

Oscillating Nernst-Ettingshausen Effect in Bismuth across the Quantum Limit

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In elemental bismuth, 10^5 atoms share a single itinerant electron. Therefore, a moderate magnetic field can confine electrons to the lowest Landau level. We report on the first study of metallic thermoelectricity in this regime. The main thermoelectric response is off-diagonal with an oscillating component several times larger than the nonoscillating background. When the first Landau level attains the Fermi energy, both the Nernst and the Ettingshausen coefficients sharply peak, and the latter attains a temperature-independent maximum. These features are yet to be understood. We note a qualitative agreement with a theory invoking current-carrying edge excitations.

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Bismuth crystallizes in a rhombohedral structure, which can be obtained by distorting a cubic lattice along its body diagonally. As a consequence of this slight deviation from higher symmetry, it becomes a semimetal with a Fermi surface occupying a tiny fraction of the Brillouin zone [1]. It has frequently played a pioneer role in the history of metal physics. Phenomena such as giant magnetoresistance, de Hass–van Alphen effect, Shubnikov–de Hass effect, and oscillatory magnetostriction were all first observed in bismuth [1]. The Nernst effect was also originally discovered in bismuth [2]. The archetypical semimetal and its unique properties are attracting new attention [3], following the recent observation of Dirac Fermions in graphite [4,5] and graphene [6,7]. Electronic transport in bismuth is indeed fascinating: A magnetic field can enhance resistivity by a factor of 10^6 without saturation. The quasilinear magneto-resistance is incompatible with the semiclassical theory [8]. Possible occurrence of exotic field-induced instabilities ranging from an excitonic insulator [9] to a topological insulator driven by spin-orbit interactions [10] continues to fuel research and debate [11–14].

Previous studies of thermoelectric response of bismuth in quantizing magnetic fields were performed decades ago and restricted to temperatures above 4.2 K and to relatively low fields ($B < 5$ T) [15,16]. Here, we present a study of thermoelectricity extending down to 0.28 K and up to 12 T. This allows us to attain the quantum limit, when the magnetic length equals the Fermi wavelength. Both transverse (Nernst) and longitudinal (Seebeck) components of the thermoelectric tensor display quantum oscillations, but the Nernst response dominates by far. The Nernst signal is large when a Landau level meets the chemical potential, and is damped when the latter lies between two successive Landau levels. The change in the magnitude is much larger than the quantum oscillations of the density of states monitored by the Shubnikov–de Hass effect. These experimental findings are a challenge to our understanding

of weakly-interacting electrons in the quantum limit. Qualitatively, they are strikingly similar to a recent theoretical predication invoking edge states in the context of the Quantum Hall Effect [17]. We also note, but fail to explain, that the Ettingshausen coefficient becomes temperature-independent in the low-temperature limit. This offers another notable constraint for theory.

A longitudinal thermal gradient can produce a longitudinal electric field (Seebeck effect), a transverse electric field (Nernst effect), and a transverse thermal gradient (Righi-Leduc effect). We used a one-heater-three-thermometers setup to measure the two thermoelectric coefficients, the thermal and electric conductivities as well as the Righi-Leduc coefficient. Because of the dominance of the lattice thermal conductivity [18], the latter was found to be very small. Therefore, the measured adiabatic Nernst coefficient is identical to the isothermal one. The physical properties of the bismuth single crystal used in this study ($RRR = 47$, dimensions $0.8 \times 2.2 \times 4$ mm³) were presented in a recent communication by the authors [18]. In all measurements detailed below, the direction of the applied current is along the binary axis.

