

Supplementary Figure 1: Magnetic susceptibility and specific heat. (a) Temperature dependence of the zero-field cooled (ZFC)and field-cooled (FC) susceptibility $\chi_{\rm m}$ obtained in an applied magnetic field of $\mu_0H = 10$ mT for optimally doped $Ba_{0.65}Rb_{0.35}Fe_2As_2$. (b) Specific heat C_p/T as a function of temperature of $Ba_{0.65}Rb_{0.35}Fe₂As₂$. Inset shows the temperature dependence of the quantity $\Delta C_{\rm p}/T$ with $\Delta C_{\rm p}$ = ($C_{\rm p}$ - $C_{\rm p,n}$), where $C_{\rm p,n}$ represent the phonon dominated background. The arrow denotes the superconducting transition temperature T_c . Error bars indicate the statistical uncertainty of the data.

Supplementary Figure 2: Specific heat scaling. (a) Specific heat jump ΔC_p at the superconducting transition vs T_c for $Ba_{0.65}Rb_{0.35}Fe₂As₂$, plotted together with literature data for various FeAs-based superconductors. The line corresponds to $\Delta C_{\rm p} \propto T^3$ (after [1]).

Supplementary Figure 3: Magnetic signal at ambient pressure. (a) The $ZF-\mu SR$ time spectra for $Ba_{0.65}Rb_{0.35}Fe₂As₂ recorded above and below T_c. Error bars indicate the statistical uncertainty$ of the data. The solid line represent the fits to the data by means of Supplementary Equation 1. (b) Temperature dependence of the magnetic fraction of $Ba_{0.65}Rb_{0.35}Fe_2As_2$, extracted from the $ZF-\mu SR$ experiments. Error bars give the fit errors.

Supplementary Figure 4: Pressure independent magnetic signal. $ZF-\mu SR$ time spectra for $Ba_{0.65}Rb_{0.35}Fe₂As₂$ at various applied pressures recorded at the base temperature $T = 1.4$ K. Error bars indicate the statistical errors of the data. The solid line represents the fit to the data by means of the sum of the Eq. (1) and a damped Kubo-Toyabe depolarization function to account for the pressure cell signal.

Supplementary Figure 5: Three pocket model used in our calculations. It is assumed that the system has one hole pocket h centered around $\Gamma = (0,0)$ and two electron pockets e_1 and e_2 centered around $M_1=(\pi,0)$ and $M_2=(0,\pi)$.

Supplementary Figure 6: Analysis of the temperature dependence of the penetration depth using microscopic model. The temperature dependence of $\lambda^{-2}(T)/\lambda^{-2}(0)$ measured at various applied hydrostatic pressures of $Ba_{0.65}Rb_{0.35}Fe₂As₂$. The square symbols are experimental data and the red curves are the theoretical functions. Error bars give the fit errors. (a) Fitting for the $P = 0$ data, which suggests a nodeless state. (b) and (c) Fitting for $P = 1.57$ GPa and $P = 2.25$ GPa. The fitting suggests that nodes exists on the two electron pockets at the angles $\theta_{\rm e} = \pm \pi/4$ and $\pm 3\pi/4$.

Supplementary Figure 7: Effect of the electron pocket gap anisotropy on the penetration depth at $p = 2.25$ GPa. (a) The electron gap is nodal if $r < 1$, and becomes nodeless if $r > 1$. The low temperature data clearly shows that the gap is nodal, but the data near T_c seems to be better described by a nodeless state. (b) Fitting for the zero pressure case with the Fermi velocity ratio v_h/v_e being a free parameter. The fitting improves with respect to Fig. 6a, but the values of $v_{\rm h}/v_{\rm e}$ and $\rho_{\rm h}/\rho_{\rm e}$ seem to be too large or too small.

