Superconducting gap structure in ambient-pressure-grown \( \text{LaO}_0.5\text{F}_{0.5}\text{BiS}_2 \)

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We have performed transverse-field muon spin relaxation (TF-\( \mu \)SR) measurements on ambient-pressure-grown polycrystalline \( \text{LaO}_0.5\text{F}_{0.5}\text{BiS}_2 \). From these measurements, no signature of magnetic order is found down to 25 mK. The value of the magnetic penetration depth extrapolated to 0 K is 0.89 (5) \( \mu \)m. The temperature dependence of superconducting penetration depth is best described by either a multigap \( s + s' \)-wave model with \( \Delta_1 = 0.947 (7) \) meV and \( \Delta_2 = 0.22 (4) \) meV or the anisotropic \( s \)-wave model with \( \Delta(0) = 0.776 \) meV and anisotropic gap amplitude ratio \( \Delta_{\text{min}}/\Delta_{\text{max}} = 0.34 \). Comparisons with other potentially multigap Bi\( \text{S}_2 \)-based superconductors are discussed. We find that these Bi\( \text{S}_2 \)-based superconductors, including Bi\( \text{O}_4\text{S}_3 \) and the high-pressure synthesized \( \text{LaO}_0.5\text{F}_{0.5}\text{BiS}_2 \), generally conform to the Uemura relation.

I. INTRODUCTION

The discovery of superconductivity in the Bi\( \text{S}_2 \) layered compounds \( \text{LnO}_0.5\text{F}_{0.5}\text{BiS}_2 \) (\( \text{Ln} = \text{La, Ce, Pr, Nd, Yb} \) and Bi\( \text{O}_4\text{S}_3 \), with the highest \( T_c = 10.6 \) K observed in the La-member, has attracted considerable attention. In this new family, the superconductivity arises from the Bi\( \text{S}_2 \) layers, analogous to the Cu\( \text{O}_2 \) layers in the high-\( T_c \) cuprates and the FeAs/FeSe layers in the iron-based superconductors (IBS). General similarities in the electronic structures are also found between the Bi\( \text{S}_2 \) family and the cuprates/IBS. Electron/-hole doping is often necessary to induce superconductivity in IBS, such as oxygen-fluorine (O-F) doping in the well-studied LaFeAs\( \text{O}_{1-x}\text{F}_x \) or hole doping in Bi\( \text{O}_4\text{S}_3 \). For the Bi\( \text{S}_2 \) compounds, electron doping was shown to induce superconductivity through fluorine substitution of oxygen or tetravalent substitution of Lanthanum. Also, some members of the Bi\( \text{S}_2 \)-based superconductors exhibit exotic properties, such as the coexistence of ferromagnetic order and superconductivity in Ce\( \text{O}_0.5\text{F}_{0.5}\text{BiS}_2 \). Extensive efforts on studying IBS show that the delicate interplay between magnetism and superconductivity is rather complicated, such as either competition/microscopic coexistence between static antiferromagnetic order and superconductivity in Bi\( \text{O}_4\text{S}_3 \). Therefore, the Bi\( \text{S}_2 \) family presents a new avenue to better understand the underlying physics of lower-dimensional superconductivity, crucial to efforts in uncovering higher \( T_c \)'s.

The first member of the superconducting Bi\( \text{S}_2 \)-based materials to be discovered was Bi\( \text{O}_4\text{S}_3 \), suggesting that the superconductivity arises from the Bi\( \text{S}_2 \) layer. This was confirmed soon after superconductivity was observed in \( \text{LaO}_0.5\text{F}_{0.5}\text{BiS}_2 \). Other aspects of the superconductivity in Bi\( \text{S}_2 \) family remain unsettled. For example, the superconducting energy gap structure remains unresolved in spite of numerous investigations.