The raw data obtained for a field along the trigonal axis at $T = 0.28$ K is shown in Fig. 1. The longitudinal and the transverse voltages created by applying a constant heat current (which, since the thermal conductivity is field-independent implies a constant $\nabla_x T$) are plotted as a function of magnetic field swept from -12 T to $+12$ T. The transverse signal is by far larger (reflecting the dominance of the Nernst over the Seebeck response) and contaminates the longitudinal voltage as evidenced by the sign reversal of the latter when the field is reversed. Panel (b) of the same figure shows the Nernst ($S_{xy} = E_y/\nabla_x T$) and Seebeck ($S_{xx} = E_x/\nabla_x T$) coefficients extracted from the odd (even) component of $V_y(B)$ [$V_V(B)$]. Both show oscillations, but the transverse component is clearly dominant.

The sheer magnitude of the oscillations in S_{xy} compared to the nonoscillating background is remarkable. In bis-

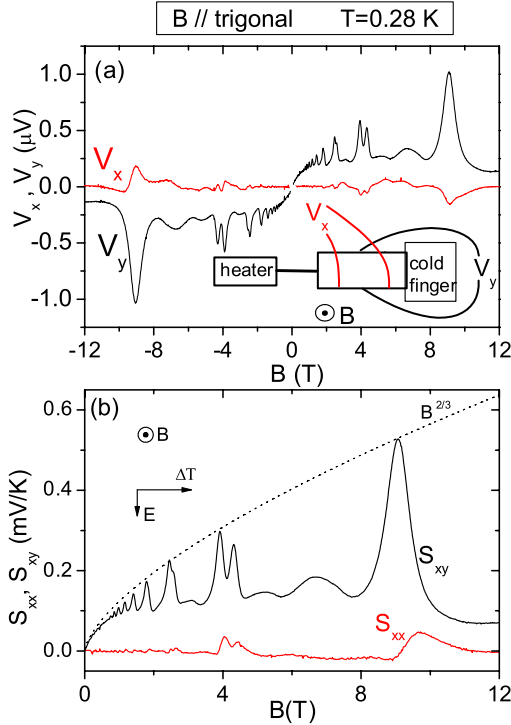


FIG. 1 (color online). (a) Experimental setup and the longitudinal voltage obtained in presence of a constant heat current as a function of magnetic field swept from -12 T to $+12$ T. (b) Nernst and Seebeck coefficients extracted from the odd (even) component of V_y (V_x). The dot line representing a $B^{2/3}$ behavior is just a guide for eye. Arrows representing the three relevant vectors define the sign of the measured Nernst signal.

math, Shubnikov–de Hass oscillations of resistivity are easily observable [11,19]. However, even at 20 mK, only a fraction of $\rho_{xx}(B)$ is oscillating and the total resistivity is dominated by a large nonsaturating magneto-resistance [11]. Figure 2 displays both resistivity and the Nernst response of our sample as a function of the inverse of the magnetic field, B^{-1} . In the case of resistivity, the passage of successive Landau levels is marked by dips in a monotonous background (the scale is semilogarithmic). In the case of the Nernst response, on the other hand, the oscillations dominate a flat background. In other words, the Nernst anomalies reveal two distinct regimes: when the chemical potential is located between two successive Landau levels, S_{xy} is small and is barely field-dependent. When a Landau level is precisely at the chemical potential, S_{xy} peaks to a value which is an increasing function of the magnetic field. The peaks become sharper as one moves towards the lower Landau levels. The inset of Fig. 2 compares S_{xy} and the electric conductivity ($\sigma \approx 1/\rho_{xx}$ since $\rho_{xy} \ll \rho_{xx}$) in the vicinity of the first peak in a semilog plot. Both peak at the same field. However, while σ is enhanced by 20%, the increase in S_{xy} is multifold. Therefore, the Nernst peak is *not* simply reflecting the behavior of a transport coefficient when the effective density of states oscillates in quantizing magnetic fields.

