Transport study of the Berry phase, resistivity rule, and quantum Hall effect in graphite

Aruna N. Ramanayaka and Ramesh G. Mani
Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303, USA
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Transport measurements indicate strong oscillations in the Hall, R_{xy}, and the diagonal, R_{xx}, resistances and exhibit Hall plateaus at the lowest temperatures, in three-dimensional (3D) highly oriented pyrolytic graphite. At the same time, a comparative Shubnikov-de Haas-oscillations-based Berry phase analysis indicates that graphite is unlike the GaAs/AlGaAs two-dimensional electron system, the 3D *n*-GaAs epilayer, semiconducting $Hg_{0.8}Cd_{0.2}Te$, and some other systems. Finally, we observe the transport data to follow $B \times dR_{xy}/dB \approx -\Delta R_{xx}$. This feature is consistent with the observed relative phases of the oscillatory R_{xx} and R_{xy}.

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Single layers of carbon atoms known as graphene are an interesting future electronic material.[1–4] At the same time, graphene is a novel two-dimensional electronic system (2DES) with remarkable features providing for a solid-state realization of quantum electrodynamics, massless Dirac particles, an anomalous Berry or Pancharatnam-Berry phase,[5–7] and an unconventional quantum Hall effect in a strong magnetic field.[1–7] The *ABAB* bernal stacking of graphene helps to produce graphite, an anisotropic electronic material exhibiting a large difference between the in-plane and perpendicular transport. Since graphite may be viewed as stacked graphene, one wonders whether remnants of the remarkable properties of graphene might also be observable in graphite.

Graphite exhibits a ≈0.03 eV band overlap, in contrast to the zero gap in graphene.[8–10] In the recent past, concepts proposed for graphene have also been invoked for graphite. For example, Ref. 11 argues for the observation of massive majority electrons with a three-dimensional (3D) spectrum, minority holes with a 2D parabolic massive spectrum, and majority holes with a 2D Dirac spectrum. Hall plateaus in σ_{xy} extracted from van-der-Pauw measurements on highly oriented pyrolytic graphite (HOPG) have been cited as evidence for the integral quantum Hall effect (IQHE) in graphite in Ref. 12. Luk'yanchuk *et al.* in Ref. 13 suggested simultaneous quantum Hall effects for the massive electrons and massless Dirac holes with Pancharatnam-Berry phase of $\beta=0$ and $\beta=1/2$, respectively. β is given here in units of 2π. Finally, an angle-resolved photoemission spectroscopy (ARPES) study has reported massless Dirac fermions coexisting with finite-mass quasiparticles in graphite.[14]

Plateaus at Hall resistance at $R_{xy}=h/ie^2$ with $i=1,2,3,\ldots$ and $R_{xx}\rightarrow 0$ are typically viewed as a characteristic of IQHE in the 2DES. At the same time, a "thick" specimen consisting of quantum wells separated by wide barriers exhibits plateaus at $R_{xy}=h/ije^2$, where j counts the number of layers in the specimen. Indeed, introducing a dispersion in the z direction does not modify the observability of IQHE.[15] Thus, IQHE can also be a characteristic of anisotropic 3D systems such as graphite. Here, in graphite, the IQHE can be of the canonical variety or of the unconventional type reported in graphene. In addition, theory has predicted the existence of a single, true bulk 3D QHE in graphite in a large magnetic field parallel to the c axis at $\sigma_{xy}=(4e^2/\hbar)(1/c_0)$, where $c_0=6.7$ Å is the c-axis lattice constant.[16]

Thus, there are reasons for carrying out quantum Hall transport studies in graphite. The possibility of both the canonical and unconventional IQHE in graphite motivates also a study of the Pancharatnam-Berry phase. Finally, one wonders whether 3D graphite might also satisfy the resistivity rule observed in 2D quantum Hall systems.[17]