I. SUPPLEMENTARY NOTE 1: SAMPLE CHARACTERIZATION

The temperature dependence of the zero field-cooled (ZFC) and field-cooled (FC) diamagnetic susceptibility of $Ba_{0.65}Rb_{0.35}Fe₂As₂ measured in a magnetic field of $\mu_0H = 1 \text{ mT}$ is$ shown in Supplementary Figure 1a. From the diamagnetic response the SC transition temperature T_c is determined from the intercept of the linearly extrapolated zero-field cooled (ZFC) susceptibility curve with $\chi_{\rm m} = 0$ line, and it is found to be $T_{\rm c} = 36.8(5)$ K. The temperature-dependent heat capacity data for this sample plotted as C_p/T vs T is shown in Supplementary Figure 1b. The jump associated with the SC transitions is clearly seen. Here the anomaly at the transition has been isolated from the phonon dominated background by subtracting a second order polynomial $C_{p,n}$ fitted above T_c and extrapolated to lower temperature. The quantity $\Delta C_{\rm p}/T$ with $\Delta C_{\rm p} = (C_{\rm p} - C_{\rm p,n})$ is presented as a function of temperature in the inset of Supplementary Figure 1b. Although there may be some uncertainty in using this procedure over an extended temperature range, the lack of appreciable thermal SC fluctuations, as evidenced by the mean-field-like form of the anomaly, means that there is very little uncertainty in the size of ΔC_p . Bud'ko et. al. [1] found that in many 122 Fe-based superconductors the specific heat jump $\Delta C_{\rm p}$ at $T_{\rm c}$ follows the empirical trend, the so-called BNC scalling $\Delta C_{\rm p} \propto T^3$. This has been interpreted as either originating from quantum critically or from strong impurity pair breaking. A violation of the BNC scaling was observed for $Ba_{1-x}K_xFe_2As_2$ for $x > 0.7$ [1] and in addition a change of the SC gap symmetry was observed. The specific heat jump data for $Ba_{0.65}Rb_{0.35}Fe₂As₂ obtained in$ this work is added in Supplementary Figure 2 to the BNC plot taken from Ref. [1]. Our data point lies perfectly on the BNC line.

II. SUPPLEMENTARY NOTE 2: RESULTS OF THE ZERO-FIELD µSR EXPER-IMENTS

It is well known that undoped $BaFe₂As₂$ is not superconducting at ambient pressure and undergoes a spin-density wave (SDW) transition of the Fe-moments far above T_c [2]. The SC state can be achieved either under pressure [3, 4] or by appropriate charge carrier doping of the parent compound [5], leading to a suppression of the SDW state. Magnetism, if present in the samples, must be taken into account in the $TF-\mu SR$ data analysis. Therefore,

we have carried out $ZF-\mu SR$ experiments above and below T_c to search for magnetism in $Ba_{0.65}Rb_{0.35}Fe₂As₂$. As an example, ZF- μ SR spectra recorded at $T = 5$ K and 50 K of $Ba_{0.65}Rb_{0.35}Fe₂As₂$ are shown in Supplementary Figure 3a. There is no preccession signal, indicating that there is no long-range magnetic order. On the other hand, we observed a significant drop of the asymmetry, taking place within 0.2 μ s. This is caused by the presence of diluted Fe moments as discussed in previous μ SR studies [6]. In order to quantify the magnetic fraction, the $ZF-\mu SR$ data were analyzed by the following function:

$$
A_{\rm ZF}(t) = \Omega A_0 \left[\frac{2}{3} e^{-\lambda_T t} + \frac{1}{3} e^{-\lambda_L t} \right]
$$

$$
+ (1 - \Omega) A_0 \left[\frac{1}{3} + \frac{2}{3} (1 - \sigma^2 t^2 - \Lambda t) e^{-\frac{\sigma^2 t^2}{2} - \Lambda t} \right].
$$

