



UvA-DARE (Digital Academic Repository)

Muon spin rotation study of the topological superconductor $\text{Sr}_x\text{Bi}_2\text{Se}_3$

Leng, H.; Cherian, D.; Huang, Y.K.; Orain, J.-C.; Amato, A.; de Visser, A.

DOI

[10.1103/PhysRevB.97.054503](https://doi.org/10.1103/PhysRevB.97.054503)

Publication date

2018

Document Version

Final published version

Published in

Physical Review B

[Link to publication](#)

Citation for published version (APA):

Leng, H., Cherian, D., Huang, Y. K., Orain, J.-C., Amato, A., & de Visser, A. (2018). Muon spin rotation study of the topological superconductor $\text{Sr}_x\text{Bi}_2\text{Se}_3$. *Physical Review B*, 97(5), Article 054503. <https://doi.org/10.1103/PhysRevB.97.054503>

General rights

It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

Disclaimer/Complaints regulations

If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: <https://uba.uva.nl/en/contact>, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.

Muon spin rotation study of the topological superconductor $\text{Sr}_x\text{Bi}_2\text{Se}_3$ H. Leng,^{1,*} D. Cherian,¹ Y. K. Huang,¹ J.-C. Orain,² A. Amato,² and A. de Visser^{1,†}¹*Van der Waals–Zeeman Institute, University of Amsterdam, Science Park 904, 1098 XH Amsterdam, The Netherlands*²*Laboratory for Muon-Spin Spectroscopy, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland*

(Received 20 September 2017; published 5 February 2018)

We report transverse-field (TF) muon spin rotation experiments on single crystals of the topological superconductor $\text{Sr}_x\text{Bi}_2\text{Se}_3$ with nominal concentrations $x = 0.15$ and 0.18 ($T_c \sim 3$ K). The TF spectra ($B = 10$ mT), measured after cooling to below T_c in field, did not show any additional damping of the muon precession signal due to the flux line lattice within the experimental uncertainty. This puts a lower bound on the magnetic penetration depth $\lambda \geq 2.3 \mu\text{m}$. However, when we induce disorder in the vortex lattice by changing the magnetic field below T_c , a sizable damping rate is obtained for $T \rightarrow 0$. The data provide microscopic evidence for a superconducting volume fraction of $\sim 70\%$ in the $x = 0.18$ crystal and thus bulk superconductivity.

DOI: [10.1103/PhysRevB.97.054503](https://doi.org/10.1103/PhysRevB.97.054503)**I. INTRODUCTION**

$\text{Sr}_x\text{Bi}_2\text{Se}_3$ belongs to the new family of Bi_2Se_3 -based superconductors, which is reported to exhibit unconventional superconducting properties. The parent compound Bi_2Se_3 is a well-documented, archetypal topological insulator [1–3]. Recently, it was demonstrated that by doping Cu [4], Sr [5], Nb [6], or Tl [7] atoms, Bi_2Se_3 can be transformed into a superconductor with $T_c \sim 3$ K. Theory predicts the superconducting state to have a topological character, which is based on the close correspondence of the Bogoliubov–de Gennes Hamiltonian for the quasiparticles of the superconductor and the Bloch Hamiltonian for the insulator (for recent reviews on topological superconductivity, see Refs. [8,9]). In a topological superconductor, the condensate is expected to consist of Cooper pairs with odd-parity symmetry, while at the surface of the material, gapless Andreev bound states form that host Majorana zero modes. This provides an excellent motivation to thoroughly examine the family of Bi_2Se_3 -based superconductors. These centrosymmetric compounds (D_{3d} point group, $R\bar{3}m$ space group) belong to the symmetry class DIII [10]. Calculations within a two-orbital model show that odd-parity pairing, favored by strong spin-orbit coupling, can be realized [11]. In the case of $\text{Cu}_x\text{Bi}_2\text{Se}_3$, specific heat [12], upper critical field [13], and soft-point contact experiments [14] lend support to an odd-parity superconducting state. However, scanning tunneling microscopy (STM) measurements were interpreted to be consistent with s -wave pairing symmetry [15]. Clearly, further studies are required to solve this issue.

Superconductivity in $\text{Sr}_x\text{Bi}_2\text{Se}_3$ was discovered by Liu *et al.* [5]. Transport and magnetic measurements on $\text{Sr}_x\text{Bi}_2\text{Se}_3$ single crystals with $x = 0.06$ show $T_c = 2.5$ K. The resistivity is metallic with a low carrier concentration $n \approx 2 \times 10^{25} \text{ m}^{-3}$. Evidence for topological surface states was extracted from Shubnikov–de Haas oscillations observed in large magnetic

fields [5]. The persistence of topological surface states upon Sr doping was confirmed by angle-resolved photoemission experiment (ARPES) measurements, which showed a topological surface state well separated from the bulk conduction band [16,17]. The superconducting state was further characterized by Shruti *et al.* [18], who reported $T_c = 2.9$ K for $x = 0.10$ and a large Ginzburg–Landau parameter, $\kappa \approx 120$, pointing to extreme type II superconducting behavior. A surprising discovery was made by Pan *et al.* [19] by performing magnetotransport measurements on crystals with nominal concentrations $x = 0.10$ and 0.15 : the angular variation of the upper critical field, $B_{c2}(\theta)$, shows a pronounced *twofold anisotropy for field directions in the basal plane*, i.e., the rotational symmetry is broken. Magnetotransport measurements under high pressures show that the twofold anisotropy is robust up to at least $p = 2.2$ GPa [20].

