Magnetostriction and magnetotransport in bismuth

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It is shown that magnetotransport properties of bismuth cannot be interpreted without taking into account the concomitant diamagnetic magnetostriction which implies a change of carrier density as well as a change of symmetry. Here, we emphasize the lack of consistency of the usual phenomenological theory of magnetotransport effects based on space–time symmetry restrictions, which implicitly assumes the conservation of crystallographic symmetry under magnetic field. This is demonstrated by Hall voltage measurements exhibiting an unexpected asymmetry when the magnetic field lies in the bisectrix direction.

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Transport properties of bismuth, which crystallizes in the $\bar{3}m$ space group, have been intensively studied in the past[1] because of its unique properties which belong to metals as well as to semiconductors; it is a semimetal. At all temperatures it contains a small equal number of electrons N and holes P ($N=P\sim 3\times 10^{17}$ cm^{-3} at 0 K). Under a magnetic field **B**, the commonly accepted structure of the magnetoresistivity tensor $\rho_{ij}(\mathbf{B})$ is determined by the phenomenological transport theory based on the spatial symmetry operations (Neuman principle) and the Onsager relations $\rho_{ij}(\mathbf{B})=\rho_{ji}(-\mathbf{B})$, or briefly "space–time symmetry restrictions."[2] The components $\rho_{ij}(\mathbf{B})$ can be expressed as the sum of an even and odd function of **B**. Figure 1 describes the tensor structure for Bi where indices 1, 2, 3 refer, respectively, to the binary, bisectrix, and trigonal direction. It appears that the off-diagonal components $\rho_{23}(B_1)$ as well as $\rho_{32}(B_1)$ are composed of an even and an odd part. This means that an asymmetry in the Hall field occurs under sign reversal of the magnetic field only when it lies in the binary direction; this is the so-called Umkehr effect.[3] The Umkehr is not specific to $R\bar{3}m$ point group but to all conductive anisotropic crystals. Moreover, it can happen for different field directions for lower symmetry point groups.[2]

From the microscopic point of view of the Boltzmann transport equation, this Umkehr effect, for Bi, is explained by the particular shape of the Fermi surface depicted in Fig. 2, which consists of a set of ellipsoids located at the L and T points of the Brillouin zone: six half electron ellipsoids (L points) and two half hole ellipsoids of revolution around the trigonal axis (T point). One can show that the tilt angle φ of the electron ellipsoids in the plane (2,3) is responsible for the Umkehr effect.[2]

In a previous paper,[4] the asymmetry of the Hall voltage observed on polycrystalline Bi has been explained by the Umkehr effect in grains randomly distributed. Oddly enough, early experimental results never published and recently reviewed by one of the authors, clearly show the existence of an Umkehr effect for the magnetic field in the bisectrix direction. This is, in principle, in contradiction with the theory. In order to clarify this puzzling situation, it was essential to carefully measure the Hall voltage on well orientated single crystals. Moreover, still now, no systematic measurements of the Umkehr effect on single crystals have been performed.

Hall voltages have been measured on two high purity Bi

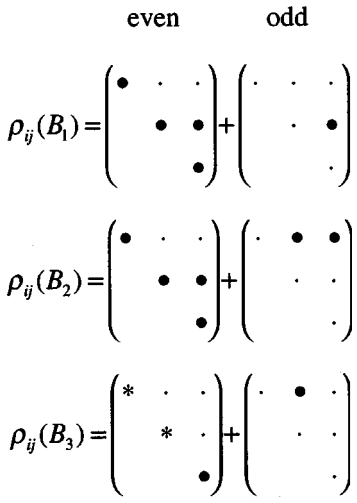

FIG. 1. Space–time symmetry-restricted form of the magnetoresistivity tensor $\rho_{ij}(\mathbf{B})$ for the trigonal class $\bar{3}m$ (from Akgöz and Saunders—Ref. 2). Key to notation: (·) zero component, (●) nonzero component, (*) equal component. The co-existence of even and odd terms in $\rho_{23}(B_1)$ and $\rho_{32}(B_1)$ gives rise to the Umkehr effect (the indices 1, 2, and 3 refer to the binary, bisectrix, and trigonal axis, respectively).

