PHYSICAL REVIEW B 79, 245124 (2009)

Hall plateaus at magic angles in bismuth beyond the quantum limit

Benoît Fauqué,[1] Huan Yang,[1] Ilya Sheikin,[2] Luis Balicas,[3] Jean-Paul Issi,[4] and Kamran Behnia[1]
[1]*LPEM (UPMC-CNRS), ESPCI, F-75231 Paris, France*
[2]*Grenoble High Magnetic Field Laboratory (CNRS), F-38042 Grenoble, France*
[3]*National High Magnetic Field Laboratory, FSU, Tallahassee, Florida 32306, USA*
[4]*CERMIN, Université Catholique de Louvain, B-1348 Louvain-la-Neuve, Belgium*
(Received 25 May 2009; published 22 June 2009)

We present a study of the angular dependence of the resistivity tensor up to 35 T in elemental bismuth complemented by torque magnetometry measurements in a similar configuration. For at least two particular field orientations a few degrees off the trigonal axis, the Hall resistivity was found to become field independent within experimental resolution in a finite field window corresponding to a field which is roughly three times the frequency of quantum oscillations. The Hall plateaus rapidly vanish as the field is tilted off these magic angles. We identify two distinct particularities of these specific orientations, which may play a role in the emergence of the Hall plateaus.

DOI: 10.1103/PhysRevB.79.245124 PACS number(s): 71.70.Di, 71.18.+y, 72.15.Gd, 73.43.−f

I. INTRODUCTION

The quantum limit is attained when the magnetic field is strong enough to confine electrons to their lowest Landau level. Beyond this limit, an interacting two-dimensional electron gas can display the fractional quantum-Hall effect (FQHE).[1] In three dimensions, on the other hand,[2] the fate of the electron gas pushed to this ultraquantum regime is barely explored. Because of its low carrier concentration, elemental bismuth[3] provides a unique opportunity to attain the extreme quantum limit in a bulk metal with laboratory magnetic fields. Recent studies on bismuth has uncovered a rich and poorly understood physics beyond the quantum limit.[4,5] One central question is to determine if the band picture, which treats electrons as noninteracting entities, remains valid in such an extreme limit, where the interactions and their associated instabilities are enhanced[2] and the dimensionality is reduced.[6,7]

A first study[4] of high-field Nernst and Hall coefficients in bismuth-resolved unexpected anomalies at fields exceeding 9 T for a field roughly oriented along the trigonal axis. In this configuration, transport properties and their quantum oscillations are dominated by the holelike ellipsoid of the Fermi surface.[8,9] Since the quantum limit of these carriers occur at 9 T, the detected anomalies were attributed to interacting holelike quasiparticles at fractional filling factors.[4] Following this observation, a study of torque magnetometry[5] detected the quantum oscillations of the three electron pockets and their angular variation. In addition to the anomalies caused by the passage of successive Landau levels, this study resolved a field scale with a sharp angular variation and identified it as a phase transition of the quasiparticles of the electron pocket, which, in contrast to holes, present a Dirac spectrum.[10] The link between these two sets of observations was yet to be clarified. These experimental results initiated new theoretical investigations regarding the possible occurrence of FQHE in a bulk system[11,12] as well as the high-field electronic spectrum of bismuth.[13,14]

Here we present a study of Hall and longitudinal resistivities in the presence of a strong rotatable magnetic field. The magnetic field was tilted off the trigonal axis either in the (trigonal, bisectrix) or the (trigonal, binary) plane of the crystal. These transport measurements were preceded by a study of torque magnetometry in the same configuration, which confirms the observations reported by Li *et al.*,[5] and provided supplementary insight to the transport data.

The results presented in this paper allow us to conclude that (i) the Hall response is dominated by the carriers of the hole pocket of the bulk Fermi surface; (ii) the contribution of the quasiparticles of the electron pocket are visible as a perturbation to the overall conductivity (both longitudinal and transverse); and (iii) there are particular orientations of magnetic field (dubbed "magic angles") for which the Hall resistivity becomes field independent in the vicinity of 20 T.

Such a field-independent Hall resistivity has not been previously observed in any bulk quasi-isotropic material. They are concomitant with a sizable finite longitudinal resistivity. This latter feature is in sharp contrast with what is expected for a gapped quantum-Hall fluid in the well-understood case of two-dimensional quantum-Hall effect. On the other hand, it is yet to be demonstrated that the one-particle picture can explain a field-independent Hall resistivity in an ambipolar multivalley electron system.

In the final part of this paper, we use the magnetometry data to identify two particularities of these orientations. When the field is oriented along one these particular orientations, two remarkable features occur: (i) the cross section of the hole ellipsoid becomes equal to the cross section of one or two electron ellipsoids and (ii) the Landau level of three electron pockets empty at the same field. One or both of these peculiarities may be relevant to the emergence of the Hall plateaus.

