave symmetry of the order parameter in the pnictide. However, the I_cR_n product is very small $\simeq 3\mu V$, which is not consistent with the sign-preserving s_{++} symmetry either. We argue that the small I_cR_n value, along with its unusual temperature dependence, provides evidence for the sign-reversal $s_{\pm}$ symmetry of the order parameter in $Ba_{1-x}Na_xFe_2As_2$. We conclude that it is the phase sensitivity of our junctions that leads to an almost complete (below a subpercent) cancellation of supercurrents from sign-reversal bands in the pnictide.

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Symmetry of the order parameter provides one of the main clues about the mechanism of superconductivity. Attractive electron-phonon interaction leads to a simple s-wave symmetry in conventional low-T_c superconductors. Unconventional superconductivity in cuprates and iron pnictides is characterized by a proximity to an antiferromagnetic state, suggesting the importance of spin interactions. The corresponding direct electron-electron interaction is nonretarded and, therefore, repulsive. It was predicted that this could favor superconductivity with a sign-reversal symmetry [1–3]. In single band cuprates, the sign-reversal can be only achieved with a d-wave symmetry [1,4]. However, in multiband pnictides the sign change may also take place between different bands, resulting in the $s_{\pm}$ symmetry [2,3,5–7]. On the other hand, the presence of nematic order [8–14] suggests the importance of charge-orbital interactions [15,16], which could lead to a sign-preserving s_{++} symmetry [17]. Thus, establishing of the gap symmetry provides key evidence towards the mechanisms of unconventional superconductivity.

At present, determination of the gap symmetry in iron pnictides remains ambiguous. For example, a resonant peak observed in inelastic neutron scattering [18–20] can be due to either a zero in the denominator of the dynamic spin susceptibility, caused by the sign-reversal order parameter, or to the numerator (Lindhard function) [21], if one takes into account quasiparticle damping [22]. Gap nodes, deduced from angular resolved photoemission spectroscopy [23] and heat conductance [24], may indicate either $s_{\pm}$ or d-wave symmetry. Alternatively, the strong reduction of the gap can be related with the change of the orbital character from $d_{xz/yz}$ to d_{z^2-1} [25].

The Josephson effect facilitates a phase-sensitive probe of the order parameters [1,4–7]. So far, few reliable phase-sensitive experiments have been reported for pnictides [26–29]. Both integer and half-integer flux-quantum transitions were observed [26] and large variations of the I_cR_n product, where I_c is the critical current and R_n is the junction resistance, were reported [27]. Interpretation of such results is ambiguous because half-flux quantum transitions may occur even in conventional s/s junctions due to the influence of Abrikosov vortices [30], and for $s/s_{\pm}$ junctions, phase shifts depend on the tunneling direction and the I_cR_n depends on tunneling probabilities from the two bands [5–7,28]. Evidence for the $s_{\pm}$ symmetry in pnictides was obtained via impurity dependence of the London penetration depth [31] and quasiparticle interference patterns in STM [32,33]. Yet, for understanding of the unconventional superconductivity in iron pnictides there is a need of unambiguous phase-sensitive experiments. This in turn requires solution of a technological problem of fabrication of high-quality, homogeneous, and reproducible Josephson junctions.

In this Letter, we study small, reproducible, and high-quality Josephson junctions between a conventional s-wave superconductor Nb and a c-axis oriented single crystal of $Ba_{1-x}Na_xFe_2As_2$ (BNFA). Junctions exhibit a clear Fraunhofer modulation of the critical current. The presence of significant Josephson current precludes the d-wave symmetry in BNFA. However, the eI_cR_n product of junctions is very small $\sim\mu eV$, several hundred times smaller than the corresponding energy gaps. This is inconsistent with s_{++} symmetry and provides strong evidence for the $s_{\pm}$ symmetry in BNFA. We conclude that there is an almost complete cancellation of opposite supercurrents from the sign-reversal bands in the pnictide [5–7,28,34]. This conclusion is also supported by observation of a specific temperature dependence of I_cR_n.

