Linear Magnetoresistance in Topological Insulator Thin Films: Quantum Phase Coherence Effects at High Temperatures

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In addition to the weak antilocalization cusp observed in the magnetoresistance (MR) of topological insulators at low temperatures and low magnetic fields, we find that the high-field MR in Bi₂Te₂Se is linear in field. At fields up to B=14T the slope of this linear-like MR is nearly independent of temperature over the range T=7 to 150K. We find that the linear MR arises from the competition between a logarithmic phase coherence component and a quadratic component. The quantum phase coherence dominates up to high temperatures, where the coherence length remains longer than the mean free path of electrons.

Three-dimensional (3D) topological insulators have been predicted to host metallic surface states surrounding an insulating bulk as a result of a band inversion in compounds containing elements having a large spin-orbit interaction.¹,² These electron states have been shown to exhibit strong spin-momentum locking that is attractive for potential applications in spintronic devices. Bi₂Se₃ and Bi₂Te₃ have been experimentally confirmed to have these topological properties through the observation of the surface Dirac dispersion using angle-resolved photoelectron spectroscopy (ARPES).³

Considerable effort has thus been focused on probing the surface states with magnetotransport experiments. Quantum coherence effects,⁴,⁵,⁶,⁷ quantum oscillations⁸,⁹,¹⁰ as well as some non-linear Hall signatures⁹,¹⁰ have been observed and well-studied in many cases. An interesting observation was made by several groups recently; the magnetoresistance (MR) shows a non-saturating positive, linear-like trend at high fields⁷,⁸,⁹,¹¹. It has been suggested⁷,⁸ that this linear-like MR response arises from the linear Dirac surface dispersion, which according to the quantum theory of linear MR (LMR) developed by Abrikosov should result in such a phenomenon¹² Some studies have also suggested that the LMR behavior becomes suppressed as the bulk is depleted of carriers.¹¹ This raises the question of the origin of the LMR.

We report on magnetotransport measurements of Bi₂Te₂Se thin films grown by molecular beam epitaxy (MBE). At low magnetic fields the MR exhibits a weak antilocalization (WAL) cusp that becomes suppressed when the temperature is increased. In addition, at high fields the MR increases with increasing field in a nonsaturating, linear trend up to B = 14 T. We account for MR(B) over the entire range of fields and temperatures with a modified Hikami, Larkin and Nagaoka (HLN) quantum interference model.¹³ It simultaneously accounts for the quantum phase interference cusp at low fields as well as the linear-like MR at high fields. It is shown that an additional quadratic term - consisting of both quantum and classical components - compensates the logarithmic dependence of the quantum interference at high fields, leading to an intermediate linear-like MR field dependence. However, we show that even at high temperatures the quantum interference dominates, indicating that electrons retain quantum phase coherence at high temperatures, possibly up to room temperature.

Thin films of Bi₂Te₂Se were grown on Si (111) at 200 °C using MBE. The base pressure in the MBE chamber was ~7x10⁻¹⁰ torr prior to the growth. Flux monitors in the MBE system allow the evaporated thickness to be monitored in real time during the growth.¹⁴ The stoichiometry was calibrated prior to the growth and monitored during the growth to achieve a 2:2:1 ratio of Bi:Te:Se. This ratio was later confirmed by energy dispersive X-ray spectrometry. X-ray diffraction data showed good c-axis alignment for all films, having (000ℓ) diffraction peaks up to ℓ = 21 as shown in Fig. 1. We confirmed the formation of a single phase and found no evidence of peak splitting. The inset of Fig. 1 shows the (0006) reflection peak with Kiessig fringes in the vicinity of the Bragg peak corresponding to the 15 nm thickness of the film. Magnetotransport measurements were performed on photolithography-patterned Hall bars (1 mm long and 0.3 mm wide). The samples were measured at temperatures down to T = 7 K and in magnetic fields up to B = 14 T. The Hall voltage was found to be linear in field and the electron density was ~5x10¹⁹ cm⁻³.¹³
Although MBE-grown Bi$_2$Te$_2$Se has not been previously reported, it is expected to be the most insulating among the Bi-based topological insulator materials as it lies between n-type Bi$_2$Se$_3$ and p-type Bi$_2$Te$_3$ when grown in bulk. Also, the ordering of the Se and Te atomic layers presumably inhibits Se from vacating the lattice.\textsuperscript{10,15} Unfortunately, the present MBE-grown films have not yet exhibited strong insulating characteristics possibly due to the fact that excess Se was not used during the growth. The present sample quality is similar to that previously reported for Bi$_2$Te$_2$Se nanoplatelets.\textsuperscript{16}

