Additional Evidence for the Surface Origin of the Peculiar Angular-Dependent Magnetoresistance Oscillations Discovered in a Topological Insulator Bi$_{1-x}$Sb$_x$

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Abstract. We present detailed data on the unusual angular-dependent magnetoresistance oscillation phenomenon recently discovered in a topological insulator Bi$_{0.91}$Sb$_{0.09}$. Direct comparison of the data taken before and after etching the sample surface gives compelling evidence that this phenomenon is essentially originating from a surface state. The symmetry of the oscillations suggests that it probably comes from the (111) plane, and obviously a new mechanism, such as a coupling between the surface and the bulk states, is responsible for this intriguing phenomenon in topological insulators.

1. Introduction

Topological insulators [1-3] are an emerging class of materials that host a new quantum-mechanical state of matter [4-7] where an insulating bulk state supports an intrinsically metallic surface state that is “topologically protected”, meaning that it is stable against any disorder that does not break time-reversal symmetry. Recently, angle-resolved photoemission spectroscopy (ARPES) studies on a cleaved trigonal surface of Bi$_{1-x}$Sb$_x$ have revealed that the energy dispersions of its surface states possess the distinctive character to qualify this material as a topological insulator [8-10]. However, to directly probe the unique properties of the surface states and to elucidate whether they could really be exploited on a macroscopic level, transport and magnetic studies of high-quality single crystals are indispensable. Unfortunately, in “real-life” samples of topological insulators available today, there is always some bulk conductivity due to residual carriers, and separating the contributions from two-dimensional (2D) and three-dimensional (3D) states turns out to be challenging [11-13].

In this context, we have recently succeeded in observing both the de Haas-van Alphen (dHvA) oscillations [14] and the Shubnikov-de Haas (SdH) oscillations [15] in high-quality bulk single crystals of Bi$_{1-x}$Sb$_x$ alloy in the “insulating” regime (0.07 ≤ x ≤ 0.22), which is the first material to be known as a 3D topological insulator. These observations became possible by growing highly pure and homogeneous single crystals of this alloy and achieving the bulk carrier density of the order of 10$^{16}$ cm$^{-3}$. The dHvA and SdH oscillations signified a previously-unknown Fermi surface (FS) with a clear 2D character that coexists with a 3D bulk FS [14,15]. Since Bi$_{1-x}$Sb$_x$ is a 3D material, the observed 2D FS is naturally assigned to the “surface”, which could be internal surfaces such as twin boundaries.

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Furthermore, we have extended our measurements to the angular dependence of the magnetoresistance (MR), which was successfully applied to the studies of quasi-2D organic conductors in the 1980s and lead to the discovery of the celebrated angular-dependent magnetoresistance oscillations (AMRO) [16-18]. Intriguingly, in our angular-dependent MR studies, we found oscillatory angular dependences in both the resistivity $\rho_{xx}$ and the Hall resistivity $\rho_{yx}$ [15]. The oscillations observed at lower fields were elucidated to be essentially a manifestation of the SdH oscillations, whereas the ones observed at higher fields were obviously of novel origin [15]. We proposed that the latter originates from the topological surface state on the cleaved (111) surface of Bi$_{1-x}$Sb$_x$, where the coupling between the surface and bulk states are probably playing a key role in the peculiar MR oscillation phenomenon [15].

In this paper, we present additional evidence to demonstrate that the “high-field” oscillations are most likely due to the exposed surface, by directly comparing the oscillations before and after the sample surface was chemically etched.

2. Experimental

High-quality Bi$_{0.91}$Sb$_{0.09}$ single crystals were grown from a stoichiometric mixture of high-purity (6N) Bi and Sb elements by a zone melting method. The resistivity was measured by a standard four-probe method on a rectangular sample, where the current was directed along the C$_1$ axis. Continuous rotations of the sample in constant magnetic fields were used to measure the angular dependence of the MR within the trigonal-binary (C$_3$-C$_2$) crystallographic plane. Magnetic fields up to 16 T were applied using a dc superconducting magnet in a $^4$He cryostat. All the data shown here were taken at the lowest temperature of 1.5 K. To obtain a fresh surface on the Bi$_{1-x}$Sb$_x$ single crystals, we applied the following chemical etching procedure:

1) Keep for several minutes in the 1:4:9 mixture of HNO$_3$, CH$_3$COOH, and H$_2$O.
2) Rinse in distilled water.
3) Keep for several minutes in the diluted (50 mol-%) HCl.
4) Rinse in distilled water and dry.

3. Results

3.1. Angular-Dependent MR Oscillations

Figures 1(a) and 1(b) show the angular dependences of the transverse MR and the Hall resistivity,
respectively, measured in magnetic fields rotated within the trigonal-binary \((C_3-C_2)\) plane perpendicular to the current. The magnetic-field strength was kept constant during each rotation. As can be seen in Figs. 1(a) and 1(b), both \(\rho_{xx}\) and \(\rho_{yx}\) present an oscillatory behavior as a function of the rotation angle \(\theta\). As we have elaborated in our previous paper [15], two different types of oscillations can be distinguished: The first type [though not very clearly seen in Fig. 1] consists of oscillations appearing at lower fields, while the second one becomes prominent at higher fields \((B \geq 10 \ T)\). The former “low-field” oscillations were demonstrated [15] to be essentially due to the SdH oscillations originating from the 2D state residing on the \((2,-1,-1)\) plane, which was unambiguously observed in the \(\rho_{xx}(B)\) data for magnetic-field sweeps in fixed magnetic-field directions.

