Electrical Magnetochiral Anisotropy

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Electrical conductors can be chiral, i.e., can exist in two forms where one is the other's mirror image. Thus far, no effect of chirality on magnetotransport has been observed. We argue that the electrical resistance of any chiral conductor should depend linearly both on the external magnetic field and the current through the conductor and on its handedness. We suggest two mechanisms to carry this effect and show experimentally on model systems that both are effective.

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Quite recently, a new polarization-independent optical effect was discovered; magnetochiral anisotropy (MCHA) [1–3]. On the basis of symmetry arguments, it was shown that an extra term exists in the dielectric constant of a chiral (from the Greek $\chi\varepsilon\iota\rho$ = hand) medium which is proportional to $\mathbf{k} \cdot \mathbf{B}$, where $\mathbf{k}$ is the wave vector of the light and $\mathbf{B}$ is the external magnetic field [4,5]. Additional features of MCHA are the dependence on the handedness of the chiral medium and the *independence* of the polarization state of the light. The symmetry arguments used for the optical case may also be applied to the case of electrical transport, and the question naturally comes to mind if an analogous effect exists for electronic magnetotransport in chiral conductors. In this Letter we show both theoretically and experimentally that electrical magnetochiral anisotropy (EMCHA) indeed exists, and we identify and demonstrate two microscopic mechanisms that cause such an effect.

An electrical conductor may be chiral because of several reasons. The material may crystallize in a chiral space group, such as tellurium or β-manganese [6], or be composed of chiral subunits such as chiral conducting polymers [7], DNA molecules [8,9], and Langmuir-Blodgett films [10] or vapors [11] of chiral molecules. Even if the material itself is nonchiral, it may still be formed into a chiral shape, similar to a helix. In all these cases, the conductor can exist in two forms, each of which is the mirror image of the other and which we shall call right (D) or left (L) handed. In some chiral conductors, spin-polarized electronic transport has been studied [10,11] and effects similar to natural circular dichroism in optical absorption have been observed. Note that spin polarization is not synonymous with chirality, as spin polarization, as any form of magnetization, is odd under time reversal and even under parity reversal. Only particles having a nonzero drift velocity *and* a longitudinal angular momentum are chiral [12]. As charged particles in a magnetic field acquire angular momentum due to their cyclotron motion, charge carriers moving parallel to the magnetic field fulfill this condition and form a chiral system.

Onsager was the first to consider the symmetry properties of kinetic coefficients [13]. (For a discussion, see, e.g., [14,15].) He showed that, for a generalized transport coefficient σ_{ij} (e.g., the electrical conductivity tensor), close to thermodynamic equilibrium one can write

$$\sigma_{ij} = \int_{-\infty}^{0} \langle y_i(0) y_j(t) \rangle \, dt = \sigma_{ji}^{\dagger}, \tag{1}$$

where $\dagger$ denotes time reversal and the y_i denote microscopic parameters describing the system. If y_i and y_j have the same time-reversal symmetry, one finds $\sigma_{ij}(\mathbf{B}) = \sigma_{ji}(-\mathbf{B})$. This is equivalent to the statement that any two-terminal resistance can have only an even magnetic field dependence [16]. The frequently employed term "linear magnetoresistance" [17] refers in fact always to a magnetic field dependence where R varies linearly with B for large B, but which is still even in B. In chiral systems, symmetry allows all microscopic properties to have, in principle, an odd dependence on the wave vector $\mathbf{k}$ of the moving particles [18]. (The only symmetry allowed $\mathbf{k}$ dependence in nonchiral systems is even.) As the wave vector is also odd under time reversal, Eq. (1) gives $\sigma_{ij}(\mathbf{k}, \mathbf{B}) = \sigma_{ji}(-\mathbf{k}, -\mathbf{B})$. More specifically, we find

$$\sigma_{ij}(\mathbf{k} \cdot \mathbf{B}) = \sigma_{ji}(-\mathbf{k} \cdot -\mathbf{B}) = \sigma_{ji}(\mathbf{k} \cdot \mathbf{B}), \tag{2}$$

and so there are no time-reversal symmetry objections against a linear dependence of σ_{ii}, and therefore of any two-terminal resistance, on $\mathbf{k} \cdot \mathbf{B}$. The claims that a two-terminal resistance should always be even in the applied magnetic field [16] are based on an incomplete view of the basic Onsager relation Eq. (1), neglecting the symmetry properties of the wave vector. As $\langle \mathbf{k} \rangle \propto \mathbf{J}$, the electrical current density, we therefore conjecture that the two-terminal electrical resistance of a chiral conductor subject to a magnetic field $\mathbf{B}$ is of the form

$$R^{D/L}(\langle \mathbf{k} \rangle, \mathbf{B}) = R_o\{1 + \beta B^2 + \chi^{D/L} \mathbf{I} \cdot \mathbf{B}\}, \tag{3}$$

where $\mathbf{I}$ is the electrical current, and parity reversal symmetry requires that $\chi^D = -\chi^L$. Therefore such a $\mathbf{I} \cdot \mathbf{B}$

term can exist only for chiral conductors. The parameter β describes the normal magnetoresistance that is allowed in all conductors (we neglect higher even orders in **B** that are also allowed). We call the effect corresponding to the linear **B** dependence in Eq. (3) electrical magnetochiral anisotropy, in direct analogy to the optical case. The existence of this effect is the direct consequence of the simultaneous breaking of time-reversal symmetry by a magnetic field and of parity by chirality and is therefore fundamental and universal. The magnetic field dependence of EMCHA is strictly linear, including the sign of **B**. Below we will discuss two microscopic mechanisms that may determine the magnitude of MCHA, namely, chiral scattering and the magnetic self-field.

