

Weak antilocalization and electron-electron interaction effects in Cu-doped Bi₂Se₃ films

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We investigate the low-temperature transport properties in Cu_xBi_{2-x}Se₃ films prepared by a hot-wall-epitaxy growth of Bi₂Se₃ layers on Cu-deposited substrates. We observe a positive magnetoresistance due to the weak antilocalization effect and a classical magnetoresistance that exhibits a power-law dependence on the magnetic field. The resistance increases logarithmically with lowering temperature regardless of the strength of the magnetic field. The electron-electron interaction effect is thus evidenced to be strong. While the magnitude of the weak antilocalization effect is in reasonable agreement with theory, the correction to the conductivity due to the electron-electron interaction effect is unaccountably larger than the theoretical prediction. The discrepancy may indicate that the contribution from the bulk state is as large as that from the surface states, at least, for the interaction effect.

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I. INTRODUCTION

The Dirac fermion surface states in topological insulators (TIs) are protected from nonmagnetic scattering by time reversal symmetry.¹⁻⁵ One may, therefore, conceive that the weak localization effect, which originates from the quantum interference between the forward and backward propagating waves,⁶ is absent in TIs.⁷⁻¹⁰ [Due to the strong spin-orbit coupling in TIs, the quantum interference would, to be precise, result in the weak antilocalization (WAL) effect].⁶ As it turns out, the π Berry phase specific to the Dirac fermions gives rise to a quantum correction to the conductivity whose temperature and magnetic-field dependencies are identical in form to those for the WAL effect.¹¹⁻¹⁴ (We thus henceforth refer to the Berry phase effect as the WAL effect.)

In thin layers of the three-dimensional TIs Bi₂Se₃ and Bi₂Te₃, positive magnetoresistance at weak magnetic fields was indeed observed at low temperatures.¹⁵⁻²¹ However, in spite of the WAL effect, the resistance increased as the temperature was lowered. Similar behavior implicating an insulating ground state was reported several decades ago for, for instance, Bi (Ref. 22) and Au-Pd (Ref. 23) films. The electron-electron interaction (EEI) effect was, therefore, speculated to be significant in Bi₂Se₃ and Bi₂Te₃ (Refs. 19 and 20), as has been established for the Bi and Au-Pd films.

Although the WAL and EEI effects were both assumed to be associated with the surface states, the assumption needs to be justified. That is, the Se and Te vacancies in Bi₂Se₃ and Bi₂Te₃ generate carriers, and so the bulk state also participates in the electrical conduction. The existence of the surface conductive states was demonstrated by the Aharonov-Bohm oscillations observed for Bi₂Se₃ nanowires.²⁴ As expected for a two-dimensional system, the magnetoresistance in thin layers attributed to the WAL effect was confirmed to depend on the normal component of the magnetic field.¹⁷ In addition, the WAL effect was more pronounced when the layers were thinner.²⁰ The surface states hence appear to be responsible for the WAL effect. In contrast, no such proof has been presented so far for the EEI effect. We emphasize that the contributions from the bulk state can also be two dimensional as the measurements were performed on thin films to take advantage of their large surface-to-volume ratio. We point out that Lükermann *et al.*,²⁵ for instance, associated the WAL

effect and the classical magnetoresistance in Bi films with, respectively, the bulk and surface states, opposite to the usual interpretation for TI films.

In this paper, we analyze the WAL and EEI effects observed in Cu-doped Bi₂Se₃ films. We find that the contribution from the EEI effect is too large to be accounted for by theory. This may be an indication that the quantum corrections originating from the surface and bulk states can be comparable in magnitude.