II. SAMPLES AND METHODS

The samples were all cut from a large single crystal of bismuth several cm long.[15] While the residual resistivity ratio (RRR) [$\mathrm{RRR}=\rho(300\ \mathrm{K})/\rho(4.2\ \mathrm{K})$] of the mother crystal was 300, the RRR of the tailored samples with a typical

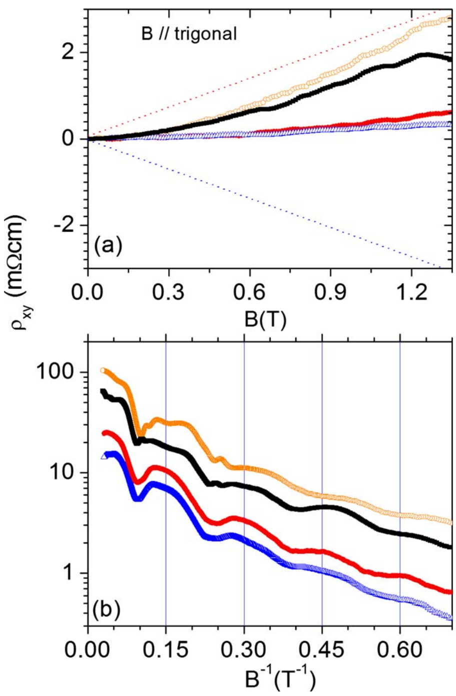

FIG. 1. (Color online) (a) The low-field dependence of the Hall resistivity, ρ_{xy}, in several bismuth crystals. All sets of data were obtained with the field oriented along the trigonal, the current applied along the bisectrix, and the voltage measured along the binary. The red (blue) dotted line corresponds to the classical Hall response expected for holes (electrons). (b) The same data are plotted as a function of inverse of magnetic field B^{-1} in a semilogarithmic plot. The period of quantum oscillations (0.15 T^{-1}) is similar but not the fine structure of the field dependence.

thickness of 0.8 mm was found to be much lower (~100) pointing to a mean-free path long enough to be affected by sample dimensions.[16] Resistivity and Hall effect were measured with a standard six-contact setup. For each orientation, the Hall data were collected after reversing the orientation of the magnetic field. Torque magnetometry was measured using a cantilever and a high-resolution capacitance bridge.

III. HALL EFFECT IN BISMUTH

A small magnetic field ($B > 0.1$ T) is sufficient to put bismuth in the strong-field limit ($\omega_c \tau \gg 1$). In this limit, the magnitudes of the Hall resistivity, ρ_{xy}, for a field applied along the trigonal and a current applied along the bisectrix was found to be different among various simples cut from the same single crystal. This striking variation can be seen in Fig. 1. In contrast, the magnetoresistance in these samples was found to be almost the same.

Bismuth is a compensated metal (where the density of electronlike carriers is equal to the concentration of holelike carriers: $n_e = n_h = 3 \times 10^{17}$ cm^{-3}. In a first approximation, its Hall response is expected to be zero. As seen in Fig. 1, for all samples studied in fields smaller than 0.1 T, the Hall response is vanishingly small in agreement with previous reports for this configuration.[17–19] In the high-field regime (i.e., for $B > 0.1$ T), a positive signal emerges which remains smaller than $\rho_{xy} = B/(n_h e)$, the expected signal for holes. The variety observed in the amplitude of this signal can be attributed to two uncontrolled features.

First of all, because of the extreme anisotropy of the electron pockets, the available phase space for scattering the quasiparticles of the electron pocket is not uniform, and therefore the electron mobility is drastically angular dependent. In the weak-field limit, the electronic mobility along the binary and bisectrix directions differ by a factor of 37.[19] This anisotropy persists in the high-field regime ($\omega_c \tau \gg 1$) as evidenced by the large angular variation observed in transverse magnetoresistance for a field rotating in the bisectrix, binary plane 1.[17] The mean-free path of both holes and electrons is comparable with sample dimensions. Therefore, the overall contribution of electronlike carriers to Hall response can crucially depend on contact geometry.

A second relevant feature is the extreme anisotropy of the conductivity tensor. Previously reported data[17–19] indicate that at $B = 1$ T, ρ_{xz} (that is the Hall resistivity for a field applied along binary) is 2 orders of magnitude larger than ρ_{xy} (the one measured here for a field along trigonal). In such a context, any small misalignment between the three presumably perpendicular vectors (B, J, and E) would lead to a contamination of ρ_{xy} with ρ_{xz}. Since the latter is also an off-diagonal component of the conductivity tensor, this contamination cannot be eliminated by reversing the field.

With this variety in the magnitude of the measured Hall resistivity in mind, let us note that all samples presented a positive signal (when $B > 1$ T) and all of them showed the same period of quantum oscillations (0.15 T^{-1}), as seen in Fig. 1(b). Both these features point to holelike carriers as the dominant contributor to the Hall response. As detailed below, this is confirmed by the angular dependence of the measured ρ_{xy}.

The main result reported in this paper—the existence of particular orientations of the magnetic field, for which the Hall response becomes flat in a finite field window—was found to be experimentally robust in spite of the large variation in both the magnitude and the fine structure of the Hall coefficient across the samples.

