Angular Dependent Magnetostriction Measurements of Bismuth Close to the Quantum Limit

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The magnetostriction Δl(B), i.e. the length change of a sample induced by an applied magnetic field, is a thermodynamic quantity that provides important information on ground state properties and is ideally suited to investigate horizontal lines in T-B phase diagrams. By using a newly developed low-temperature capacitance dilatometer designed for high-resolution measurements of the thermal expansion and magnetostriction, we show here that Δl(B) can additionally be used to study the quantum oscillations associated with Landau quantization. An angular dependent study has been carried out on bismuth single crystals showing that the magnetostriction is a unique tool for direct measurements of carrier density close to the quantum limit, where only one Landau level remains occupied. This is on account of the magnetostrictive strain being a linear function of the field-induced changes ΔN of the carrier densities in each band. Each time a Landau level crosses the Fermi energy, the magnetostriction is sharply peaked due to discontinuities in ΔN. All the experimentally resolved peaks and their complex angular dependence are in very good agreement with the one-particle theory.

The outstanding feature of the capacitance method is the sensitivity ($\Delta l/l = 10^{-9} - 10^{-10}$), which is orders of magnitude better than the sensitivity of all other methods (X-ray diffraction: $\Delta l/l = 10^{-5}$, optical interferometry: $\Delta l/l = 10^{-7} - 10^{-8}$)[1]. In the first part of this paper, we describe the design of the new cell and the experimental setup. In the second part we present the first study of angular-resolved magnetostriction measurements in bismuth and map the angular variation of the Landau level crossings close to the quantum limit. The last section compares the measured and the calculated angular dependences of the magnetic fields, at which the electron and hole Landau levels cross the Fermi energy.

The construction of the cell is based on the design of Ref. [2] and is shown in Figure 1. This compact cell has overall dimensions of 33 mm length and 26 mm diameter. Samples from 1 mm to 10 mm length can be measured. All parts except some isolating spacers are machined out of high-purity beryllium-copper that ensures good thermal conductivity while minimizing the "dynamical cell effect", whereby eddy currents in the cell material are induced by the change of the magnetic field. The induced moment interacts with the applied field and produces a torque on the moveable part of the cell resulting in an unwanted displacement of the capacitor plates. The two capacitor plates are electrically isolated and surrounded by guard rings to avoid stray electric fields. While the lower plate is mounted to the frame, the upper plate is fixed to the moveable part, which is held in the frame by two springs. In this parallelogram suspension the upper plate can only move vertically. The sample is fixed by means of an adjustment screw between the outer frame and the moveable part. In this construction, a length change of the sample causes an equivalent displacement of the upper plate with

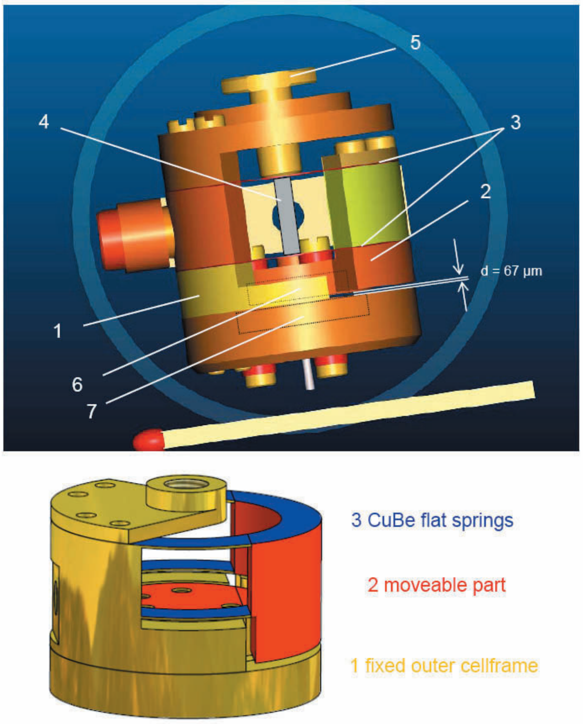

Fig. 1: Drawing of the cell. 1: cell frame, 2: moveable part, 3: CuBe flat springs, 4: sample, 5: adjustment screw, 6: upper capacitor plate, 7: lower capacitor plate.

