Penetration depth and gap structure in the antiperovskite oxide superconductor \( \text{Sr}_3\text{SnO} \) revealed by \( \mu\text{SR} \)

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We report a \( \mu\text{SR} \) study on the antiperovskite oxide superconductor \( \text{Sr}_3\text{SnO} \). By using transverse-field \( \mu\text{SR} \), we observed an increase of the muon relaxation rate upon cooling below the superconducting transition temperature \( T_c = 5.4 \) K, evidencing bulk superconductivity. The exponential temperature dependence of the relaxation rate \( \sigma \) at low temperatures suggests a fully gapped superconducting state. We evaluated the zero-temperature penetration depth \( \lambda(0) \propto 1/\sqrt{\sigma(0)} \) to be around 320–1020 nm. Such a large value is consistent with the picture of a doped Dirac semimetal. Moreover, we established that the ratio \( T_c/\lambda(0)^2 \) is larger than those of ordinary superconductors and is comparable to those of unconventional superconductors. The relatively high \( T_c \) for small carrier density may hint at an unconventional pairing mechanism beyond the ordinary phonon-mediated pairing. In addition, zero-field \( \mu\text{SR} \) did not provide evidence of broken time-reversal symmetry in the superconducting state. These features are consistent with the theoretically proposed topological superconducting state in \( \text{Sr}_3\text{SnO} \), as well as with ordinary \text{s-wave} superconductivity.

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I. INTRODUCTION

Antiperovskite (inverse perovskite) oxides \( A_2\text{BO} \) are materials crystallizing in the same structure as the ordinary perovskite oxides, but with the reversed positions of the metal and oxygen \([1]\); in antiperovskite oxides, oxygen is at the center of \( \text{OA}_3 \) octahedra. When \( A \) is an alkaline-earth element and \( B \) is a group 14 element, one can expect the ionic configuration \((A^{2+})_3B^{2+}\text{O}^{2-}\) with a metallic \( B \) anion such as \( \text{Sn}^{4+} \) or \( \text{Pb}^{4+} \), which is rare in oxides. Indeed, this unusual metallic anion is directly observed in \( \text{Sr}_3\text{SnO} \) by recent studies of Mössbauer spectroscopy \([2,3]\).

There has been a number of investigations toward clarifying their peculiar electronic band structure \([2–18]\). Theoretical calculations suggest a band inversion between the valence \( B-p \) and conduction \( A-d \) bands in some antiperovskite oxides containing heavy elements such as \( \text{Ca}_3\text{PbO} \) and \( \text{Ba}_3\text{SnO} \). Due to this band inversion, these materials belong to the Dirac semimetals, or to the topological crystalline insulators with a metallic band structure in \( \text{Sr}_3\text{SnO} \) can lead to topological superconductivity \([9]\). Theoretical analyses suggested that the inverted band structure in \( \text{Sr}_3\text{SnO} \) can lead to topological superconductivity with a high winding number originating from electrons with a total angular momentum \( J = 3/2 \) \([15]\). In this theory, besides the ordinary \text{s-wave} superconductivity, unconventional superconductivities with a full gap and a point-nodal gap are suggested.

For investigations of the symmetry of the superconducting order parameter, the muon-spin relaxation/rotation (\( \mu\text{SR} \)) technique is a powerful tool. The London penetration depth \( \lambda \) can be evaluated from the muon-spin depolarization rate \( \sigma \), measured in the vortex state of a type-II superconductor. We can deduce the structure of the superconducting gap from the temperature dependence of \( \lambda \) at temperatures much below the superconducting critical temperature \( T_c \); in fully gapped superconductors, \( \Delta \lambda^{-2}(T) = \lambda^{-2}(0) - \lambda^{-2}(T) \) exhibits an exponential temperature \( T \) dependence, whereas in nodal superconductors this quantity exhibits a power-law temperature dependence at low temperature \([19]\). Moreover, time-reversal symmetry breaking of superconductivity can be detected by a change in the zero-field muon-spin depolarization rate \( \Lambda \), below \( T_c \), measured in zero magnetic field \([20]\).