Figure 1(a) shows a sketch of the Brillouin zone of $Ba_{1-x}K_xFe_2As_2$ [35], which is similar to BNFA [36]. The Fermi surface consists of three barrels in the center and propellerlike sheets at the corners [37]. Contributions to superfluid density of corner sheets and central barrels are comparable [38]. Josephson current between an s-wave superconductor and c-axis oriented BNFA is sensitive to the signs of the gaps in different electronic bands of the pnictide. If the gaps are of the same sign (s_{++} case), the eI_cR_n should be of order of the energy gap $\Delta \sim$ meV in the s superconductor [39]. However, in the sign-reversal $s_{\pm}$ case, contributions from the bands with different signs would oppose each other [5–7,28,34] and we expect a smaller eI_cR_n. For the pure d-wave case, the first harmonic Josephson current should be zero [40].

Figure 1(b) represents a top view of the studied sample. Initially, six Nb/BNFA junctions (J1–6) $\sim 6 \times 5$ μm^2 with two contacts each are made on top of the BNFA crystal. After the initial test, the sample was transferred into a focused ion beam (FIB) machine and some junctions were trimmed down to submicron sizes, as shown in Fig. 1(c). This is done in order to study scaling of the junction characteristics with area. All junctions have approximately a square shape with areas from 32 to ~ 0.1 μm^2. More experimental details can be found in the Supplemental Material [41].

A multiterminal configuration allows simultaneous measurements of junctions and in-plane characteristics of the base crystal, as described in Refs. [41,45]. Figure 1(d) shows resistive transition of the junction $J4$ (6.4×5 μm^2) measured in a quasi-four-probe configuration (I^+ and V^+ at two separate contacts at the same Nb electrode and I^- and V^- at two different contacts). With decreasing T, we first observe a drop of resistance at $T_c(\mathrm{BNFA}) \simeq 30$ K, when the underlying BNFA crystal becomes superconducting. The drop is small because only a small volume immediately beneath the junction is probed since the crystal contribution is measured via two different electrodes. The drop occurs at the same temperature and has the same width $\Delta T \sim 1$ K as the in-plane resistive transition of the crystal [41]. This proves that the crystal at the junction interface is not deteriorated (no proximity effect). The resistance drops to zero below $T_c(\mathrm{Nb}) \simeq 8$ K, indicating the appearance of the supercurrent through the junction. The inset in Fig. 1(d) shows $R(T)$ at $T \lesssim T_c(\mathrm{Nb})$ at zero field and at 6 mT applied parallel to the junction interface. At this field, the supercurrent is fully suppressed. Therefore, the red curve in the inset represents junction resistance $R_n(T)$. It is slightly increasing with decreasing T.

Figure 1(e) shows current-voltage ($I - V$) characteristics of a small FIB-trimmed junction (0.95×0.85 μm^2). The blue curve is zero field $I - V$. It is nonhysteretic and has the