Most interestingly, rotational symmetry breaking appears to be a common feature of the Bi_2Se_3 -based superconductors when the dopant atoms are intercalated. In $\text{Cu}_x\text{Bi}_2\text{Se}_3$ it appears in the spin-system below T_c , as was established by the angular variation of the Knight shift measured by nuclear magnetic resonance (NMR) [21]. Moreover, specific-heat measurements show the basal-plane anisotropy in B_{c2} is a thermodynamic bulk feature [22]. In $\text{Nb}_x\text{Bi}_2\text{Se}_3$, rotational symmetry breaking was demonstrated by torque magnetometry that probes the magnetization of the vortex lattice [23]. These recent experiments put important constraints on the superconducting order parameter. Notably, it restricts the order parameter to an odd-parity two-dimensional representation, E_u , with Δ_4 pairing [24–26]. Moreover, the superconducting state involves a nematic director that breaks the rotational symmetry when pinned to the crystal lattice, hence the label “nematic superconductivity.” The odd-parity Cooper pair state implies that these Bi_2Se_3 -derived superconductors are topological superconductors.

Here we report a muon spin rotation study on $\text{Sr}_x\text{Bi}_2\text{Se}_3$. Muon spin rotation is an outstanding technique to determine the temperature variation of λ , as well as its absolute value, via the Gaussian damping rate, σ_{TF} , of the μ^+ precession signal in

*h.leng@uva.nl

†a.devisser@uva.nl

a transverse field experiment. Below T_c , an increase of σ_{TF} is expected because the muon senses the additional broadening of the field distribution due to the flux line lattice [27,28]. The measurements show, however, that the increase of σ_{TF} is smaller than the experimental uncertainty in field-cooling experiments, which tell us λ is very large ($\geq 2.3 \mu\text{m}$ for $T \rightarrow 0$). On the other hand, when we induce disorder in the vortex lattice by changing the magnetic field below T_c , a sizable damping rate $\sigma_{\text{SC}} \approx 0.36 \mu\text{s}^{-1}$ ($T \rightarrow 0$) is obtained. These results provide microscopic evidence for a superconducting volume fraction of $\sim 70\%$ in the crystal with nominal Sr content $x = 0.18$ and thus bulk superconductivity.

II. EXPERIMENT

Single-crystalline samples $\text{Sr}_x\text{Bi}_2\text{Se}_3$ with nominal values $x = 0.15$ and 0.18 were synthesized by melting high-purity elements at 850°C in sealed evacuated quartz tubes. The crystals were formed by slowly cooling to 650°C at a rate of $3^\circ\text{C}/\text{h}$. Powder x-ray diffraction confirmed the $R\bar{3}m$ space group. The single-crystalline nature of the crystals was checked by Laue backreflection. Thin (thickness 0.4 mm) flat rectangular crystals were cut from the single-crystalline batch by a scalpel and/or spark erosion. The sample plane contains the trigonal basal plane with the a and a^* axes. The sample area for the incident muon beam is 8×12 and $3 \times 10 \text{ mm}^2$ for $x = 0.15$ and 0.18 , respectively. The characterization of the single-crystalline batch with $x = 0.15$ is presented in Ref. [19]. ac-susceptibility measurements show a superconducting shielding fraction of 80% . For the $x = 0.18$ batch, we obtain a slightly lower screening fraction, 70% .

Muon spin rotation (μSR) experiments were carried out with the Multi Purpose Surface Muon Instrument DOLLY installed at the πE1 beamline at the $S\mu\text{S}$ facility of the Paul Scherrer Institute. The technique uses spin-polarized muons that are implanted in a sample. Taking into account the density of $\text{Sr}_x\text{Bi}_2\text{Se}_3$, we calculate that muons typically penetrate over a depth of $230 \mu\text{m}$ and thus probe the bulk of the sample. If there is a local or applied field at the sample position, the muon spin will precess around the field direction. The subsequent asymmetric decay process is monitored by counting the emitted positrons by scintillation detectors that are placed at opposite directions in the muon spin precession plane [27,28]. The parameter of interest is the muon spin asymmetry function, $A(t)$, which is determined by calculating $A(t) = [N_1(t) - \alpha N_2(t)]/[N_1(t) + \alpha N_2(t)]$, where $N_1(t)$ and $N_2(t)$ are the positron counts of the two opposite detectors, and α is a calibration constant. In our case, α is close to 1. In the transverse-field (TF) configuration, the damping of the muon spin precession signal is a measure for the field distribution sensed by the muon at its localization site. For a superconductor below T_c , in a small TF of typically 10 mT , the vortex lattice is expected to produce a Gaussian damping, $\sigma_{\text{SC}} = \gamma_\mu \sqrt{\langle(\Delta B)^2\rangle}$, with $\gamma_\mu = 2\pi \times 135.5 \text{ MHz/T}$ the muon gyromagnetic ratio and $\langle(\Delta B)^2\rangle$ the second moment of the field distribution. TF experiments were performed for a field along the a axis and the c axis. In the first case, the muon spin is horizontal, i.e., along the beam direction, and the positrons are collected in the forward and backward detectors. In the second case, the muon spin is vertical (spin rotated mode),

the field is applied along the beam, and the positrons are collected in the left and right detectors. The crystals were glued with General Electric (GE) varnish to a thin copper foil that was attached to the cold finger of a helium-3 refrigerator (HELIOX, Oxford Instruments). μSR spectra were taken in the temperature interval $T = 0.25\text{--}10 \text{ K}$. The μSR time spectra were analyzed with the software packages WIMDA [29] and MUSRFIT [30].