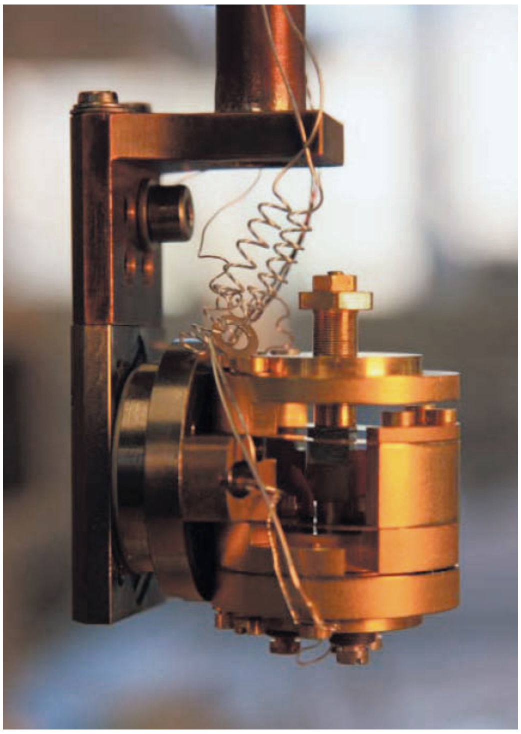

Fig. 2: Photograph of the cell mounted on a rotator.

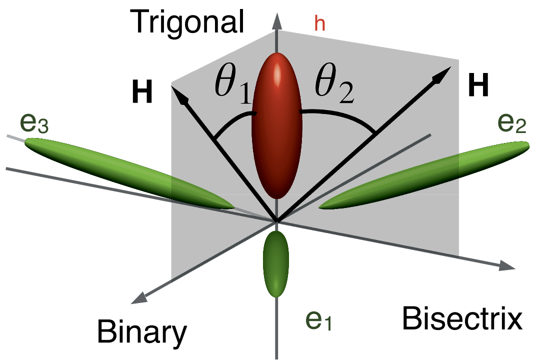

Fig. 3: The Fermi surface of bismuth consists of a hole pocket (red) and three electron pockets (green). *θ1 (θ2)* show angles between the magnetic field and the trigonal axis in the trigonal-binary (trigonal-bisectrix) rotating plane (taken from [12]).

respect to the lower, and therefore a change in capacitance. Before mounting the dilatometer, the capacitor plates were polished within their frames. A uniform surface of the plates within their frames is necessary to achieve best parallel orientation of the plates. In its rest position the capacitance of the dilatometer is 5.4 pF, corresponding to a distance of 0.25 mm between the capacitor plates. After mounting the sample the adjustment screw is used to reduce this distance to 0.067 mm which corresponds to a capacitance of about 20 pF. Utilizing an ultrahigh-resolution Andeen-Hagerling capacitance bridge, the absolute value of the capacitance is measured within the resolution of 10^{-6} pF and accordingly a length change of 0.02 Å can be resolved. The cell has been designed for the use at low temperatures (down to 10 mK) and in magnetic fields as high as 20 T. It has also been miniaturized so that it can be mounted on a rotator and fit inside the inner vacuum chamber (40 mm diameter) of a dilution refrigerator. Figure 2 shows a photograph of the apparatus. The dilatometer can be rotated using a piezoelectric rotator provided by Attocube. The rotator is equipped with a vertical mounting plate to rotate the cell around a horizontal rotation axis. The one-axis rotation setup allows a rotation window of 25 degrees. The rotator is made from non-magnetic materials enabling usage in very strong magnetic fields (current max. tested field: 31 T). The angle between the magnetic field and the sample can be determined using a variable resistor. The relative accuracy of angular determination is about 0.05 degrees.