IV. ANGULAR DEPENDENCE OF RESISTIVITY TENSOR

An extensive study of the field dependence of longitudinal, ρ_{xx}, and transverse, ρ_{xy}, resistivities in a Bi single crystal with different angles between the magnetic field and the trigonal axis is presented in Fig. 2. Two different planes of rotations where investigated for the same crystal with the same electrodes. The field was rotated either in the (trigonal, binary) or in the (trigonal, bisectrix) plane. In the first (second) rotating plane, θ_1 (θ_2) designates the angle between the field and the trigonal axis.

In both planes of rotation, high-field features, occurring at fields exceeding the quantum limit, are particularly visible in ρ_{xy}. They rapidly evolve as the magnetic field is tilted a few degrees off the trigonal axis. The rapid angular evolution indicates that at least some of the ultraquantum transport anomalies, observed in both bismuth[4] and in $Bi_{0.96}Sb_{0.04}$,[20] are extremely sensitive to the precise orientation of the magnetic field.

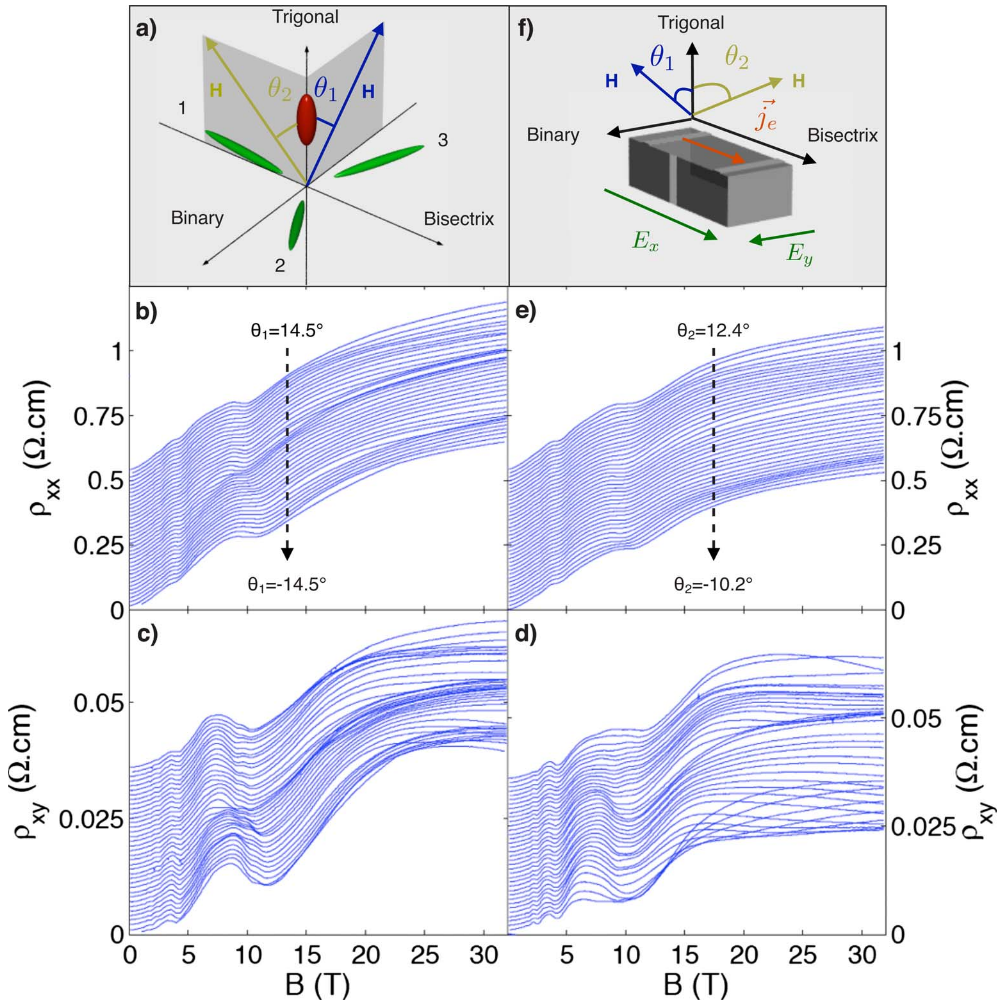

FIG. 2. (Color online) (a) The Fermi surface of bismuth. The magnetic field was applied along an orientation tilted off the trigonal axis by sweeping either θ_1 or θ_2. Panels (b) and (c) present the Hall and resistivity data obtained in the first configuration at $T=1$ K. Panels (d) and (e) present the same for second configuration. Curves are shifted for clarity. (f) The sample geometry and the orientation of various relevant vectors.