This magnetotransport study of HOPG shows strong oscillations in the Hall resistance, R_{xy}, and Shubnikov-de Haas (SdH) oscillations in the diagonal resistance, R_{xx}, while manifesting Hall plateaus at the lowest temperatures. A Fourier transform of the R_{xx} SdH oscillations indicates a single set of carriers, namely, electrons, in these HOPG specimens. A Pancharatnam-Berry phase analysis of the SdH data suggests that these carriers in graphite are unlike those in GaAs/AlGaAs heterostructures, *n*-GaAs epilayers, bulk semiconducting $Hg_{0.8}Cd_{0.2}Te$, the HgTe quantum well, 3D AlGaN, and InSb systems. Further, a resistivity rule study of graphite indicates $R_{xx}\sim -B\times dR_{xy}/dB$, in variance with observations in canonical quantum Hall systems while a phase analysis suggests a new classification (type IV)[18] of the phase relation between the oscillatory R_{xx} and R_{xy}. The latter observation is consistent with $R_{xx}\sim -B\times dR_{xy}/dB$.

Our 25-μm-thick graphite specimens were exfoliated from bulk HOPG, and the measurements were carried out using standard lock-in techniques with the B parallel to the c axis. Measurements of R_{xx} and R_{xy} are shown in Figs. 1(a) and 1(b), respectively, for $1.5\leq T\leq 154$ K and $0\leq B\leq 5$ T. At low T, SdH oscillations in R_{xx} and R_{xy}, and plateaus in

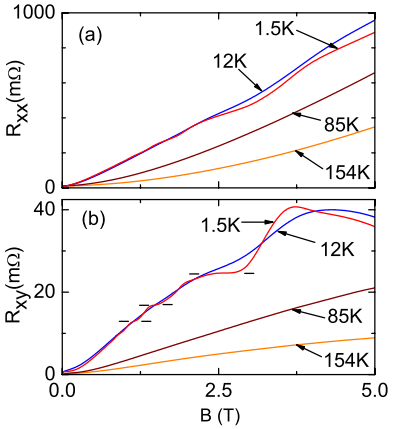

FIG. 1. (Color online) (a) The diagonal resistance (R_{xx}) and (b) Hall resistance (R_{xy}) of sample S1 are exhibited vs the applied magnetic field, B, between $154\leq T\leq 1.5$ K. R_{xy} plateaus and SdH oscillations in R_{xx} are manifested at low T.

R_{xy}, appear manifested, see Figs. 1(a) and 1(b), as in Figs. 2 and 3 of Ref. 19. Three Hall plateaus are observable in Fig. 1(b). The Hall plateau resistance observed at $B=2.5$ T and $T=1.5$ K is consistent, within a factor of 2, with viewing the specimen as a stack of uncoupled quantum Hall layers. Note that $R_{xx}>0$ through the Hall plateaus. The finite tilt of the highest-B plateau at $T=1.5$ K might be due to the incipient R_{xy} saturation, possibly due to multiband transport.

The SdH effect has been used to probe the Pancharatnam-Berry phase.[5,6] For the graphene system, the oscillatory R_{xx} has been written as $R(B,T)\cos[2\pi(B_0/B+1/2+\beta)]$,[7] where $R(B,T)$ is the SdH oscillation amplitude, B_0 is the SdH oscillation frequency, and β is the Pancharatnam-Berry phase in units of 2π. The observation of an anomalous Pancharatnam-Berry phase in graphene has generated interest in the study of the same in graphite. Hence, we set out to examine our data for graphite from a similar perspective.

Graphite is a semimetal with majority electrons and holes exhibiting elongated Fermi surfaces along the c axis.[9] The Fermi surface of graphite has been studied via the oscillatory magnetization [de Haas–van Alphen (dHvA)] effect, and also the oscillatory magnetoresistance (SdH) effect.[9] Graphite exhibits a strong dHvA effect, and the Fourier transform of the associated oscillatory magnetization readily exhibits two spectral peaks corresponding to the two sets of majority carriers. The SdH data, on the other hand, can often exhibit just a single broad peak in the Fourier spectrum.[13] It has been suggested that the scattering lifetimes manifested by the carriers in the two experiments might not be the same.[9] Further, that one set of carriers might suffer more broadening, and as a consequence, the associated oscillatory contribution due to this band might vanish in the SdH effect.

