Superconducting properties of Cu intercalated Bi$_2$Se$_3$ studied by Muon Spin Spectroscopy

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We present muon spin rotation measurements on superconducting Cu intercalated Bi$_2$Se$_3$, which was suggested as a realization of a topological superconductor. We observe a clear evidence of the superconducting transition below 4 K, where the width of magnetic field distribution increases as the temperature is decreased. The measured broadening at mK temperatures suggests a large London penetration depth in the $ab$ plane ($\lambda_{\text{eff}} \sim 1.6 \mu$m). We show that the temperature dependence of this broadening follows the BCS prediction, but could be consistent with several gap symmetries.

KEYWORDS: low energy muons spin spectroscopy, topological superconductivity, Cu$_x$Bi$_2$Se$_3$

Similar to topological insulators which have an insulating bulk and topologically protected conducting surface states, a topological superconductor features a bulk superconducting gap with gapless boundary states [1]. The quasiparticle excitations in these surface states can act as Majorana fermions, which are particles that are their own antiparticles [2]. In addition, it has been predicted that spin triplet $p$-wave superconductivity can be produced at the interfaces between an $s$-wave superconductor and a topological surface state [3–6]. A superconducting vortex at such an interface is expected to contain a Majorana bound state, that is the basic element in a recent proposal for fault-tolerant quantum computing [7]. The prototypical topological insulator Bi$_2$Se$_3$ has been reported to become superconducting upon carrier doping by either Sr, Nb or Cu intercalation [8–10]. It has been shown with angle resolved photoemission spectroscopy (ARPES) and torque magnetometry that the Dirac surface states of the parent material are inherited by Cu$_x$Bi$_2$Se$_3$ [11–13]. This and the strong spin orbit coupling in Bi$_2$Se$_3$ have made this material a promising candidate for a topological superconductor. Theoretically, it is proposed that odd-parity pairing, a full gap and an odd number of time reversal symmetry (TRS) invariant momenta enclosed by the Fermi surface are sufficient to make the superconductivity in Cu$_x$Bi$_2$Se$_3$ topologically nontrivial [14]. It is worth noting here that the strong spin orbit coupling prevents the usual characterization of the superconducting state with the symmetry of the gap function into $s$, $p$, $d$, ... wave [1].

Despite much work that has been dedicated to optimize the preparation procedure of Cu$_x$Bi$_2$Se$_3$ the observed diamagnetic shielding fraction remains below 70%, which indicates partial volume superconductivity [15–17]. First indications for topological superconductivity in Cu$_x$Bi$_2$Se$_3$ were reported by point contact spectroscopy, where a zero bias conductance peak (ZBCP) indicated the presence of Majorana fermions at the surface [18–20]. In contrast, no ZBCP was found with scanning tun-
nel microscopy [21]. Furthermore, ARPES measurements and the observation of quantum oscillation revealed that the Fermi surface becomes two-dimensional-like with increasing carrier concentration, thereby enclosing two TRS invariant momenta [13, 22]. Recently, new evidence of topologically non-trivial, nematic superconductivity has been found using NMR measurements [23]. The $^{77}$Sc Knight shift in the superconducting state exhibits a two-fold rotation symmetry when the external field is rotated in the $ab$-plane [23]. Similar features were also observed for the specific heat and the upper critical field [24]. Since the rotation symmetry of the crystal in the $ab$-plane is hexagonal, this is a strong indication that the superconductivity is topologically nontrivial.

In this paper we present results from muon spin rotation ($\mu$SR) measurements on Cu intercalated Bi$_2$Se$_3$ in the vortex state, where the magnetic field is applied along the $c$-axis of the crystal. From these measurements, we estimate the effective London penetration depth in the $ab$-plane at mK temperatures to be $\sim 1.6 \mu m$. Our measurements reveal that the superconducting gap in the bulk exhibits BCS temperature dependence, which could be consistent with various pairing symmetries.

The $\mu$SR measurements were performed on the High-field spectrometer (HAL-9500) at the Paul Scherrer Institute, Switzerland. In these measurements, fully spin polarized positive muons ($\mu^+$) are implanted into the studied sample. Muons decay into a positron with a lifetime of $\tau_{\mu} \sim 2.2 \mu s$, which is emitted preferentially in the direction of the muon’s spin at the time of decay. The ensemble average of the polarization of the implanted muons is proportional to the asymmetry, $A(t)$, which is monitored via the asymmetric decay using appropriately positioned detectors. $A(t)$ can be used to extract information regarding the internal magnetic fields at the muon site.

The measurements reported here were performed on high quality single crystals of Cu$_x$Bi$_2$Se$_3$. These samples were prepared using electrochemical Cu intercalation in single crystals of Bi$_2$Se$_3$. The samples are similar to those used in Ref. [25] and have a superconducting volume fraction of 40–60\% as estimated from magnetization measurements. The crystals were glued (using Apiezon grease) onto an 8 mm diameter silver rod, which serves as a cold finger of a dilution fridge. The magnetic field (applied along the $c$-axis of the Cu$_x$Bi$_2$Se$_3$ crystals) is generated using a superconducting warm bore magnet (0-9.5 Tesla).