\[
\Delta G(B) = -\frac{\alpha e^2}{2\hbar} \left[ \psi \left( \frac{B_B}{B} + \frac{1}{2} \right) - \ln \left( \frac{B_{SO} + B}{B} \right) \right] - \frac{e^2}{\hbar} \left[ \psi \left( \frac{B_{SO} + B_B}{B} + \frac{1}{2} \right) - \ln \left( \frac{B_{SO} + B_B}{B} \right) \right] + \frac{3\alpha e^2}{2\hbar} \left[ \psi \left( \frac{4/3B_{SO} + B_B}{B} + \frac{1}{2} \right) - \ln \left( \frac{4/3B_{SO} + B_B}{B} \right) \right].
\]

Here, $B_i$ are the characteristic fields of each respective scattering channel ($i = \varphi, SO, e$) given by $B_i = \hbar/(4eL_i^2)$. $L_{SO}$ is the phase coherence length, $L_{SO}$ is the spin-orbit scattering length and $L_e$ is the elastic scattering length (or the mean free path). For small fields the phase coherence scattering term (first term) dominates since the coherence length is the longest of the three lengths. The spin-orbit and elastic scattering lengths yield characteristic fields of the order of several tesla. A maximum field of 14 T would therefore allow us to cover the low-field regime of the two latter terms in Eq. (1). It can be shown that the latter two terms containing spin-orbit scattering and elastic scattering can thus be approximated by a $B^2$ parabola.\textsuperscript{13} These approximations lead to

\[
\Delta G(B) = -\frac{\alpha e^2}{2\hbar} \left[ \psi \left( \frac{\hbar}{4eL^2B} + \frac{1}{2} \right) - \ln \left( \frac{\hbar}{4eL^2B} \right) \right] + \beta B^2.
\]

$\alpha$ is included to account for 2D bulk effects and possible contributions from the top and bottom surfaces of the film. $L$ is the phase coherence length and $\beta$ is the quadratic coefficient arising from the additional scattering terms. The first term in Eqs (1) and (2) will be referred to as the simple HLN model. One must also keep in mind that conventional cyclotronic MR might have to be taken into account. $\beta$ would therefore be composed of a classical cyclotronic component $\beta_c$ in addition to the quantum scattering terms arising from the last two terms in the HLN model that will be referred to as $\beta_q$.

There is, however, a requirement that must be met in using this model at high temperatures. Quantum interference requires that the phase coherence length be longer than the elastic mean free path ($\ell$) of the scattering electrons. In the present samples the phase coherence length is about an order of magnitude larger than the elastic mean free path at low temperatures. At $T = 7$ K, $L = 58$ nm and $\ell = 3.9$ nm, while at $T = 147$ K, $L = 9.4$ nm and $\ell = 3.5$ nm, where $\ell$ is obtained from the measured Hall mobilities and the Fermi wavevector $k_F = 0.060$ Å$^{-1}$ measured in Bi$_2$Te$_2$Se. The ratio $L/\ell$ is thus 15 at low temperatures and at least 2.7 at high temperatures. When $L/\ell$ is large the electrons make enough elastic collisions before losing phase coherence. We fit $\Delta G(B)$ to the

Figure 1. (Color Online) 0-20 X-ray diffraction spectra of a 15 nm thick Bi$_2$Te$_2$Se film on Si acquired using Cu K$_\alpha$ radiation. The gray dashed lines correspond to the calculated diffraction peaks of the hexagonal structure with $a = 0.431$ nm and $c = 3.001$ nm, labeled by their hexagonal axis indices. The inset shows an enlarged view of the (0006) reflection peak to clarify the Kissing fringes in the vicinity of the Bragg peak. The crystal unit cell is shown on the right.
experimental data for the entire range of fields up to \( B = 14 \) T. The fitted curves from Eq. (2) and the data for \( \Delta G(B) \) are shown in Fig. 3 for the \( \text{Bi}_2\text{Te}_3\text{Se} \) film at \( T = 7 \) and 147 K. There is excellent agreement over the entire field range. Similar fits were also carried over to the inverted \( MR(B) \) data and plotted as solid curves in Fig. 2. Equation (2) was found to describe the experimental data at all measured fields and temperatures.

