

Hall plateaus at magic angles in bismuth beyond the quantum limit

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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.

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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 but poorly understood physics beyond the quantum limit[4, 5]. One central question is to determine if the band picture, which treats electrons as non-interacting 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 hole-like 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 hole-like quasi-particles 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 quasi-particles of the electron pocket, which, in contrast to holes, present a Dirac spectrum[10]. The link between these two sets of observation remained unclear. These experimental results initiated new theoretical investigations regarding the possible occurrence of FQHE in a bulk system[11] as well as the high-field electronic spectrum of bismuth[12, 13].

Here we present a study of Hall and longitudinal resis-

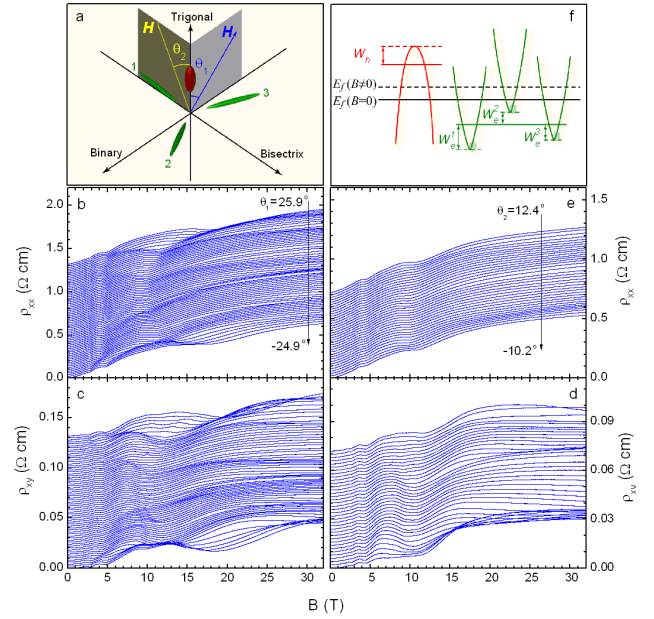


FIG. 1: a) The Fermi surface of bismuth. The magnetic field was applied along an orientation tilted off the trigonal axis by sweeping either θ_1 or θ_2 . Panels b and c (d and e) present the Hall and resistivity data obtained in the first (second) case at $T = 1$ K. Curves are shifted for clarity. f) Schematic view of the band structure. In presence of magnetic field along an arbitrary direction, the Fermi level, the top of the hole band and the bottom of the electron bands shift to new positions (dotted lines) in order to maintain charge neutrality.

tivities in presence of a strong rotatable magnetic field. 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 provide supplementary insight to the transport data. The results 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 quasi-particles of the electron pocket are visible as a perturbation to the overall conductivity (both longitudinal and transverse); 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 Hall plateau has not been previously observed in any bulk quasi-isotropic material and its explanation is a challenge for the one-particle picture. Two distinct features of these orientations, which may be relevant to the emergence of the plateaus can be readily identified in our data.

The samples were all cut from a large single crystal of bismuth several cm long[14]. While the Residual Resistivity Ratio ($RRR = \rho(300K)/\rho(4.2K)$) of the mother crystal was 300, the RRR of the tailored samples with a typical 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[15]. Resistivity and Hall effect were measured with a standard 6-contact set-up. Torque magnetometry was measured using a cantilever and a high-resolution capacitance bridge. The current was applied along the bisectrix and the magnetic field was tilted off the trigonal axis either in the (trigonal, bisectrix) or the (trigonal, binary) plane of the crystal. Two miniature Hall probes were used to determine the orientation with a relative resolution lower than 0.1 degree, but with an absolute uncertainty of about 1 degree.

Fig. 1 presents the field dependence of longitudinal, ρ_{xx} , and transversal, ρ_{xy} , resistivity. 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. This rapid angular evolution indicates that the ultra-quantum transport anomalies which were reported in both bismuth[4] and in $\text{Bi}_{0.96}\text{Sb}_{0.04}$ [16] for a magnetic field roughly along the trigonal axis, are extremely sensitive to the orientation of the magnetic field.

