In situ Magnetotransport Measurements in Ultrathin Bi Films: Evidence for Surface-Bulk Coherent Transport

Masaki Aitani, Toru Hirahara,* Satoru Ichinokura, Masahiro Hanaduka, Dongyoon Shin, and Shuji Hasegawa

Department of Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

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We performed *in situ* magnetotransport measurements on ultrathin Bi(111) films [4–30 bilayers (BLs), 16–120 Å thick] to elucidate the role of bulk or surface states in the transport phenomena. We found that the temperature dependence of the film conductivity shows no thickness dependence for the 6–16 BL films and is affected by the electron-electron scattering, suggesting surface-state dominant contribution. In contrast, the weak antilocalization effect observed by applying a magnetic field shows clear thickness dependence, indicating bulk transport. This apparent inconsistency is explained by a coherent bulk-surface coupling that produces a single channel transport. For the films thicker than 20 BLs, the behavior changes drastically which can likely be interpreted as a bulk dominant conduction.

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Dirac particles have become one of the hot topics in condensed matter physics due to the emergence of graphene and topological insulators [1,2]. However, it was noticed already back in 1964 that the Hamiltonian describing the electrons in bismuth (Bi) has the Dirac form and this is the first realization of Dirac fermions in solid state physics [3]. In fact, Bi is one of the most extensively studied materials showing a variety of peculiar properties such as the three-dimensional fractional quantum Hall effect [4].

Recently, it has been shown by angle-resolved photoemission spectroscopy measurements (ARPES) that Bi has surface states with a large Fermi surface, which is in contrast to the semimetallic nature of the bulk [5,6]. It was argued that these surface states do show up in the physical properties of Bi [7–10]. However, these measurements were performed in air and it is questionable whether or not the surface states are still present. In fact, *in situ* measurements showed that the surface state conductivity can be changed easily [11–13]. Magnetotransport measurements have also been performed *in situ* for Bi films and reported to have clarified the surface-state contribution on the film transport around 10 K [13–15]. However, it is still not clear whether the surface and bulk states can be treated separately or interact with each other in the transport, which has become a crucial issue in the study of topological insulator surface states. Several experimental and theoretical works have shown that in the doped topological insulators, the weak antilocalization (WAL) phenomena can be described by a coherent single channel transport due to the bulk-surface scattering [16–19]. Although such behavior can be an obstacle to observing the peculiarity of the topological surface states, it may be an advantage for exploring intriguing conductivity properties that arise due to this bulk-surface coupling in Bi. Especially, there should be some influence on the spin Hall effect of Bi [20–22]. Although previous works mainly considered the bulk states, it should be important to include the surface states which should exhibit additional contribution based on the Rashba spin-split band structure [23,24].

In the present Letter, we performed magnetotransport measurements on single-crystalline Bi(111) films from 0.8 to 8 K and applied magnetic field as high as 7 T perpendicular to the surface. The temperature dependence of the film conductivity changed from a metallic behavior to an insulating one at 4 K due to the electron-electron interaction, which did not quantitatively change for the thickness from 6 to 16 bilayers (BL, 1 BL = 3.9 Å). This suggests that the surface-state contribution is dominant in the film transport. On the other hand, we observed WAL behavior which showed clear thickness dependence in terms of phase coherence, suggesting that the bulk states dominate the film transport. This seemingly contradictory behavior can be understood by considering the bulk-surface scattering that leads to a formation of a single coherent transport channel. The films thicker than 20 BLs exhibit a qualitatively different behavior which should be dominated by the bulk transport.

The experiments were performed *in situ* in ultrahigh vacuum with our micro-four-point-probe conductivity measurement system, in which the sample and the probe can be cooled down to 0.8 K and a magnetic field as high as 7 T can be applied perpendicular to the surface [25]. A reflection high-energy electron diffraction (RHEED) system was used for sample fabrication. A clean Si(111)-(7×7) surface was prepared on an n-type substrate (P-doped, 1–10 $\Omega \cdot$ cm at room temperature, RT) by a cycle of resistive heat treatments. Then Bi was deposited on the 7×7 surface at RT followed by annealing at ~380 K. Such a procedure results in the formation of high-quality, single-crystalline, epitaxial

Bi(111) films thicker than 6 BLs (25 Å) as reported in Refs. [26,27]. Below 6 BLs, the Bi films grow in the {012} direction with rotational disorder [26]. So the 4 BL film in the following have different atomic and electronic structures compared to the thicker films.