The HOPG specimens examined in this study are also characterized by a single broad peak in the Fourier transform of the SdH data, see Fig. 2(b). This feature suggests that the SdH effect in our HOPG specimen is also dominated by one type of carriers, namely, electrons, and this feature indicates that this single-carrier type may be subjected to a traditional Pancharatnam-Berry phase analysis as in graphene.

In order to examine and compare the Pancharatnam-Berry phase, plots of the Landau-level index, n (left axis), and R_{xx} (right axis) versus B^{-1} are shown in Figs. 2(a) and 2(c)–2(e) for graphite, the GaAs/AlGaAs 2D electron system, an n-GaAs epilayer,[20] and bulk semiconducting $Hg_{0.8}Cd_{0.2}Te$,[21] respectively. For all four material systems, minima of oscillatory R_{xx} have been assigned with n and maxima of the oscillatory R_{xx} have been assigned with $n+1/2$, as in Ref. 7. In such an analysis, an intercept $n_0=0$ would normally indicate a Pancharatnam-Berry phase $\beta=0$ while $n_0=1/2$ would normally correspond to $\beta=1/2$, as in graphene.[7] For the three graphite samples S1, S2, and S3 [see Fig. 2(a)], the Landau-level index intercept of a linear fit to n vs B^{-1} yields 0.47, i.e., 1/2. Panels (c)–(e) give intercepts of 0.05, 0.06, and −0.04, i.e., zero, for the GaAs/AlGaAs, n-GaAs epilayer, and bulk $Hg_{0.8}Cd_{0.2}Te$ systems. These fit-extracted intercepts, n_0, and B_0 have been summarized in Table I.

A similar analysis was carried out with other published data and these results are also summarized in Table I for the HgTe quantum well,[22] 3D AlGaN,[23] InSb,[14] and $C_{9.3}AlCl_{3.4}$—a first-stage graphite intercalation

