Pairing Symmetry and Magnetic Relaxation in Topological Superconductor CuₓBi₂Se₃

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Abstract. Topological insulators are materials with a bulk-insulating gap, exhibiting a quantum-Hall-effect-like behavior in the absence of a magnetic field. The experimental as well as theoretical study show Bi₂Se₃ has a single surface Dirac cone associated with the topologically protected surface state against time reversal symmetry. CuₓBi₂Se₃ is of particular interest because of the signature of superconductivity found at low temperatures. Here we report the growth and the observation of bulk superconductivity from dc magnetization measurements in a cylindrical single crystal of CuₓBi₂Se₃. The magnitude of the magnetization in the Meissner state is very small and the magnetic field dependence of the magnetization just above the lower critical field \( H_{c1} \) is very different from those of usual type II superconductors. We believe that superconductivity observed in CuₓBi₂Se₃ is consistent with the spin-triplet pairing superconductivity with odd parity. We also observed a rapid relaxation phenomenon of the diamagnetic magnetization, indicating the flexible motion of the vortices in that temperature and field regime.

1. Introduction

Recently physicists and material scientists are interested in a new type of material known as topological insulator due to its unusual property: a bulk insulating gap along with the conduction surface state. Topological insulators exhibit a quantum-Hall-effect-like behavior in the absence of a magnetic field. A large band gap (~ 0.3 eV) and a single surface Dirac cone associated with the topologically protected surface state against time reversal symmetry. CuₓBi₂Se₃ is of particular interest because of the signature of superconductivity found at low temperatures. Here we report the growth and the observation of bulk superconductivity from dc magnetization measurements in a cylindrical single crystal of CuₓBi₂Se₃. The magnitude of the magnetization in the Meissner state is very small and the magnetic field dependence of the magnetization just above the lower critical field \( H_{c1} \) is very different from those of usual type II superconductors. We believe that superconductivity observed in CuₓBi₂Se₃ is consistent with the spin-triplet pairing superconductivity with odd parity. We also observed a rapid relaxation phenomenon of the diamagnetic magnetization, indicating the flexible motion of the vortices in that temperature and field regime.
sample [13, 16, 17, 18]. Here we are presenting the growth and magnetization properties of single crystalline Cu$_x$Bi$_2$Se$_3$. We observe the bulk superconductivity with small anisotropy and a rapid relaxation in Cu$_x$Bi$_2$Se$_3$. We believe that the nature of vortex state in this superconductor is exotic with odd-parity, i.e., spin triplet-superconducting state owing to the strong spin-orbit coupling in the superconductor, very different from conventional or cuprate high-$T_c$ superconductors.

2. Experimental

Bi$_2$Se$_3$ and Cu$_{0.15}$Bi$_2$Se$_3$ single crystals were prepared by reacting pressed pellets of a thoroughly mixed powder of Bi, Se, and Cu sealed in an evacuated quartz tube at 1123 K for 24 hours and then cooled at a rate of 2.5 K/min. to 893 K following liquid nitrogen quenching. The silver colour platelet crystals were obtained and easily cleaved off from the ingots of the obtained single crystal. Good crystallinity is proved by the sharp $c$-axis (00l) Bragg reflections in x-ray diffraction. For the magnetization measurement, we have chosen a cylindrical shaped crystal (diameter ~ 6 mm and height ~ 1mm) of Cu$_{0.15}$Bi$_2$Se$_3$ single crystal. The dc magnetization was measured with a SQUID magnetometer (Quantum Design Inc., USA).

3. Results & Discussion

Figure 1 shows representative plots of ZFC (zero field cooled) & FC (field cooled) temperature variation of dc magnetization for the Cu$_{0.15}$Bi$_2$Se$_3$ crystal with $H \parallel$ [001] in fixed fields 5Oe, 50 Oe, and 250 Oe as a function of temperature. The ZFC data at 5 Oe show the clear diamagnetic signal from the normal to superconducting transition at ~ 3.64 K. The normal state is diamagnetic with $\chi_{dia} = -2.2 \times 10^{-6}$ emu/mol without significant temperature dependence up to at least room temperature. This behavior is nearly the same for the mother compound Bi$_2$Se$_3$ as well. The superconducting signal down to 2 K is not fully saturated, indicating the whole sample may not be superconducting, but a very small fractional part of the obtained sample may be superconducting down to the lowest achievable temperature in our experiment. The ZFC and FC curves do not follow the same path, showing the typical behavior for type-II superconductors with pinning.