$$
(1)
$$

the first and the second terms describe the magnetic and nonmagnetic part of the signals, respectively. A_0 is the initial asymmetry, Ω is the magnetic volume fraction, and λ_T (λ_L) is the transverse (longitudinal) depolarization rate of the μ SR signal, arising from the magnetic part of the sample. The second term describing the paramagnetic part of the sample is the combination of a Lorentzian and a Gaussian Kubo-Toyabe depolarization functions [7, 8]. σ and Λ are the depolarization rates due to the nuclear dipole moments and randomly oriented diluted local electronic moments, respectively. The temperature dependence of the magnetic fraction obtained for $Ba_{0.65}Rb_{0.35}Fe_2As_2$ is plotted in Supplementary Figure 3b. The magnetic fraction at the base temperature was found to be only 8 %. Bearing in mind that the signal from the magnetically ordered parts vanishes within the first $0.2 \mu s$ in the whole temperature region, the analysis of transverse field data was restricted to times $t >$ $0.2 \mu s$.

Supplementary Figure 4 shows the ZF- μ SR time spectra for $Ba_{0.65}Rb_{0.35}Fe_2As_2$ at various applied pressures. The ZF relaxation rate stays nearly unchanged between $p = 0$ GPa and 2.25 GPa, implying that there is no sign of pressure induced magnetism in this system.

III. SUPPLEMENTARY NOTE 3: MICROSCOPIC MODEL FOR ANALYZING THE PENETRATION DEPTH DATA OF $BA_{0.65}RB_{0.35}FE_2AS_2$

Model for s^{+-} pairing:

As a minimal model that accounts for the different superconducting states of the iron

pnictides (nodeless s^{+-} , nodal s^{+-} , and d-wave), we consider a two-dimensional system with three isotropic Fermi pockets [9]: one hole pocket h centered around $\Gamma = (0,0)$ and two electron pockets e_1 and e_2 centered around $M_1 = (\pi, 0)$ and $M_2 = (0, \pi)$ (see Supplementary Figure 5). To describe the s^{+-} state, the pairing interaction between the hole pocket h and the electron pocket e_1 is assumed to be angular dependent with the form:

$$
V_{\text{he}_1} = V_0(r - \cos 2\phi)h_{\uparrow}^{\dagger}(\mathbf{k})h_{\downarrow}^{\dagger}(-\mathbf{k})e_{1\downarrow}(-\mathbf{p})e_{1\uparrow}(\mathbf{p}) + h.c. , \qquad (2)
$$

where ϕ is the polar angle measured relative to the center of the electron pocket, V_0 is the interaction energy scale, and r is the relative amplitude of the angular-independent and the angular-dependent pairing interactions. Due to the tetragonal symmetry of the system, the pairing interaction between h and e_2 is:

$$
V_{\text{he}_2} = V_0(r + \cos 2\phi)h_1^{\dagger}(\mathbf{k})h_1^{\dagger}(-\mathbf{k})e_{2\downarrow}(-\mathbf{p})e_{2\uparrow}(\mathbf{p}) + h.c. \tag{3}
$$

Furthermore, to minimize the number of free parameters, we assume that the three pockets have the same Fermi velocity v_f , while the density of states can in principle be different $\rho_h/\rho_e = \eta$. Within this model, we obtain an s⁺⁻ state, where the SC gap of the hole pocket is a constant, Δ_h , and the gap on the electron pockets is of the form $\Delta_{\mathbf{e}_1} = \Delta_{\mathbf{e}}(r - \cos 2\phi)$ and $\Delta_{\mathbf{e}_2} = \Delta_{\mathbf{e}}(r + \cos 2\phi)$. Accidental nodes appear in the electron pockets if $r < 1$. Introducing the energy cutoff Λ_c , we can write down the corresponding BCS-like gap equations:

$$
\Delta_{\rm h} = -\rho_{\rm e}V_0\Delta_{\rm e}\int_{-\Lambda_{\rm c}}^{\Lambda_{\rm c}} d\epsilon \int \frac{d\phi}{2\pi} \left(\frac{(r + \cos 2\phi)^2}{2E_{\rm e_1}(\mathbf{k})} \tanh \frac{\beta E_{\rm e_1}(\mathbf{k})}{2} + \frac{(r - \cos 2\phi)^2}{2E_{\rm e_2}(\mathbf{k})} \tanh \frac{\beta E_{\rm e_2}(\mathbf{k})}{2} \right)
$$
\n
$$
\Delta_{\rm e} = -\rho_{\rm h}V_0\Delta_{\rm h} \int_{-\Lambda_{\rm c}}^{\Lambda_{\rm c}} \frac{d\epsilon}{2E_{\rm h}(\mathbf{k})} \tanh \frac{\beta E_{\rm h}(\mathbf{k})}{2} \tag{5}
$$

where $E_{e_1}(\mathbf{k}), E_{e_2}(\mathbf{k}),$ and $E_h(\mathbf{k})$ are the quasi-particle energy dispersions:

$$
E_{e_1}(\mathbf{k}) = \sqrt{\epsilon_e^2 + \Delta_e^2 (r - \cos 2\phi)^2} \,, \quad E_2(\mathbf{k}) = \sqrt{\epsilon_e^2 + \Delta_e^2 (r + \cos 2\phi)^2} \,, \quad E_h(\mathbf{k}) = \sqrt{\epsilon_h^2 + \Delta_h^2} \,.
$$

To determine T_c , we linearize the gap equations, yielding:

$$
\begin{cases} \Delta_{\rm h} = -\Delta_{\rm e}\rho_{\rm e}V_0(2r^2+1)\int_0^{\Lambda_{\rm c}}\frac{\mathrm{d}\epsilon}{\epsilon}\tanh\frac{\beta_{\rm c}\epsilon}{2} \\ \Delta_{\rm e} = -\Delta_{\rm h}\eta\rho_{\rm e}V_0\int_0^{\Lambda_{\rm c}}\frac{\mathrm{d}\epsilon}{\epsilon}\tanh\frac{\beta_{\rm c}\epsilon}{2} \end{cases} \Longrightarrow \rho_{\rm e}V_0 = \left[\sqrt{\eta(2r^2+1)}\int_0^{\Lambda_{\rm c}}\frac{\mathrm{d}\epsilon}{\epsilon}\tanh\frac{\beta_{\rm c}\epsilon}{2}\right]^{-1}
$$

To perform the fitting, we set T_c to be fixed, and set the energy cutoff $\Lambda_c = 86 \text{meV}$ (the results do not depend significantly on the choice of the cutoff). This provides a constraint on $\rho_e V_0$, η , and r. When $T < T_c$, the gaps are calculated based on the BCS Eqs. (4) and (5).

The expression for the penetration depth of a single-band system is:

$$
\lambda_{\mu\mu}^{-2}(T) = \frac{4\pi}{cV} \sum_{\mathbf{k}} \left[\langle \frac{\partial^2 \epsilon}{\partial k_{\mu}^2} \rangle + \left(\frac{\partial \epsilon}{\partial k_{\mu}} \right)^2 \frac{\partial f}{\partial E_{\mathbf{k}}} \right] \to \frac{1}{V} \sum_{\mathbf{k}} \left(\frac{\partial \epsilon}{\partial k_{\mu}} \right)^2 \left[\frac{\partial f}{\partial E_{\mathbf{k}}} - \frac{\partial f}{\partial \epsilon_{\mathbf{k}}} \right] ,
$$

where f is the Fermi distribution function, ϵ is the energy of the non-interacting system, and E_k is the quasi-particle energy dispersion. Applying this formula to our three pocket model, we obtain