In this paper, we report \( \mu\text{SR} \) results on the antiperovskite oxide superconductor \( \text{Sr}_3\text{SnO} \). We observe a clear increase of the transverse-field muon spin depolarization rate \( \sigma \) below \( T_c \), which exhibits exponential behavior at low temperatures, indicating the absence of nodes in the superconducting gap. The deduced London penetration depth for \( T \rightarrow 0 \) is

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320–1020 nm and the ratio $T_c/\lambda(0)^{-2}$ is comparable to those of high-temperature superconductors. This fact possibly indicates an unconventional pairing mechanism. We also performed $\mu$SR at zero field and did not detect a breaking of the time-reversal symmetry. These results are consistent with the theoretically proposed unconventional superconductivity belonging to the same symmetry as the Balian-Werthamer (BW) state [21], as well as with the ordinary $s$-wave superconductivity [15].

II. EXPERIMENT

A. Sample preparation and characterization

Polycrystalline samples of Sr$_{3-x}$SnO were prepared from Sr (Sigma-Aldrich, 99.99%) and SnO (Sigma-Aldrich, 99.99%). Three samples were used for the $\mu$SR studies: sample A for the measurements down to 1.5 K and samples B and C to 0.26 K. They were synthesized in the procedure same as method B described in Ref. [13]. We confirmed that the samples are dominated by Sr$_{3-x}$SnO [7] with the lattice parameters of $a = 0.51429(4)$ nm in sample A, $a = 0.51435(3)$ nm in sample B, and $a = 0.51421(4)$ nm in sample C. These lattice parameter values were extracted from the powder x-ray diffraction (PXRD) patterns, as described in our previous report [2]. Although it is not easy to determine the actual composition of the samples accurately, we expect that $x$ is close to 0.5 because our previous study shows that the average Sr/Sn ratio is roughly 2.5 based on energy-dispersive x-ray spectroscopy [13]. The sample for the specific-heat measurement (sample D) was prepared from the stoichiometric ratio of Sr (Furuuchi, 99.9%) and SnO (Furuuchi, 99.9%) in the procedure same as method A described in Ref. [13].

B. Magnetic susceptibility and specific heat

Direct-current (dc) magnetization was measured with a commercial magnetometer using the superconducting quantum interference device (Quantum Design, MPMS-XL). We used powder samples that were sealed in plastic capsules inside an argon-filled glovebox. Alternating-current (ac) magnetic susceptibility was measured via the mutual inductance method using a lock-in amplifier (Stanford Research Systems, SR830) and using an adiabatic demagnetization refrigerator on a commercial apparatus (Quantum Design, PPMS) [22]. Heat capacity of a sintered chunk was measured using a $^3$He refrigerator on PPMS.

C. $\mu$SR

For $\mu$SR, we used pellets with a diameter of 10 mm. Measurements on sample A were performed at the GPS spectrometer ($\pi M3$ beamline) at the Paul Scherrer Institute (PSI) down to 1.5 K. The pellet was sealed in a polyethylene bag with a thickness of 0.1 mm under argon atmosphere in order to avoid direct contact to air. Measurements on samples B and C were carried out at the Dolly spectrometer ($\pi E1$ beamline) at PSI down to 0.26 K. Sample pellets were placed on 25-$\mu$m-thick Cu foils with grease (Apiezon, N Grease) to achieve a good thermal contact and sealed with a polyimide film (Du Pont, Kapton) under nitrogen atmosphere. The pellets of samples B and C were mounted on the cryostat using a portable glovebox with a continuous flow of nitrogen. The typical integration time of $\mu$SR was 1.5 h for each temperature and field condition.