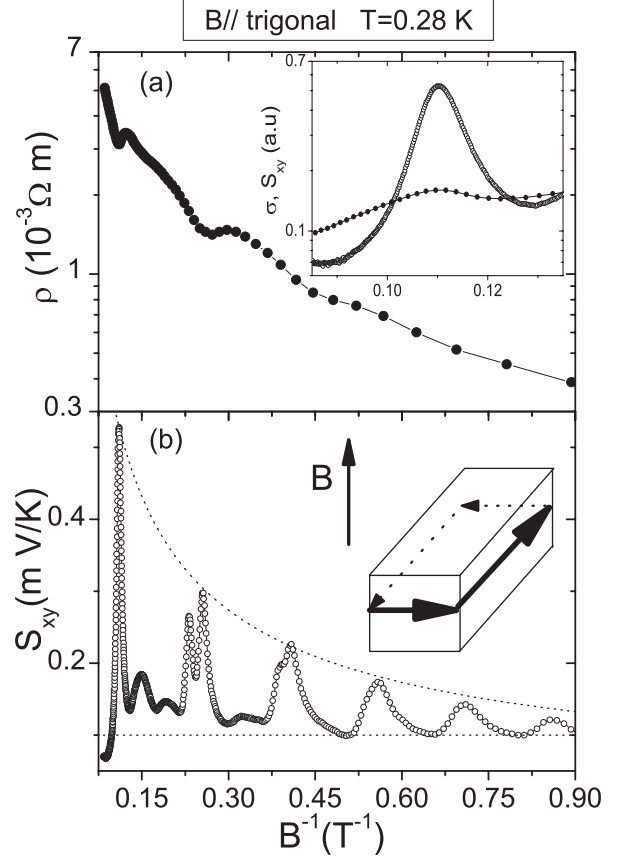


FIG. 2. (a) The resistivity of the sample as a function the inverse of the magnetic field. (b) The Nernst signal as a function of the inverse of the magnetic field. Dotted lines (constant and following $B^{2/3}$) are guide for eyes. The upper inset compares the changes in the electric conductivity and the Nernst signal in the vicinity of the first peak in a semilog scale. The lower inset represents wrapping currents in a bulk sample.

Figure 3 presents the Nernst signal at three different temperatures in a semilog. plot. The decay in temperature is faster than linear and roughly T^2 . We presented and analyzed the temperature dependence of the thermal conductivity, κ , of the same sample in Ref. [18] and concluded that heat is almost entirely transported by ballistic phonons and $\kappa \propto T^3$ from the lowest temperature up to $T \sim 3.8$ K. Thanks to the absolute dominance of the lattice contribution, κ remains insensitive to the magnetic field. The field dependence of the Ettingshausen coefficient at these three temperatures can be obtained using our data and the Bridgman relation [20], which links Ettingshausen (ϵ) and Nernst (S_{xy}) coefficients via thermal conductivity (κ). Namely,

$$S_{xy} = \frac{\kappa}{T} \epsilon. \quad (1)$$

This relation allows us to compute ϵ using S_{xy} and κ . The results are displayed in the main panel of Fig. 3. The Ettingshausen coefficient is virtually temperature-independent between 0.28 K and 1.2 K. More precisely,