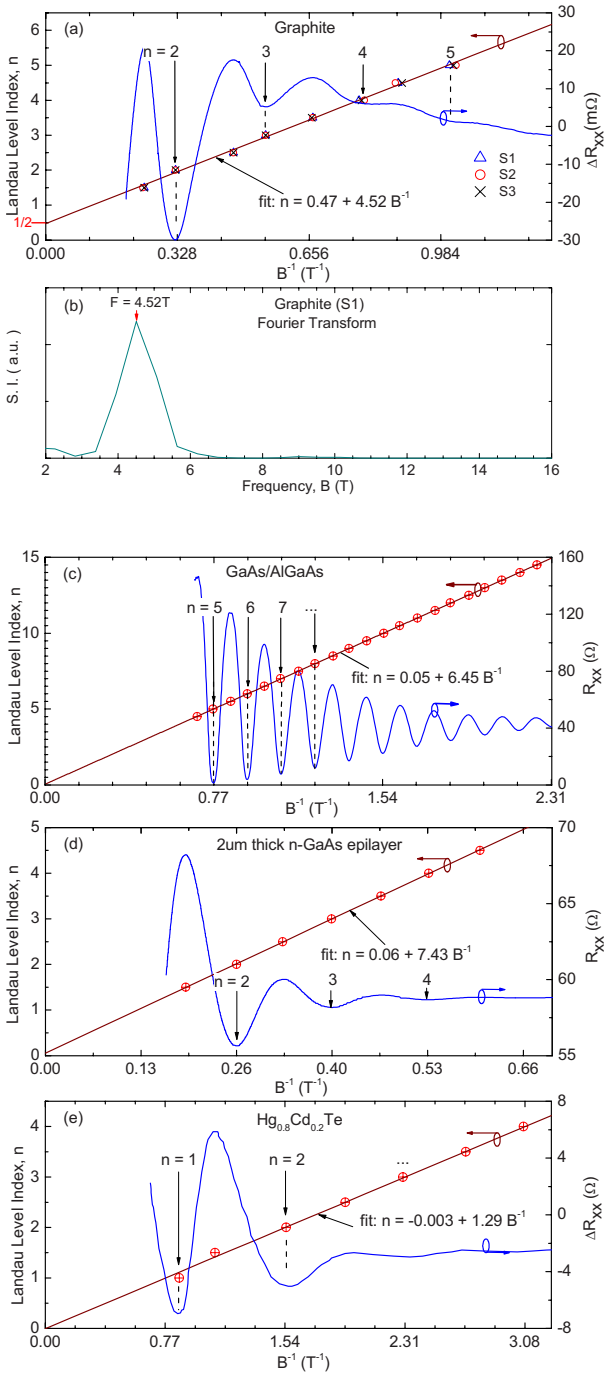

FIG. 2. (Color online) (a) Shown here are plots of Landau-level index (n) (left) of samples S1, S2, and S3 and R_{xx} (right) of sample S1 vs B^{-1}. The slope of the n vs B^{-1} plot indicates the SdH frequency, $F=4.52$ T, and the intercept, $n(B^{-1}=0)=0.47$. (b) This plot shows the spectral intensity of the Fourier transform of $\Delta R_{xx}(1/B)$ of sample S1. A single peak in the Fourier spectrum confirms that the SdH effect in these graphite specimens is basically dominated by one type of carrier with $F=4.52$ T. (c) n and R_{xx} are shown vs B^{-1} for the GaAs/AlGaAs 2D electron system. In (d) and (e) n and R_{xx} are shown for the 2-μm-thick n-GaAs epilayer and 3D $Hg_{0.8}Cd_{0.2}Te$ systems. Linear fit of n vs B^{-1} intersects the ordinate at 0.47, 0.05, 0.06, and −0.04 for HOPG, GaAs/AlGaAs, 2-μm-thick n-GaAs epilayer, and bulk $Hg_{0.8}Cd_{0.2}Te$ systems, respectively.

TABLE I. Intercept n_0 and the slope B_0 of (n) vs B^{-1} plot. β is the suggested Pancharatnam-Berry phase in units of 2π.

Material	n_0	B_0 (T)	β
Graphite	0.47 ± 0.02	4.52	1/2
GaAs/AlGaAs	0.05 ± 0.01	6.45	0
GaAs (Ref. 20)[a]	0.06 ± 0.02	7.43	0
$Hg_{0.8}Cd_{0.2}Te$ (Ref. 21)[b]	-0.003 ± 0.022	1.298	0
HgTe (Ref. 22)[c]	0.06 ± 0.03	26.14	0
3D AlGaN (Ref. 23)	-0.01 ± 0.03	35.39	0
InSb (Ref. 24)	0.05 ± 0.03	19.80	0
$C_{9.3}AlCl_{3.4}$ (Ref. 25)[d]	0.48 ± 0.02	11.7	1/2

[a]2-μm-thick n-GaAs epilayer.
[b]Bulk semiconducting $Hg_{0.8}Cd_{0.2}Te$.
[c]n-type HgTe quantum wells.
[d]First-stage graphite intercalation compound.

compound.[25] Notice that the Table I represents eight different systems, yet only graphite and $C_{9.3}AlCl_{3.4}$ show $n_0=1/2$ in the $B^{-1}\rightarrow 0$ limit as observed in graphene.[7] This is a principal experimental finding of this work.

Does this experimental finding signify an anomalous Pancharatnam-Berry phase for the observed carriers in graphite? If the experimental results shown here in Fig. 1 had indicated $R_{xy} > R_{xx}$ as in Ref. 7, then, certainly, the conclusion would immediately follow that electrons in graphite exhibit an anomalous Pancharatnam-Berry phase as in graphene. The fact that Fig. 1 indicates $R_{xy} \ll R_{xx}$ complicates the issue since many would reason that $R_{xy} \ll R_{xx}$ implies $\rho_{xy} \ll \rho_{xx}$ given the geometrical factors in these graphite