Figure 1(a) shows the normalized magnetoresistance (NoMR) $\{\rho(B)-\rho(0)\}/\rho(0)$ at 0.8 K as a function of the magnetic field for ultrathin Bi(111) films (4, 6, 9, 12, 16, and 20 BLs thick), where $\rho(B)$ is the two-dimensional resistivity under the magnetic field B. All of the films show positive magnetoresistance, but three different behaviors can be noticed. (i) For the films of 6–16 BLs, the NoMR increases nearly quadratically (classical magnetoresistance) with hardly any thickness dependence. (ii) For the 20 BL film, the NoMR is much more pronounced reaching nearly 70% at 7 T and has linear dependence at high fields. Finally, (iii) the behavior of the 4 BL film is quantitatively different showing a negative curvature, probably due to the different atomic structure as mentioned above. Thus it is clear that a different type of conduction is taking place by slightly changing the thickness even for the same single-crystalline Bi(111) films [(i) and (ii)].

The inset in Fig. 1(a) shows the magnified view of the data for low fields. There is a sharp cusp near the zero field

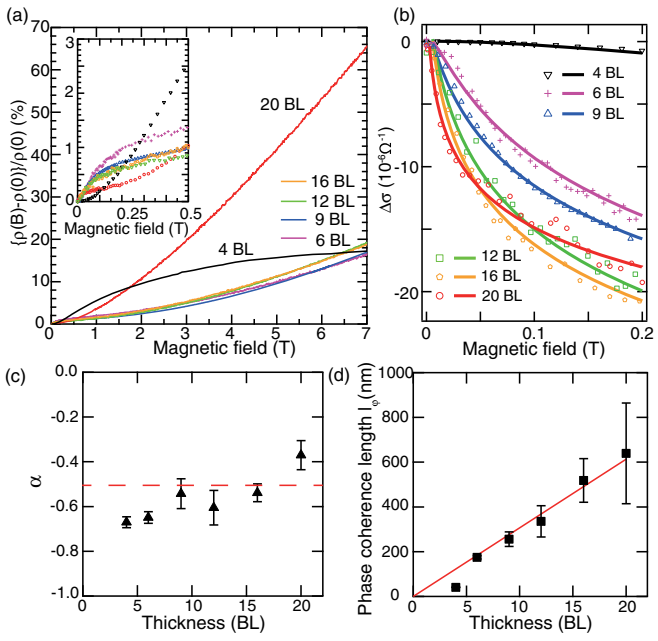

FIG. 1 (color online). Normalized magnetoresistance $\{\rho(B)-\rho(0)\}/\rho(0)$ at 0.8 K as a function of the magnetic field for ultrathin Bi(111) films with various thicknesses. The inset shows the magnification for small magnetic field. (b) WAL analyses (conductivity change as a function of the magnetic field) from the raw data of (a). The solid lines show the fitted curves to Eq. (1). (c), (d) Thickness dependence of α (c) and l_ϕ (d) in Eq. (1), respectively. The dashed line in (c) shows the expected value for the single channel WAL and the solid line in (d) shows the fitted curve to a linear function.

as has often been reported for Bi [13,14] and other Bi-based compounds [18,28]. This can be attributed to the WAL effect. By converting the resistivity to conductivity [29], we can fit the data with the Hikami-Larkin-Nagaoka (HLN) formula expressed as

$$\Delta\sigma = \sigma(B)-\sigma(0) = \frac{\alpha e^2}{2\pi^2\hbar}\left[\psi\left(\frac{1}{2}+\frac{\hbar}{4el_\phi^2 B}\right)-\ln\left(\frac{\hbar}{4el_\phi^2 B}\right)\right], \quad (1)$$

with α and l_ϕ (phase coherence length) as the fitting parameters (ψ is the digamma function). Figure 1(b) plots $\Delta\sigma$ together with the results of the HLN fitting [30]. The derived values of α and l_ϕ are plotted in Figs. 1(c) and 1(d), respectively. The value of α should ideally be -0.5 for a single channel WAL, but the obtained values deviate slightly; for the 4–16 BL films, it ranges from -0.52 to -0.64 whereas for the 20 BL film, it is -0.35, suggesting that something has changed for the thickest film. Such deviation from the ideal value has been previously reported for topological insulators [17,18]. We will discuss this point in detail later. The value of l_ϕ becomes larger as the film thickness increases. Especially, there is a simple relation $l_\phi = 75d$ (nm, d is the actual thickness) for the 6–20 BL thick films as shown by the solid line in Fig. 1(d). Therefore, it suggests that l_ϕ is determined by the film thickness in these films which also indicates that the bulk carriers are involved in the WAL effect although the surface states have a much higher carrier density [6,11].