II. SAMPLE PREPARATION

The Cu-doped Bi₂Se₃ films were grown using a hot-wall-epitaxy method. Hot wall epitaxy was employed previously for producing Bi₂Se₃ and Bi₂Te₃ films at low cost for thermoelectric applications.²⁶ The growth in hot wall epitaxy takes place under thermodynamical equilibrium conditions.²⁷ As demonstrated in Ref. 28, the Bi atoms in the Bi₂Se₃ crystal are substituted by Cu atoms in the hot-wall-epitaxy process to the degree only limited by the supply of the Cu atoms. To utilize this enormous Cu incorporation, we grew Bi₂Se₃ layers on Si(001) substrates, where a Cu layer was deposited on the Si surface by sputtering prior to the growth. The purity of the source materials was 99.99 and 99.999% for Cu and Bi₂Se₃, respectively. The hot-wall-epitaxy growth of the Bi₂Se₃ layers was carried out at a substrate temperature of ~ 250 °C. The source temperature was 500 °C. In the following, we show results from a film whose thickness was about 80 nm after a 5-h growth.

The growth of the Cu-doped Bi₂Se₃ films is anticipated to take place as follows. The Bi₂Se₃ layer at the initial stage of the growth turns into CuSe crystallites as the Cu atoms from the predeposited layer completely substitute the Bi atoms.²⁸ As the growth continues, the amount of the Cu atoms becomes insufficient to replace all the Bi atoms. Consequently, a Cu_xBi_{2-x}Se₃ layer is eventually produced. The distribution of the Cu atoms in the Bi₂Se₃ layer is expected to be homogeneous due to the extremely long diffusion of Cu (Ref. 28). The diffusion length is expected to be, at least, an order of magnitude larger than the thickness of the Cu_xBi_{2-x}Se₃ layer that we investigate below.²⁹ The Cu atoms will be, therefore, diluted in the Bi₂Se₃ matrix when the growth further proceeds.

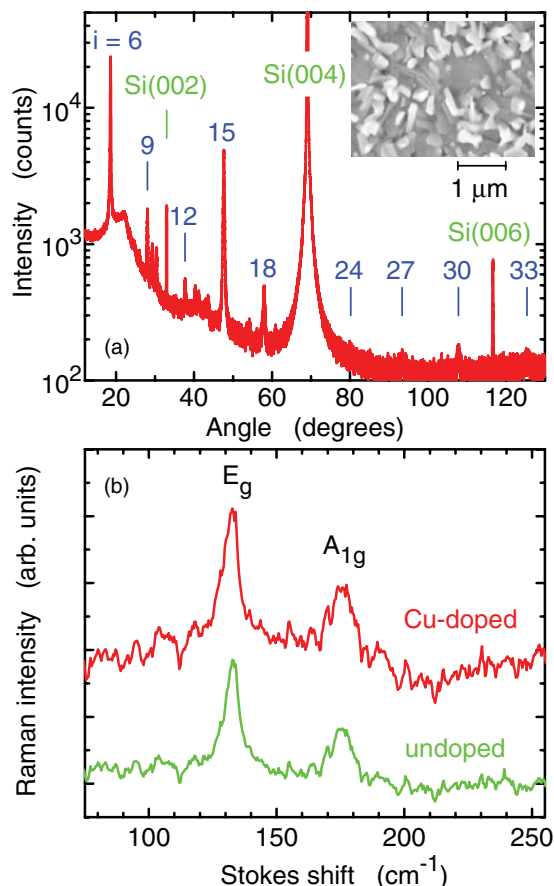


FIG. 1. (Color online) (a) ω - 2θ x-ray diffraction curve of a Cu-doped Bi_2Se_3 film grown on $\text{Si}(001)$. The main peaks are associated with the $(000i)$ reflections from Bi_2Se_3 and the (002) , (004) , and (006) reflections from Si. The c lattice parameter is estimated to be 2.8653 nm. The inset shows a scanning electron micrograph of the film surface. (b) Raman spectra of the Cu-doped Bi_2Se_3 layer (top curve) and undoped Bi_2Se_3 layer (bottom curve). The peaks are associated with the E_g and A_{1g} modes of Bi_2Se_3 . Curves are offset for clarity.

We show a scanning electron micrograph of the film in the inset of Fig. 1(a). The formation of the CuSe microcrystallites at the initial stage of the growth as well as the large lattice mismatch between the Bi_2Se_3 layers and the $\text{Si}(001)$ substrates are responsible for the grainy film morphology.³⁰ The substrate surface is covered entirely by a continuous $\text{Cu}_x\text{Bi}_{2-x}\text{Se}_3$ layer. Free-standing disks that were generated spontaneously during growth³⁰ are also seen to be present on the continuous layer. We emphasize that the free-standing disks are unlikely to play a significant role in the transport properties as they are attached to the underlying continuous layer merely at their base.