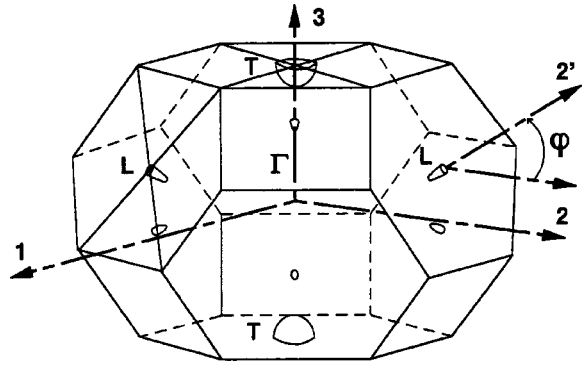

FIG. 2. Fermi surfaces of bismuth in the Brillouin zone from Issi (Ref. 1). Electrons are located at *L* points and holes at *T* points. Axes 1, 2, and 3 are parallel to the binary, bisectrix, and trigonal axes, respectively. φ is the tilt angle of the electron ellipsoids.

$(6N)$ samples coming from an original long (20 mm) parallepipedic single crystal whose length is in the trigonal direction and edges parallel to the binary and bisectrix directions. The parallelism was not ideal, but the deviation never exceeds 5°. Current and potential probes were attached to the crystals with a low melting solder (60 °C).

As Bi is known to have a high thermoelectric figure of merit, special care has been taken to realize electrical measurements under strict isothermal conditions. The use of an alternating current, instead of a direct current, eliminates the problem of an undesirable thermomagnetic contribution to the voltage. An ac current of 40 Hz excitation frequency and 10 mA amplitude was used. The Hall voltages (five-wire configuration) and the diagonal component of the resistivity were measured by an ac transport measurement system option of the PPMS (Quantum Design) over the temperature range of 5–300 K and with magnetic field ranging up to 7 T. The potentiometer was balanced with accuracy at room temperature (where the resistance of the samples is the highest) in such a way that the ratio between the longitudinal voltage and the Hall voltage at null magnetic field was better than 10^2. The experiment was carried out with the cell immersed in helium gas to guarantee thermal stability.

Figure 3 displays the behavior of the Hall voltages for **B** lying in the binary and bisectrix direction at selected temperatures (5, 80, 300 K) versus magnetic fields. Note that in the representation of the graphs the opposite of the Hall voltage was used for positive magnetic fields. While the shape of the three curves looks quite different, it varies continuously versus temperature. An Umkehr effect is well observed for **B** lying in the binary direction [Fig. 3(a)] in accordance with the phenomenological theory but an unexpected Umkehr effect is evidenced when **B** lies in the bisectrix direction [Fig. 3(b)]. In both cases, the Umkehr is more and more pronounced as temperature decreases because the even component of the total Hall voltage becomes dominant in such a way that the sign of the Hall voltage does not change under reversal of **B**. Of course, at low temperatures quantum oscillations are superimposed. It was verified that the diagonal component of the resistivity was an even function of the field whatever the temperature in agreement with the structure of $\rho_{ij}(\mathbf{B})$ reported in Fig. 1.

The main point to emphasize is the occurrence of an Umkehr effect for **B** lying in the bisectrix direction, confirming early unpublished data. This anomalous behavior can be explained, quite simply, by the phenomenon of diamagnetic magnetostriction, which modifies the crystallographic symmetry as well as the distribution of carriers in the Fermi pockets. While this magnetostriction is known for a long time, it is never taken into account in the interpretation of magnetotransport measurements.

Kapitza[5] discovered the magnetostriction of Bi in 1932 for temperatures ranging from 77 to 300 K. Oscillatory magnetostriction, at low temperature, has been observed later,[6–8] proving the essential role of free carriers in this diamagnetic magnetostriction. From thermodynamic arguments, it follows that magnetostriction is directly related to the stress dependence of the magnetic susceptibility and that it is enhanced if many valleys or bands are present, like in Bi or in degenerate semiconductors like Si or Ge.[9] Owing to their different magnetic susceptibilities, some bands or valleys will have higher magnetization energies and a strain is induced, in a magnetic field, in order to shift the valley extrema and redistribute the carriers until the minimization of the free energy is reached. This thermodynamic theory of magnetostriction leads to the conclusion that the induced strain is proportional to the change of carrier density ΔN and ΔP.[10] It is valid in the whole range of fields. The detailed tensorial expression of the magnetostrictive strain can be found in Michenaud *et al.*[8] It is worth emphasizing that the total change of the electron density is shared between the three ellipsoids (which is equivalent to the six half ellipsoids described previously):

$$\Delta N = \sum_{i=1}^{3} \Delta N_i , \tag{1}$$

and the condition $\Delta N = \Delta P$ must be respected. Due to the small carrier density in Bi, $\Delta N/N$ (or $\Delta P/P$) is far from being negligible. Then, the magnetostrictive strain induces a drastic effect on magnetotransport properties.