During the first decades of the 20^{th} century, the bulk semi-metal bismuth has been extensively studied. As early as 1930, studies of bismuth led to the discovery of quantum oscillations in both magnetization (de Haas-van Alphen effect) [3] and resistivity (Shubnikov-de Haas effect) [4]. Bismuth was also the first metal whose Fermi surface was experimentally identified [5]. This remarkable historical role owes much to the peculiar Fermi surface of bismuth. The tiny size of the Fermi surface is itself a direct consequence of the extremly low electron concentration. This tiny Fermi surface in bismuth consists of a hole ellipsoid with its long axis along the trigonal, and three electron ellipsoids slightly tilted out of the equatorial plane (see Fig. 3). The volume of the hole ellipsoid (which equals the total volume of the three electron ellipsoids) is only 10 − 5 of the Brillouin zone [6]. Because of the low carrier density, the cyclotron energy becomes comparable to the Fermi energy, and as a conseqence confines the electrons as well as the holes to their lowest Landau levels in easily accessible magnetic fields. As the magnetic field increases, these orbits one after the other cross the Fermi surface and generate quantum oscillations in various physical properties. The so-called quantum limit is reached when all but the lowest Landau level have already crossed the Fermi level. For a typical metal, several hundred Tesla would be necessary to reach such a limit, which current technology cannot deliver [6]. In contrast, when the magnetic field in bismuth is aligned along the high-symmetry crystalline axis, known as the trigonal, an accessible field of 9 T allows to reach the quantum limit. This means that if the field is applied exactly along the trigonal axis and exceeds 9 T [7], no more crossing of the Fermi energy by any known Landau level is expected. However, recent experimental studies of bismuth [6,8,9] have detected a number of unknown field scales beyond this quantum limit, which cannot be understood within the single particle picture [11].

Two independent groups [10,11] have both calculated that the angle-dependent one-particle spectrum of bismuth is remarkably complex due to the implications of the charge neutrality in a compensated system, the particular Fermi surface topology and the relatively large Zeeman energy. According to these calculations, the field scales of the three electron ellipsoids are expected to present a very sharp angular dependence when the field is slightly tilted off the trigonal axis [10,11] as found by torque magnetometry experiments [8]. On the other hand, no other field scale above 11 T is expected in the one-particle picture, if the field is strictly oriented along the trigonal axis. In this context, the three new resolved anomalies in the field dependence of the Nernst coefficient beyond the quantum limit [6], which are still present in a misalignment of a few degrees [12], cannot be explained [10,11].

In the next section, we report the results of angle-dependent magnetostriction measurements performed on a bismuth single crystal for a field applied in the trigonal-bisectrix plane. Figure 4 presents the oscillatory behaviour of the magnetostriction measured at $T = 0.03$ K and 4.2 K along the high-symmetry crystalline axis for fields up to 10 T. Displacements as small as $2 \cdot 10^{-12}$ m (0.02 Å) have been resolved, owing to the great sensitiv ity of the new dilatometer. As one can see, the oscillations grow in amplitude as the temperature is lowered. This occurs because finite temperatures 'smear out' the edge of the Fermi-Dirac distribution function, which separates filled and empty states [13]. At T = 0.03 K, the Landau tubes pop out of a very sharp Fermi surface. In contrast at T = 4.2 K the surface is 'fuzzy'. As a consequence, the modulation of the density of states around the Fermi