Transverse-field muon spin relaxation (TF-\( \mu \)SR) measurements on high-pressure synthesized (HP) \( \text{LaO}_0.5\text{F}_{0.5}\text{BiS}_2 \) find that the temperature dependence of the superfluid density, derived from the measured magnetic penetration depth, \( \lambda \), is best described by an anisotropic single-gap \( s \)-wave model due to two-dimensional Fermi surface nesting at \((\pi, \pi, 0)\) with strong electronic correlation. This is also consistent with theoretical work for a single extended \( s \)-wave band based upon electron-electron correlations. However, electrical resistivity measurements on ambient-pressure synthesized (AP) \( \text{LaO}_0.5\text{F}_{0.5}\text{BiS}_2 \) and \( \text{CeO}_0.5\text{F}_{0.5}\text{BiS}_2 \) under applied pressure display behavior consistent with a two-gap model. Additionally, TF-\( \mu \)SR measurements on Bi\( \text{O}_4\text{S}_3 \) found evidence for multigap superconductivity. To complicate matters further, theoretical functional renormalization group (FRG) studies on the spin-orbital coupling claim that pairing in the Bi\( \text{S}_2 \)-based superconductors is a mixture of singlets and triplets. As the Bi\( \text{S}_2 \) family shares similarities with the IBS, and multigap superconductivity has been observed in several IBS, such as Bi\( \text{O}_4\text{S}_3 \), and Fe\( \text{As}_1-y\text{Te}_y\text{Se}_{2+y} \), elucidating the origins of this potential multigap superconductivity in the Bi\( \text{S}_2 \) family of superconductors is crucial. We have performed TF-\( \mu \)SR measurements on \( \text{LaO}_0.5\text{F}_{0.5}\text{BiS}_2 \) (AP). From the temperature dependence of the superfluid density, we find that \( \text{LaO}_0.5\text{F}_{0.5}\text{BiS}_2 \) (AP) is well described by a two-gap model. However, the anisotropic \( s \)-wave model cannot be ruled out. Furthermore, an analysis based upon the Uemura relation for unconventional superconductors finds that a number of Bi\( \text{S}_2 \)-based superconductors conform to this relation, similar to that observed in several IBS and cuprate superconductors.
II. EXPERIMENTAL METHODS

TF-μSR has been widely utilized to probe superconductivity in type-II superconductors at the microscopic level, including the magnetic penetration depth obtained from the muon spin depolarization ratio. 100% spin-polarized positive muons, each with a momentum of 29.8 MeV/c and kinetic energy of 4.12 MeV, are injected one at a time into the sample in an external magnetic field \( H_{ext} \) applied perpendicularly to the initial muon spin polarization. Each muon spin precesses about the local magnetic field \( B_{loc} \) at the muon stopping site with the Larmor frequency \( \omega = \gamma_{\mu} B_{loc} \), where \( \gamma_{\mu} / 2\pi = 135.53 \) MHz/T is the muon gyromagnetic ratio. The muons decay with an average life-time of \( \tau_{\mu} = 2.2 \) μs, predominantly emitting a positron along the direction of the muon spin. Measurements of the anisotropic distribution of the decay positrons as well as the lapse time between muon implantation and positron detection for an ensemble of muon decay events yield the time evolution of asymmetry \( A(t) \), which is proportional to the muon depolarization.

TF-μSR experiments on an AP unaligned powder sample of LaO\(_{0.5}\)F\(_{0.5}\)BiS\(_{2}\) were carried out in an applied field of 266 Oe in the LAMPF spectrometer at the M20 beamline and in the DR spectrometer at the M15 beamline, TRIUMF, Vancouver, Canada. Details of the synthesis method are described in a previous report. Heat capacity measurement gives a single sharp specific heat jump at 2.9 K, with entropy conserved under the superconducting specific heat curve, evidence of high sample homogeneity. The samples were mounted on a silver holder in the DR spectrometer. The LAMPF spectrometer only requires very thin silver tape to hold the sample. The TF-μSR data was analyzed with the software MUSRFIT.

III. RESULTS

Figure 1 shows the TF-μSR spectrum in an applied field \( H \) of 266 Oe for LaO\(_{0.5}\)F\(_{0.5}\)BiS\(_{2}\) at 25 mK (squares) and 3.96 K (circles). For clarity, a rotating reference frame corresponding to a magnetic field of 220 Oe is used to display the TF-μSR spectrum. Slightly faster damping is observed at base temperature compared to 3.96 K, consistent with an enhanced field inhomogeneity in the vortex state. The Fourier transform of the asymmetry spectrum (not shown here) is not purely Gaussian-shaped; thus, a single Gaussian term along with a background signal does not describe the spectra well. Instead, we find that an additional Lorentzian term along with the Gaussian term is required to best fit the TF-μSR spectrum, giving the following functional form:

\[
A(t) = A_0 \left[ f_s \exp\left( -\frac{1}{2} \sigma_s^2 t^2 \right) \cos(\omega_s t + \phi) + (1 - f_s) e^{-\Lambda_0 t} \cos(\omega_0 t + \phi) \right],
\]

where the first and second terms correspond to muons that stop in the sample and silver sample holder, respectively \( f_s \) represents the fraction of muons stopping in the sample). The second term is not necessary for the LAMPF spectrometer as no muon stops in the thin sample-holding silver tape. \( \Lambda_0 \) is the initial asymmetry of the signal. The Gaussian relaxation rate \( \sigma \), which appears below \( T_c \), is proportional to the rms width of the internal field distribution, which is due to the emergence of flux-line-lattice (FLL) field inhomogeneity in the superconducting state. The exponential damping rate \( \Lambda \) represents the nuclear dipolar field distribution. The observation of an exponential relaxation rate for a static nuclear dipolar field is unusual. One possible cause of the origin of Lorentzian-like nuclear relaxation \( \Lambda \) is the formation of fluoride-μ or fluoride-μ-fluorine states. However, the typical well-defined shape of precession signals from fluoride-μ-fluorine “hydrogen bonding” is not seen in our TF-μSR spectrum. The two relaxation terms with the Gaussian rate \( \sigma \) and the Lorentzian rate \( \Lambda \) are multiplied as the FLL field and nuclear dipole field are completely decoupled. \( \omega_s \) is the internal precession frequency of muons stopping in the sample, which is used to determine the internal magnetic field. In the DR spectrometer, the background frequency \( \omega_{bg} \) and the background relaxation rate \( \Lambda_{bg} \) are constant (from the fits \( \Lambda_{bg} \) is determined to be \( \sim 0.0624 \) (2) \( \mu s^{-1} \)). No extra damping component is found in the TF-μSR spectra down to 25 mK, suggesting no magnetic order in the LaO\(_{0.5}\)F\(_{0.5}\)BiS\(_{2}\) (AP).

![FIG. 1. (Color online) TF-μSR spectra from LaO\(_{0.5}\)F\(_{0.5}\)BiS\(_{2}\) (AP) in the normal (circles) and superconducting (squares) states with an external magnetic field of \( H_{ext} = 266 \) Oe. Solid curves are fits to the raw data with Eq. (1). For clarity, the spectra are shown in a rotating reference frame corresponding to a field of 220 Oe.](image)

The temperature dependences of \( \Lambda \) and \( \sigma \) obtained from fits of Eq. (1) to the data are given in Figure 2. \( \Lambda \) exhibits a nearly temperature-independent behavior, with an average value of \( \Lambda = 0.049 \) (3) \( \mu s^{-1} \) in the normal state. \( \Lambda \) is also expected not to change when entering the superconducting state, and thus is fixed to its normal state average value. A noticeable upturn in \( \sigma \) develops below 2.9 K, consistent with \( T_c \) determined from measurements of heat capacity and electrical resistivity. The temperature dependence of \( \sigma \) can then be fit with:

\[
\sigma(T) = \sigma(0)[1 - (T/T_c)^{n}], \quad (T < T_c),
\]

where \( \sigma(0) \) is the amplitude of the exponential term.
yielding $\sigma(0) = 0.11 (1) \, \mu\text{s}^{-1}$ and $n = 1.84 (1)$ in LaO$_{1.5}$F$_{0.5}$BiS$_2$. The exponent $n < 2$ suggests structure within the superconducting energy gap. Among candidates for this structure are gap nodes\textsuperscript{35,36}, multiple gaps\textsuperscript{37,38}, and $s$-wave anisotropy\textsuperscript{39}, as we discuss below.

