Unconventional Large Linear Magnetoresistance in Cu$_{2-x}$Te

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We report a large linear magnetoresistance in Cu$_{2-x}$Te, reaching $\Delta \rho / \rho (0) = 250\%$ at 2 K in a 9 T field. This is observed for samples with $x$ in the range 0.13 to 0.22, and the results are comparable to the effects observed in Ag$_2$X materials, although in this case the results appear for a much wider range of bulk carrier density. Examining the magnitude vs. crossover field from low-field quadratic to high-field linear behavior, we show that models based on classical transport behavior best explain the observed results. The effects are traced to misdirected currents due to topologically inverted behavior in this system, such that stable surface states provide the high mobility transport channels. The resistivity also crosses over to a $T^2$ dependence in the temperature range where the large linear MR appears, an indicator of electron-electron interaction effects within the surface states. Thus this is an example of a system in which these interactions dominate the low-temperature behavior of the surface states.

Considerable attention has recently been devoted to systems exhibiting linear magnetoresistance (MR), starting with the effects observed in Ag$_2$X materials, although in this case the results appear for a much wider range of bulk carrier density. Examining the magnitude vs. crossover field from low-field quadratic to high-field linear behavior, we show that models based on classical transport behavior best explain the observed results. The effects are traced to misdirected currents due to topologically inverted behavior in this system, such that stable surface states provide the high mobility transport channels. The resistivity also crosses over to a $T^2$ dependence in the temperature range where the large linear MR appears, an indicator of electron-electron interaction effects within the surface states. Thus this is an example of a system in which these interactions dominate the low-temperature behavior of the surface states.

Weak antilocalization as a bulk rather than surface effect$^{12}$ may also occur in these layered systems in the presence of spin-orbit coupling. Furthermore it was recently demonstrated$^{12}$ that even very weak disorder may lead to such effects in 3D high-mobility systems such as the Dirac semimetals.

Cu$_2$Te has been of significant interest for potential applications including thermoelectric and solar energy conversion, as well as a variety of nano-device$^{21-24}$, and it has been connected to a topologically nontrivial band configuration$^{25-26}$. Synthesized materials in bulk have a Cu$_{2-x}$Te stoichiometry, with the Cu deficit corresponding to vacancies which lead to $p$-type semiconducting behavior. Here we present magnetotransport properties of materials in the range $x = 0.13$ to 0.22, exhibiting a large linear MR which can be traced to surface states, reinforcing the topological insulator nature of this system, and occurring in a regime of high carrier density and with strong electron interactions distinct from what has been observed in other systems.

The three polycrystalline Cu$_{2-x}$Te samples were obtained by solid state reaction and vacuum annealing. Their properties have been described in more detail in Ref. $^{27}$. Compositions from electron microprobe measurements are Cu$_{1.87}$Te, Cu$_{1.82}$Te, and Cu$_{1.78}$Te ($x = 0.13$ to 0.22), with Hall measurements showing them to be heavily-doped $p$-type semiconductors with room temperature carrier densities 3.6, 4.1, and 6.5 $\times 10^{21}$ cm$^{-3}$, respectively. The results along with NMR measurements are consistent with a Fermi level in the bulk which is pulled below the valence band edge due to Cu deficit$^{28}$, with room temperature Hall results matching the expected bulk carrier densities. The structure for Cu$_{1.87}$Te and Cu$_{1.82}$Te is a superstructure of the hexagonal Nowotny structure$^{27,28}$, with a somewhat different superstructure for the Cu$_{1.78}$Te case. Measurements reported here utilized a Quantum Design PPMS system and a Quantum Design MPMS combined with an AC
bridge. Transport measurements were performed on bar-
shaped samples cut from the polycrystalline ingots, with
magnetoresistance measured in the geometry with the
field perpendicular to the current direction.

Figure 1 exhibits resistivities below 60 K. The behav-
ior is quadratic in the low-temperature limit, particularly
for the lowest-vacancy composition: fitting to $\rho_0 + A T^n$
below 30 K yielded $n = 2.02, 2.13$ and $2.34$ for increasing $x$.
Extended to room temperature, the residual resistiv-
ity ratios for the three samples are 32, 31, and 14 for
Cu$_{1.87}$Te, Cu$_{1.82}$Te, and Cu$_{1.78}$Te respectively. The in-
set of Fig. 1 also shows carrier densities derived from
the Hall resistances, shown vs. temperature for Cu$_{1.87}$Te
and Cu$_{1.82}$Te. These show a low temperature downturn
where the $T^2$ resistivity sets in, apparently a result of
parallel conduction paths rather than a change in the
bulk carrier density, since as noted below it is in this
range that the surface states are believed to influence
the transport properties.