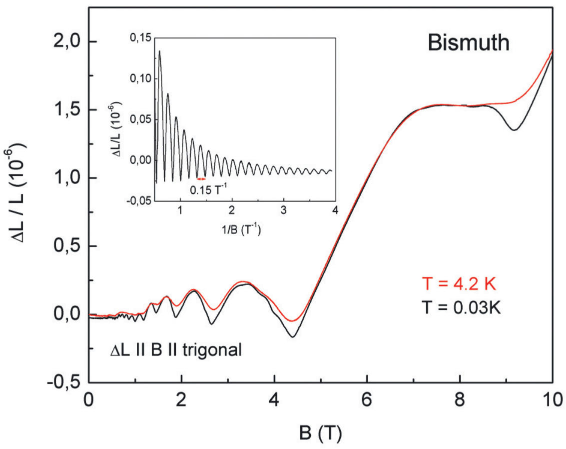

Fig. 4: Quantum oscillations of the magnetostriction for a field along trigonal at T = 0.03 and 4.2 K. Inset: Low-field oscillatory magnetostriction at 0.03 K, up to 2 T.

energy caused by the Landau tubes is less significant at higher temperature. The thermal smearing implies that the cyclotron energy $\hbar\omega_c \sim B$ has to be greater than $\sim k_B T$ for oscillations to be observed, i.e. low temperatures are needed [13]. Therefore all further measurements were performed at the lowest accessible temperature of 30 mK. At low temperature, quantum oscillations become visible in presence of a magnetic field as small as 0.2 T. In the smaller field range ≤ 2 T, more than twenty full periods were identified (see inset of Fig. 4). The de Haas-van Alphen period was found to be 0.15 T^{-1}, in good agreement with the results obtained from other oscillatory phenomena [6,8]. This period corresponds to the maximum cross section of the hole ellipsoid. This group of carriers is well known primarily to be responsible for the oscillatory behavior, when the field is applied along the trigonal axis [6]. As the field is swept and the number of the filled Landau levels decreases, the amplitude of the oscillations increases and dominates by far the non-oscillatory background (see Fig. 4). This occurs because scattering causes the Landau levels to have a finite energy width $\sim \hbar\,\tau^{-1}$. As B increases, $\hbar\omega_c$ will increase, so that the broadened Landau level will become better resolved [13]. The last and most prominent peak at 9 T is associated with the quantum limit of the hole ellipsoid.

Next, we analyze the dependence of the peaks on the tilt angle of the magnetic field with respect to the trigonal direction. These angular dependences enable to distinguish between the peaks that appear even in the one-electron approximation and the

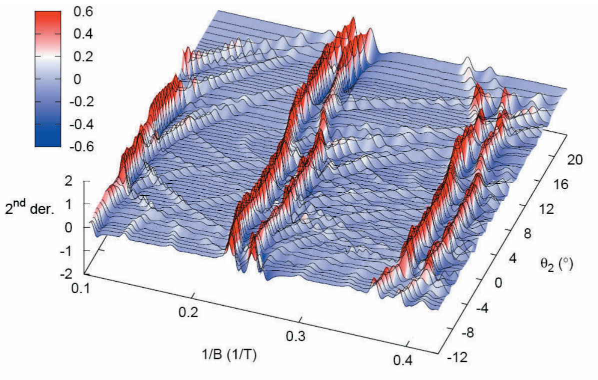

Fig. 5: Quantum oscillations observed in the 2nd derivative of the magnetostriction data, divided by B^2, with a tilted magnetic field (trigonal to bisectrix) up to 10 T at T = 30 mK.