V. QUANTUM OSCILLATIONS IN CHARGE TRANSPORT AND IN TRANSVERSE MAGNETIC SUSCEPTIBILITY

Charge transport in this configuration is dominated by the contribution of holelike carriers. This conclusion is based on the sign of the Hall effect and the fact that the period of quantum oscillations in both ρ_{xx} and ρ_{xy} of all samples ($0.15\ \mathrm{T}^{-1}$) corresponds to what is reported for the hole pocket of the *bulk* Fermi surface by the de Haas-van Alphen[21] and the Shubnikov-de Haas[8] studies as well as the quantum oscillations of the Nernst coefficient.[9]

The moderate anisotropy of the holelike ellipsoid implies that tilting the magnetic field a few degrees off the trigonal axis does not significantly modify the period of oscillations. This is indeed the case as seen in Fig. 3, which compares this feature with the sharp angular variation in the quantum oscillations of the transverse magnetic susceptibility ($\chi_\perp = \tau/B^2$, where τ is the magnetic torque). The torque response is dominated by the more anisotropic and threefold degenerate electron pockets whose diamagnetic response is accentuated by their Dirac dispersion.[5] As the field is tilted, the quantum oscillations of the torque response rapidly vary as expected for the electron pockets. As seen in the upper panel, the period of quantum oscillations of Hall and torque data is in rather good agreement with the expected periods for electrons and holes.[21]

VI. FIELD SCALES OF ELECTRON POCKETS AND MAGIC ANGLES

The contribution of the electronlike carriers to charge transport can be resolved by putting under scrutiny the angular dependence of the magnetoresistance, ρ_{xx} as seen in Fig. 4. The rotating magnetic field generates sharp minima in the angular dependence of ρ_{xx}. When the field was rotated in the (trigonal, bisectrix) plane, the field dependence of these anomalies define quasivertical field scales in the (B, θ_1) plane, which are symmetrical with respect to the $\theta_1=0$ line. The two central lines lie very close to the field scale reported by Li and co-workers[5] and identified as a phase transition involving the electron pockets. Note that this field scale

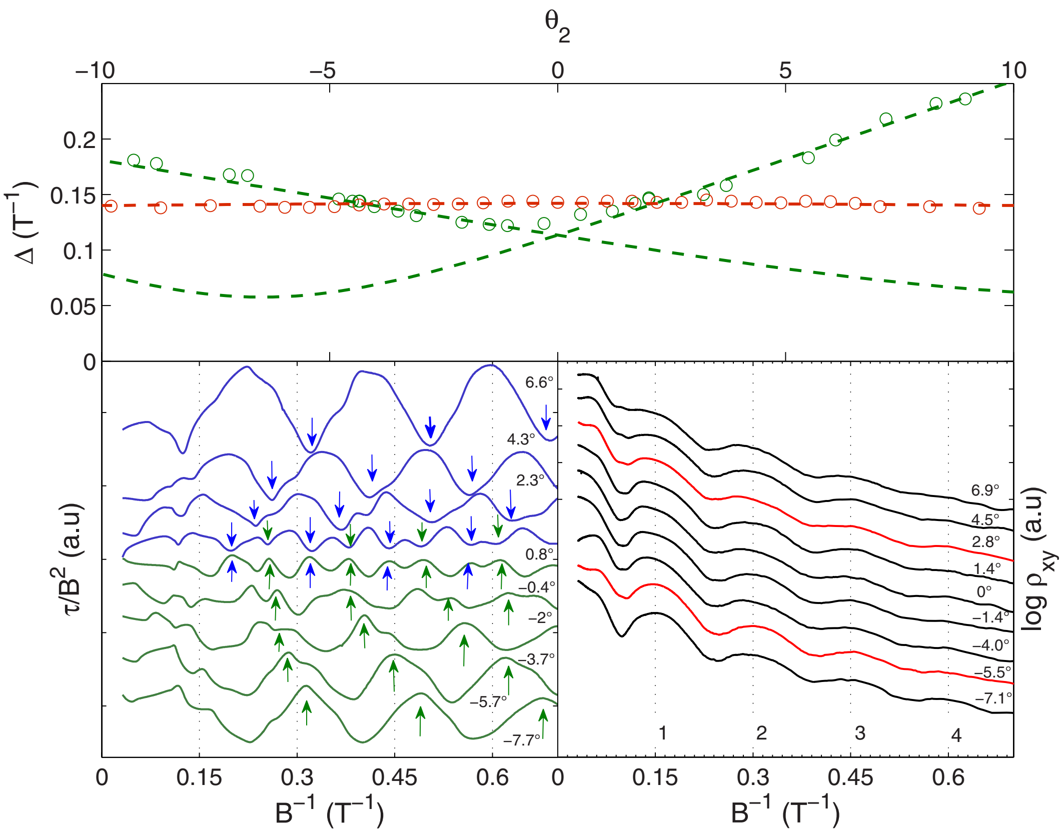

FIG. 3. (Color online) Lower panels: transverse susceptibility (left) and Hall resistivity (right) as functions of the inverse of the magnetic field for different θ_2 tilt angles slightly off the trigonal axis. Curves are shifted for clarity. The red Hall curves correspond to magic angles. Arrows allow to follow the displacement of an extremum in the torque data. Upper panel: the angular dependence of the period of quantum oscillations seen in the Hall (red symbols) and torque (green symbols) data. Green (red) dotted lines correspond to the expected angular dependence of electron (hole) ellipsoids.

tracks the 0_e^+ Landau level of two electron pockets according to calculations.[13,14]

In this plane of rotation, the minima in $\rho_{xx}(\theta_1)$ and in $\rho_{xy}(\theta_1)$ are concomitant; the Hall response does not present any additional structure and does not become field independent in any finite field window, at least up to 28 T. In particular, as seen in panel (b) of Fig. 4, there is no field independent ρ_{xy} in the vicinity of 20 T for any θ_1 angle.