We have confirmed using the x-ray diffraction that the crystal structure of the layer is rhombohedral, as it should be for Bi_2Se_3 . (CuSe is a hexagonal crystal.) The ω - 2θ scan plotted in Fig. 1(a) indicates that the layer is predominantly (0001) oriented. The Cu content was estimated to be several percent by energy-dispersive x-ray spectroscopy. Importantly, the Bi content was found there to decrease with the Cu incorporation. Our anticipation that the Cu atoms were introduced substitutionally instead of intercalationally is hence

supported. We compare the Raman spectra obtained from the Cu-doped and undoped samples in Fig. 1(b). Due to the small Cu content, the spectra are indistinguishably similar. They thus provide other evidence that growing a Bi_2Se_3 layer on the predeposited Cu did not alter the crystal structure of Bi_2Se_3 .

III. TRANSPORT PROPERTIES

In Fig. 2, we show the dependence of the sheet resistivity ρ_{xx} and Hall resistivity ρ_{xy} on a magnetic field B applied perpendicular to the film at a temperature of $T = 0.3$ K. The transport coefficients were determined by the van der Pauw method to avoid surface degradations that may occur in the process of fabricating a Hall bar. For this reason, we also bonded Au wires directly to the film without preparing Ohmic contact pads. The size of the sample was approximately 5×2.5 mm. The resistances were measured using the lock-in technique with an excitation current of 10 nA. Negligible Joule heating was attested by the logarithmic temperature dependence of ρ_{xx} observed over the entire temperature range of measurements, as we will show below.

The magnetic-field dependence of ρ_{xy} is almost completely linear, suggesting that only a single type of electrons is involved in the transport process. We note that at least two types of electrons were identified in undoped films.³⁰ The mobility and concentration for the electrons in the bulk state are estimated to be $0.082 \text{ m}^2/\text{Vs}$ and $8.4 \times 10^{23} \text{ m}^{-3}$, respectively. The electron mobility is seen to be drastically reduced by the Cu incorporation in correspondence to the degradation in the crystallinity of the film evidenced by the small peak amplitude in Fig. 1(a).

Kim *et al.*²¹ pointed out a universal relationship between the electron concentration n and the layer thickness d . Assuming that this relationship, $n \propto d^{-1/2}$, is applicable also to our samples, the electron concentration in our samples is found to be about a factor of 2 larger for the $0.7\text{-}\mu\text{m}$ -thick undoped layer³⁰ and more than one order of magnitude smaller for the

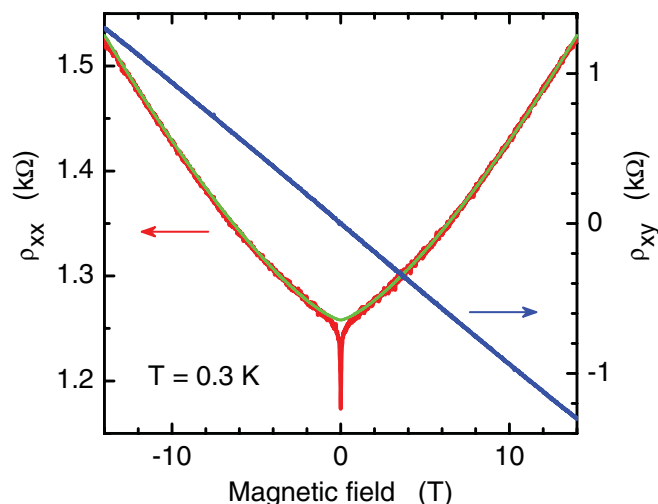


FIG. 2. (Color online) Dependence of the sheet resistivity ρ_{xx} and Hall resistivity ρ_{xy} in a $\text{Cu}_x\text{Bi}_{2-x}\text{Se}_3$ film ($x = 0.02$ – 0.03) on magnetic field B at a temperature of 0.3 K. The field was applied perpendicular to the film. The green curve shows a power-law behavior $\propto |B|^{1.37}$.