In Fig. 2 (a) we show magnetization hysteresis loops (only part of the first quadrant is shown for clarity) recorded at 2 K with $H \parallel$ [001] and at 2 K & 4 K with $H \perp$ [001], respectively. From the initial portion of the $M-H$ curve, we determine the lower critical field at 2 K is ~ 5 Oe. $M-H$ loop is reversible above 1500 Oe. The signal is diamagnetic at very high field, consistent with isofield measurements. The in-plane measurement at 2 K is similar to the one for the out-of-plane and the difference is very little at low fields around the turnaround field value.

The magnetization sharply increases in a small range of the magnetic field just above the lower critical magnetic field $H_{c1}$ with the increasing external magnetic field and then finally merges with the normal state diamagnetic curve at $H_{c2}$. This behavior is very different from those of usual type II superconductors. The situation can be understood qualitatively as follows, the vortex current induces a non-uniform magnetic field $b(x)$ in the vortex. This magnetic field polarizes the spins of the spin-triplet pairs and produces a non-uniform spin magnetization $m(x)$ in the crystal. Along with the vortex current, the spin magnetization contributes to the magnetic flux $b(x)$ in the superconductor. The magnetic flux is given by a sum of the magnetic field and the spin magnetizations.
As the total magnetic flux of a single vortex is quantized, the vortex current is drastically affected by the spin magnetization and the current inversion occurs in some portion of the vortex. The current inversion leads to an attractive force between the vortices. The magnetization curve is strongly influenced by the attractive interaction as we observed the drastic increase of the magnetization just above $H_{c1}$. In Fig. 2 (b) we schematically show the variation of the magnetic field with distance to visualize the situation. The green dotted upper line and the red dotted lower line show the magnetic field for the spin-singlet and triplet-pairing superconductor, respectively. The magnetic field $h(x)$ is always positive for all the region of $x$ for the singlet case, indicating that the interaction force between the vortices is always repulsive. This is the usual magnetization curve for type II superconductors. In the case of triplet state, the negative part of $h(x)$ appears in its tail and exerts the attractive force between the vortices. The attractive force helps the vortices to enter into the crystal although the vortex density is small at the external magnetic field just above $H_{c1}$. This fact explains the experimental results that the magnetization drastically increases in the magnetic field range as seen in Fig. 2(a). When the external magnetic field is increased furthermore, the vortex density increases and the distance between the vortices become shorter. In this regime, the repulsive interaction begins to work as seen in Fig. 2(b), and then the increase of the magnetization becomes gradual in consistency with the experimental results shown in Fig. 2 (a). The results discussed above strongly support that the superconductivity in the topological superconductor Cu$_{0.15}$Bi$_2$Se$_3$ is due to the spin-triplet pairing with odd parity, not singlet pairing. We found small separation between the magnetization signal in the $M$-$H$ data between the in-plane and out-of plane measurements, pointing towards the anisotropy presents in the system and corroborating that the superconductivity is in bulk nature.

We investigate the magnetic relaxation experiments as a function of applied magnetic field by choosing a particular field value where the sample is in the superconducting region in the field-temperature phase space. The measurement method in our experiment is as follows, at first the sample was cooled from the normal state in zero field condition to the chosen temperature (2 K) and after that, the magnetic field was switched on at a constant rate of 100 Oe/s. The measurement is started when
the field reaches the desired value and is continued for 3600 s. Figure 3 shows the time variation of the magnetic moment plot at 2 K with \( H// [001] \) and \( H\perp [001] \) for 50 Oe. We noticed the rapid relaxation of the magnetization signal and then nearly after 100 second the magnetization signal is almost stable in the duration of the experiment. The observed fluctuations in the data are due to the rapid vortex motion in that temperature and field regime.

In summary, we have investigated the magnetization measurements in a single crystal of \( \text{Cu}_x\text{Bi}_2\text{Se}_3 \) and observed that the superconductivity occurs in the bulk and the difference between the magnetization curves for parallel and perpendicular to the \( c \)-axis is very small. We also noticed the rapid relaxation of the diamagnetic magnetization, indicates the flexible motion of the vortices. From the experimental data and theoretical understanding of the magnetic field evaluation of the magnetization just above \( H_{c1} \), we presume that the vortices are those in the superconducting state of spin-triplet pairing.

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