Fixing the low-temperature exponent to $n = 2$, the fit-
ted resistivity pre-factors are $A = 0.019, 0.009,$ and $0.010$
$\mu\Omega\text{cm}/K^2$ for the three samples with increasing $x$.
These compare to the lower end of the range for Fermi liquid
behavior in heavy-Fermion materials$^{29}$, although with
the low-temperature behavior attributed to high-mobility
threading states, this implies considerably smaller effective $A$
values for these states, comparable for example to elemental transition metals. Mobilities derived from
the resistivities and room-temperature Hall-effect carrier
densities are $\mu = 10 \text{ cm}^2/\text{Vs}$ or less at room temperature,
increasing to 170, 280, and 55 cm$^2$/Vs for Cu$_{1.87}$Te,
Cu$_{1.82}$Te, and Cu$_{1.78}$Te respectively at low temperature.
These are not unexpected for semiconductors with large
vacancy densities and large hole band mass$^{25}$ close to 0.5
$m_e$, although the temperature dependences are large for
such a case, apparently due to threading states.

Figure 2 displays the magnetoresistance, $MR = \Delta \rho/\rho(0)$, where $\Delta \rho = [\rho(B) - \rho(0)]$, and $\rho(B)$ denotes
the resistivity measured in applied field $B$. The 2 K mag-
nitudes reach 250%, 180%, and 37% at 9 T for Cu$_{1.87}$Te,
Cu$_{1.82}$Te, and Cu$_{1.78}$Te respectively. The largest of
these are comparable to effects observed in Ag$_2$Te, al-
though differing in that the Ag$_2$Te results are observed in
a much narrower composition window for carrier densities
near zero, and decrease more slowly vs. temperature.

The insets of Fig. 2 also display Kohler plots, often
used to understand whether a single scattering process controls the magnetoresistance. The curves deviate from a common line at 30 K and below, showing that there are parallel scattering processes corresponding to the low-temperature conduction mechanism.

Fig. 3(a) shows crossover fields ($B_c$), where the MR changes from quadratic to linear. $B_c$ was obtained by fitting the resistivity in the low-field limit to a $B^2$ dependence, and linear at higher fields, and extracting the fields where these curves cross. The highest-temperature curves were excluded since the small response makes the fits unreliable.

In the Parish and Littlewood (PL) classical transport model, $B_c$ corresponds to a condition $B\langle \mu \rangle = 1$. In Fig. 3(a) values of the average mobility are given on the right axis according to this condition. For the two lowest-$x$ samples these are close to $7500 \text{ cm}^2/\text{Vs}$ at 2 K, considerably larger than the Hall mobilities extracted for these samples. This confirms that there must be high mobility regions within the overall low mobility material, reinforcing the evidence for threading conduction due to topological surface states, as was noted above.

For carriers of one sign, in the PL model the magnitude of the linear MR should also scale with average mobility, and in some cases a direct proportionality has been observed for linear MR vs. measured mobility. As a measure of the linear MR we plotted the 9 T values (MR Max) vs. $T$ in Fig. 3(b). In the inset these are plotted vs. the normalized inverse resistivities measured at the corresponding temperatures, a measure of the mobilities for the case of constant carrier densities as we have here. However the plots are not linear, reflective of the discontinuous mobility distribution in these samples, and the crossover in resistivity mechanisms at low temperatures.

A more direct measure of the connection between the crossover field and the high mobility carriers responsible for the linear MR is shown in the plot of $1/B_c$ vs. inverse resistivity, Fig. 3(c). There is a linear relation below 30 K for the two lowest-$x$ samples, which exhibit the largest MR and low-$T$ resistivities closest to $T^2$ behavior. Thus there is strong evidence that the high mobility threading carriers for these samples can be traced directly to the measured resistivity in the low temperature limit, and these carriers apparently dominate the resistivity in this limit. The $1/(a + bT^2)$ curves in Fig. 3(a) and (b) are also drawn to correspond to this relation, with both $1/B_c$ and MR Max connected to the inverse of the mobility for low temperatures.

A plot of $1/B_c$ vs. MR max is also given in Fig. 3 and we see that there is a universal scaling between these quantities for all samples. A straight-line relationship is expected for a classical transport model, but with zero intercept in the PL model. This result is also similar to the universal scaling identified in Ref. [31] for nanoparticle films, although again in the present case there is a large offset. Note however that other systems have been observed to exhibit such an offset, for example in results for Ag$_2$Se films one can see that the corresponding offset has a much larger value of about $-23 \text{ T}^{-1}$. The reasons are not clear, although in the present materials the source of the linear MR appears more likely to be misdirected currents rather than a broad distribution of mobilities, with the threading nature of the high mobility currents a distinguishing feature relative to more continuous models.