The Fermi surface of bismuth consists of a hole ellipsoid and three electron pockets. In a first approximation, the Hall response of such a compensated metal, with strictly equal concentration of electrons and holes, should be zero. A finite signal is expected when the mobility of one type of carriers exceeds the mobility of the other. In the case of bismuth, due to a large interband contribution, the Hall response of the electron pockets is expected to be non-trivial even in the weak-field limit[17]. In our experimental configuration (field along trigonal and current along bisectrix axes), the Hall response of all samples studied was found to be vanishingly small or slightly negative in the weak-field limit (below 0.1 T) in agreement with previous reports for this configuration[18, 19, 20]. Moreover, in all of them a positive Hall signal emerged when the field exceeded 0.2 T. The magnitude of this large-field Hall coefficient, R_H , was found to be sample dependent but always smaller or of the order of $\frac{1}{n_{he}}$ ($n_h = 2.7 \times 10^{-17} \text{ cm}^{-3}$ is the density of hole-like carriers). The positive sign of the Hall response indicates that

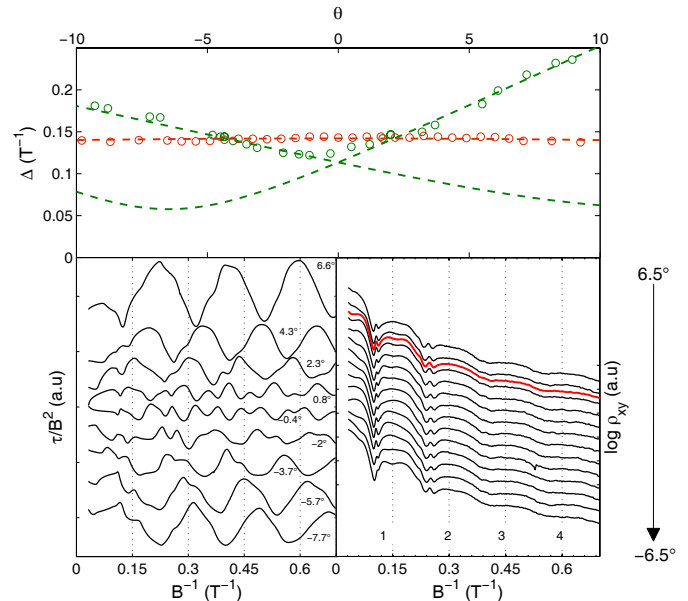


FIG. 2: Transverse susceptibility (lower left) and Hall resistivity (lower right) as a function of the inverse of the magnetic field for different $[\theta_2]$ tilt angles slightly off the trigonal axis. Curves are shifted for clarity. The red Hall curve corresponds to a magic angle. Upper panel shows 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 periods for electron (hole) ellipsoids.

holes dominate the Hall response as a result of higher hole mobility in this configuration. This conclusion is supported by the fact that the period of quantum oscillations in all samples (0.15 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 hole-like 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. 2, which compares this feature with the sharp angular variation of 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 three-fold degenerate electron pockets whose diamagnetic response is accentuated by their Dirac dispersion. 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.