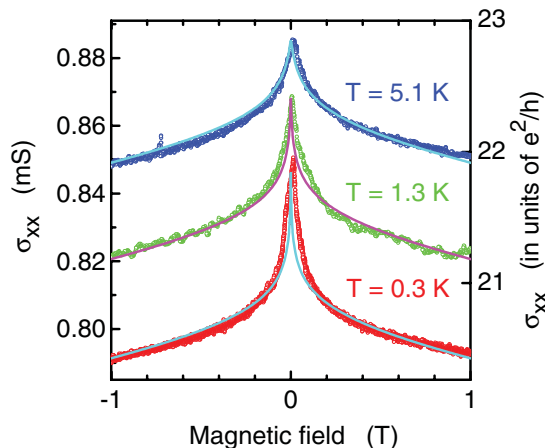


FIG. 3. (Color online) Low-magnetic-field behavior of the sheet conductivity $\sigma_{xx} = 1/\rho_{xx}$ at temperatures of $T = 0.3, 1.3,$ and 5.1 K. The solid curves show fits to Eq. (3). Here, we have taken into account the power-law dependence of the background magnetoconductance ($\propto B^{1.18}$).³⁴ The fit parameters are $\alpha = -0.55$ and $l_\phi = 0.3, 0.7,$ and $1.2 \mu\text{m}$ for $T = 5.1, 1.3,$ and 0.3 K, respectively.

Cu-doped layer. The small electron concentration manifests that the Cu atoms predominantly acted as acceptors. This is in agreement with the fact that substitutional Cu atoms on the Bi sites are acceptors as they carry a double negative charge.³¹ Owing to the reduced electron concentration in the bulk, the quantum effects associated with the surface states are considerably large in magnitude despite the relatively large film thickness.³² Moreover, we can avoid ambiguities arising from the participation of multiple types of carriers in analyzing the transport phenomena, which was not the case in some previous reports.¹⁹

The positive magnetoresistance in Fig. 2 contains two components, a sharp dip around zero magnetic field and a gradually varying background. As shown in Fig. 3, the temperature dependence is strong for the weak-field component, suggesting its quantum origin. The origin for the high-field component appears to be classical as the temperature dependence is negligibly small. However, the magnetic-field dependence for the high-field component is rather linear than parabolic. Similar magnetic-field dependencies intermediate between linear and parabolic behaviors were reported also by other groups.^{19,33} In fact, we find that the entire field dependence obeys a power law, as shown by the green curve in Fig. 2.³⁴ It may be noteworthy that a power-law dependence ascribed to the coexistence of multiple carriers was observed in MnAs films.³⁵ Influences from the surface states and/or the presence of additional carriers in the bulk state might be suggested although $\rho_{xy}(B)$ exhibited the linear dependence.

The quantum correction to the conductivity resulting from the π Berry phase is given at zero magnetic field as^{12,14}

$$\delta\sigma_L(T) = -\alpha \frac{e^2}{\pi h} \ln\left(\frac{\tau_\varphi}{\tau}\right) = \alpha p \frac{e^2}{\pi h} \ln\left(\frac{T}{T_L}\right), \quad (1)$$

where τ is the elastic scattering time and T_L is a characteristic temperature at which the quantum correction vanishes. The phase coherence time τ_φ typically varies with temperature T as $\tau_\varphi \propto T^{-p}$. The prefactor α was derived to be $-\frac{1}{2}$ for the TI