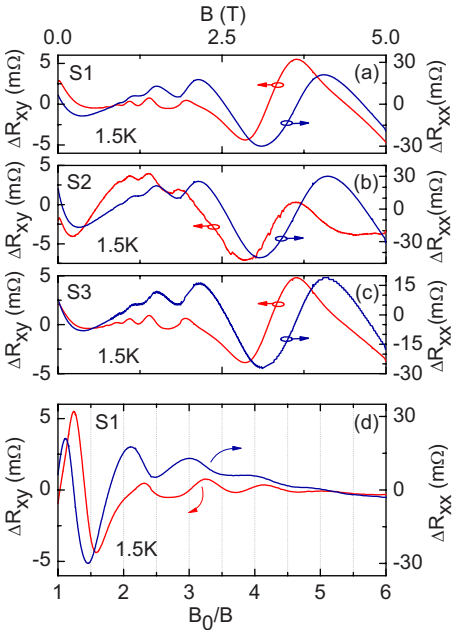

FIG. 3. (Color online) The oscillatory Hall (ΔR_{xy}) and diagonal (ΔR_{xx}) resistances are shown for samples (a) S1, (b) S2, and (c) S3 vs B. (d) ΔR_{xy} and ΔR_{xx} are shown vs the normalized inverse magnetic field B_0/B. This plot shows a $\pi/2$-phase shift between ΔR_{xx} and ΔR_{xy}.

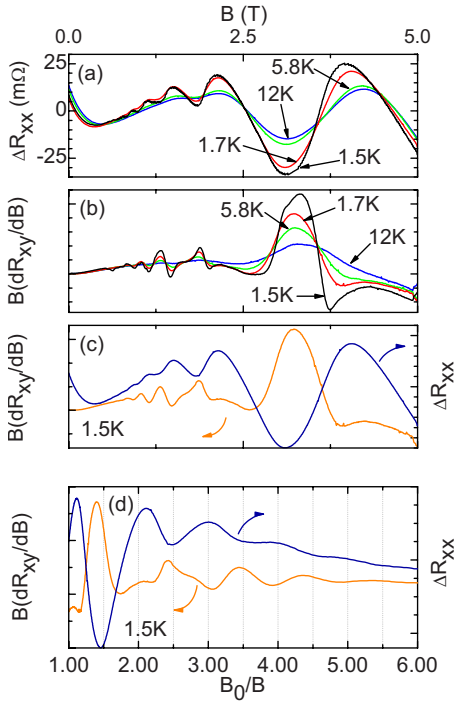

FIG. 4. (Color online) Panels (a) and (b) show that the phase of the oscillatory ΔR_{xx} and $B \times dR_{xy}/dB$ does not change with T. (c) Comparison of ΔR_{xx} and $B \times dR_{xy}/dB$ at $T=1.5$ K. (d) ΔR_{xx} and $B \times dR_{xy}/dB$ are shown vs B_0/B. Both (c) and (d) suggest a phase shift of approximately π between ΔR_{xx} and $B \times dR_{xy}/dB$.

specimens, and as a consequence, $n_0=1/2$ in graphite should be taken to indicate normal carriers,[13] not Dirac carriers. Some features should, however, serve as caution against following this line of reasoning. First, strong SdH oscillations, as in Fig. 1, are typically observed only when $\rho_{xy} > \rho_{xx}$, i.e., $\omega\tau > 1$, for the associated carriers. Thus, perhaps, the observation of $R_{xy} \ll R_{xx}$ in graphite need not imply $\rho_{xy} \ll \rho_{xx}$. Next, it appears worth pointing out that some experiments in the GaAs/AlGaAs system have clearly shown that there need not be a simple relation between R_{xx} and ρ_{xx}.[26] Finally, it is also known that certain types of longitudinal specimen thickness variations, density variations, and/or current switching between graphene layers in graphite can introduce a linear-in-B component into R_{xx} originating from the Hall effect; such an effect can produce experimental observations of $R_{xx} \gg R_{xy}$ even in systems that satisfy $\rho_{xx} < \rho_{xy}$. Due to such possibilities in graphite, the results shown Fig. 2 and Table I could plausibly identify an anomalous Pancharatnam-Berry phase for electrons in graphite and $C_{9.3}AlCl_{3.4}$.[25] Here, it is worth noting that semiconducting $Hg_{0.8}Cd_{0.2}Te$ specimen also exhibits $R_{xx} > R_{xy}$ (Ref. 21) and yet shows $n_0=0$, unlike graphite. Clearly, the experimental finding is unambiguous: the infinite field phase extracted from the SdH oscillations for graphite is unlike the results for a number of canonical semiconductor systems, see Table I.