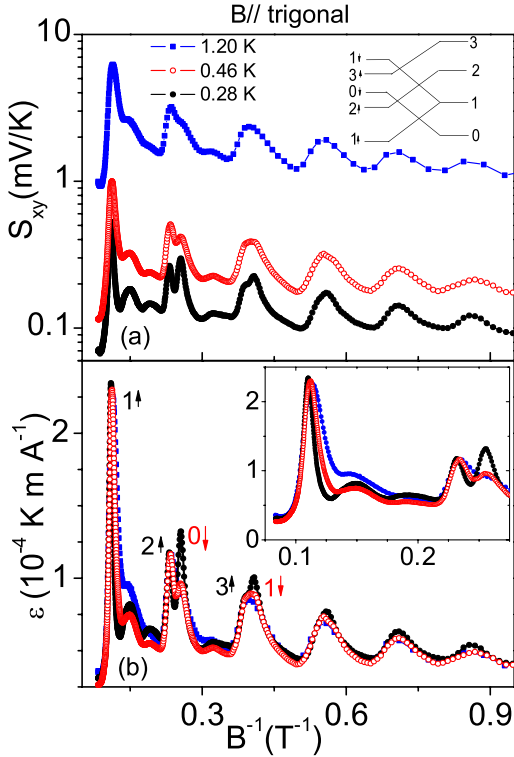


FIG. 3 (color online). (a) Nernst effect as a function of inverse magnetic field along the trigonal for three different temperatures. The inset shows the hierarchy of energy levels. (b) The Ettingshausen coefficient obtained using the same data (see text). The quantum numbers of first peaks are given. The inset is a zoom on the high-field regime.

the temperature dependence of some the peaks saturates below 1.2 K. As discussed below, these peaks are identified as those associated with the Landau level of holes with spins parallel to field. Explaining their magnitude appears as a well-defined challenge to the theory.

In contrast to metals, the thermoelectric response of semiconductors has been extensively studied beyond the quantum limit. In particular, the thermoelectricity of a two-dimensional electron gas (2DEG) in a quantizing magnetic field was studied both theoretically [21–23] and experimentally [24,25]. Early theoretical studies [21,22] highlighted the role played by the edge states [26] and predicted sharp features in the thermoelectric response when a Landau Level attains the Fermi energy. However, in contrast to what is observed here, the main thermoelectric response was expected to be diagonal (S_{xx}). Only in presence of disorder, the off-diagonal component (S_{xy}) would display a feature [21–23]. Experimentally, both S_{xx} [24,25] and S_{xy} [24] were found to show quantum oscillations of comparable magnitude. More recently, Nakamura *et al.* presented a new theoretical study of a ballistic 2DEG in the quantum Hall regime [17]. They concluded that a circulating edge current would realize a nonequilibrium steady state, and the principal thermoelectric response would be off-diagonal. In their picture, when the chemical

potential lies between two Landau levels, the edge currents of ballistic electrons from the hot bath to the cold one (and vice versa) suppress the Nernst voltage. But when the chemical potential is precisely located at a Landau level, a finite Nernst response is restored. Qualitatively, the features observed in our study are compatible with these predictions.

There is ample evidence for current-carrying edge states in two dimensions [27]. In the case of a three-dimensional crystal, Quantum Hall Effect has been observed in a number of bulk systems [28–30] and the associated “wrapping currents” [31] are experimentally plausible. Note, however, that in these cases, the metallic surface states emerge in presence of (and in distinction to) a bulk insulator. In spite of its exceptionally large magneto-resistance, bismuth is believed to remain a metal at high magnetic fields and low temperature [8,13,14]. Moreover, there has been no report on quantized Hall effect in this three-dimensional system. Careful studies, designed to distinguish between the bulk and surface states, are required to shed light on this issue.

An open question is the contribution of phonon drag to the Nernst signal. A scenario invoking phonons cannot be ruled out. Decades ago, Gurevich *et al.* [32] argued that the absorption of acoustical phonons by electrons should sharply peak when a Landau Level is located at the chemical potential. This is experimentally confirmed, and sound attenuation coefficient of bismuth displays giant quantum oscillations [33].