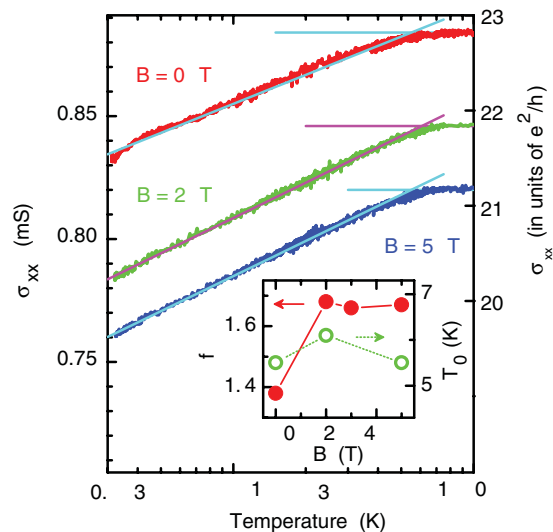


FIG. 4. (Color online) Temperature dependence of the sheet conductivity σ_{xx} in a 80-nm-thick $\text{Cu}_x\text{Bi}_{2-x}\text{Se}_3$ film ($x = 0.02-0.03$). The magnetic field applied perpendicular to the film was set to $B = 0, 2,$ and 5 T. The horizontal lines indicate the temperature-independent saturation conductivity values. Logarithmic temperature dependence is displayed by the inclined lines. Parameters f and T_0 determined by a fit to $[e^2/(\pi h)]f \ln(T/T_0)$ are plotted in the inset.

surface states.¹⁴ With respect to the weak localization effect, Eq. (1) corresponds to the limit of strong spin-orbit scattering.⁶

Although the conductivity should increase as the temperature is lowered for the WAL effect, the opposite was observed for the $\text{Cu}_x\text{Bi}_{2-x}\text{Se}_3$ film, as shown in Fig. 4. The EEI effect is, therefore, manifested to be dominant rather than the WAL effect. If we assume that the temperature dependence of the EEI effect for the TI surface states is identical to that for the conventional EEI effect, the correction to the conductivity in thin films is given by³⁶

$$\delta\sigma_1(T) = -\frac{e^2}{\pi h} \left(1 - \frac{3}{4}F\right) \ln\left(\frac{T}{T_1}\right), \quad (2)$$

where F is the screening factor and T_1 is the characteristic temperature for the EEI effect.

As one finds in Fig. 4, the conductivity changes logarithmically at low temperatures regardless of the strength of the magnetic field. We plot the parameters f and T_0 determined by a fit to $\delta\sigma_{xx}(T) = [e^2/(\pi h)]f \ln(T/T_0)$ in the inset of Fig. 4. The parameter f becomes independent of magnetic field for $|B| \geq 2$ T. As the magnetic-field dependence of the EEI effect is considerably weaker than that of the WAL effect, we assume that the almost constant f value in high magnetic fields is given by the EEI effect.^{22,23} The difference in f between the absence and presence of magnetic field is then attributed to the WAL effect, which is almost completely quenched at $|B| = 2$ T. We note that T_0 was roughly independent of magnetic field. The two effects appear to provide quantum corrections in almost the same temperature range (below 6 K), that is, $T_L \approx T_1$.

Having assigned the WAL and EEI contributions as we mentioned above, we obtain $\alpha p = -0.3$ from Eq. (1). For the dephasing by two- and three-dimensional electron-electron scatterings, $p = 1$ and $\frac{3}{2}$ are expected, respectively. The value of α evaluated from the $\ln T$ dependence is thus in

reasonable agreement with the value expected for the surface states. As a matter of fact, the value of $|\alpha|$ being somewhat smaller than the theoretical prediction is consistent with other measurements.^{17,19,20} Such agreement implies that only one surface of the film is relevant for the WAL effect as α should be doubled if the states at both surfaces of the film provide identical quantum corrections. The seeming absence of the WAL effect for one of the surfaces was a common finding in previous reports.^{15,17,19,20}

In contrast to the satisfactory agreement of the magnitude of the WAL correction with theory, the EEI effect is too large to be explained by Eq. (2). Specifically, we obtain $1 - \frac{3}{4}F = 1.67$ although the value of F should lie between 0 and 1. The large coefficient of the $\ln T$ term may suggest a contribution from the surface that does not seem to provide the WAL effect. The excess contribution may also originate from the bulk state. For the three-dimensional EEI effect, the temperature dependence is given by $\propto T^{1/2}$. However, the EEI effect becomes two dimensional if the thermal diffusion length $L_T = (\hbar D/k_B T)^{1/2}$, where D is the diffusion constant, exceeds the layer thickness. Assuming the effective electron mass to be $m^* = 0.15m_e$, we obtain L_T of the bulk state to be 95 nm at $T = 1$ K, which is comparable to the film thickness. Thus, the EEI effect in the bulk state may also be two dimensional.