![Fig. 1. Typical spectra in one of the detectors. There is no visible difference in the spectra at temperatures above and below the superconducting transition. The lines show fits with a Gaussian damped oscillation.](image)

In the normal state of Cu$_x$Bi$_2$Se$_3$ the internal magnetic field sensed by the implanted muons is primarily the applied magnetic field with the addition of a small contribution from the magnetic moments of the nuclei in the system. Below $T_c$, a vortex lattice is formed in SC Cu$_x$Bi$_2$Se$_3$ which
results in an inhomogeneous broadening of the field distribution sensed by the muons. Typical asymmetry spectra are shown in Fig. 1. The difference between measurements above and below $T_c$ is not visible by eye. To allow for an accurate fitting of this data a total of more than 100 million events have been collected at each temperature. The spectra were fitted to a Gaussian damped oscillation using the \texttt{Musrfit} software package [26]. The damping rate, $\sigma$, which reflects the width of the internal field distribution, as a function of temperature is shown in Fig. 2. We detect a clear increase in the broadening below $T_c$, with $\sigma$ saturating at very low temperatures. This is an indication that the superconducting state in Cu$_x$Bi$_2$Se$_3$ is fully gapped. The change in $\sigma$ is very small and on the limit of what is detectable with $\mu$SR. The broadening is expected to slightly decrease with increasing field, as the spacing between the vortices decreases [27]. This is in agreement to what we observe for measurements in different fields in Fig. 2.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{Fig_2}
\caption{The damping rate of the precessing asymmetry measured in Cu$_x$Bi$_2$Se$_3$ as a function of temperature at 10 mT (circles) and 50 mT (squares). The solid lines show a fit of an anisotropic gap with a BCS temperature dependence.}
\end{figure}

In the limit of a small applied field, $\sigma$ is related to the London penetration depth in a superconductor, such that [28]

$$\sigma_{sc}(T) = \sqrt{\sigma(T)^2 - \sigma_{ns}^2} = 0.0609\gamma_{\mu}\Phi_0/\lambda_{\text{eff}}^2(T),$$  \hspace{1cm} (1)

where $\gamma_{\mu} = 2\pi \times 135.5$ MHz/T is the gyromagnetic ratio of the muon, $\Phi_0$ is the magnetic flux quantum, $\sigma_{ns}$ is the broadening in the normal state determined above $T_c$ and $\lambda_{\text{eff}}$ is the effective London penetration depth under the assumption that the whole sample is superconducting. The temperature dependence of $\lambda_{\text{eff}}$ can be modelled within BCS theory [29]. The results of fits of this temperature dependence for various gap symmetries are summarized in Table I. Due to the large statistical error on the damping rate, with regard to the change at the superconducting transition, all listed gap symmetries are consistent with the observed signal. The inferred size of the superconducting gap is in agreement with the 0.6 meV reported by other techniques [11, 18, 19, 30]. The penetration depth is found to be $\lambda_{\text{eff}} \approx 1.6 \mu$m (for the measurements in 10 mT, which is closer to the small field limit assumed in Eq. 1). Note that this value will be reduced if the assumption of a fully superconducting sample is removed. The best fit results in terms of the reduced $\lambda_{\text{req}}^2$ are obtained for a full, anisotropic gap. This is consistent with the recently proposed fully gapped $\Delta_{4y}$ case, where the superconducting gap has minima along the $k_x$ direction [24].
Table I. Results of fits to different gap symmetries using a BCS temperature dependence. The small $\chi^2_{\text{red}}$ show that all models are consistent with the observed increase in $\sigma$.

| Model                        | applied Field [mT] | Gap [meV]     | $\lambda_{\text{eff}}$ [$\mu$m] | $\chi^2_{\text{red}}$ |
|------------------------------|--------------------|---------------|----------------------------------|-------------------------|
| isotropic ($s$-wave) gap     | 10                 | 0.64(9)       | 1.61(3)                          | 1.74(6)                 | 0.208 |
|                              | 50                 | 0.5(1)        |                                  |                         |       |
| anisotropic gap              | 10                 | 0.9(3)        | 1.59(3)                          | 1.71(4)                 | 0.052 |
|                              | 50                 | 0.8(5)        |                                  |                         |       |
| nodal ($d$-wave) gap         | 10                 | 1.2(3)        | 1.58(3)                          | 1.68(7)                 | 0.070 |
|                              | 50                 | 1.0(5)        |                                  |                         |       |

In conclusion, we have characterized the superconducting state of Cu$_x$Bi$_2$Se$_3$ with $\mu$SR. We find a very long effective penetration depth in the $ab$-plane, $\lambda_{\text{eff}} \approx 1.6 \mu$m, which is on the limit of what can be detected by $\mu$SR. A fit to a full and anisotropic gap to the temperature dependence of the superconducting state gives the lowest $\chi^2_{\text{red}}$. However, an isotropic or nodal gap cannot be ruled out.

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