$$
\lambda^{-2}(T) \propto \rho_{\rm h} \frac{v_{\rm f}^{2}}{2} \int_{-\Lambda_{\rm c}}^{\Lambda_{\rm c}} \mathrm{d}\epsilon \left(\frac{\partial f}{\partial E_{\rm h}} - \frac{\partial f}{\partial \epsilon_{\rm h}} \right) + \rho_{\rm e} v_{\rm f}^{2} \int_{-\Lambda_{\rm c}}^{\Lambda_{\rm c}} \mathrm{d}\epsilon \int \frac{\mathrm{d}\phi}{2\pi} \cos^{2}\phi \left(\frac{\partial f}{\partial E_{\rm e_{1}}} - \frac{\partial f}{\partial \epsilon_{\rm e}} \right) + \rho_{\rm e} v_{\rm f}^{2} \int_{-\Lambda_{\rm c}}^{\Lambda_{\rm c}} \mathrm{d}\epsilon \int \frac{\mathrm{d}\phi}{2\pi} \cos^{2}\phi \left(\frac{\partial f}{\partial E_{\rm e_{2}}} - \frac{\partial f}{\partial \epsilon_{\rm e}} \right) \lambda^{-2}(T) \propto \rho_{\rm e} v_{\rm f}^{2} \left[\frac{2 + \eta}{2} \left(1 - 2f(\Lambda_{\rm c}) \right) + \eta \int_{0}^{\Lambda_{\rm c}} \mathrm{d}\epsilon \frac{\partial f}{\partial E_{\rm h}} + 2 \int_{0}^{\Lambda_{\rm c}} \mathrm{d}\epsilon \int \frac{\mathrm{d}\phi}{2\pi} \frac{\partial f}{\partial E_{\rm e}} \right] \tag{6}
$$

In the fittings, we will focus on the normalized penetration depth $\lambda^{-2}(T)/\lambda^{-2}(0)$.

Model for d-wave pairing:

To describe the d-wave superconducting state within our three band model, we consider the following form of the pairing interaction:

$$
V_{\text{he}_1} = V_0(r - \cos 2\theta)h_+^{\dagger}(\mathbf{k})h_+^{\dagger}(-\mathbf{k})e_{1\downarrow}(-\mathbf{p})e_{1\uparrow}(\mathbf{p}) + h.c.
$$

$$
V_{\text{he}_2} = V_0(r + \cos 2\theta)h_+^{\dagger}(\mathbf{k})h_+^{\dagger}(-\mathbf{k})e_{2\downarrow}(-\mathbf{p})e_{2\uparrow}(\mathbf{p}) + h.c.
$$

where θ is the angle around the hole pocket. The gap functions can then be written as:

$$
\Delta_{\mathbf{e}_1} = -\Delta_{\mathbf{e}_2} = \Delta_{\mathbf{e}} , \quad \Delta_{\mathbf{h}}(\mathbf{k}) = \Delta_{\mathbf{h}} \cos 2\theta .
$$

resulting in the BCS-like gap equations:

$$
\Delta_{\rm h} = 2\Delta_{\rm e}\rho_{\rm e}V_0 \int_{-\Lambda_{\rm c}}^{\Lambda_{\rm c}} \frac{\rm d\epsilon}{2E_{\rm e}} \tanh\frac{\beta E_{\rm e}}{2}
$$

$$
\Delta_{\rm e} = \Delta_{\rm h}\eta \rho_{\rm e}V_0 \int_{-\Lambda_{\rm c}}^{\Lambda_{\rm c}} \rm d\epsilon \int \frac{\rm d\theta}{2\pi} \frac{\cos^2 2\theta}{2E_{\rm h}} \tanh\frac{\beta E_{\rm h}}{2}
$$

Here, $\eta = \rho_h/\rho_e$, $E_e = \sqrt{\epsilon_e^2 + \Delta_e^2}$, and $E_h = \sqrt{\epsilon^2 + \Delta_h^2 \cos^2 2\theta}$. Repeating the same steps as for the s^{+-} case, we obtain the penetration depth:

$$
\lambda^{-2}(T) \propto \rho_{\rm e} v_{\rm f}^2 \left[\frac{2+\eta}{2} (1 - 2f(\Lambda_{\rm c})) + \eta \int_0^{\Lambda_{\rm c}} d\epsilon \int \frac{d\theta}{2\pi} \frac{\partial f}{\partial E_{\rm h}} + 2 \int_0^{\Lambda_{\rm c}} d\epsilon \frac{\partial f}{\partial E_{\rm e}} \right] \tag{7}
$$