III. RESULTS

A. Magnetic susceptibility and specific heat

First, we show the dc magnetic susceptibility of the Sr$_{3-x}$SnO samples in Fig. 1. All samples exhibit superconductivity at around 5.4 K. We call this primary superconducting phase the 5-K transition but a sharper 0.8-K transition than sample B.
seen in PXRD patterns. For sample A, the superconducting volume fraction evaluated from the dc susceptibility without the demagnetization correction reaches 11% at 1.8 K. In the case of sample B, the volume fraction is 9% at 5 mT and the fraction decreases with increasing the field. Considering that the fields applied during the measurements of sample B are stronger, we expect that the superconducting volume fraction of the 5-K phase is almost the same in both samples. The volume fraction smaller than 100% is attributable to the phase separation into multiple compositions with different amounts of Sr deficiency [2,16]. The onset of the transition decreases to 4.9 K at 30 mT. Figure 1(b) shows the ac susceptibility down to 0.2 K of samples B and C measured at zero dc field. Both samples exhibit additional superconducting transition at around 0.8 K as reported [9]. We hereafter call this second superconducting phase the 0.8-K phase. The origin of the 0.8-K phase is not clear yet, but the separation into two phases with slightly different amounts of the Sr deficiency may cause such splitting of the transitions [2]. From these results, we infer that sample B contains more 5-K phase than the 0.8-K phase, whereas sample C contains more 0.8-K phase. Sample D has a diamagnetic ratio between 5-K and 0.8-K phases similar to sample C, or sample D is dominated by the 0.8-K phase. Thus, comparing the \( \mu \)SR results of these samples, we may unveil the difference in the superconducting properties of the 5-K and 0.8-K phases.

Next, we present in Fig. 2 the electronic specific heat \( C_e \) divided by temperature \( T \) of sample D at low temperature. We observed a clear anomaly at around 0.85 K. We fitted the temperature dependence using the Bardeen-Cooper-Schrieffer (BCS) theory [24] and the phononic contribution, which is already subtracted in Fig. 2. We fixed \( T_c = 0.85 \) K to satisfy the entropy balance. We obtained the Sommerfeld coefficient \( \gamma = 5.67(3) \) mJ mol\(^{-1}\)K\(^{-2}\), the Debye temperature \( \Theta_D = 119.9(2) \) K, and the volume fraction of 23%. This fact evidences the bulk nature of the 0.8-K phase.

FIG. 2. Electronic specific heat divided by temperature \( C_e/T \). We observed a clear superconducting transition at 0.85 K with a volume fraction of 23%, evidencing bulk superconductivity. The solid curve represents the fitting with the BCS theory.

B. \( \mu \)SR at 30 mT

Figure 3 compares the \( \mu \)SR time spectra, recorded above and below \( T_c \), measured under an applied field of 30 mT for sample A. The presence of the randomly oriented nuclear moments causes a weak relaxation of the \( \mu \)SR signal above \( T_c \). The relaxation rate is enhanced below \( T_c \), which is caused by the formation of a flux-line lattice in the superconducting state, giving rise to an inhomogeneous magnetic field distribution. Assuming the Gaussian distribution for the probability \( n(B) \) that a muon stops at a position with a local flux density of \( B \), we fitted the data with the Gaussian cosine function

\[
A(t) = A(0) \exp \left(-\frac{\sigma t^2}{2}\right) \cos(2\pi \gamma B_0 t + \phi), \tag{1}
\]

where \( A(0) \) and \( \phi \) are the asymmetry and phase at \( t = 0 \), respectively, \( \gamma \) is the gyromagnetic ratio of muon, and \( B_0 \) is the mean flux density inside the sample. \( \sigma \) changes from 0.084(6) \( \mu s^{-1} \) at 7 K to 0.160(4) \( \mu s^{-1} \) at 1.6 K. This change in \( \sigma \) is readily seen in the raw data, namely the faster damping at 1.6 K. The change in the relaxation rate across \( T_c \) indicates that a large portion of the muons stopped at the superconducting region of the sample.

We measured \( \sigma \) as a function of the applied field at 1.5 K and 3.5 K (see Fig. 4). Each point was obtained by field cooling the sample from above \( T_c \) to each measurement temperature. First, \( \sigma \) strongly increases with increasing magnetic field until reaching a maximum at 15 mT and then above it continuously decreases up to the highest field (100 mT) investigated. The observed field dependence of \( \sigma \) implies that for a reliable determination of the penetration depth, the applied field must be just above the peak. Thus we measured the temperature dependence of \( \sigma \) at 30 mT.