Furthermore, in order to separate out the quantum phase-coherent contribution to the MR, Fig. 3 plots the field-dependence of the two terms of Eq. (2) separately. Note that the decreasing slope \( (d\Delta G/dB) \) of the phase-coherent term is compensated by the increasing slope of the quadratic term, leading to a constant slope. The phase-coherent term in the simple HLN model is seen to deviate from the data beginning at around 4 T. Below 4 T, fitting the data to either the simple or modified HLN model yields similar fitting parameters. This indicates that the modified model accurately accounts for the WAL cusp. The LMR on the other hand is seen to arise at high fields as an intermediate regime where the quadratic term compensates the saturating logarithm in the simple HLN model. Thus, the quadratic term makes its most significant contribution in the high field regime. It is interesting that the quantum coherent contribution to the MR is clearly much larger than the quadratic contribution at temperatures as high as 147 K. This reveals that electrons are able to retain phase information at high temperature, which can be technologically relevant if it is shown to hold at room temperature.

The fitting parameters \( \alpha(T) \) and \( L(T) \) were extracted from the \( \text{Bi}_2\text{Te}_3\text{Se} \) MR data and plotted in Fig. 4. At \( T = 7 \) K we find \( \alpha = 0.85 \), which matches previous reports on WAL in topological insulators. \(^4\,^5\,^6\) An increase in \( \alpha \) is observed with increasing temperature. This has been attributed to a coherent surface-to-bulk scattering channel in the regime where the surface-to-bulk scattering length is smaller than the coherence length. \(^5\) The behavior of the phase coherence length \( L(T) \) shows a decaying \( T^{-\gamma} \) power law trend, with \( \gamma = 0.75 \pm 0.05 \). This temperature dependence is complicated and can have contributions from several effects. At low temperatures a decay can arise from inelastic electron-electron collisions, which would result in \( \gamma = 0.5 \) for 2D, and \( \gamma = 0.75 \) for 3D. \(^7\,^8\,^9\) However, electron-phonon interactions can have the same effect when the temperature is higher, resulting in a faster decay with \( \gamma = 1 \). \(^8\) It is difficult to attribute the decay to a specific mechanism since our measurements span a large temperature range where various contributions result in different temperature variations.

Next, we consider the origin of the quadratic component \( \beta \). As discussed previously, spin-orbit and elastic terms in the HLN model as well as cyclotronic MR all have to be taken into account, as \( \beta = \beta_q + \beta_e \). The quantum part of \( \beta \) can be approximated by the following:\(^13\)

\[
\beta_q B^2 = \frac{e^2}{2A} \left[ \frac{B}{B_{so} + B_c} \right]^2 + \frac{3e^2}{48\pi h} \left[ \frac{B}{(4/3)B_{so} + B_e} \right]^2
\]

\[
\beta_q B^2 = \frac{e^2}{2A} \left[ \frac{B}{B_{so} + B_c} \right]^2 + \frac{3e^2}{48\pi h} \left[ \frac{B}{(4/3)B_{so} + B_e} \right]^2
\]  

(3)
Since $L_{SO} > L_e$, $\beta_q$ provides a positive quantum correction to $\Delta G$. However, the experimental data requires a total negative correction to $\Delta G$. We thus need to incorporate a conventional negative cyclotronic term in $\Delta G$ of classical origin given by

$$\beta_c B^2 = -\mu_{MR}^2 G_0 B^2$$

(4)

where $\mu_{MR}$ is the MR mobility$^{19}$ and $G_0$ is the zero-field conductance. We extract the value $L_{SO} = 6$ nm at $T = 7$ K from $\beta = -1.1 \times 10^{-8}$ Ω$^{-1}$ T$^{-2}$ and the assumption that $\mu_{MR} \approx \mu_r$. This is a reasonable estimate for $L_{SO}$ compared to values reported in the literature for Bi$_2$Se$_3$.\(^5\)

Note added in proof: Recent measurements of the MR in Bi$_2$Te$_3$ have shown a linear-like trend up to 60 T.$^{23}$ The full HLN model (our Eq. (1)) yielded a good fit to that data. This agrees precisely with our assertion that the spin-orbit and elastic terms in the full HLN model yield a significant contribution to the parabolic term. Terms with characteristic fields of the order of the maximum applied field will produce a parabolic functional form (Eq. (3)) that compensates the phase coherence of the simple HLN formula at high fields. This is evidently the case with their term containing $B_{SO}$ and $B_e$. It is unclear, however, how much classical MR contributes to the compensation in their case, nevertheless, the fact that $L_{SO} < L_e$ causes the contributions from $\beta_q$ and $\beta_c$ to have the same negative sign.

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