When the field rotates in the other plane, additional features emerge. Figure 5 presents the data obtained for the same crystal with the same contacts for a field rotating in the (trigonal, bisectrix) plane. Here also minima in $\rho_{xx}(\theta_2)$ track quasivertical field scales in the (B, θ_2) plane. However, the angular separation between these lines exceeds what is expected according to the calculated Landau levels of electrons.[14] More strikingly, the angular dependence of the Hall response presents an additional structure. There are two narrow angular windows, 8.4° apart, in which $\rho_{xy}(\theta_2)$ becomes field independent in the vicinity of 20 T. As seen in the inset, the Hall plateau rapidly vanishes as the field is tilted a fraction of degree away from these magic angles.

Figure 6 presents the field dependence of ρ_{xy} for two orientations for which ρ_{xy} does not vary with magnetic field in a finite field window. As seen in the inset, the flatness of the Hall resistivity in this plateau window is comparable with the experimental noise ($\frac{\delta\rho}{\rho} \sim \times 10^{-3}$).

We have confirmed the robustness of this observation by studying different crystals and in particular the same crystal with new sets of electrodes in different magnet laboratories. Figure 7 presents another set of data obtained in a different experiment with the same single crystal with a different set of electrodes. Both the magnitude and the fine structure of ρ_{xy} are visibly different. However, a flat ρ_{xy} in the vicinity of 20 T for two different orientations 8.4° apart are detectable. A similar conclusion was drawn from other similar sets of study.

To sum up, we rotated the same sample with the same electrodes in two different ways. When the field was rotated in the (trigonal, binary) plane, no Hall plateaus were observed. When it was rotated in the (trigonal, bisectrix) plane, there are two specific orientations of magnetic field, for which ρ_{xy} presents 20 T plateaus. The latter observation was reproduced with different electrodes and different samples.

Note that such a plateau cannot be attributed to a possible contamination of our ρ_{xy} data with a finite ρ_{xz} contribution. Indeed, the latter is expected to change sign when θ_2 changes sign. Therefore, the contaminating signal should have opposite signs for negative and positive angles. A plateau at a negative θ_2 may occur as a result of an accidental cancellation of two opposing slopes. However, in such a case, no such cancellation is expected to occur for a positive θ_2. In the next section, we proceed to discuss the possible sources of a flat Hall resistivity for specific orientations of magnetic field.

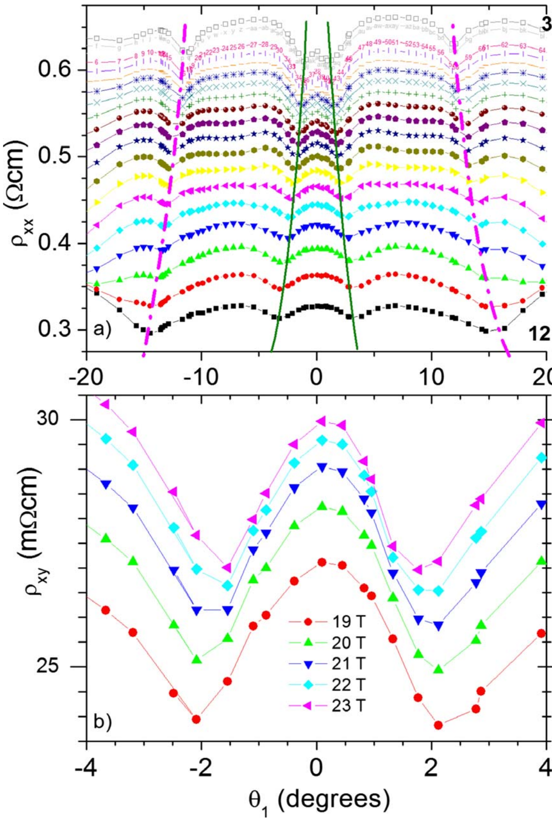

FIG. 4. (Color online) (a) The angular dependence of the longitudinal resistivity as a function of θ_1 for different magnetic fields. Lines are guide to lines to follow the field dependence of the minima. (b) The same for the Hall resistivity in a restricted field window around 20 T. The Hall resistivity does not become field independent for any orientation of magnetic field.