In order to gain more insight into the contribution of the bulk or surface states in the WAL, we have changed the temperature and performed the analyses for the 6 and 20 BL films which are shown in Figs. 2(a) and 2(b), respectively [31]. The deduced values of α and l_ϕ are plotted in Figs. 2(c) and 2(d). We can see from Fig. 2(c) that there is a systematic difference in the α value between the 6 and 20 BL films, which is also indicated in Fig. 1(c); the absolute value for the 20 BL film ($\alpha \sim -0.2$ to -0.3) is smaller than that for the 6 BL film ($\alpha \sim -0.5$ to -0.6) in the whole temperature range. In Fig. 2(d), it can be seen that l_ϕ decreases by raising the temperature. Furthermore, the obtained data can be fitted to $l_\phi \propto T^{-P/2}$, which gives $P = 1.04$ for the 6 BL film. For the 20 BL film, the data cannot be fitted by a single line and $P = 1.08$ (2.23) is obtained for 0.8 K $< T <$ 2 K (2 K $< T <$ 4 K). From these analyses, it is possible to determine the phase decoherence scattering mechanism; namely, $P = 2$ is expected for electron-phonon (e-ph) scattering in two dimensions, while $P = 1$ for the electron-electron (e-e) scattering [32,33]. Thus we can say that the scattering that determines l_ϕ is e-e scattering for the 6 BL film, while it changes from e-e to e-ph scattering as the temperature is raised above 2 K for the 20 BL film. Therefore, Fig. 2 clearly shows that the transport mechanism changes drastically as the film thickness is increased.

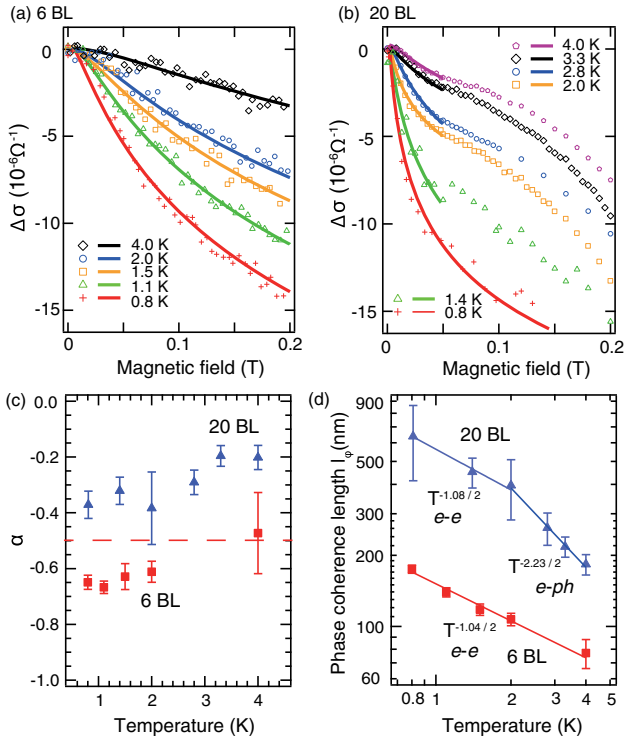

FIG. 2 (color online). (a), (b) The temperature dependence of WAL for the 6 (a), and 20 BL (b) Bi(111) film, respectively. The solid lines show the fitted curves to Eq. (1). (c), (d) Temperature dependence of α (c) and l_ϕ (d) for the 6 and 20 BL films. The dashed line in (c) shows the expected value for the single channel WAL and the solid line in (d) shows the fitted curve to $l_\phi \propto T^{-P/2}$.

To further investigate the intriguing difference between the thin and thick Bi(111) films, we have performed temperature dependent conductivity measurements at zero field, as shown in Fig. 3(a). The raw data in the inset of Fig. 3(a) show that there is significant thickness dependence concerning the absolute value, but the temperature dependence is quite weak for all the film thicknesses. However, upon close inspection, we notice that the films above 6 BLs basically behave metallically from 4 K to 8 K (conductivity increases by lowering the temperature) due to the *e-ph* scattering, but then become insulating below 4 K (conductivity decreases by lowering the temperature). To emphasize this temperature dependence, we have plotted $\sigma - \sigma_{2.5\,\mathrm{K}}$ in Fig. 3(a). Surprisingly, we notice that the curves for the 6, 9, 12, and 16 BL film exhibit quantitatively identical behavior. This absence of thickness dependence strongly suggests that this behavior originates from the surface states. For the 4 and 20 BL films, although the insulating behavior at the lowest temperature is the same with the 6–16 BL films, there is qualitative difference at higher temperatures; the 4 BL film shows insulating behavior throughout the whole temperature range from 0.8 to 8 K, whereas for the 20 BL film, the metallic temperature dependence above 4 K seems to be much stronger (the slope is steeper) compared to that found for the 6–16 BL films. In fact, the metallic behavior is seen until 2 K. This is consistent with the behavior of l_ϕ shown in Fig. 2(d). We have also performed the temperature dependence for the 30 BL film and the data shows quantitative consistency with that of the 20 BL film. Therefore, Fig. 3(a) again suggests that there is a clear difference in the transport mechanism between the thick and thin films.