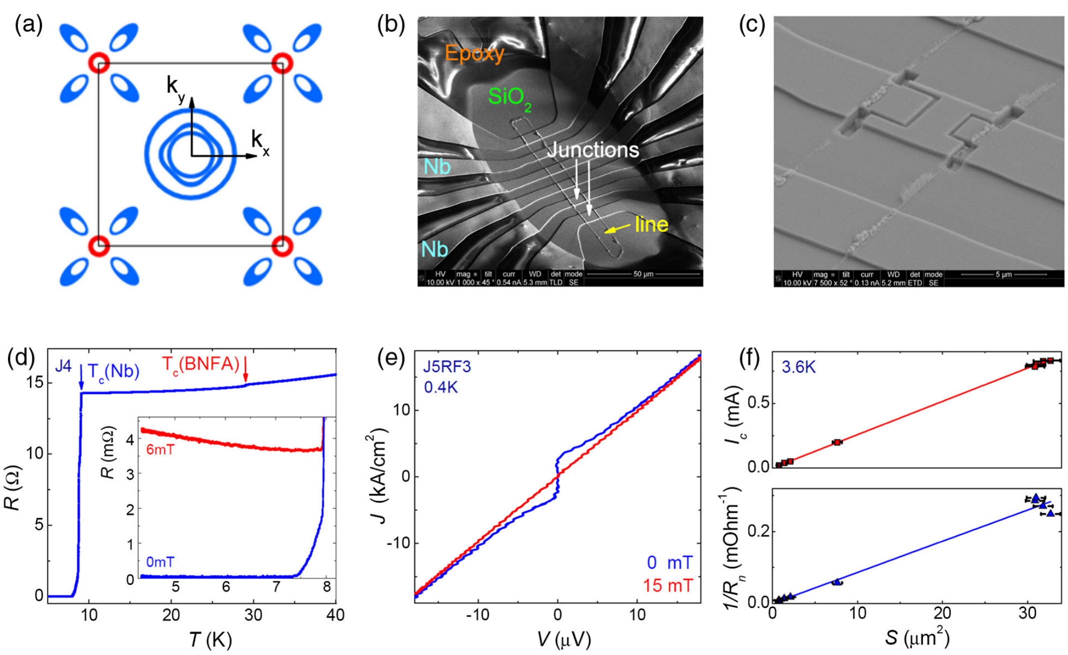

FIG. 1. (a) A sketch of the Fermi surfaces: (blue) hole type, (red) electron type sheets. (b),(c) Scanning electron microscopy images of the studied sample (b) before and (c) after FIB trimming. (c) Temperature dependence of the junction resistance. Inset shows resistance below the T_c of Nb. Blue curve at zero field. R drops to zero due to appearance of the Josephson current. The red curve is measured at field 6 mT parallel to the junction plane, at which the critical current is fully suppressed. Therefore, it represents junction resistance $R_n(T)$. (e) Current-voltage characteristics of a small FIB trimmed junction at $T \simeq 0.4$ K at zero field (blue) and at 15 mT in-plane field (red). (f) Critical current (top) and junction conductance (bottom) versus junction area for junctions at the same chip.

shape typical of resistively shunted junctions (RSJs) with a clear critical current and an asymptotic Ohmic behavior at $I \gg I_c$. The red curve shows the $I - V$ measured at in-plane field of 15 mT. The small parallel field totally suppresses I_c and the $I - V$ becomes Ohmic. We emphasize that it is fully linear, without excess current. This again implies that there is no deteriorated interface with reduced T_c (no proximity effect), nor pinholes in the junction interface [46]. This is important because it shows that we probe bulk properties of the BNFA crystal, rather than some obscure surface layer.

Figure 1(f) shows critical currents and conductances $1/R_n$ versus junction area. Linear scaling demonstrates excellent reproducibility of junction characteristics.

The main fingerprint of the dc-Josephson effect is Fraunhofer modulation of I_c versus the in-plane magnetic field. Figure 2(a) shows $I_c(H)$ dependencies for the $J5$ junction with the length perpendicular to the field $L = 6.35$ μm. A clear Fraunhofer modulation is seen, indicating good uniformity of the junction [47]. The period $\mu_0 \Delta H = \Phi_0 / L\Lambda$ corresponds to a flux quantum Φ_0 in the junction. Here Λ is the magnetic thickness of the junction [41]. ΔH should be inversely proportional to the junction size L. Figure 2(b) shows Fraunhofer modulations (see the Supplemental Material [41]) for junctions with $L = 6.35$ μm (blue) and 2.52 μm (red). It is seen that the period of modulation depends on L. Figure 2(c) shows the period versus inverse junction length (error bars reflect the uncertainty, ~100 nm, of determination of junction sizes). Linear dependence $\Delta H \propto 1/L$, together with scaling of I_c and $1/R_n$ with area [see Fig. 1(f)], confirms that we indeed probe junction characteristics.

Figure 2(d) shows temperature evolution of the $I - V$ curves at zero field. The shapes of $I - V$'s are in perfect agreement with the RSJ model, as demonstrated by the solid black line. It is seen that both the critical current and the junction resistance R_n increase with decreasing T. The corresponding variation of $R_n(T)$ can be seen from the $R(T)$ curve at $\mu_0 H = 6$ mT in the inset of Fig. 1(d).

Figure 2(e) shows temperature dependencies of critical currents. The I_c vanishes sharply with a negative curvature $d^2 I_c / dT^2 < 0$ at $T \to T_c$(Nb). This is the third indication that there is no deteriorated surface layer at the BNFA crystal interface. Indeed, a deteriorated nonsuperconducting (normal metal) layer would lead to the appearance of the proximity effect, which usually leads to a positive curvature of $I_c(T)$ at $T \to T_c$ [46,48–50]. The observed negative curvature, along with the sharp resistive transition of the BNFA interface beneath the junction at the bulk T_c(BNFA) [see Fig. 1(d)], and the absence of the excess