III. RESULTS AND ANALYSIS

A. Field-cooled spectra

A first set of experiments was carried out for $x = 0.15$. The crystal with $T_c = 2.8 \text{ K}$ was slowly cooled in a TF of 10 mT ($B \parallel a$) to $T = 0.25 \text{ K}$, after which μSR spectra were recorded at fixed temperatures, during stepwise increasing the temperature up to 3.0 K . The measured spectra at 0.25 and 3.0 K are shown Fig. 1, where we have plotted the decay asymmetry as a function of time. The initial asymmetry $A(0) = 0.24$ is the full experimental asymmetry (A_{tot}). As can be noticed, the spectra at 0.25 and 3.0 K are very similar. We have fitted the spectra with the muon depolarization function,

$$A(t) = A_{\text{tot}} \exp\left(-\frac{1}{2}\sigma_{\text{TF}}^2 t^2\right) \cos(2\pi\nu t + \phi). \quad (1)$$

Here σ_{TF} is the Gaussian damping rate, $\nu = \gamma_\mu B_\mu/2\pi$ is the muon precession frequency, B_μ is the average field sensed by the muon ensemble, and ϕ is a phase factor. The resulting temperature variation $\sigma_{\text{TF}}(T)$ is shown in Fig. 2. In the normal phase, $\sigma_{\text{TF}} = 0.089 \pm 0.002 \mu\text{s}^{-1}$, which we attribute to the field distribution due to nuclear moments considered

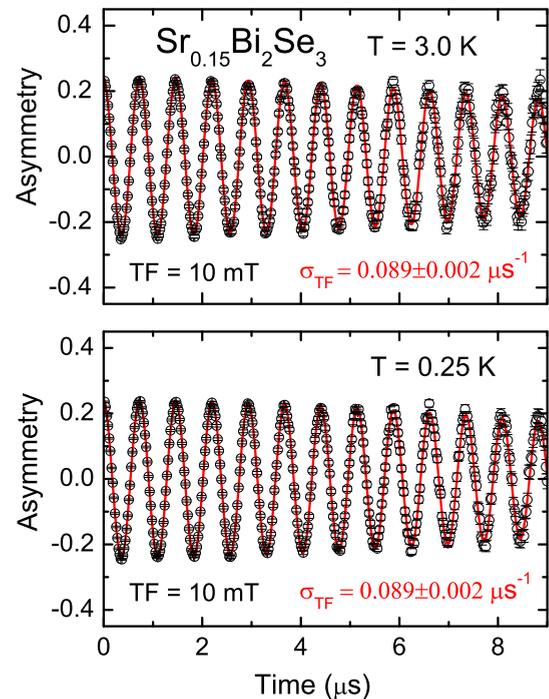


FIG. 1. μSR spectra for $\text{Sr}_{0.15}\text{Bi}_2\text{Se}_3$ measured in a transverse field of 10 mT ($B \parallel a$) at $T = 3.0 \text{ K}$ (upper panel) and $T = 0.25 \text{ K}$ (lower panel). The red lines are fits using the muon depolarization function Eq. (1). The spectra are taken after field cooling in 10 mT .

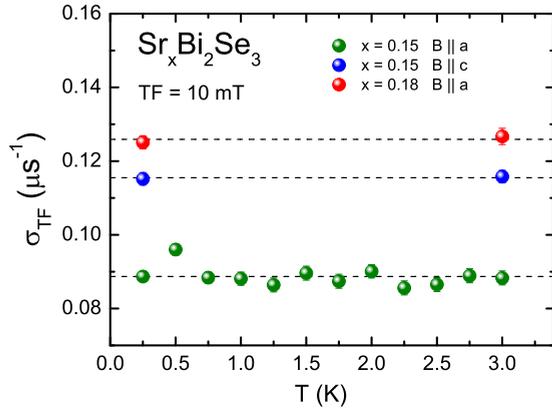


FIG. 2. Temperature variation of the Gaussian damping rate σ_{TF} extracted with help of Eq. (1) from TF μSR spectra taken after field cooling in 10 mT. Green symbols: for $x = 0.15$ and $B \parallel a$. Blue symbols: for $x = 0.15$ and $B \parallel c$. Red symbols: for $x = 0.18$ and $B \parallel a$. The dashed horizontal lines show σ_{TF} is temperature-independent.

static within the μSR time window. No additional damping is observed below T_c within the experimental resolution, and we conclude σ_{SC} is very small. An upper bound for σ_{SC} can be derived with help of the equation [31]

$$\sigma_{\text{SC}} = (\sigma_{\text{TF}, T < T_c}^2 - \sigma_{\text{TF}, T > T_c}^2)^{1/2}. \quad (2)$$

With the experimental uncertainty in σ_{FL} of $\pm 0.002 \mu\text{s}^{-1}$, we obtain $\sigma_{\text{SC}} \leq 0.02 \mu\text{s}^{-1}$. This allows us to determine a lower bound for the London penetration depth. In the vortex state of an extreme type II superconductor with a trigonal flux line lattice, λ can be estimated from the second moment of the field distribution for $B > B_{c1}$ via the relation $\langle(\Delta B)^2\rangle = 0.003706 \times \Phi_0^2/\lambda^4$, where Φ_0 is the flux quantum [32], or

$$\lambda = (0.0609\gamma_u\Phi_0/\sigma_{\text{SC}})^{1/2}. \quad (3)$$

With $\sigma_{\text{SC}} = 0.02 \mu\text{s}^{-1}$ we calculate $\lambda \geq 2.3 \mu\text{m}$ for $T \rightarrow 0$.