As explained above, a longitudinal external magnetic field will make moving charge carriers chiral. In a chiral medium, scatterers such as crystal defects, phonons, or other charge carriers will, in general, be chiral. The scattering probabilities of the chiral charge carriers will be dependent on the relative handedness of these carriers and the scatterers. This will combine to a magnetically induced change in the carrier scattering rate in a chiral conductor, and therefore lead to a change of its electrical resistance. Such a mechanism was considered in Refs. [19,20] for the scattering of free electrons by chiral molecules in a magnetic field. Indeed a difference in scattering rate proportional to $\mathbf{k} \cdot \mathbf{B}$ between D and L molecules was calculated. Below, we will show that, in a solid state conductor into which chiral scattering centers are induced, EMCHA can be observed experimentally.

The second microscopic mechanism is based on the magnetic self-field. In general, a current carrying chiral conductor will possess a magnetization, the sign of which depends on the direction of the current and the handedness of the conductor. An analogous magnetization has been predicted for the propagation of unpolarized light in chiral media [21]. Consider a nonchiral material, the resistivity of which is given by $\rho(B) = \rho_0\{1 + \beta B^2\}$. A D or L helix made of this material, carrying a current I, will generate an axial magnetic field at the position of the charge carriers $B_a = \alpha^{D/L} I$, where $\alpha^{D/L}$ depends on the geometry of the helix, and $\alpha^D = -\alpha^L$. If an external field B_{ext} is applied parallel to the helix axis, the charge carriers in the conductor feel $B_{\mathrm{ext}} + B_a$. It is easily seen that the resistance of such a helix is given by

$$R^{D/L}(I, B_{\mathrm{ext}}) = R_o\{1 + \beta B_{\mathrm{ext}}^2 + 2\alpha^{D/L}\beta I B_{\mathrm{ext}} + O(I^2)\}. \quad (4)$$

So, although the resistance is an even function of the total magnetic field, for a chiral conductor, due to the self-field, a term linear in the external magnetic field and the current exists. This term describes EMCHA. Equation (4), although certainly an oversimplification for a microscopic chiral conductor, illustrates that, for this self-field mechanism to be strong, conductors with a large quadratic magnetoresistance are favorable. Promising microscopic chiral conductors to investigate would be chiral nanotubes, for which a large magnetoresistance was observed [22] and a strong current induced axial magnetic field was calculated [23]. Unfortunately, thus far no control over the chirality of nanotubes has been obtained and an experiment to test Eq. (4) on such conductors has to await further progress in this area. However, a metallic helix can be regarded as a simplified macroscopic model of a chiral molecule with given chirality. This has been demonstrated for the optical properties by Tinoco [24]. We have used such helices to study experimentally the self-field effect as a source for EMCHA. 99.999% bismuth was used as the wire material, as it has high β values, in particular at low temperature [25]. We have fabricated helices by injecting molten bismuth into helical molds, followed by annealing. The axial magnetic self-field, averaged over the cross section of the conducting part of such a helix, can be easily calculated by means of finite element methods [26]. Typical values of α for our helices are 0.5 mT/A. We perform two-terminal resistance measurements because four-terminal measurements may be affected by off-diagonal, Hall-like resistivity tensor components, induced by geometrical imperfections, that are also odd in the magnetic field and the current. As the contact resistances are found to be very low, the two-terminal method measures the truly dissipative resistivity of the helices. The two-terminal magnetochiral anisotropy of these helices was experimentally determined as $\Delta R(I, B_{\mathrm{ext}}) \equiv R(I, B_{\mathrm{ext}}) - R(-I, B_{\mathrm{ext}})$ by means of standard phase-sensitive detection techniques. Figure 1 shows this magnetochiral anisotropy ΔR for a D and a L helix of the same dimensions, as a function of magnetic field, at 300 and 77 K. Clearly a linear magnetoresistance is found, and of opposite slopes for the opposite handedness. To our knowledge, this is the first time that a truly linear two-terminal magnetoresistance has been observed. Also shown are the theoretical predictions based on Eq. (4) and the calculated α and measured β values. Good agreement in sign and magnitude is obtained with the experimental results. At higher fields, the observed field dependence becomes nonlinear. This is due to irreversible plastic deformations of the helices by the Lorentz force, which at even higher fields resulted in breaking of the helices. Figure 2 shows the angular dependence and the current dependence of the EMCHA, which are also in good agreement with Eq. (4). Our experimental findings on macroscopic chiral conductors therefore quantitatively verify the self-field mechanism as a source for EMCHA in electronic transport. The self-field effect will be operative at all length scales and will therefore induce EMCHA in all chiral conductors that show quadratic magnetoresistance.