In order to elucidate the origin of the insulating behavior found below 4 K (2 K for the 20 and 30 BL films), we have measured the temperature dependence while applying a magnetic field of 0.1 and 1 T for the 16 BL film as shown in Fig. 3(b). It can be noticed that the slope of the insulating part becomes steeper when the magnetic field is applied. Basically, all the curves can be fitted to $\Delta\sigma \propto (-\kappa e^2/2\pi^2\hbar)\ln T$. At zero field, κ is nearly 0.5 with hardly any thickness dependence, as shown in Fig. 3(c). When the magnetic field is applied, κ becomes $\sim$1.1 [Fig. 3(d)]. This can be understood as follows. At first glance the, $\ln T$ correction should be a result of the weak localization (WL) behavior. However, we have seen in Figs. 1 and 2 that instead of WL, WAL is observed due to the strong spin-orbit coupling (SOC). But the WAL should have a positive contribution (conductivity should increase as the temperature is lowered), which is opposite to our finding in Fig. 3. Therefore, we need to think about alternative factors that can cause this negative correction, which is the *e-e* interaction effect [34,35]. The *e-e* effect overcomes the WAL effect to produce the negative contribution in Fig. 3(a) at zero field [36]. When the magnetic field is applied, the WAL is suppressed, as we have seen in Figs. 1 and 2. Thus only the *e-e* interaction comes into play which results in the larger conductivity decrease. The nearly identical value of κ observed for $B = 0.1$ and 1 T is due to the strong SOC of Bi and $\kappa \simeq 1$ is theoretically expected [18,34,37], which agree nicely with our observation of $\kappa \sim 1.1$. When $B = 0$, the combined effect of WAL and *e-e* interaction should lead to $\kappa = 1\text{–}1/2 = 1/2$, which is nearly equal to what we have found in Figs. 3(c) and 3(d). Thus the insulating behavior below 4 K can be safely ascribed to the *e-e* interaction effect.

Now let us discuss the surface and bulk contributions in more detail. The NoMR data in Fig. 1(a) shows that there was hardly any thickness dependence for the 6–16 BL films. Furthermore, there was no thickness dependence in the temperature dependence of the conductivity change also for the 6–16 BL films [Fig. 3(a)]. These two points strongly suggest that the surface state transport is dominant in these films. However, the analyses of the thickness dependence of WAL showed that l_ϕ is proportional to the film thickness for films thicker than 6 BLs [Fig. 1(d)], meaning this should originate from the bulk carriers. This apparent contradiction can be resolved when we consider that there is no distinction between the surface and the bulk; i.e., the surface and the bulk states are