Next we obtain the zero-temperature penetration depth $\lambda(0)$ from the relaxation rate $\sigma(0) = 0.11 (1) \, \mu\text{s}^{-1}$. In LaO$_{1.5}$F$_{0.5}$BiS$_2$ (AP) the upper critical field $H_{c2}(0)$ is estimated to be $1.9 \, \text{T}$, giving a reduced applied magnetic field $b = H/H_{c2}(0) \approx 0.014 < 1$ ($H$ is the applied field). For intermediate values of $b$ (see below), $\sigma(T)$ is related to $\lambda(T)$ by\textsuperscript{40,41}

$$\sigma(T) = \frac{\lambda(T)}{\mu} = A(b)\Phi_0 \lambda^{-2}(T),$$

where $\Phi_0 = 2.07 \times 10^{-15} \, \text{Wb}$ is the magnetic-flux quantum, and

$$A(b) = 0.172(1-b)[1 + 1.21(1 - \sqrt{b^3})]/2\pi.$$  \hspace{1cm} (4)

In our case $A(b) = 0.0494 (3)$, which yields a value $\lambda(0) = 0.89 (5) \, \mu\text{m}$. Equations (3) and (4) are valid provided\textsuperscript{41}

$$0.13/\kappa^2 \ll b \ll 1,$$

where $\kappa = \lambda_{ab}(0)/\xi(0)$ is the Ginzburg-Landau parameter\textsuperscript{41}. Here $\lambda_{ab}$ is the in-plane penetration depth and $\xi$ is the Ginzburg-Landau coherence length, given by $\xi(0) = (\Phi_0/2\pi H_{c2}(0))^{1/2}$. Since polycrystalline LaO$_{1.5}$F$_{0.5}$BiS$_2$ is a layered compound and is expected to be highly anisotropic, we can estimate $\lambda_{ab}$ from the relation $\lambda = 3^{1/4}\lambda_{ab}$ (Ref. [42] and [43]); $H_{c2}(0)$ was previously determined to be $1.9 \, \text{T}$\textsuperscript{39}, and using this we obtain $\xi(0) = 13.2 \, \text{nm}$ and $\kappa = 46.6$. Then $0.13/\kappa^2 \approx 6 \times 10^{-5}$, thus Eq. (5) is easily satisfied. We note that $A(b)$ is about 20% smaller than $A(b = 0) = 0.0605$. The latter estimate is often used, but is only applicable when $b$ is sufficiently small.

The London penetration depth of Bi$_2$O$_3$S$_3$ measured by TF-$\mu$SR suggests multigap superconductivity\textsuperscript{44}, and a two-gap $s$-wave model (the $\alpha$ model) describes the gap structure in Bi$_2$O$_3$S$_3$. This model is widely used to characterize many canonical multiband superconductors such as MgB$_2$\textsuperscript{45}. Applying the same model here, the temperature dependence of $\lambda^{-2}(T)/\lambda^{-2}(0)$ for LaO$_{1.5}$F$_{0.5}$BiS$_2$ is fit by the following functional form within the London approximation\textsuperscript{46,47,48}:

$$\lambda^{-2}(T)/\lambda^{-2}(0) = a \rho[\Delta_1(0), T] + (1 - a) \rho[\Delta_2(0), T],$$

and the equation reverts to the more common form for the isotropic single gap BCS $s$-wave model and the anisotropic $s$-wave model with $a = 1$. $\rho[\Delta_i(0), T]$ is defined by\textsuperscript{49}:

$$\rho[\Delta_i(0), T] = 1 + \frac{1}{\pi} \int_0^{2\pi} \frac{\partial f}{\partial E} \frac{E dE d\phi}{E^2 - \Delta_i(T, \phi)^2},$$

where $\Delta_i(T, \phi) = \Delta_i(0)\delta(T/T_c)g(\phi)$, $(\Delta_i(0) = 0.947 (7) \, \text{meV}$ and $\Delta_2(0) = 0.220 (4) \, \text{meV}$ with a weighting factor $a = 0.45 (2)$. From these results, we then determine $2\Delta_1(0)/k_B T_c = 7.58 (6)$ for the large gap, indicating strong coupling, and $2\Delta_2(0)/k_B T_c = 1.76 (4)$ for the second energy gap, below the BCS prediction of 3.74 for weak coupling. This is consistent with theoretical predictions, which find the large band to be strongly coupled and the smaller band to be weakly coupled\textsuperscript{50}. The anisotropic $s$-wave model gives $\Delta(0) = 0.776 (2) \, \text{meV}, with 2\Delta(0)/k_B T_c = 4.15 (3)$ (obtained by averaging gap value over $[0,2\pi]$), which is compatible with the BCS prediction in the weak-coupling limit. The parameters obtained from the different models for representative Bi$_2$S$_3$-based superconductors are summarized in Table III.