While the origin and the magnitude of the giant oscillations observed here are yet to be understood, their period can be compared to the well-known topology of the Fermi surface in bismuth [1]. It consists of a hole ellipsoid and three electron ellipsoids. The volume of the former is equal to the total volume of the latter. The inset of Fig. 4 shows a projection in the plane normal to the bisectrix (see Ref. [19] for a 3-D representation). The tiny dimensions of these ellipsoids set frequencies of the order of a few teslas for quantum oscillations. The period of the oscillations observed here for a field along the trigonal (0.147 T^{-1}) corresponds to the hole ellipsoid. It is very close to what was reported by Bompadre *et al.* [11] (0.146 T^{-1}). Moreover, it yields an equatorial radius of 0.144 nm^{-1} very close to what is expected (0.146 nm^{-1}) by band calculations. As seen in Fig. 3, at high B/T values, the peaks show an additional structure. The structure can be understood thanks to a careful study by Bompadre *et al.* [11], who concluded that the Zeeman energy is more that twice the cyclotron energy ($E_z = 2.16\hbar\omega_c$ [1,11]). This puts the $|n, \downarrow\rangle$ energy level above $|n+2, \uparrow\rangle$ (see the inset of Fig. 3). Remarkably, the Ettingshausen peaks associated with the minority spins continue to evolve even at 0.28 K.

For a field along trigonal, the electron ellipsoids were not detectable in previous studies of quantum oscillations [11,35]. The lower mobility of electrons compared to holes is attested by their threefold higher Dingle temperature [35]. Moreover, in this configuration, the cross section of

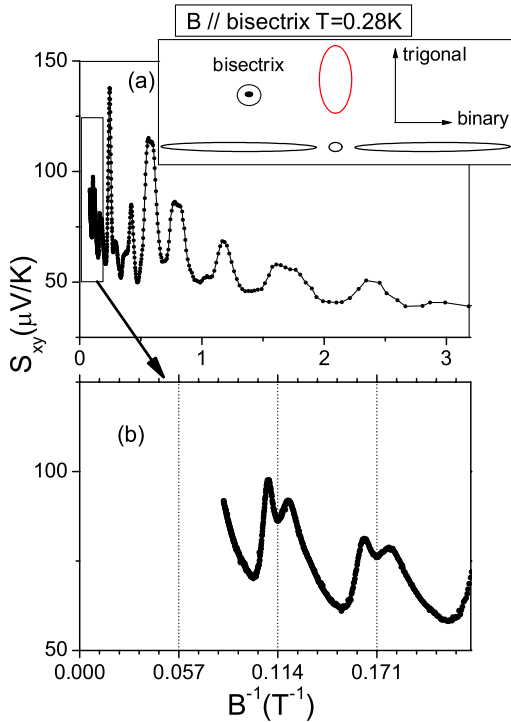


FIG. 4 (color online). (a) Nernst effect as a function of inverse magnetic field along the bisectrix. The cross section of the three elongated electron ellipsoids and the single hole ellipsoid in this geometry are presented in the inset. (b) A zoom on high-field data which isolates peaks corresponding to the hole ellipsoid.

electron and hole ellipsoids become accidentally resonant, making the detection of the electron frequency very difficult.

We also studied the Nernst effect for a field oriented in the plane normal to trigonal and almost along the bisectrix (the misalignment was less than 10 degrees). The data are presented in Fig. 4. For this geometry, the cross section of the hole ellipsoid is larger than the electron ellipsoids, and long-period oscillations of the latter dominate the Nernst response at lower fields below 6 T. Above this field, electrons have been all pushed to their lowest Landau levels, and the residual oscillations are due to holes. As seen in the lower panel of Fig. 4, four peaks corresponding to two successive Landau levels and their Zeeman splitting can be clearly identified. For $B < 6$ T, the oscillations due to the three electron ellipsoids dominate the Nernst response. The longest detected period ($\sim 0.8 \text{ T}^{-1}$) corresponds to a cross section with an average radius of 0.06 nm^{-1} , comparable to the reported values for the two shorter radii of the cigarlike electronic ellipsoid ($0.05\text{--}0.07 \text{ nm}^{-1}$ [1,34]).

Scrutinizing the *phase* of the quantum oscillations would be instructive for probing the Berry phase of electrons. According to Mikitik and Sharlai [36], when the electrons orbit around a line of band contact, the semiclassical quantization condition would be modified. Extending the study to fields larger than 12 T and well in to the

ultraquantum limit could help to address this point in bismuth.

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