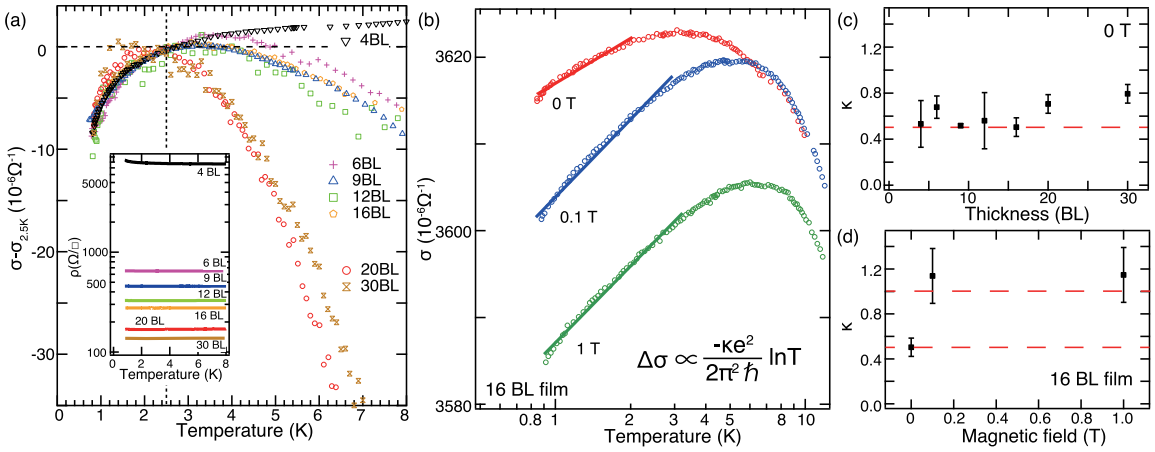

FIG. 3 (color online). Temperature dependence of the change in sheet conductivity with respect to the value at 2.5 K for Bi(111) films with various thicknesses. No magnetic field is applied. The inset shows the raw data of the resistivity as a function of the temperature. (b) Temperature dependence of the sheet conductivity for the 16 BL film when applying a magnetic field. The solid lines show the fitted curves to $\Delta\sigma \propto -\kappa \ln T$ for the data below 2 K. (c) Thickness dependence of κ from the data of (a). The dashed line shows the value expected (0.5) from the combination of electron-electron interaction (EEI) and WAL. (d) The magnetic field dependence of κ from the data of (b). The two dashed lines show the values expected from EEI and WAL ($\kappa = 0.5$) and EEI only ($\kappa = 1.0$).

coherently coupled and form a single conduction channel. This idea was originally proposed for topological insulator surfaces and is reflected in α of Eq. (1) [16]. Several experimental works actually showed that α changes from $-1/2$ to -1 by tuning the Fermi level [17–19]. This was explained as a transition from a state where the surface-bulk or the top-bottom surface scattering dominates (interchannel coupled state) to a regime where the top and bottom surface states contribute to the WAL separately. In our case, since the surface states overlap with the bulk states near the $\bar{\Gamma}$ and $\bar{M}$ points in the surface Brillouin zone [6], a surface-bulk scattering without the loss of coherence can be expected to take place. Furthermore, the existence of the bottom surface states has been indicated by spin ARPES measurements [38] and first-principles calculations [39]. All of these facts together with the analyses of $\alpha \sim -0.5$ [Figs. 1(c) and 2(c)] suggest that the surface-bulk or the top-bottom surface coupling can occur to produce a single-channel behavior.

Another interesting question to ask is what happens between the 16 and 20 BL films which show qualitatively different behavior both in the magnetic-field and temperature-dependent measurements. Namely, the NoMR of the 20 BL film shows a much larger value of 70% compared to the thinner films. Since single crystal Bi films are known to show a giant positive magnetoresistance [40], the transport in 20 BL film is most likely dominated by the bulk states. This is also corroborated by the fact that the magnetoresistance seems to be linear in B, a manifestation of the Dirac character of bulk Bi [41]. Another evidence of the bulk transport is the α, which is different for the 20 BL film (~ -0.3) compared to the other films [Fig. 2(c)]. It is shown in Ref. [16] that massive Dirac fermions can show WL behavior instead of the WAL even in the presence of strong SOC. Thus the WAL of the surface states may be compensated by the WL of the Bi Dirac bulk states and decrease $|\alpha|$. The third important point is that the phase decoherence process changes to *e-ph* scattering above 2 K for the 20 BL film instead of the *e-e* scattering [Fig. 2(d)], which is also reflected in the temperature dependence of the 20 and 30 BL films [Fig. 3(a)]. This probably reflects the fact that the Debye temperature is smaller at the Bi(111) surface (81 K) than the bulk value (119 K) [27].

However, our previous ARPES measurements showed that there is no drastic change of the band structure between 16 and 20 BL Bi(111) films [42]. One possible explanation for this discrepancy between ARPES and magnetotransport measurements may be that the phase-coherent transport phenomena is much more sensitive to the hybridization between the top and bottom surfaces as reported for Bi_2Se_3 films [17,43]. Therefore, a small gap which is undetectable with ARPES may still be present for the 16 BL Bi(111) film and is closed for 20 BLs.

In conclusion, we have performed *in situ* magnetotransport measurements on ultrathin Bi(111) films. We observed a thickness-dependent WAL effect as well as thickness independent *e-ph* and *e-e* interaction effects for the 6–16 BL films. This suggests a coherent coupling between the surface and bulk states. For the films thicker than 20 BLs, it is likely that the bulk channel dominates the film transport.

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*Present address: Department of Physics, Tokyo Institute of Technology, 2-12-1 Ookayama, Meguro-ku, Tokyo 152-8551, Japan.
hirahara@phys.titech.ac.jp

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