The temperature dependence of \( \sigma \) for sample C under an applied field of 30 mT is shown in Fig. 5. An increase of \( \sigma \) was observed below \( T_c = 6 \) K. The relaxation rate is related
to the London penetration depth $\lambda(T)$ via
\[ \frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = 1 + \frac{1}{2\pi} \int_0^{\infty} \frac{d\epsilon}{E_k} \frac{\partial \epsilon}{\partial E_k} dk, \]
where $\sigma_{SC}(T) = \sqrt{0.00371 \times 2\pi g_\mu B \Phi_0 / \lambda^2(T)}$. The temperature dependence of $\lambda$ for the isotropic as well as anisotropic superconducting gaps can be calculated using the BCS theory:
\[ \frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = 1 + \frac{1}{2\pi} \int_0^{\infty} \frac{d\epsilon}{E_k} \frac{\partial \epsilon}{\partial E_k} dk, \]
where $f_k = (1 + \exp[(E_k/(\hbar k_B T))]^{-1}$ is the Fermi distribution function, $k_B$ is the Boltzmann constant, and $E_k = \sqrt{\epsilon^2 + \Delta^2_k(T)}$ is the quasiparticle excitation energy of the superconducting state with the kinetic energy $\epsilon$ relative to the Fermi energy $\epsilon_F$ and the superconducting gap $\Delta_k(T) = \Delta_0(T) g(\hat{k})$ [24]. The temperature dependence of $\Delta_0$ is obtained by solving the gap equation [26]
\[ \frac{1}{4\pi} \int_0^{\epsilon_c} \frac{d\epsilon}{E_k} \tanh \left( \frac{E_k}{2k_B T} \right) \gamma^2(\hat{k}) \epsilon d\epsilon d\hat{k} = \text{const}, \]
where $\epsilon_c$ is a cutoff energy. The constant value in the right-hand side is numerically obtained by substituting $T = T_c$ and $\Delta_0 = 0$. We simply set the cutoff energy and the Fermi energy as $\epsilon_c = 100k_B T_c$ and $\epsilon_F = 2000k_B T_c$. These values are compared to the Debye temperature $\epsilon_F = 35k_B T_c$ measured by $^{119}$Sn-Mössbauer spectroscopy [3] and $\epsilon_F = 2000k_B T_c$ estimated from the band structure calculation [12].

We calculated the temperature dependence of the relaxation rate assuming two superconducting gap structures on a spherical Fermi surface: $Y(\hat{k}) = 1$ for a fully gapped and $Y(\hat{k}) = \sqrt{k_x^2 + k_y^2}$ for a point-nodal state. While $\sigma$ saturates at low temperatures for a fully gapped state (the solid curve in Fig. 5), $\sigma$ continues to increase as lowering temperature for a point-nodal state (the dashed curve). Both fitting curves reasonably match the experimental result within the experimental error, but the root mean square error is smaller for the fully gapped state (1.2758) than for the point-nodal state (1.3933). Although it is theoretically suggested that the superconductivities with a full gap and point nodes have similar transition temperatures [15], this fitting analysis suggests that the fully gapped superconductivity such as the ordinary $s$-wave superconductivity or the topological superconductivity resembling the BW state is more likely to be realized in Sr$_x$SnO.

Figure 6 compares $\sigma(T)$ of all samples measured at 30 mT. We fitted $\sigma(T)$ assuming a fully gapped superconducting wave function. The resulting values of $T_c$ and $\lambda(0)$ are summarized in Table I. For all samples, we obtained higher $T_c$ than those in the magnetic measurements in Fig. 1, probably because $\mu$SR detects the superconducting transition of a small...
part of the sample with slightly higher $T_c$. Using the coherence length of $\xi(0) = 27$ nm evaluated from the upper critical field $[9]$, the Ginzburg-Landau parameter $\kappa$ is estimated to be $\kappa = \lambda(0)/\xi(0) = 32–38$, suggesting a strong type-II superconductivity.