In the experimental configuration used to measure the data in Fig. 2 ($B \parallel a$), we probe the penetration depths orthogonal to the field direction, or rather the product $\lambda_c\lambda_{a^*}$. We have also carried out field-cooled (10 mT) measurements for $B \parallel c$ (muon spin rotated mode; here $A_{\text{tot}} = 0.19$) to probe the product $\lambda_a\lambda_{a^*}$. The extracted σ_{TF} values at 0.25 and 3.0 K are slightly larger than those for $B \parallel a$, but they are equal within the experimental resolution, as shown in Fig. 2. Finally, we have measured field-cooled μSR spectra on the $x = 0.18$ crystal for $B \parallel a$ at 0.25 and 3.0 K. The analysis shows $\sigma_{\text{TF}} = 0.126 \pm 0.002 \mu\text{s}^{-1}$ and, again, no significant temperature variation in σ_{TF} is observed, as shown in Fig. 2. We conclude that for both crystals the London penetration is very large and a conservative lower bound is $\lambda = 2.3 \mu\text{m}$.

B. Vortex lattice with disorder

The standard procedure, used above, to extract λ from the μSR spectra for a type II superconductor relies on cooling the crystal in a small magnetic field $> B_{c1}$, which tends to produce a well-ordered flux line lattice. Equation (3) can then be used to calculate λ once σ_{SC} is determined [32]. It is well

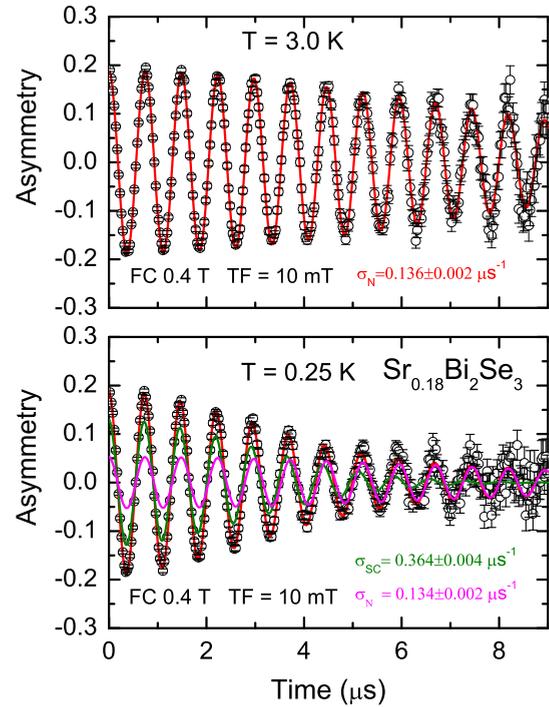


FIG. 3. μSR spectra for $\text{Sr}_{0.18}\text{Bi}_2\text{Se}_3$ measured in a transverse field of 10 mT at $T = 3.0$ K (upper panel) and $T = 0.25$ K (lower panel). The crystal was cooled in a strong field of 0.4 T ($B \parallel c$), after which the field was reduced to 10 mT. The red lines are fits to the muon depolarization function Eq. (4). The green and magenta lines in the lower panel represent the contributions to the μSR signal from the superconducting and normal state volume fractions, respectively.

known that inducing disorder in the vortex lattice increases the distribution of the internal magnetic fields, and hence σ_{SC} [33–35]. In this case, λ can no longer be calculated with the help of Eq. (3), because the calculation of λ from the field distribution has become an intricate problem [33–35]. Inducing disorder in the vortex lattice provides, however, an appealing route to probe the superconducting volume fraction of our crystals.

A standard procedure to induce disorder in the flux line lattice is to cool the sample to below T_c in zero field and then sweep the field to the desired TF value (ZFC mode). Examples in the literature that show a pronounced increase of σ_{SC} due to disorder can be found in Refs. [31,36]. Here we followed a different procedure and cooled the $x = 0.18$ crystal in a strong magnetic field ($B \parallel c$) of 0.4 T to $T = 0.25$ K, after which the field was reduced to 10 mT. Decreasing the applied field causes the flux lines to move. Pinning of flux lines at crystalline defects and inhomogeneities generates magnetic disorder. We remark that for an applied field of 0.4 T, the lattice parameter of the trigonal vortex lattice is $a_\Delta = (4/3)^{1/4}(\Phi_0/B)^{1/2} = 0.08 \mu\text{m}$. After decreasing the field to 10 mT, $a_\Delta = 0.49 \mu\text{m}$. Next, TF = 10 mT μSR spectra were taken in the temperature range 0.25–5 K by stepwise increasing the temperature. In Fig. 3 we show the data taken at 0.25 and 3.0 K. As expected, a pronounced damping now appears in the superconducting state. We first fitted the spectrum at 0.25 K to Eq. (1), but it appeared a better fit can be made with the two-component