Comparing the expressions for the d-wave case to the expressions we derived for the s^{+-} case, Eqs. (4) and (6), we note that they can be mapped onto each other if $r = 0$. In this extreme case, changing $\eta_d \to 4/\eta_s$, $V_{0,d} \to \eta V_{0,s}/2$, and $\Delta_h \leftrightarrow \Delta_e$ leads to the same gap equations and penetration depth expression. With these replacements, both s and d pairing give the same $\lambda^{-2}(T)/\lambda^{-2}(0)$. Therefore, we conclude that the penetration depth cannot distinguish between nodal- s^{+-} and d-wave if the nodal- s^{+-} is the extreme case with $r = 0$.

Fitting Results:

We now fit the experimental data $\lambda^{-2}(T)/\lambda^{-2}(0)$ of optimally-doped $Ba_{1-x}Rb_xFe_2As_2$ to find the values of $\rho_e V_0$, η , and r for different pressures. Note that the value of T_c imposes another constraint on these three parameters, as explained above. Supplementary Figures 6a, b and c show the fitting for the s^{+-} model for $P = 0$, $P = 1.57$ GPa, and $P = 2.25$ GPa, respectively. For the $P = 0$ case, we find equal gap amplitudes and no nodes, as seen by ARPES experiments in the related compound $Ba_{1-x}K_xFe_2As_2$. We see that the fitting is not as good in the region immediately below T_c . We will discuss this issue in more details below. For the pressurized samples, the fitting is overall better and indicates a nodal state $(r < 1)$. The value of the density of states ratio ρ_h/ρ_e is little affected by pressure (as expected, since no charge carriers are introduced), and is consistent with the value of a nearly compensated metal.

Surprisingly, the best fittings for both the $P = 1.57$ GPa and $P = 2.25$ GPa cases give $r = 0$, where the nodes on the electron pockets are fixed at $\theta = \pm \pi/4$. This is a very special case of the accidentally nodal s^{+-} state, since by symmetry there is no reason for r to vanish. To make this point more transparent, in Supplementary Figure 7a we plot the non-zero pressure data and the theoretical urves for the penetration depth for various values of r – keeping all the other parameters constant. Clearly, $0 < r < 1$ gives worst fittings than $r = 0$. What we also found is that $r = 10 - i.e.$ a nodeless superconducting state – describes the data better near T_c , on the expense of a very bad fitting at low temperatures – where the nodal behavior is evident.

As we discussed in the previous section, a nodal- s^{+-} state with $r = 0$ is indistinguishable – for fitting purposes – from a d -wave state. Since there is no symmetry reason to have $r = 0$ in our simple model, or even $r \ll 1$ over a wide pressure range, we interpret this result as an indirect indication that a d-wave state is more likely to be the state of the pressurized samples.

Finally, we comment on the difficulty of the fittings to capture the behavior near T_c – particularly for the sample at ambient pressure (see Supplementary Figure 6a). One reason could be the presence of inhomogeneities, which would require a distribution of gaps to be taken into account, instead of a single gap value. Another reason could be related to our choice of fixing the Fermi velocities to be the same for both the electron and hole pockets. To investigate this possibility, we lift this restriction and allow v_h/v_e to also be a fitting parameter. The result is shown in Supplementary Figure 7b. Clearly, we obtain a better fitting, but not only $\rho_e V_0$ is relatively large, but the ratios ρ_h/ρ_e and v_h/v_e are very large or very small, which is difficult to reconcile with the Fermi surface of these materials. Most likely, additional pockets are necessary to capture the full temperature dependence of the penetration depth. Nevertheless, our microscopic model provides results that agree with those obtained from the α -model fitting, particularly in the low-temperature regime, suggesting that a d-wave state is more likely to be realized than a nodal s^{+-} state.

IV. SUPPLEMENTARY REFERENCES

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