$\lambda^{-2}$ is related to the superfluid density $n_S$ and the effective mass $m^*$ through the equation

$$\lambda^{-2} = \frac{\mu_0 n_S e^2}{m^*} \times \frac{1}{1 + \xi/l}, \quad (5)$$

where $\mu_0$ is the magnetic permeability in vacuum, $e$ is the elementary charge, and $l$ denotes the mean free path $[24]$. Since the density of states at the Fermi energy $D(\varepsilon_F)$ is evaluated to be $D(\varepsilon_F) = 1.203(7) \text{ eV}^{-1}$ per unit cell per spin from $\gamma$, the superfluid density is estimated to be $n_S = \Delta_0 \times D(\varepsilon_F) \approx 1.76 k_B T_c D(\varepsilon_F) = 1.7 \times 10^{25} \text{ m}^{-3}$. Therefore, the effective mass is calculated to be $m^* = \mu_0 n_S e^2\lambda^2/(1 + \xi/l) < \mu_0 n_S e^2\lambda^2$ in the clean limit. The effective mass normalized by the rest mass of the electron $m^*/m_e$ for each sample is listed in Table I. If the samples are in the dirty limit, $m^*$ is further reduced.

It has been known that $T_c$ and $\lambda(0)^{-2}$ of various materials exhibit a scaling behavior with the ratio $T_c/\lambda(0)^{-2}$ depending on the class of superconductors, as summarized in the Uemura plot $[27,28]$. For some classes of superconductors, $T_c$ is very high despite a small $n_S$. For example, hole-doped cuprates and iron-based superconductors satisfy the relation $T_c/\lambda(0)^{-2} \approx 4 \text{ K} \mu\text{m}^2 [27]$, whereas elemental superconductors exhibit much smaller $T_c/\lambda(0)^{-2}$ of less than 0.02 K $\mu\text{m}^2 [29]$. For the low-carrier superconductors SrTi$_{1-x}$Nb$_x$O$_3$ $[30]$ and YPtBi $[31]$, the ratio is $T_c/\lambda(0)^{-2} = 0.267–0.496$ and 2.0 $\mu\text{m}^2$, respectively. Interestingly, the obtained values of Sr$_{1-x}$SnO ($T_c = 6 \text{ K}$ and $\lambda(0) = 0.9–1 \mu\text{m}$) give a $T_c/\lambda(0)^{-2}$ ratio close to those of high-temperature or unconventional superconductors as presented in Table I and Fig. 7.

These values of $T_c/\lambda(0)^{-2}$ are between those of the high-temperature superconductors and of the topological superconductor Cu-intercalated Bi$_2$Se$_3$, for which $T_c/\lambda(0)^{-2} = 10 \mu\text{m}^2$ has been reported $[32]$. The relatively high $T_c$ for a small number of carriers implies an unconventional superconducting mechanism in Sr$_{3-x}$SnO.

We also performed analyses to take into account the volume fraction of the superconductivity. To do this, we fitted the time-dependent asymmetry with two Gaussian cosine functions

$$A \left[ \alpha \exp \left( -\frac{\sigma_1^2 t^2}{2} \right) + \exp \left( -\frac{\sigma_2^2 t^2}{2} \right) \right] \cos(2\pi \gamma_B t + \phi), \quad (6)$$

where $\sigma_1$ and $\phi$ are fixed to the values above $6 \text{ K}$ and $\alpha$ is fixed to 0.1 for samples A and B and to 0.04 for sample C corresponding to the volume fraction of 9% and 4% estimated from the susceptibility measurements in Fig. 1, respectively. The results of the two-component fits are summarized in Table I. The $T_c/\lambda^{-2}$ ratios are comparable to those of the low-carrier superconductors. This fact again indicates the relatively high $T_c$ for a small number of carriers in Sr$_{3-x}$SnO. Thus the qualitative conclusion is still valid even if the phase separation is taken into account.