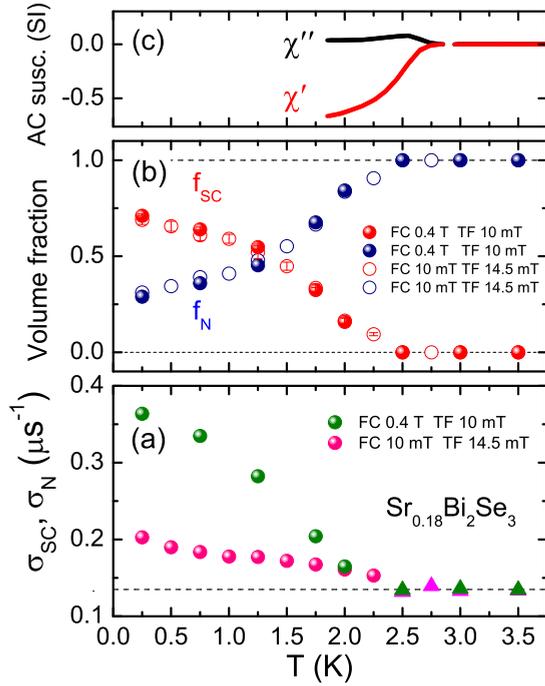


FIG. 4. Fit parameters of the two-component analysis [Eq. (4)] of TF μ SR spectra for $\text{Sr}_{0.18}\text{Bi}_2\text{Se}_3$. Disorder in the vortex lattice is induced by changing the field below T_c . (a) $\sigma_{SC}(T)$ (round symbols) and $\sigma_N(T)$ (triangles). Green symbols: field-cooling in 0.4 T, spectra measured after reducing the field to TF = 10 mT ($B \parallel c$). Magenta symbols: cooling in 10 mT, spectra measured after sweeping the field first to zero and then up to TF = 14.5 mT ($B \parallel a$). (b) Superconducting f_{SC} and normal-state f_N volume fraction for cooling in 0.4 T (closed symbols) and 10 mT (open symbols). (c) ac susceptibility in SI units. The superconducting screening fraction is 0.7.

depolarization function,

$$A(t) = A_{\text{tot}} \left[f_{SC} \exp\left(-\frac{1}{2}\sigma_{SC}^2 t^2\right) \cos(2\pi\nu_{SC}t + \phi_{SC}) + f_N \exp\left(-\frac{1}{2}\sigma_N^2 t^2\right) \cos(2\pi\nu_N t + \phi_N) \right]. \quad (4)$$

Here $f_{SC} = A_{SC}/A_{\text{tot}}$ and $f_N = A_N/A_{\text{tot}}$ are the volume fractions related to the superconducting and normal phases, respectively (ν_{SC} , ϕ_{SC} , and ν_N, ϕ_N are the corresponding frequencies and phases). In the normal state, $f_{SC} = 0$ and the relaxation rate σ_N equals $0.134 \pm 0.002 \mu\text{s}^{-1}$. This value is close to the one reported in Fig. 2. The result of the two-component fit of the spectrum at 0.25 K is shown in Fig. 3. Here the total asymmetry $A_{\text{tot}} = A_{SC} + A_N$ is fixed at the experimental value $A(0) = 0.19$ (spin-rotated mode) and σ_N is fixed at $0.134 \mu\text{s}^{-1}$. We obtain $f_{SC} = 0.71$, $f_N = 0.29$, and $\sigma_{SC} = 0.36 \pm 0.02 \mu\text{s}^{-1}$. It shows that for this crystal, the superconducting volume fraction amounts to 70%, in good agreement with ac-susceptibility measurements; see Fig. 4(c). In Fig. 4, we show the temperature variation of the fit parameters f_{SC} , f_N , and σ_{SC} . The smooth variation of f_{SC} to zero indicates $T_c = 2.5$ K, which is slightly below the onset temperature for superconductivity $T_c = 2.7$ K determined by ac susceptibility on a piece of the same single-crystalline batch.

The values of σ_{SC} in Fig. 4 indicate considerable disorder in the flux line lattice. In a second run, we have field-cooled

the sample in 10 mT to 0.25 K, next reduced the field to zero, and subsequently increased it to 14.5 mT. TF spectra ($B \parallel a$) taken after this field history showed $\sigma_{TF} = 0.20 \mu\text{s}^{-1}$ at 0.25 K, which indicates a much weaker degree of disorder in the vortex lattice. The temperature variation of the fit parameters σ_{SC} , f_{SC} , and f_N obtained by using Eq. (4) for this second run are shown in Fig. 4.

The fitting procedure with a two-component muon depolarization function [Eq. (4)] is a standard and frequently used method to determine the superconducting volume fraction. Another method is to directly compare the frequencies in the normal and superconducting phases and the corresponding amplitudes of the fast Fourier transform (FFT). Frequency shifts of the asymmetry spectra for our data are reported in the supplemental material file [37]. Clear frequency shifts are detected in the superconducting phase. However, the shifts are small ($<1.1\%$) and the FFTs of the asymmetry spectra relatively broad. This hampers the determination of the superconducting volume fraction from the FFTs.