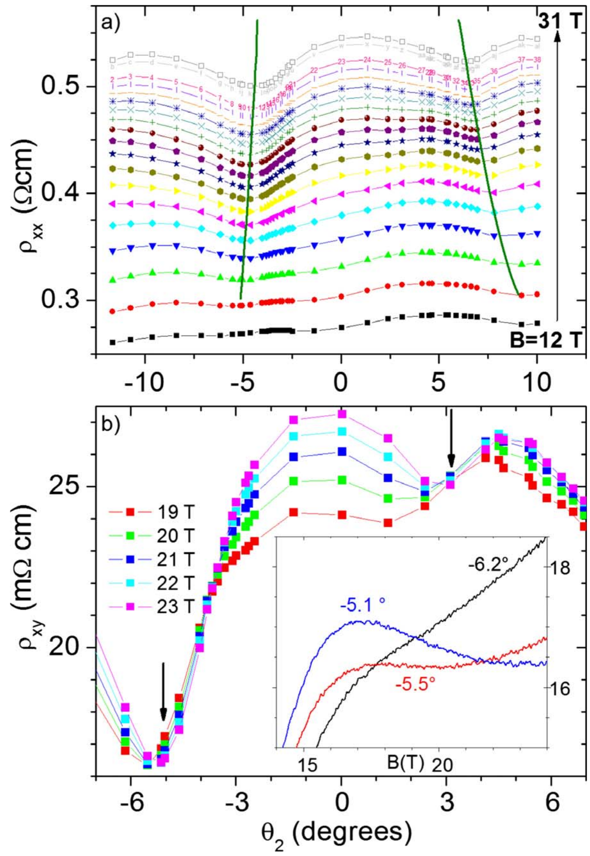

FIG. 5. (Color online) (a) The angular dependence of the longitudinal resistivity as a function of θ_2 for different magnetic fields. Lines are guide to lines to follow the field dependence of the minima. (b) The same for the Hall resistivity in a restricted field window around 20 T. Arrows indicate magic angles at which the Hall resistivity becomes flat. The inset shows a Hall plateau and its angular fragility.

VII. DISCUSSION

In the absence of magnetic field, the anisotropy of charge conductivity in elemental bismuth is less than 2. This quasi-isotropy distinguishes the context of our observation from all cases of Hall plateaus including the integer quantum-Hall effect seen in bulk-layered systems such as the Bechgaard salts.[22,23]

We note that the Hall plateaus occur in the vicinity of 20 T, which is roughly three times the main frequency of the quantum oscillations, B_0=6.67 T=0.15^{-1} T. Naively, this corresponds to a filling factor of 1/3 for holes. However, both the carrier density and the effective filling factor at high fields and arbitrary angles could be significantly different from the one estimated from the low-field spectrum.

Moreover, in contrast with the nondissipative behavior expected for an incompressible quantum-Hall fluid, longitudinal resistivity in our samples remains always finite; this appears to rule out a scenario based on the quantum-Hall effect as currently understood. We note that very recently a gapless fractional quantum-Hall state specific to three dimensions was theoretically proposed.[12]

Finding a credible scenario for a Hall plateau in a bulk system in the presence of the z-axis degeneracy remains a challenge. The presence of several subsystems adds twists to

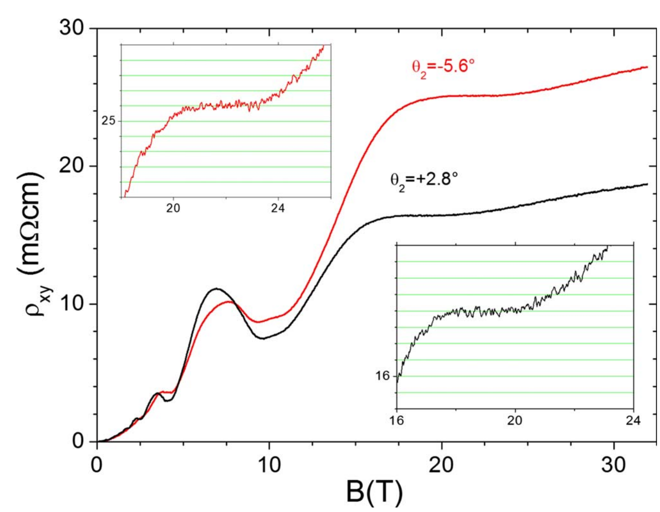

FIG. 6. (Color online) The field dependence of ρ_{xy} at magic angles. The insets are zooms on restricted windows. The angular separation between the green lines (0.05 mΩ cm) is the experimental resolution.

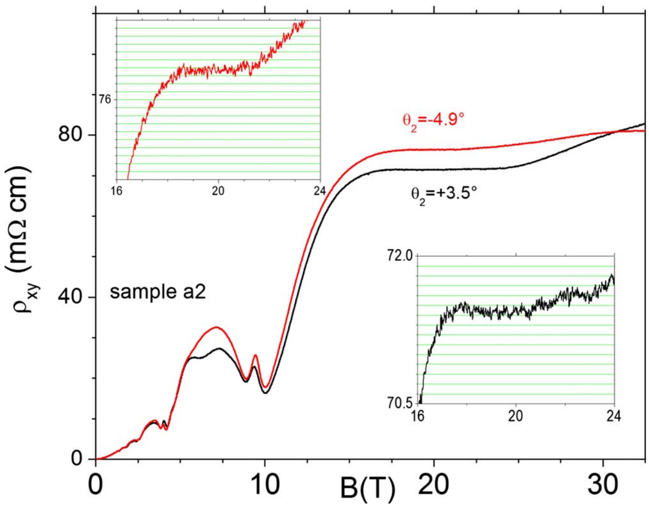

FIG. 7. (Color online) The field dependence of ρ_{xy} obtained in a different study. Both the magnitude and the fine structure of $\rho_{xy}(H)$ are different. Yet, there are still Hall plateaus at two specific orientations in two field windows close to 20 T.