The contribution of the electron-like carriers to hole-

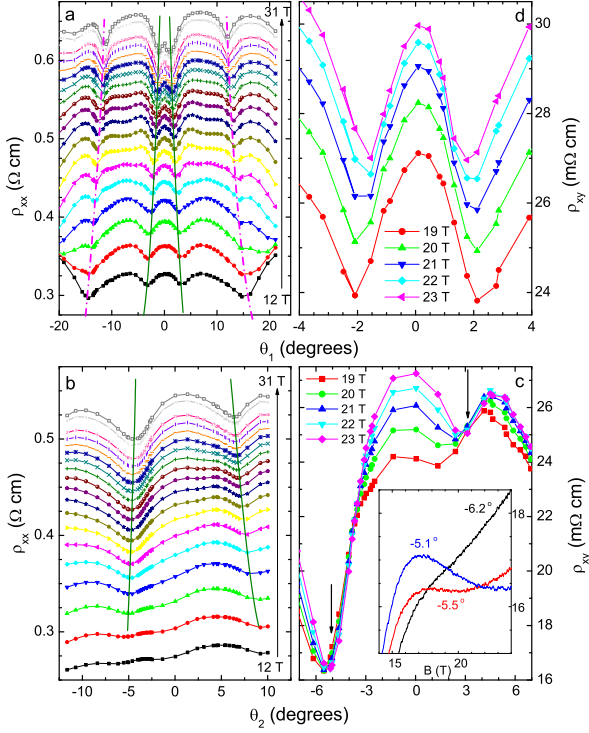


FIG. 3: a) and b) The angular dependence of the longitudinal resistivity a function of θ_1 and θ_2 for different magnetic fields. Lines are guide to lines to follow the field-dependence of the minima. c) and d) Same for the Hall resistivity in a restricted field window around 20 T. In panel (c) arrows indicate magic angles and the inset shows a Hall plateau and its angular fragility.

dominated charge transport can be resolved by putting under scrutiny the angular dependence of the magnetoresistance, ρ_{xx} as seen in Fig. 3. 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 quasi-vertical 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 tracks the 0_e^+ Landau levels of two electron pockets according to calculations [12, 13]. In this configuration, the minima in $\rho_{xx}(\theta_1)$ and in $\rho_{xy}(\theta_2)$ 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.

The lower panels of the same figure present the data obtained for the same crystal with the same contacts for a field rotating in the (trigonal, bisectrix) plane. Here also minima in $\rho_{xx}(\theta_2)$ trace quasi-vertical lines 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[13]. More strikingly,

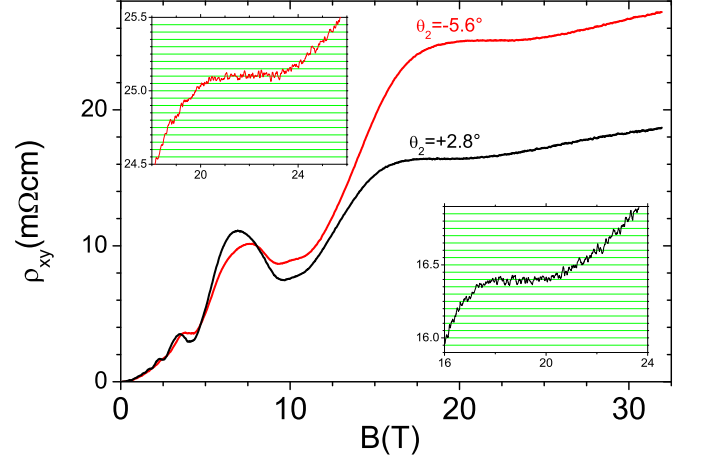


FIG. 4: The field dependence of ρ_{xy} at magic angles. The Insets are zooms on restricted windows with horizontal lines separated by $0.05 \text{ m}\Omega \text{ cm}$ indicating a flatness of $5 \cdot 10^{-3}$.

the angular dependence of the Hall response presents an additional structure. There are two narrow angular windows in which, $\rho_{xy}(\theta_2)$ becomes field-independent in the vicinity of 20 T. In other words, there are two orientations for which ρ_{xy} does not vary with magnetic field in a finite field window. As seen in the inset, the Hall plateau rapidly vanishes as the field is tilted a fraction of degree away from these “magic angles”.

Fig.4 presents the field-dependence of ρ_{xy} at two magic angles. As seen in the inset, that the flatness of the Hall resistivity is comparable with the experimental noise (3×10^{-3}). The Hall plateaus centered around 19 T and 21 T, roughly three times the main frequency of the quantum oscillations, $B_0 = 0.15^{-1} T$. 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.