IV. DISCUSSION

An important conclusion that can be drawn from the TF μ SR spectra taken in the disordered vortex lattice case is that $\text{Sr}_x\text{Bi}_2\text{Se}_3$ for $x = 0.18$ is a bulk superconductor. We remark that specific-heat experiments around T_c , which provide a thermodynamic way to demonstrate bulk superconductivity, have not been reported in the literature so far. The superconducting volume fraction of 70% obtained by μ SR agrees nicely with the superconducting screening fraction determined by ac-susceptibility measurements.

In the field-cooled case (ordered vortex lattice), we could not detect the damping of the μ^+ precession signal due to superconductivity. This puts a lower bound on the penetration depth λ of $2.3 \mu\text{m}$. Within the London model, λ is related to the superfluid density n_s via the relation

$$\lambda = \left(\frac{m^*}{\mu_0 n_s e^2} \right)^{1/2}, \quad (5)$$

where m^* is the effective mass of the charge carriers, μ_0 is the permeability of the vacuum, and e is the elementary charge. Assuming $m^* = m_e$, $\lambda = 2.3 \mu\text{m}$ translates to an extremely small value $n_s \sim 0.05 \times 10^{26} \text{ m}^{-3}$. This is difficult to reconcile with the carrier density $n = 1.2 \times 10^{26} \text{ m}^{-3}$ that we measured by the Hall effect on a crystal from the same batch at 4.2 K. In the literature, however, significant lower values for n have been reported: $0.27 \times 10^{26} \text{ m}^{-3}$ (Ref. [5]) and $0.19 \times 10^{26} \text{ m}^{-3}$ (Ref. [18]), which results in λ values of 1.0 and $1.2 \mu\text{m}$, respectively, using Eq. (5). A possible solution is that $m^* > m_e$, but this is not in accordance with quantum oscillation studies. For low-carrier-density samples of Bi_2Se_3 , Shubnikov-de Haas data ($B \parallel c$) show $m^* = 0.124m_e$ [38]. Doping may result in a slightly larger value of m^* . For instance, for Cu-doped Bi_2Se_3 , $m^* = 0.2-0.3m_e$ [39]. On the other hand, from specific-heat experiments on Cu-doped Bi_2Se_3 , a quasiparticle mass of $2.6m_e$ has been deduced [12]. Values for the effective mass of $\text{Sr}_x\text{Bi}_2\text{Se}_3$ have not been reported so far.

Very recently, TF muon spin rotation experiments on $\text{Cu}_x\text{Bi}_2\text{Se}_3$ crystals have been reported for a field of 10 mT

applied along the c axis [40]. Interestingly, the authors do find a small increase of σ_{TF} below T_c . In the normal state, $\sigma_{\text{TF}} = 0.105 \pm 0.001 \mu\text{s}^{-1}$, a value comparable to those for the Sr-doped case reported in Fig. 2. In the superconducting phase, a small but clear increase of σ_{TF} is observed to a value of $0.113 \pm 0.001 \mu\text{s}^{-1}$ for $T \rightarrow 0$. By analyzing the data with the help of Eq. (2), the authors calculate $\sigma_{\text{SC}} = 0.04 \mu\text{s}^{-1}$ and $\lambda = 1.6 \mu\text{m}$. We remark that the total increase in σ_{TF} below T_c is only $0.008 \mu\text{s}^{-1}$, which is only slightly larger than the scatter in our values of σ_{TF} (see Fig. 2). The higher precision in these experiments is partly due to very long counting times resulting in better statistics. The μSR experiments on Cu- and Sr-doped Bi_2Se_3 agree in the sense that for both materials λ is very large. Note that for $\text{Cu}_x\text{Bi}_2\text{Se}_3$ we calculate, with Eq. (5), using $\lambda = 1.6 \mu\text{m}$ and assuming $m^* = m_e$, a superfluid density $n_s = 0.11 \times 10^{26} \text{ m}^{-3}$, which is also at variance with the measured carrier concentration [12,39] (n_s is a factor 10 smaller). The recurring result that $n_s \ll n$ seems to indicate that only part of the conduction electrons participate in the superconducting condensate. A possible explanation is substantial electronic phase inhomogeneities, where the superconducting phase (volume fraction 70% for $\text{Sr}_{0.18}\text{Bi}_2\text{Se}_3$ and 40–60% for $\text{Cu}_x\text{Bi}_2\text{Se}_3$ [12,40]) has effectively a lower carrier concentration than the normal phase. On the other hand, a similar mismatch between n_s and n has recently been reported for the Nb-doped low-carrier-density superconductor SrTiO_3 notably in the over-doped, dirty regime, which is relevant in the context of high- T_c cuprates as well [41]. We remark that the discrepancy between n_s and n does not show up in the standard analysis of the Ginzburg-Landau parameter $\kappa = \lambda/\xi$, where ξ is the superconducting coherence length. The large value of $\kappa \sim 100$ and the small coherence length $\xi \sim 15 \text{ nm}$ extracted from transport and magnetic measurements [12,18] result in a substantial value $\lambda \sim 1.5 \mu\text{m}$. Here we have neglected for the purpose of simplicity the crystalline anisotropy of about a factor 1.5 in these parameters.