FIG. 3. (a) Crossover field vs. $T$ for the three samples, with mobility values corresponding to the Parish-Littlewood crossover condition shown on the right axis. (b) 9 T MR values vs. $T$, and (inset) vs. scaled inverse resistivity. Trendlines in plots (a) and (b): $1/(a + bT^2)$ curves. (c) Crossover field vs. normalized inverse resistivity, with straight line through origin as guide to the eye. Symbols are common to all plots.
Aside from a surface state mechanism, because of its layered nature 2D weak anti-localization from spin-orbit-split bulk states could play a role in Cu$_{2-\delta}$Te. Such effects can be difficult to separate from classical linear MR, and the corresponding magnetococonductance indeed works reasonably well as an alternative fitting model for the present data (not shown). These fits, with addition of a large quadratic classical magnetococonductance term, yield maximum dephasing lengths$^{11}$ at 2 K of between 19 and 30 nm, not unreasonable values. For comparison, in the analog material Cu$_2$Se weak anti-localization effects were also identified at low temperature$^{34}$, although with a rather different amplitude and field dependence than what is observed here. Thus though it seems possible that some of the observed response in Cu$_2$Te is due to such effects, the scaling with resistivity established here points to a classical model based on high mobility surface states as a more reasonable model to explain these observations.

It is also possible to attribute the results to the existence of bulk Dirac-like electronic states, such as for the guiding center mechanism recently introduced to account for inhomogeneous transport in such systems$^{35}$. However, in such cases a generally high mobility would be expected$^{36-37}$, as opposed to the situation here. Alternatively, a compensation-based mechanism due to multiple carrier pockets$^{39}$ might also explain the present results, however it has been shown$^{40}$ that hole pockets alone account well for the bulk transport and NMR behavior in Cu$_{2-\delta}$Te, and the observation of linear MR in compositions with different carrier concentrations appears inconsistent with the balance of carrier occupations required for such a mechanism.

Another consideration would be whether magnetic quantization conditions are reached here, such that a quantum MR model$^{10}$ is appropriate. Given the effective mass obtained for Cu$_2$Te and the carrier densities present in these samples, we expect$^{41}$ that the Fermi energy in the bulk corresponds to several tenths of eV. With a corresponding Fermi level for Dirac-like surface states having the Fermi velocity of graphene, a field of 100 T or more would be required to occupy only the lowest Landau level$^{38}$. Thus under the conditions used here we expect that many Landau levels will be occupied, a situation far from the quantum limit.

In a classical treatment, it is also clear that a model based on uniformly distributed weak disorder$^{12}$ cannot account for the present behavior, since the crossover fields are well out of range of the nearly-uniform high mobility needed for this to work. Indeed, as noted above it appears more reasonable to treat this system as discontinuous, with carrier behavior closer to bimodal, consisting of that of the bulk and of the polycrystal interfaces. There have been a number of works analyzing such situations, including through models based on resistor networks$^{33}$ as well as in effective medium theories$^{39,40}$. However we are not aware of specific predictions related to the behavior of Fig. 4 in which the behavior appears to go smoothly from an inhomogeneous transport-based liner MR to a purely quadratic classic behavior as the temperature and carrier density increases.

Returning to the observed $T^2$ resistivities, there has been considerable interest in understanding the electron-electron interaction behavior of topological surface states$^{41,42}$. In some cases these are predicted to have $T^2$ behavior analogous with that of ordinary Fermi liquids, through processes that should be strongly dependent on umklapp scattering, and correspondingly on the symmetry and curvature of the Fermi surface of the 2D topological states$^{43,44}$. An alternative mechanism for the observed results would be scattering between bulk and surface electronic states, although with a scattering rate proportional to the density of states$^{45}$, this mechanism would not be expected to produce the observed $T^2$ dependence. However it seems possible that such a mechanism is responsible for the much more rapid drop in linear MR with increasing bulk carrier density as observed$^{46}$ in Ag$_2$Te vs. what is seen here. Cu$_2$Te presents a case where a clear $T^2$ behavior sets in at low temperatures dominated by the surface states, a situation for which there are few experimental examples.

In conclusion, we observe a large linear magnetoresistance in samples of Cu$_{2-\delta}$Te with increasing carrier densities, with the magnitude reaching $\Delta \rho/\rho(0) = 250\%$ at 2 K in a 9 T field, comparable to the effects observed in Ag$_2$Se and Ag$_2$Te. Examining the magnitude of the effect vs. the crossover field where the behavior changes from low-field quadratic to high-field linear behavior, we demonstrated that models based on classical transport behavior best explain the observed results. We also identified a universal scaling between the MR magnitude and the crossover field independent of carrier density. The effects are traced to previously identified indications of topologically inverted behavior in this system, such that topologically stable surface states provide the high mobility transport channels. There is a crossover to a $T^2$ resistivity behavior at low temperatures where the large linear

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**FIG. 4.** Inverse crossover field plotted vs. maximum MR (9 T values) for the Cu$_{2-\delta}$Te samples at 2 K. Straight line is guide to the eye.
MR appears, which we connected to electron-electron interaction effects within the surface states, so this system also provides an experimental example of such strongly interacting surface states.

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