In absence of magnetic field, the anisotropy of charge conductivity in elemental bismuth is less than two. 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]. Moreover, in contrast with the non-dissipative behavior expected for an incompressible quantum Hall fluid, longitudinal resistivity in our samples remains always finite.

Finding a credible scenario for a Hall plateau in a bulk system in presence of the z-axis degeneracy remains a challenge. The presence of several subsystems adds twists to the problem. As the magnetic field is swept or rotated, both electrons and holes modify their zero-field band parameters in order to insure charge neutrality[23, 24]. The steady and large high-field magnetostriction[25] suggests a sizeable field-induced correction of the carrier density (Fig. 1f), which could significantly affect the trans-

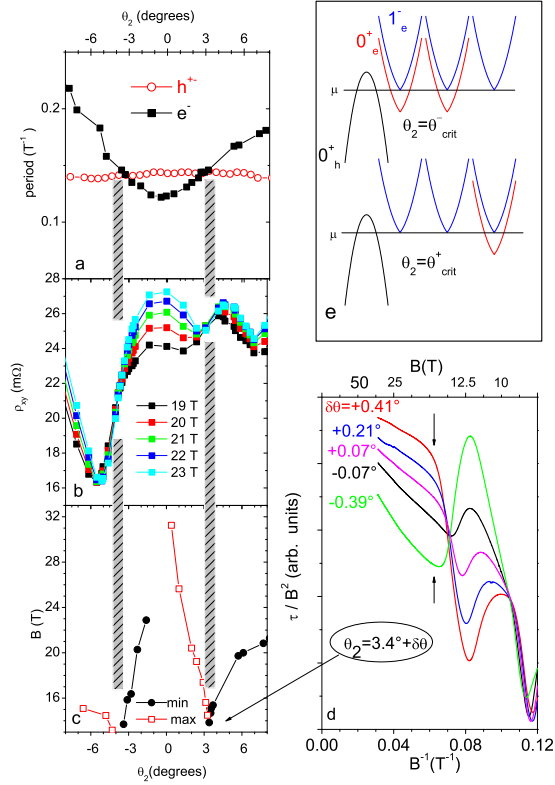


FIG. 5: : Left panel : Comparison of the angular dependence of the a) period quantum oscillations, b) ρ_{xy} and c) high-field torque anomalies. Gray vertical bars mark magic angles. d) Drastic change of transverse susceptibility near a magic angle. Arrows indicate anomalies which are tracked in panel c. e) Schematic representation of Landau levels at two critical angles possibly corresponding to the experimentally observed magic angles.

port properties[26]. The restriction of the Hall plateaus to a finite angular and field window points to a subtle balance of parameters. According to a recent theoretical work, FQHE can occur in a bulk quasi-isotropic system only if the electrons re-organize themselves in layers perpendicular to the magnetic field. It is tempting to speculate that the “magic angles” correspond to a specific re-

organization of charge distribution fulfilling the required conditions. Transport measurements along z -axis would be helpful for checking this hypothesis.

Experimentally, the angular separation between the two magic angles is 8.4 degrees. Two distinct and possibly relevant features of these two field orientations can be readily identified. The first concerns both holes and electrons. As seen in 5a, 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 possible commensurability of hole and electron wave-vectors along the magnetic field for these particular orientations may 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. 5 (panels c and d), a drastic change in torque response occurs when the field orientation crosses a magic angle. According to the theoretical phase diagram[13], this point corresponds to a simultaneous Landau level crossing of distinct electron pockets. As detailed above, 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. 5e). This is because when θ_2 is swept, two electron pockets (no 2 and 3 of Fig. 1a) remain degenerate. According to theoretical calculations, the angular distance between these two points in the (B, θ_2) plane is 8 degrees [13]. It has been argued that Coulomb interactions are significantly enhanced whenever a low Landau level crosses the Fermi level[12]. These two particularities of the magic angles appear as clues to plausible scenarios for a field-independent Hall response.

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