External pressure applied during synthesis produces a high pressure superconducting phase, with a distinctly higher
That the possible multigap superconductivity is intrinsic to the AP sample does not contain HP phases, supporting temperatures higher than $T_c$ on the AP sample has no obvious transition signal for the low-temperature regime. Data points taken at the DR and the LAMPF spectrometers are represented by the circle and diamond symbols.

$T_c$ is very small ($T_F$ is the Fermi temperature). $T_F$ can be obtained from $T_F = \varepsilon_F/k_B$, where Fermi energy $\varepsilon_F = n_s/m^*(h^2/2\pi)$ with $\gamma \propto m^*$ for two dimensional noninteracting electron gas, and $\varepsilon_F \propto \sigma^4/\gamma^{-1/4}$ for three dimensional systems (Ref. [21]). Here $\gamma = 2.53 \text{ mJ mol}^{-1} \text{ K}^{-2}$ is the Sommerfeld coefficient determined by heat capacity measurements. This gives a rough estimation of $T_F$ of the order of 100 K in LaO$_{0.5}$F$_{0.5}$BiS$_2$ (AP). Interestingly, the ratio $T_c/T_F$ is larger than for many ordinary BCS superconductors, but close to that for some exotic superconductors including the heavy fermion superconductors UPt$_3$ and UBe$_{13}$.

Even though these BiS$_2$ compounds obey the Uemura plot, which is a possible signature of unconventional superconductivity, angle-resolved photoemission spectroscopy measurements on the single crystalline NdO$_{0.5}$F$_{0.5}$BiS$_2$ concluded that it is more likely to be a conventional BCS superconductor mediated by electron-phonon coupling. It should be noted that there are many exceptions to the Uemura relation in IBS. For example, a recent study on the iron-based LaFeAsO$_{1-x}$F$_x$ system observed the breakdown of the Uemura relation with the application of external pressure. It is possible that NdO$_{0.5}$F$_{0.5}$BiS$_2$ and other members of the BiS$_2$-based superconductors do not follow the Uemura relation.

Future work is necessary to determine if the Nd-member of the BiS$_2$-based family is an exception to the Uemura relation. Additional investigations on energy gap structures in single crystals of the other rare-earth based members would be necessary to better characterize the potential unconventional superconductivity of the BiS$_2$-based layered family.

Figure 3 shows the linear dependence of $T_c$ with $\lambda_{ab}^{-2}$ for representative BiS$_2$-based superconductors which is referred to as the Uemura relation (carrier density over effective mass). The slope of the Uemura plot line for LaO$_{0.5}$F$_{0.5}$BiS$_2$ (AP), Bi$_2$O$_3$S$_3$, and LaO$_{0.5}$F$_{0.5}$BiS$_2$ (HP) is 1.47 K·μm$^{-2}$. Similar trends are observed in the iron chalcogenide superconductors such as LaFeAsO$_{1-x}$F$_x$, SmFeAsO$_{1-y}$F$_y$, and Fe$_{1+y}$Se$_{2-y}$Te$_y$, and in many hole-doped cuprate superconductors. The BiS$_2$ superconductors conform to the Uemura plot behaving as if they were unconventional. It would be intriguing if more BiS$_2$-based superconductors conform to the Uemura relation.

For many conventional BCS superconductors, the ratio of $T_c/T_F$ is very small ($T_F$ is the Fermi temperature). $T_F$ can be obtained from $T_F = \varepsilon_F/k_B$, where Fermi energy $\varepsilon_F = n_s/m^*(h^2/2\pi)$ with $\gamma \propto m^*$ for two dimensional noninteracting electron gas, and $\varepsilon_F \propto \sigma^4/\gamma^{-1/4}$ for three dimensional systems (Ref. [21]). Here $\gamma = 2.53 \text{ mJ mol}^{-1} \text{ K}^{-2}$ is the Sommerfeld coefficient determined by heat capacity measurements. This gives a rough estimation of $T_F$ of the order of 100 K in LaO$_{0.5}$F$_{0.5}$BiS$_2$ (AP). Interestingly, the ratio $T_c/T_F$ is larger than for many ordinary BCS superconductors, but close to that for some exotic superconductors including the heavy fermion superconductors UPt$_3$ and UBe$_{13}$.