Owing to the 120° layout of the electron Fermi ellipsoids it is clear that the magnetic field applied in the binary direction (B_1) does not produce the same magnetization as when applied in the bisectrix one (B_2). For B_1, we can distinguish two effective cyclotron masses called ''light binary (LBN)'' and ''heavy binary (HBN)'' and for B_2, ''light bisectrix (LBX)'' and ''heavy bisectrix (HBX)'' following the designation of Vecchi *et al.*[11] as illustrated in Fig. 4. It obviously implies a loss of symmetry of the electron Fermi surface because the magnetic energy is not equivalent in the three ellipsoids. With increasing field, in the range of high fields ($\mu B \gg 1$ where μ is the carrier mobility), the electron density is more and more unequally shared between the Fermi pockets.[8] When the magnetic field is along the trigonal axis, the symmetry is not broken and the change in electron density is equally shared.

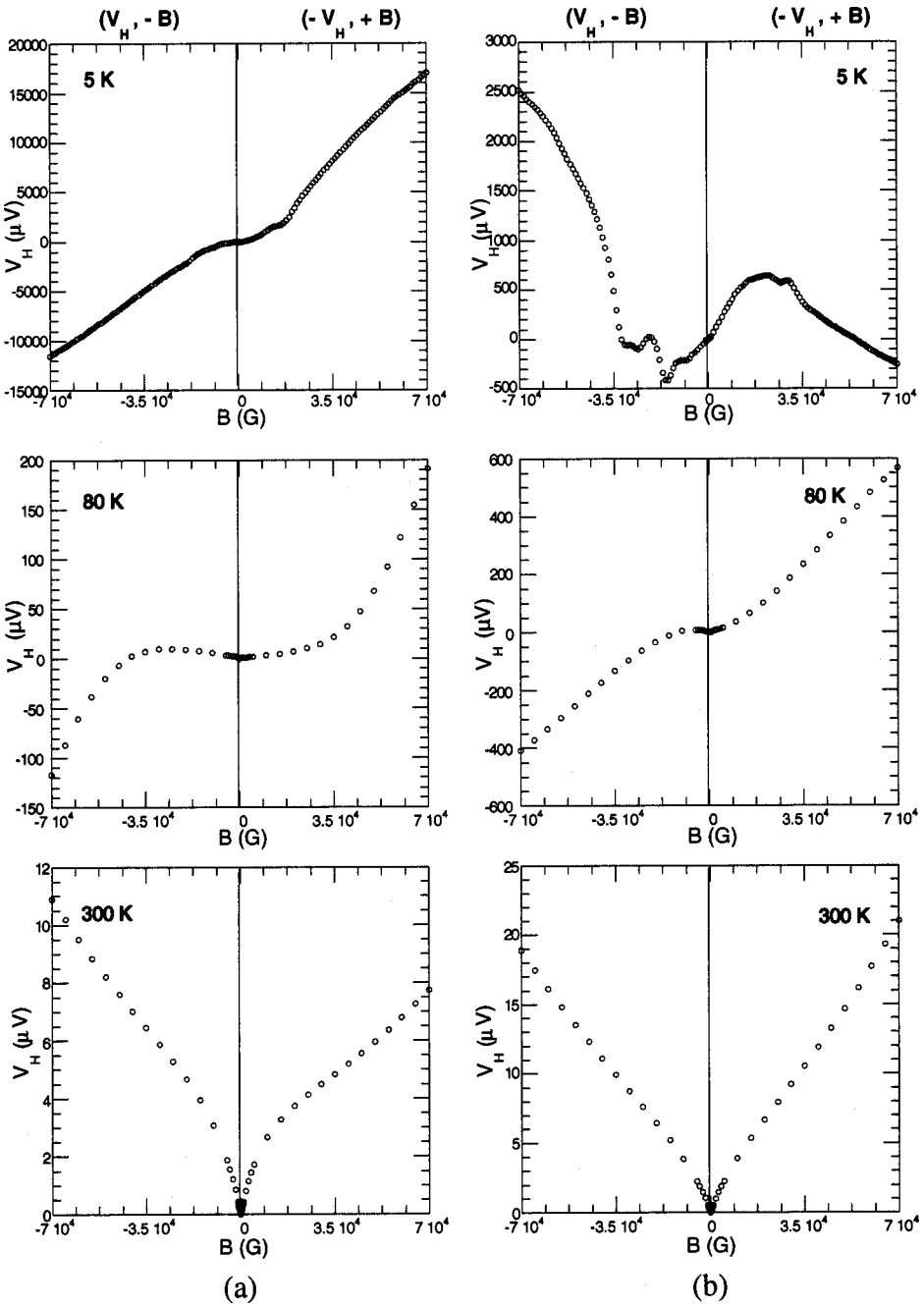

FIG. 3. Hall voltage vs magnetic field lying (a) in the binary direction and (b) in the bisectrix direction at different temperatures. Note that for positive magnetic fields it is the opposite of the Hall voltage which is represented.

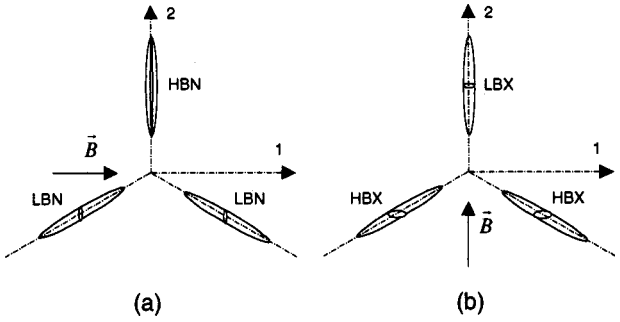

FIG. 4. 120° layout of the electron Fermi surfaces in the trigonal plane showing the different effective cyclotron masses for **B** lying (a) in the binary and (b) in the bisectrix direction. The others symbols are explained in the text.

In fact, this broken symmetry is already implicitly contained in Vecchi *et al.*[11] without reference to magnetostriction. In this paper, the authors show, from magnetoreflection experiments, that the carrier density varies versus field. The similarity between these results and the behavior of the magnetostriction was first pointed out by Michenaud *et al.*[7]