We have found an anomalous behavior in the magnetic-field dependence of the WAL effect, which may be related to the puzzling temperature dependence. That is, the experimental data cannot be fitted by theory at low temperatures. The magnetoconductance for the π Berry phase effect is given by^{12,14}

$$\Delta\sigma_L(B) = \alpha \frac{e^2}{\pi h} \left[\psi \left(\frac{1}{2} + \frac{B_\phi}{B} \right) - \ln \left(\frac{B_\phi}{B} \right) \right], \quad (3)$$

where $\psi(x)$ is the digamma function and $B_\phi = \hbar/(4el_\phi^2)$ with l_ϕ being the phase coherence length. As shown by the solid curves in Fig. 3, the experimental data can be described well using Eq. (3) at $T = 5.1$ K. Here, α was derived to be -0.55 , which is almost twice as large as the value estimated in Fig. 2 in magnitude. The value of l_ϕ ($= 0.3 \mu\text{m}$ at $T = 5.1$ K) was larger than the film thickness. It ought to be, therefore, pointed out that we cannot rule out the possibility that the WAL effect we observed in the $\text{Cu}_x\text{Bi}_{2-x}\text{Se}_3$ film originated from the bulk state as the WAL effect in such a circumstance is also two dimensional.

The experimental behavior and Eq. (3) begin to disagree with each other as the temperature is lowered. Specifically, the experimental result can no longer be compared with the theory as the magnetic-field dependence near $B = 0$ becomes

nearly linear instead of the logarithmic dependence expected from Eq. (3). (For the fit for $T = 0.3$ K in Fig. 3, l_ϕ was set to the value extrapolated from the high- T values assuming $l_\phi \propto T^{-1/2}$.) The reason for this anomalous behavior is not clear at present. Nevertheless, we would like to point out several possible explanations.

First, the WAL effect may be quenched by the magnetic field at different field scales for the two surfaces of the film. As Cu atoms were provided by depositing a layer prior to the Bi_2Se_3 growth in our sample, the possibility of the properties of the two surfaces being significantly different cannot be ignored. Alternatively, the bulk state may be partly responsible for the magnetic-field dependence.^{37,38} The magnetoconductance is thereby given by a superposition of the surface and bulk contributions that would be, in general, not identical. The value of α extracted from the temperature dependence of the WAL correction is, however, inconsistent with this interpretation as one surface of the film suffices to produce the WAL contribution.

Second, the magnetic moments of the Cu atoms might have affected the magnetotransport properties by altering the magnetic field that the electrons experience.³⁹ Elemental copper has a relatively sizable diamagnetic response. However, the paramagnetic response of copper originating from a single unpaired 4s electron outweighs its diamagnetic response.

The third possibility is the fact that Cu-doped Bi_2Se_3 can be superconductive.⁴⁰ The quenching of the superconductivity by magnetic field will give rise to a negative magnetoconductivity. The Cu incorporation, however, does not induce superconductivity when Cu atoms substitute the Bi atoms.⁴⁰ Moreover, the temperature dependence plotted in Fig. 4 shows no sign of superconductivity. The last possibility can thus be ruled out.

IV. CONCLUSIONS

In conclusion, we have investigated the WAL and EEI effects in a Cu-doped Bi_2Se_3 film, where the Cu atoms were introduced in the Bi_2Se_3 lattice substitutionally by growing Bi_2Se_3 layers on Cu-deposited substrates using a hot-wall-epitaxy method. The magnitude of the logarithmic temperature dependence in the conductance due to the WAL effect suggests that only the surface states at one side of the film contribute to the quantum correction, provided that the surface states are responsible for the effect. We have encountered significant deviations from theoretical predictions for the magnetic-field dependence of the WAL effect and the magnitude of the temperature dependence attributed to the EEI effect. The disagreements suggest that the bulk state needs to be taken into account in analyzing the quantum transport effects.

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