The μSR spectra for the $x = 0.18$ crystal, taken after cooling in 0.4 T and subsequently reducing the field to 10 mT, show a sizable depolarization due to disorder in the vortex lattice. If we assume a random distribution of flux lines, λ can be calculated using the expression $\langle(\Delta B)^2\rangle = \Phi_0 B / 4\pi \lambda^2$ (see Refs. [34,35]). With $\sigma_{\text{SC}} = 0.36 \mu\text{s}^{-1}$ [see Fig. 4(a)] we calculate $\lambda = 3.0 \mu\text{m}$, a value in line with the lower bound $2.3 \mu\text{m}$ estimated from the field-cooled experiments. It is not surprising that substantial disorder in the vortex lattice can be created. In the Cu, Sr, and Nb case, experimental evidence has been presented that the dopant atoms are intercalated

in the van der Waals gaps between the quintuple layers of the Bi_2Se_3 structure [4–6]. However, partial substitution on the Bi lattice cannot be ruled out. A detailed refinement of the crystal structure after intercalation has not been reported for these compounds so far. For $\text{Cu}_x\text{Bi}_2\text{Se}_3$ it has been inferred by analogy to related selenides that the intercalant atoms reside in the $3b$ site (Wyckoff notation) [4]. Moreover, structural investigations report considerable disorder on various length scales [42,43]. Thus the Bi-based superconductors are prone to various types of structural disorder, which in turn may provide different sources of flux pinning.

V. SUMMARY AND CONCLUSIONS

We have performed transverse field muon spin rotation experiments on single-crystalline samples of $\text{Sr}_x\text{Bi}_2\text{Se}_3$ with the aim to determine the London penetration depth, λ . Field-cooled μSR spectra measured for the ordered flux line lattice reveal, however, no additional damping of the μ^+ precession signal in the superconducting phase. From the data, we infer a lower bound for λ of $2.3 \mu\text{m}$. By changing the applied magnetic field in the superconducting phase, we are able to induce disorder in the vortex lattice. This results in a sizable value $\sigma_{\text{SC}} = 0.36 \mu\text{s}^{-1}$ for $T \rightarrow 0$. By analyzing the μSR time spectra with a two-component function, we obtain a superconducting volume fraction of 70%. This provides solid evidence for bulk superconductivity in $\text{Sr}_x\text{Bi}_2\text{Se}_3$. We signal a discrepancy between the superfluid density, n_s , calculated from λ within the London model, and the measured carrier concentration. Finally, we recall that the reported [19,21–23] breaking of rotational symmetry in the small family of Bi_2Se_3 -based superconductors deserves close examination, notably because it offers an excellent opportunity to study unconventional superconductivity with a two-component order parameter.

ACKNOWLEDGMENTS

This work was part of the research program on Topological Insulators funded by FOM (Dutch Foundation for Fundamental Research of Matter). The research of H.L. was made possible by Grant No. 201604910855 of the Chinese Scholarship Council. The authors are grateful to M. J. Graf for help with the experiments and fruitful discussions. Part of this work was performed at the Swiss Muon Source ($S\mu S$) of the Paul Scherrer Institute, Villigen, Switzerland.

- [1] H. Zhang, C.-X. Liu, X.-L. Qi, X. Dai, Z. Fang, and S.-C. Zhang, *Nat. Phys.* **5**, 438 (2009).
- [2] Y. Xia, D. Qian, D. Hsieh, L. Wray, A. Pal, H. Lin, A. Bansil, D. Grauer, Y. S. Hor, R. J. Cava, and M. Z. Hasan, *Nat. Phys.* **5**, 398 (2009).
- [3] D. Hsieh, Y. Xia, D. Qian, L. Wray, J. H. Dil, F. Meier, J. Osterwalder, L. Patthey, J. G. Checkelsky, N. P. Ong, A. V. Fedorov, H. Lin, A. Bansil, D. Grauer, Y. S. Hor, R. J. Cava, and M. Z. Hasan, *Nature (London)* **460**, 1101 (2009).

- [4] Y. S. Hor, A. J. Williams, J. G. Checkelsky, P. Roushan, J. Seo, Q. Xu, H. W. Zandbergen, A. Yazdani, N. P. Ong, and R. J. Cava, *Phys. Rev. Lett.* **104**, 057001 (2010).
- [5] Z. Liu, X. Yao, J. Shao, M. Zuo, L. Pi, S. Tan, C. Zhang, and Y. Zhang, *J. Am. Chem. Soc.* **137**, 10512 (2015).
- [6] Y. Qiu, K. N. Sanders, J. Dai, J. E. Medvedeva, W. Wu, P. Ghaemi, T. Vojta, and Y. S. Hor, [arXiv:1512.03519](https://arxiv.org/abs/1512.03519).
- [7] Z. Wang, A. A. Taskin, T. Frölich, M. Braden, and Y. Ando, *Chem. Mater.* **28**, 779 (2016).