peaks that are actually due to collective effects. To analyze the data, we plotted the magnetostriction data at all measured angles divided by B^2 and calculated the second derivative. By calculating the second derivative, the peaks keep their positions but become more pronounced. One part of our comprehensive study is shown in Figure 5. Here, the field is rotated in the trigonal-bisectrix plane. One can easily associate the detected peaks with the Landau level crossings following previous theoretical studies on bismuth [11]. As one can see, the main peaks scarcely move when the field is tilted in the trigonal-bisectrix plane of the crystal. These peaks with large amplitudes are caused by the hole ellipsoid, and their positions are in good agreement with the theoretical calculation [11]. In contrast, the smaller ripples visible between the large peaks rapidly change their field position with tilt angle. For the first time, these small peaks have been detected by high-resolution magnetostriction measurements and can be clearly identified thanks to their sharp angular variation, caused by the very anisotropic electron ellipsoids. Figure 6 again presents the experimentally detected but also the calculated angular dependences of the magnetic fields, at which the electron and hole Landau levels cross the Fermi energy. It clearly shows that by controlling the orientation of the magnetic field with subdegree accuracy we can confirm the complex theoretical one-particle picture of bismuth in its basic form. We observe the expected two different field scales. The first are more or less horizontal field scales, caused by the hole Landau levels. The second are less pronounced in the data and show a very sharp angular variation. Nevertheless, the observed electron spectrum also appears to be in quantitative agreement with the theoretical calculation. It is already known from other experiments that the positions, especially of the small maxima, vary from experiment to experiment and even from sample to sample [6–8,11]. One should also keep in mind that the theoretically used formulas are not exact [11].

In conclusion, it could be demonstrated by means of our angular-dependent magnetostriction measurements, that the band picture is quite successful in explaining the complex electronic spectrum of bismuth up to 10 T. But it is known from numerous Nernst effect studies that it becomes inadequate as the quantum limit is crossed. Beyond the quantum limit, when all carriers are confined to the first

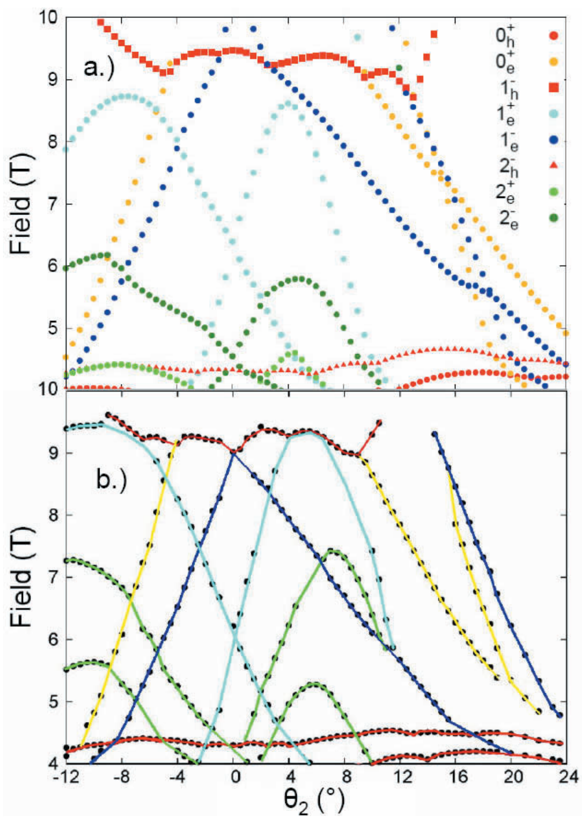

Fig. 6: The calculated (a) and experimentally detected (b) angular dependences of the magnetic fields, $B(\theta_2)$, at which the electron and hole Landau levels cross the Fermi energy; θ_2 is the angle between the magnetic field and the trigonal axis. The magnetic field tilts towards the bisectrix axis. The positions of the hole peaks are shown by red lines, while positions of the electron peaks are marked by other colours. The numbers indicate the Landau level numbers and – or + stands for the direction of the spin projection on the magnetic field.

Landau level, the Nernst effect in bismuth detects field scales which are not expected in the one-particle picture. The open question remains: Are the profiles of these anomalies of bulk origin? We hope to answer this question soon using magnetostriction measurements in magnetic fields up to 20 T for fields tilted in the trigonal-binary plane. If this field scale, manifested in horizontal lines by angular Nernst Effect studies (for instance at about 14 T), is of thermodynamic nature, it should also be detected by means of our high-resolution magnetostriction measurements.

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