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Figure 4 shows the linear dependence of $T_c$ with $\lambda_{ab}^{-2}$ for representative BiS$_2$-based superconductors which is referred to as the Uemura relation (carrier density over effective mass). The slope of the Uemura plot line for LaO$_{0.5}$F$_{0.5}$BiS$_2$ (AP), Bi$_2$O$_3$S$_3$, and LaO$_{0.5}$F$_{0.5}$BiS$_2$ (HP) is 1.47 K·μm$^{-2}$. Similar trends are observed in the iron chalcogenide superconductors such as LaFeAsO$_{1-x}$F$_x$, SmFeAsO$_{1-y}$F$_y$, and Fe$_{1+y}$Se$_{2-y}$Te$_y$, and in many hole-doped cuprate superconductors. The BiS$_2$ superconductors conform to the Uemura plot behaving as if they were unconventional. It would be intriguing if more BiS$_2$-based superconductors conform to the Uemura relation.

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**IV. CONCLUDING REMARKS**

In summary, we have performed TF-μSR measurements on ambient-pressure synthesized bulk superconducting LaO$_{0.5}$F$_{0.5}$Bi$_2$S$_2$. From fits to the temperature dependence of the penetration depth, we find LaO$_{0.5}$F$_{0.5}$Bi$_2$S$_2$ prepared at ambient pressure is well described by the $s + s$-wave model and the anisotropic $s$-wave model. The $\alpha$ model gives the two superconducting gap values of $\Delta_{s}(0) = 0.947$ meV and $\Delta_{g}(0) = 0.220$ meV with a weighting factor $\alpha = 0.45$ for $\Delta_{s}(0)$. The large-gap band is in the strongly coupled limit with $2\Delta_{s}(0)/k_B T_c = 7.58$ and the smaller-gap band is weakly coupled with $2\Delta_{g}(0)/k_B T_c = 1.76$. Fit using the anisotropic $s$-wave model results in $\Delta_{s}(0) = 0.776$ meV with anisotropic gap amplitude ratio $\Delta_{\min}/\Delta_{\max} = 0.34$. Furthermore, LaO$_{0.5}$F$_{0.5}$Bi$_2$S$_2$ is found to be consistent with the Uemura relation, along with several other Bi$_2$S$_2$-based superconductors, which is evidence for potential unconventional superconductivity.

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**TABLE I.** In-plane penetration depth $\lambda_{ab}$ and fit parameters, $2\Delta(0)/k_B T_c$ and $\chi^2$ for representative Bi$_2$S$_2$ superconductors assuming single and two-band $s$-wave energy gaps as well as the anisotropic $s$-wave paring. The parameter $\alpha$ is the weighting ratio of the two $s$-wave gaps. For anisotropic $s$ wave model, gap amplitude ratio is described by $\Delta_{\min}/\Delta_{\max} = (1 - \alpha)/(1 + \alpha)$. The data for Bi$_4$O$_4$S$_3$ and LaO$_{0.5}$F$_{0.5}$Bi$_2$S$_2$ (HP) are from Ref. [4] and Ref. [11], respectively.

| $\lambda_{ab}$ (nm) | LaO$_{0.5}$F$_{0.5}$Bi$_2$S$_2$ (AP) | Bi$_4$O$_4$S$_3$ | LaO$_{0.5}$F$_{0.5}$Bi$_2$S$_2$ (HP) |
|---------------------|-----------------------------------|-----------------|-----------------------------------|
| $\Delta_0$ (meV)    | 0.374 (5)                         | 0.88 (2)        | 1.47 (3)                          |
| $2\Delta_0/k_B T_c$ | 2.99 (7)                          | 4.50 (5)        | 3.4 (2)                           |
| $\chi^2$            | 2.5                               | 1.7             | -                                 |
| $s + s$-wave        | 0.947 (7), 0.220 (4)               | 0.93 (3), 0.09 (4) | -                                 |
| $a_s$, $\chi^2$    | 7.58 (6), 1.76 (4)                | 4.76 (7), 0.44 (9)| -                                 |
| anisotropic $s$     | 0.45 (2), 1.4                     | 0.94 (1), 1.3   | -                                 |

$^\dagger$ Obtained by averaging the gap value over $\varphi [0,2\pi]$ (see text).