### C. $\mu$SR at 5 and 0 mT

Figure 8 shows the relaxation rate of sample C at 5 mT. The jump at 3.0 K probably originates from the superconductivity of Sn. We did not detect the increase of the relaxation rate at 0.8 K, probably because the field modulation caused by the 0.8-K phase is too small at 5 mT even though the superconductivity originates from the bulk. The penetration depth of the 5-K phase at 5 mT is calculated to be $8.9(2) \times 10^{-7}$ m. This value is consistent with the one at 30 mT within the error. Since the magnetic field of 20 mT is reported to completely suppress the 0.8-K phase $[2]$, the upper critical field of the 0.8-K phase may not be large enough for $\mu$SR. Measurement

### TABLE I. Transition temperature $T_c$, penetration depth $\lambda(0)$, effective mass $m^*/m_e$, and $T_c/\lambda(0)^{-2}$ of Sr$_{3-x}$SnO extracted from $\mu$SR experiments, as resulting from fits with single and double Gaussian-cosine functions.

| Sample | Single Gaussian cosine: Eq. (1) | Double Gaussian cosines: Eq. (6) |
|--------|---------------------------------|----------------------------------|
|        | A | B | C | A | B | C |
| $T_c$ (K) | 6.3(2) | 6.2(2) | 6.2(2) | 5.6(3) | 6.05(7) | 6.04(5) |
| $\lambda(0)$ (nm) | 875(9) | 1018(11) | 878(10) | 316(23) | 340(11) | 322(15) |
| $m^*/m_e$ | 0.46 | 0.61 | 0.46 | 0.053 | 0.067 | 0.060 |
| $T_c/\lambda(0)^{-2}$ (K $\mu$m$^2$) | 4.8 | 6.4 | 4.8 | 0.56 | 0.70 | 0.62 |
FIG. 7. Plot of $T_c$ vs $\lambda(0)^{-2}$ (Uemura plot) for cuprate (black) [33], iron pnictide (brown) [34–38], low-carrier or unconventional (green) [30–32,39], transition-metal-dichalcogenide (orange) [19,40,41], elemental s-wave (blue) [29], and compound isotropic (purple) [42–45] superconductors. The solid and dashed lines represent the relations for the hole- and electron-doped cuprates and the transition metal dichalcogenides, namely $T_c/\lambda(0)^{-2} = 4$, 1, and 0.4 K $\mu$m$^2$, respectively [27,46]. The large six- and five-spoked asterisks represent the data for Sr$_{3-x}$SnO obtained from analyses with single and double Gaussian cosine functions, respectively.

of the field dependence of the relaxation rate below 0.8 K remains as a technical challenge in a future work.

Finally, we present in Fig. 9 the time dependence of the asymmetry at zero field to test a possible spontaneous time-reversal-symmetry breaking. The data were fitted with the exponential function $\Lambda \exp(-\Lambda t)$, and we obtained $\Lambda$ to be 62(3) ms$^{-1}$ at 6 K, 58(3) ms$^{-1}$ at 1.5 K, and 63(2) ms$^{-1}$ at 0.27 K. Thus temperature dependence of $\Lambda$ is quite weak, as shown in the inset of Fig. 9. The possible maximum spontaneous flux density due to superconductivity is evaluated to be $(\Lambda_{0.27 K} - \Lambda_{6 K})/(2\pi\gamma_B) = 7 \mu$T. This maximum value is seven times smaller than that observed in Sr$_2$RuO$_4$ (50 $\mu$T) [20]. Thus we conclude that the time-reversal symmetry is very likely to be preserved in the superconducting state of Sr$_{3-x}$SnO. The preserved time-reversal symmetry is consistent with the theoretically proposed topological phase as well as the ordinary s-wave superconductivity.

IV. CONCLUSION

In this paper, we reported a $\mu$SR investigation of the antiperovskite oxide superconductor Sr$_{3-x}$SnO. The temperature dependence of the muon spin depolarization rate, measured in the vortex state of Sr$_{3-x}$SnO at low temperatures, suggests a fully gapped superconductivity. The London penetration depth is estimated to be around 320–1020 nm
(depending on samples and type of analysis), and the ratio \( T_c/\lambda(0)^{−2} \) is similar to those of high-temperature superconductors or unusual low-carrier superconductors. This fact may hint at an unconventional superconducting mechanism in \( \text{Sr}_3\text{SnO} \). In addition, we did not detect any signs of broken time-reversal symmetry. These features are consistent with the theoretically proposed topological phase, namely superconductivity similar to the B phase of \( ^3\text{He} \), or with the conventional \( s\)-wave superconductivity.

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