- [8] T. Mizushima, Y. Tsutsumi, T. Kawakami, M. Sato, M. Ichioka, and K. Machida, *J. Phys. Soc. Jpn.* **85**, 022001 (2016).
- [9] M. Sato and Y. Ando, *Rep. Progr. Phys.* **80**, 076501 (2017).
- [10] A. P. Schnyder, S. Ryu, A. Furusaki, and A. W. W. Ludwig, *Phys. Rev. B* **78**, 195125 (2008).
- [11] L. Fu and E. Berg, *Phys. Rev. Lett.* **105**, 097001 (2010).
- [12] M. Kriener, K. Segawa, Z. Ren, S. Sasaki, and Y. Ando, *Phys. Rev. Lett.* **106**, 127004 (2011).
- [13] T. V. Bay, T. Naka, Y. K. Huang, H. Luigjes, M. S. Golden, and A. de Visser, *Phys. Rev. Lett.* **108**, 057001 (2012).
- [14] S. Sasaki, M. Kriener, K. Segawa, K. Yada, Y. Tanaka, M. Sato, and Y. Ando, *Phys. Rev. Lett.* **107**, 217001 (2011).
- [15] N. Levy, T. Zhang, J. Ha, F. Sharifi, A. A. Talin, Y. Kuk, and J. A. Stroscio, *Phys. Rev. Lett.* **110**, 117001 (2013).
- [16] C. Q. Han, H. Li, W. J. Chen, F. Zhu, M.-Y. Yao, Z. J. Li, M. Wang, B. F. Gao, D. Guan, C. Liu, C. L. Gao, D. Qian, and J.-F. Jia, *Appl. Phys. Lett.* **107**, 171602 (2015).
- [17] M. Neupane, Y. Ishida, R. Sankar, J.-X. Zhu, D. S. Sanchez, I. Belopolski, S.-Y. Xu, N. Alidoust, S. Shin, F. Chou, M. Z. Hasan, and T. Durakiewicz, *Sci. Rep.* **6**, 22557 (2016).
- [18] Shruti, V. K. Maurya, P. Neha, P. Srivastava, and S. Patnaik, *Phys. Rev. B* **92**, 020506(R) (2015).
- [19] Y. Pan, A. M. Nikitin, G. K. Araizi, Y. K. Huang, Y. Matsushita, T. Naka, and A. de Visser, *Sci. Rep.* **6**, 28632 (2016).
- [20] A. M. Nikitin, Y. Pan, Y. K. Huang, T. Naka, and A. de Visser, *Phys. Rev. B* **94**, 144516 (2016).
- [21] K. Matano, M. Kriener, K. Segawa, Y. Ando, and G.-Q. Zheng, *Nat. Phys.* **12**, 852 (2016).
- [22] S. Yonezawa, K. Tajiri, S. Nakata, Y. Nagai, Z. Wang, K. Segawa, Y. Ando, and Y. Maeno, *Nat. Phys.* **13**, 123 (2017).
- [23] T. Asaba, B. J. Lawson, C. Tinsman, L. Chen, P. Corbae, G. Li, Y. Qiu, Y. S. Hor, L. Fu, and L. Li, *Phys. Rev. X* **7**, 011009 (2017).
- [24] Y. Nagai, H. Nakamura, and M. Machida, *Phys. Rev. B* **86**, 094507 (2012).
- [25] L. Fu, *Phys. Rev. B* **90**, 100509(R) (2014).
- [26] J. W. F. Venderbos, V. Kozii, and L. Fu, *Phys. Rev. B* **94**, 180504 (2016).
- [27] A. Amato, *Rev. Mod. Phys.* **69**, 1119 (1997).
- [28] S. J. Blundell, *Contemp. Phys.* **40**, 175 (1999).
- [29] F. L. Pratt, *Physica B* **289-290**, 710 (2000).
- [30] A. Suter and B. Wojek, *Phys. Proc.* **30**, 69 (2012).
- [31] G. M. Luke, A. Keren, L. P. Le, W. D. Wu, Y. J. Uemura, D. A. Bonn, L. Taillefer, and J. D. Garrett, *Phys. Rev. Lett.* **71**, 1466 (1993).
- [32] E. H. Brandt, *Phys. Rev. B* **37**, 2349 (1988).
- [33] E. H. Brandt, *J. Low Temp. Phys.* **73**, 355 (1988).
- [34] Y. M. Belousov, V. N. Gorbunov, V. P. Smilga, and V. I. Fesenko, *Usp. Fiz. Nauk* **160**, 55 (1990).
- [35] C. M. Aegerter and S. L. Lee, *Appl. Magn. Res.* **13**, 75 (1997).
- [36] W. D. Wu, A. Keren, L. P. Le, B. J. Sternlieb, G. M. Luke, Y. J. Uemura, P. Dosanjh, and T. M. Riseman, *Phys. Rev. B* **47**, 8172 (1993).
- [37] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.97.054503> for the frequency shifts of the asymmetry spectra in the superconducting state.
- [38] H. Köhler, *Phys. Status Solidi B* **58**, 91 (1973).
- [39] E. Lahoud, E. Maniv, M. S. Petrushevsky, M. Naamneh, A. Ribak, S. Wiedmann, L. Petaccia, Z. Salman, K. B. Chashka, Y. Dagan, and A. Kanigel, *Phys. Rev. B* **88**, 195107 (2013).
- [40] J. A. Krieger, A. Kanigel, A. Riback, E. Pomjakushina, K. B. Chashka, K. Conder, E. Morenzoni, T. Prokscha, A. Suter, and Z. Salman, *J. Phys. Soc. Conf. Proc.* (to be published), [arXiv:1707.09189](https://arxiv.org/abs/1707.09189).
- [41] C. Collignon, B. Fauqué, A. Cavanna, U. Gennser, D. Mailly, and K. Behnia, *Phys. Rev. B* **96**, 224506 (2017).
- [42] M. Kriener, K. Segawa, Z. Ren, S. Sasaki, S. Wada, S. Kuwabata, and Y. Ando, *Phys. Rev. B* **84**, 054513 (2011).
- [43] C. Mann, D. West, I. Miotkowski, Y. P. Chen, S. Zhang, and C.-K. Shih, *Phys. Rev. B* **89**, 155312 (2014).