Coming back to magnetotransport properties of Bi, as the symmetry is not conserved, we can assert that the structure of $\rho_{ij}(\mathbf{B})$ (Fig. 1) is not suitable because space–time symmetry restrictions implicitly assume that the symmetry does not change under field. The unexpected Umkehr effect for **B** lying in the bisectrix direction is thus to be attributed to this loss of symmetry.

In summary, the Hall voltage data of Bi single crystals

clearly show an Umkehr effect forbidden by the space–time symmetry restriction theory. It is due to the broken symmetry induced by the field itself while the usual phenomenological theory implies the conservation of the symmetry.

A last comment concerns the measurement of the Hall effect. The usual method of averaging the Hall voltage data for positive and negative **B**, in order to eliminate undesirable effects, is not always suitable and can lead to wrong values of the Hall constants.

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[1] See, for instance, J.-P. Issi, Aust. J. Phys. **32**, 585 (1979), and references therein.
[2] Y. C. Akgöz and G. A. Saunders, J. Phys. C **8**, 1387 (1975).
[3] This term was first introduced by E. Gruneisen and J. Gielessen, Ann. Phys. (Leipzig) **27**, 243 (1936).
[4] B. Lenoir, F. Brochin, and J.-P. Michenaud, Europhys. Lett. **58**, 93 (2002).
[5] P. Kapitza, Proc. R. Soc. London, Ser. A **135**, 568 (1932).
[6] B. S. Chandrasekhar, Phys. Lett. **6**, 27 (1963).
[7] J.-P. Michenaud, J. Heremans, J. Boxus, and C. Haumont, J. Phys. C **14**, L13 (1981).
[8] J.-P. Michenaud, J. Heremans, M. Shayegan, and C. Haumont, Phys. Rev. B **26**, 2552 (1982).
[9] R. W. Keyes, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1967), Vol. 20, p. 37.
[10] The basic structural equation of the magnetostrictive strain e is given by $e=\Xi\Delta N/c$ where Ξ is a deformation potential and c an elastic constant. ΔN depends on **B**.
[11] M. P. Vecchi, J. R. Pereira, and M. S. Dresselhaus, Phys. Rev. B **14**, 298 (1976).