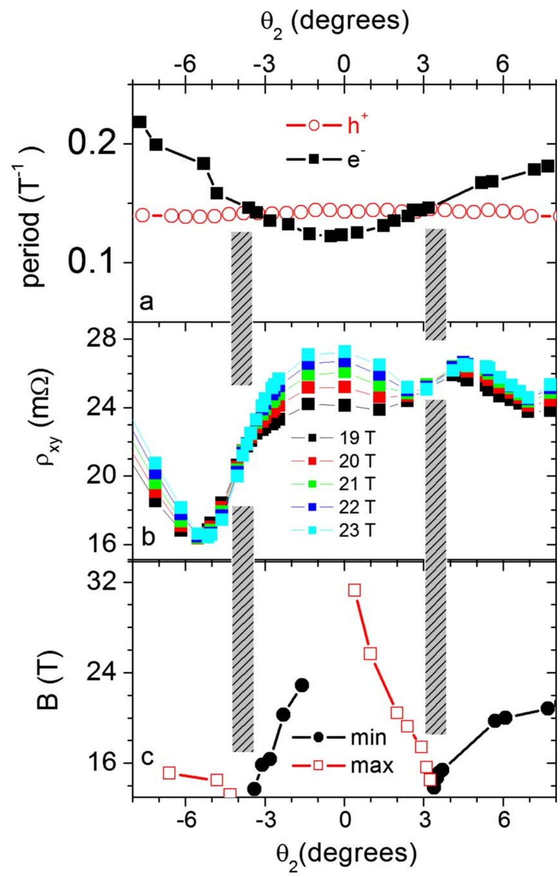

FIG. 9. (Color online) Left panel: comparison of the angular dependence of the (a) period quantum oscillations, (b) ρ_{xy}, and (c) high-field torque anomalies. The gray vertical bands are guides for the eye and point to the two specific orientations resolved in these sets of data.

the problem. As the magnetic field is swept or rotated, both electrons and holes modify their zero-field band parameters in order to ensure charge neutrality.[24,25] The steady and large high-field magnetostriction[26] suggests a sizable field-induced correction of the carrier density, which could significantly affect the transport properties.[27] The restriction of the Hall plateaus to a finite angular and field window points to a subtle balance of unidentified parameters. According to a recent theoretical work, FQHE can occur in a bulk quasi-isotropic system only if the electrons reorganize themselves in layers perpendicular to the magnetic field.[11] It is tempting to speculate that the magic angles correspond to a specific reorganization of charge distribution fulfilling the required conditions. Measurements of electric conductivity along the magnetic-field orientation would be helpful in this context.

What is particular about these magic angles? As seen in Fig. 8, a drastic change in torque response occurs when the field orientation crosses such an angle. In particular, a maximum in transverse susceptibility becomes a minimum. The field scales traced by these anomalies follow the lowest Landau level of the electron ellipsoids according to the theoretical phase diagram.[13,14]

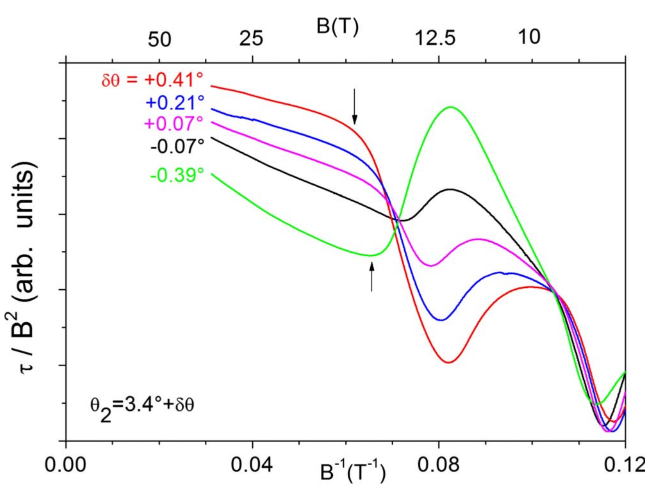

FIG. 8. (Color online) Transverse susceptibility as a function of the inverse of magnetic field near a magic angle. Note the drastic change in a maximum to a minimum (arrows) across this specific orientation.

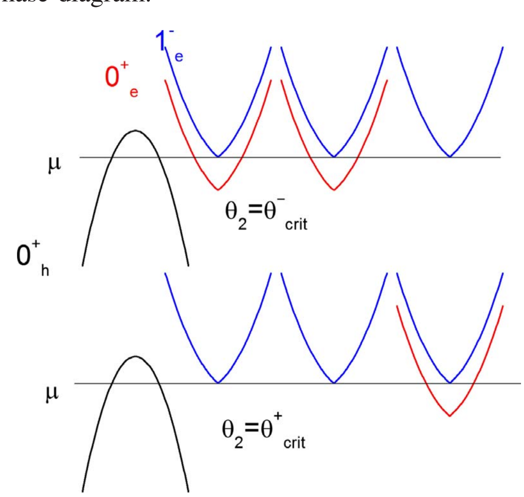

FIG. 10. (Color online) Schematic representation of Landau levels at two critical angles possibly corresponding to the experimentally observed magic angles.