Next, we examine more closely the phase relation in the oscillations of R_{xx} and R_{xy} since this is a striking feature of the data. Thus, background subtracted R_{xx} and R_{xy}, i.e., ΔR_{xx} and ΔR_{xy}, are shown in the Figs. 3(a)–3(c), for samples S1–S3. Thus far, the phase relations between ΔR_{xx} and ΔR_{xy}

have been classified into three types in the high-mobility GaAs/AlGaAs system.[18] Namely, type III, where the oscillations of ΔR_{xx} and ΔR_{xy} are approximately 180° out of phase. Type II, where the oscillation of the ΔR_{xx} and ΔR_{xy} are in phase. And, type I, where the peaks of ΔR_{xx} occur on the low-B side of the ΔR_{xy} peaks with a $\pi/2$-phase shift. Figure 3 shows, however, that the peaks of ΔR_{xx} appear on the high-B side of the ΔR_{xy} peaks, unlike the cases mentioned above. This suggests a fourth phase relation (type IV) between ΔR_{xx} and ΔR_{xy} in (HOPG) graphite. In the B^{-1} plot, see Fig. 3(d), the peaks of ΔR_{xx} are shifted toward the low B_0/B side with respect to the peaks of ΔR_{xy} with an approximately $\pi/2$-phase shift between ΔR_{xx} and ΔR_{xy}, confirming this conjecture.

For the resistivity rule study,[17,18,27,28] the temperature dependence of ΔR_{xx} and $B \times dR_{xy}/dB$ are shown in Figs. 4(a) and 4(b), respectively. Figure 4 indicates a progressive change, where the oscillatory amplitude increases with decreasing T while the B values of the extrema remains unchanged with T. Figure 4(c) shows the ΔR_{xx} (right axis) and $B \times dR_{xy}/dB$ (left axis) for the sample S1 at 1.5 K. A direct comparison [see Fig. 4(c)] reveals a π-phase difference between $B \times dR_{xy}/dB$ and ΔR_{xx}, i.e., $B \times dR_{xy}/dB \approx -\Delta R_{xx}$. Plots of ΔR_{xx} and $B \times dR_{xy}/dB$ vs B_0/B [see Fig. 4(d)] confirm an approximately π-phase shift between ΔR_{xx} and $B \times dR_{xy}/dB$. This variance from the canonical resistivity rule is consistent with the observed type-IV phase relation between the oscillatory R_{xx} and R_{xy}, see Fig. 3.

In summary, a study of (HOPG) graphite indicates strong oscillatory R_{xy} and R_{xx} with Hall (R_{xy}) plateaus coincident with a nonvanishing R_{xx} at the lowest T. On the other hand, a comparative SdH Pancharatnam-Berry phase study of graphite with other systems such the GaAs/AlGaAs 2D electron system, the 3D n-GaAs epilayer, bulk $Hg_{0.8}Cd_{0.2}Te$, HgTe quantum well, 3D AlGaN, InSb, and $C_{9.3}AlCl_{3.4}$—a graphite intercalation compound—reveals a value of $n_0 = 1/2$ in the $B^{-1} \to 0$ limit only for the graphene-based systems. As in graphene, this feature might indicate that one-half of a conduction-band Landau level is pinned together with one-half of a hole Landau level in graphite and might reflect a nontrivial Pancharatnam-Berry phase ($\beta = 1/2$) and Dirac carriers in graphite. A close study of the oscillatory diagonal and Hall resistances reveals also an anomalous (type-IV) phase relation between oscillatory R_{xx} and R_{xy}, over the entire T range. This result is consistent with the observation of $B \times dR_{xy}/dB \approx -\Delta R_{xx}$ in the resistivity rule analysis of the transport.

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