The focus of the present paper is the “high-field” oscillations, which develop on a smooth field-dependent background coming from the anisotropy of \(\rho_{xx}(B)\) along the different axes. An example of the fitting of the background for \(B = 14 \ T\) is shown by the dashed line in Fig. 1(a). Because of a large MR background in strong magnetic fields, only largest peaks in \(\rho_{xx}(\theta)\) can be clearly seen in the raw data [Fig. 1(a)].

The \(\rho_{yx}(\theta)\) data shown in Fig. 1(b) also present pronounced angular-dependent oscillations at high fields. The “background” for \(\rho_{yx}(\theta)\) is simply the angular dependence of the Hall effect, \(R_H B \cos \theta\), where \(R_H\) is the Hall coefficient. As can be clearly seen in Fig. 1(b), low-filed \(\rho_{yx}(\theta)\) data follow this expected angular dependence very closely (we use \(R_H = -37 \ \text{cm}^3/\text{C}\), obtained from the Hall measurements), and the large deviation from this simple behavior is observed only in magnetic fields above 10 T. Figure 2 shows “pure” oscillations in \(\Delta \rho_{yx}(\theta)\) after subtracting the \(R_H B \cos \theta\) contribution from \(\rho_{yx}(\theta)\). One can clearly see a set of peaks, which are marked by short vertical ticks in Fig. 2. They are symmetric with respect to the \(C_3\) axis and show a rather complicated magnetic-field dependence.

3.2. Surface-Condition Dependence

The data shown in Figs. 1 and 2 were taken on a refreshed surface immediately after chemically etching the sample. To investigate the effect of etching, we had conducted the same angular-dependent MR measurements on the same sample before etching. Incidentally, this sample was characterized 7 months before those measurements, and it was kept in a desiccator, exposing its surface to dry air for 7 months. Our intention was to see how such an “old” sample behaves. Intriguingly, the angular-dependent MR oscillations were smeared, as shown in Fig. 3 for \(\rho_{xx}\). Figure 4 presents a direct comparison of the oscillations in \(\rho_{xx}\) and \(\rho_{yx}\) before and after etching. It is evident that the refreshed surface yields more pronounced oscillations, which gives direct evidence that the peculiar angular-
dependent oscillations are essentially originating from a surface state.

4. Discussions
In the “high-field” oscillations, an important feature is that the amplitude of the peaks weakens as the magnetic field is rotated away from the C_3 axis, which is somewhat reminiscent of the behavior of the ordinary AMRO in quasi-2D systems if the conduction planes lie perpendicular to the C_3 axis [16-18]. Thus, it is probable that the “high-field” oscillations are coming from the (111) plane (which is perpendicular to the C_3 axis), where surface states are seen in photoemission [8-10] and tunnelling [19] experiments. Another distinguishable feature of the “high-field” oscillations is that they survive up to rather high temperatures [15]. For example, even at 40 K there are still visible traces of oscillations while the SdH oscillations are already gone at this temperature [15], which is reminiscent of the behaviour of the ordinary AMRO in quasi-2D systems. In spite of these similarities to the quasi-2D AMRO, the peak positions of the “high-field” oscillations apparently shift with the magnetic field, which is not expected for the ordinary AMRO. Moreover, the existence of a finite coupling between

Figure 3. Angular dependences of \( \rho_{xx} \) measured in the trigonal-binary (C_3-C_2) plane on the 7-month-old surface of the same Bi_{0.9}Sb_{0.09} sample before etching. Note that the angular-dependent MR oscillations are smeared compared to those shown in Fig. 1(a).

Figure 4. Direct comparison of the oscillations in \( \rho_{xx} \) and \( \rho_{yx} \) before and after etching the sample surface, measured on the same Bi_{0.9}Sb_{0.09} sample. It is evident that the refreshed surface yields more pronounced oscillations, which gives evidence for the surface origin of these oscillations.
conduction planes is essential for the quasi-2D AMRO [20], but there is no such inter-plane coupling for the surface states as long as the crystal is thick enough. Therefore, the observed “high-field” angular oscillations are a new phenomenon apparently specific to topological insulators.

5. Conclusion
We present detailed data on the angular-dependent MR oscillation phenomenon which was recently discovered in a topological insulator Bi$_{0.91}$Sb$_{0.09}$. Direct comparison of the data taken before and after etching the sample surface gives compelling evidence that this novel phenomenon is essentially originating from a surface state. The symmetry of the oscillations suggests that it probably comes from the (111) plane. In the surface state of a topological insulator, there is no “quasi two-dimensionality” that introduces a finite warping to the 2D cylindrical Fermi surface, whereas the existence of such a warping is essential for the ordinary AMRO to occur; therefore, it is likely that a new mechanism, such as a coupling between the surface and the bulk states, is responsible for this intriguing phenomenon in topological insulators.

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