Experimentally, the angular separation between the occurrences of the 20 T Hall plateaus is 8.4°. Two distinct and possibly relevant features of these two field orientations are identified in Fig. 9. The first concerns both holes and electrons. As seen in the figure, close to the two magic angles, the periods of quantum oscillations for holes and electrons become equal. This may be an accident. On the other hand, the equality between the cross sections of the hole ellipsoid with one or two electron ellipsoids may have significant consequences. In particular, it may lead to the possible commensurability of hole and electron wave vectors along the magnetic field for these particular orientations and lead to an instability paving the way to the emergence of the Hall plateau.

The other specificity of a magic angle concerns solely the electron pockets. As seen in Fig. 9, the structure in the angular dependence of ρ_{xy} shows a remarkable correlation with the phase diagram coming out of the torque data. A simultaneous Landau-level crossing of distinct electron pockets occurs, when the field is along one of the two specific orientations. As mentioned previously, the Hall plateaus emerge only when the field rotates in the (trigonal, bisectrix) plane. Interestingly, only in this configuration there are two specific field orientations for which the Landau levels of *all three* electron pockets cross the chemical potential at the same field (Fig. 10). This is because when θ_2 is swept, two electron pockets [Nos. 2 and 3 of Fig. 1(a)] remain degenerate. According to theoretical calculations, the angular distance between these two points in the (B, θ_2) plane is 8°.[14] It has been argued that Coulomb interactions are significantly enhanced whenever a low Landau level crosses the Fermi level.[13] Future investigations would tell if this has any correlation with the emergence of Hall plateaus.

ACKNOWLEDGMENTS

We thank J. Alicea, L. Balents, Y. Kopelevich, D. Maslov, G. Mikitik, A. J. Millis, Y. Sharlai, and V. Yakovenko for useful discussions. This work is part of DELICE, a project supported by the Agence Nationale de Recherche (Grant No. ANR-08-BLAN-0121-02). Research at the NHMFL is supported by the NSF, the State of Florida, and the DOE.

[1] T. Chakraborty and P. Pietilainen, *The Quantum Hall Effects* (Springer, Berlin, 1995).
[2] B. I. Halperin, Jpn. J. Appl. Phys., Suppl. **26**, 1913 (1987).
[3] V. S. Edel'man, Adv. Phys. **25**, 555 (1976).
[4] K. Behnia, L. Balicas, and Y. Kopelevich, Science **317**, 1729 (2007).
[5] L. Li, J. G. Checkelsky, Y. S. Hor, C. Uher, A. F. Hebard, R. J. Cava, and N. P. Ong, Science **321**, 547 (2008).
[6] V. M. Yakovenko, Phys. Rev. B **47**, 8851 (1993).
[7] C. Biagini, D. L. Maslov, M. Yu. Reizer, and L. I. Glazman, EPL **55**, 383 (2001).
[8] S. G. Bompadre, C. Biagini, D. Maslov, and A. F. Hebard, Phys. Rev. B **64**, 073103 (2001).
[9] K. Behnia, M.-A. Measson, and Y. Kopelevich, Phys. Rev. Lett. **98**, 166602 (2007).
[10] P. A. Wolff, J. Phys. Chem. Solids **25**, 1057 (1964).
[11] F. J. Burnell, B. A. Bernevig, and D. P. Arovas, Phys. Rev. B **79**, 155310 (2009).
[12] J. Alicea and L. Balents, Phys. Rev. B **79**, 241101(R) (2009).
[13] M. Levin and M. P. A. Fisher, Phys. Rev. B **79**, 235315 (2009).
[14] Y. V. Sharlai and G. P. Mikitik, Phys. Rev. B **79**, 081102(R) (2009).
[15] J. Boxus and J.-P. Issi, J. Phys. C **10**, L397 (1977).
[16] N. Garcia and Y. H. Kao, Phys. Lett. **26**, 373 (1968).
[17] S. Mase, S. von Molnar, and A. W. Lawson, Phys. Rev. **127**, 1030 (1962).
[18] A. N. Friedman, Phys. Rev. **159**, 553 (1967).
[19] R. Hartman, Phys. Rev. **181**, 1070 (1969).
[20] A. Banerjee, B. Fauqué, K. Izawa, A. Miyake, I. Sheikin, J. Flouquet, B. Lenoir, and K. Behnia, Phys. Rev. B **78**, 161103(R) (2008).
[21] R. N. Bhargava, Phys. Rev. **156**, 785 (1967).
[22] J. R. Cooper, W. Kang, P. Auban, G. Montambaux, D. Jerome, and K. Bechgaard, Phys. Rev. Lett. **63**, 1984 (1989).
[23] S. T. Hannahs, J. S. Brooks, W. Kang, L. Y. Chiang, and P. M. Chaikin, Phys. Rev. Lett. **63**, 1988 (1989).
[24] G. E. Smith, G. A. Baraff, and J. M. Rowell, Phys. Rev. **135**, A1118 (1964).
[25] M. P. Vecchi, J. R. Pereira, and M. S. Dresselhaus, Phys. Rev. B **14**, 298 (1976).
[26] J.-P. Michenaud, J. Heremans, M. Shayegan, and C. Haumont, Phys. Rev. B **26**, 2552 (1982).
[27] J.-P. Michenaud and B. Lenoir, Physica B **387**, 371 (2007).