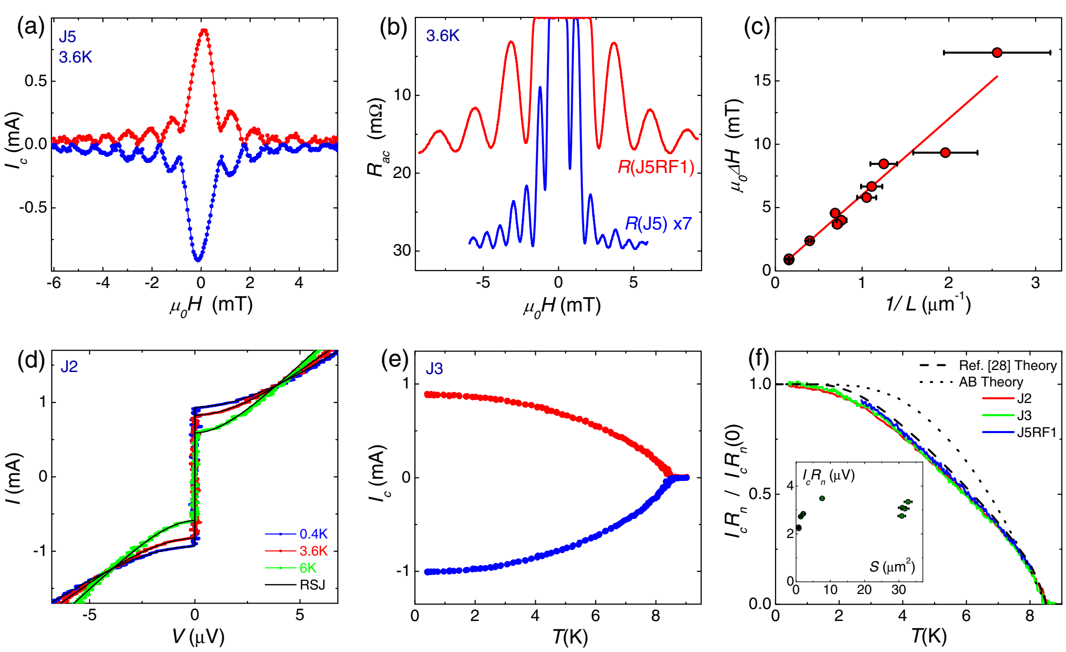

FIG. 2. (a) Measured Fraunhofer modulation $I_c(H)$ for positive and negative currents. (b) Measured Fraunhofer modulation of ac resistance for two junctions with significantly different sizes $L = 6.35$ μm (blue line) and 2.52 μm (red line). (c) Scaling of the period of Fraunhofer modulation versus the inverse length of the junction for junctions at the same chip. (d) Current-voltage characteristics of the J2 junction at different T. Note the slight decrease of the junction resistance with increasing T. The black line represents a fit in the resistively shunted junction model. (e) Temperature dependencies of the positive and negative critical currents at zero field. (f) Temperature dependencies of the $I_c R_n$ product, normalized by the $T = 0$ value, for three junctions. The dotted line and the dashed lines represent calculated dependencies for the sign-preserving s/s_{++} and sign-reversal $s/s_{\pm}$ junctions, respectively (both from Ref. [28]). Inset shows $I_c R_n$ values for junctions at the same chip. It is seen that the $I_c R_n$ is very small, confirming phase-sensitive supercurrent cancellation from the sign-reversal $s_{\pm}$ bands.

current in the $I-V$ characteristics [see Fig. 1(f)] prove the absence of surface deterioration and proximity effect in our junctions and confirm that we probe bulk order parameters in the BNFA crystal.

As follows from the scaling of I_c versus area in Fig. 1(f), all junctions have the same critical current density $J_c \simeq 3\times 10^3$ A/cm^2. It is large enough to question the d-wave symmetry of the order parameter in BNFA (see extra discussion in the Supplemental Material [41]). The s_{++} case would naturally lead to a large J_c. However, for the $s_\pm$ scenario at least a partial compensation of supercurrents from the sign-reversal bands should lead to suppression of J_c [28,34]. The critical current is not universal. The proper quantity for analysis is the I_cR_n product. For junctions between s-wave superconductors, including the s_{++} case, the I_cR_n is determined by the nearly universal (transparency independent) Ambegaokar-Baratoff expression $eI_cR_n(T=0)=a\Delta$ [39], where a is a constant of order unity. In our case, the smallest gap is $\Delta_{\mathrm{Nb}}\simeq 1.5$ meV. Thus, in the s_{++} case, we would anticipate the I_cR_n of the order of millivolts, while for $s_\pm$ it should be smaller.

The inset in Fig. 2(f) shows I_cR_n products at $T=3.6$ K. It is seen that $I_cR_n\simeq 3\ \mu$V is similar for all junctions. A systematic decrease in the smallest junctions can be explained by the influence of noise and thermal fluctuations [44,51], which suppress the switching current due to a proportional-to-area reduction of the Josephson energy. Therefore, we conclude that there is good consistency of the data. A remarkable fact is that the eI_cR_n is extremely small, ~500 times smaller than the gap in Nb. Such a small value, with no sign of the proximity effect in our junctions, essentially precludes the s_{++} symmetry in the studied pnictide.

To understand if our results are consistent with the $s_\pm$ symmetry, we refer to theoretical analysis made by Burmistrova *et al.* [28]. In the case of coherent tunneling, tunneling probability depends on the band structures and shapes of the corresponding Fermi surfaces. In pnictides, one of the bands is placed in the center, while the other is placed at the edge of the Brillouin zone [see Fig. 1(a)]. Supercurrents from the two sign-reversal $s_\pm$ bands oppose each other, reducing the total critical current. However, the extent to which they compensate each other depends on several factors. (i) Hopping probabilities from the two bands. (ii) Momentum selectivity: the tunnel current depends on the value of the in-plane momentum $k_\parallel$. Since the two bands in the pnictide do not overlap, for each $k_\parallel$, only one of the bands contributes to the electronic transport, and which one and how much depends on the size or shape of the receiving s-wave Fermi surface. (iii) The momentum selectivity depends on the transparency of the barrier: increasing the thickness of the barrier leads to the predominance of tunneling of electrons with large $k_\perp$ and thus reduces the contribution from the band with large $k_\parallel$ at the edges of the Brillouin zone.

The main panel in Fig. 2(f) shows normalized temperature dependence of I_cR_n for several junctions. It provides another clue about the gap symmetry in BNFA. The dotted line in Fig. 2(f) represents the Ambegaokar-Baratoff [39] $I_cR_n(T)$ dependence, expected for s/s' junctions (data from Ref. [28]). It is clearly different from the reported experimental $I_cR_n(T)$ dependence, which exhibits a more rapid falloff at low $T\gtrsim 2$ K. The dashed line in Fig. 2(f) represents the theoretical $I_cR_n(T)$ dependence for a $s/s_\pm$ junction from Fig. 11(c) of Ref. [28]. Apparently, it fits quantitatively to the measured $I_cR_n(T)$ dependence, including all the characteristic deviations from the Ambegaokar-Baratoff dependence. Furthermore, the theoretical curve corresponds to the case of almost complete compensation of opposite supercurrents from the two sign-reversal bands. This is fully consistent with the observed very small absolute value of $eI_cR_n(0)\simeq 3\ \mu$eV, 500 times smaller than the smallest $\Delta_{\mathrm{Nb}}\simeq 1.5$ meV. This implies that there is indeed an almost complete (to a subpercent level) cancellation of the Josephson current in our Nb/BNFA junctions.

To conclude, we fabricated and studied high-quality Josephson junctions between c-axis oriented $Ba_{1-x}Na_xFe_2As_2$ and an s-wave Nb. Junctions show a Josephson current density $\sim 10^3$ A/cm^2, which is large enough to preclude the pure d-wave symmetry of the order parameter in the pnictide. However, the I_cR_n product is very small $\simeq 3\ \mu$V, not consistent with the s_{++} symmetry either. We emphasize that the inconsistency is not marginal but is almost 3 orders of magnitude. So, a large discrepancy with no signs of the proximity effect, along with the observed unusual temperature dependence, provides strong evidence for the $s_\pm$ symmetry of the order parameter in $Ba_{1-x}Na_xFe_2As_2$. It is the phase sensitivity of our c-axis oriented Nb/BNFA junctions that leads to an almost complete (down to a subpercent) cancellation of opposite supercurrents from the sign-reversal $s_\pm$ bands in the pnictide.

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*Vladimir.Krasnov@fysik.su.se

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[41] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.120.067001 for additional information about sample fabrication, characterization, a summary of junction characteristics, and a discussion of the junction interface, which includes Refs. [42–44].

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