We have experimentally studied the chiral scattering mechanism as a source of EMCHA by measuring the two-terminal resistance of straight bismuth wires containing screw dislocations. (Two-terminal resistance was measured for the same reason as for the case of the helices.)

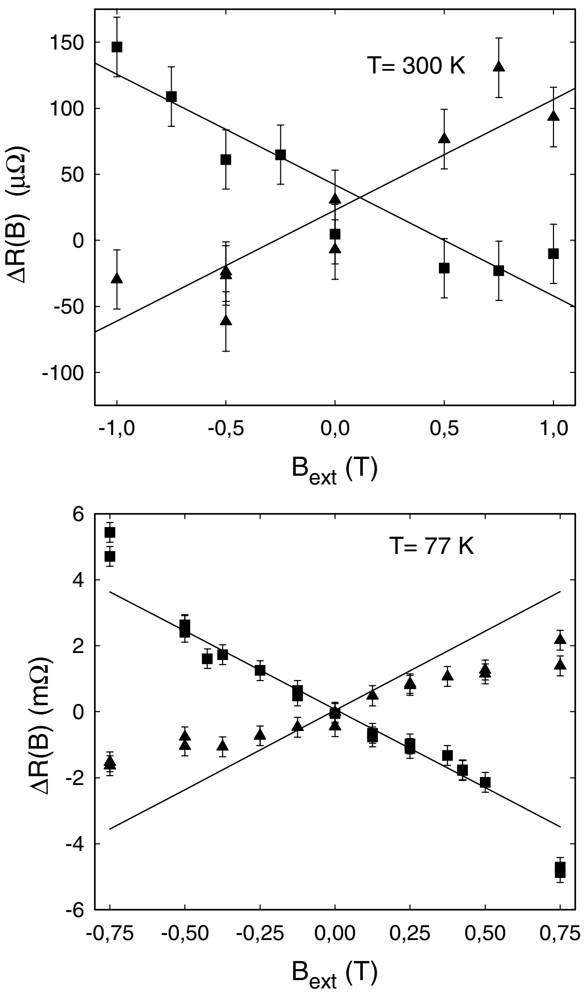

FIG. 1. Two-terminal magnetochiral resistance anisotropy $\Delta R(I, B_{\mathrm{ext}}) \equiv R(I, B_{\mathrm{ext}}) - R(-I, B_{\mathrm{ext}})$ of D (squares) and L (triangles) bismuth helices (seven turns, 8 mm diameter, and 0.8 mm pitch) with $I = 0.2$ A, as a function of the external magnetic field B_{ext}, at 300 K (top, $R_o = 0.9\ \Omega$) and 77 K (bottom, $R_o = 0.2\ \Omega$). The solid lines are the predictions based on Eq. (4) where an arbitrary vertical offset was incorporated to account for small asymmetries in contact resistances and electronics. We call a helix "D" if it has the same handedness as a normal screw.

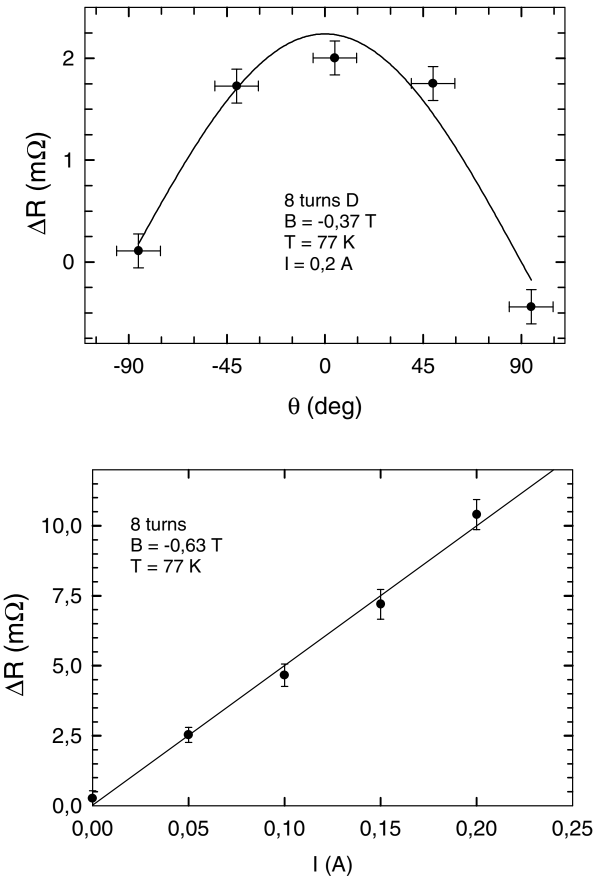

FIG. 2. Top panel: Angular dependence of the magnetochiral resistance anisotropy of the D bismuth helix from Fig. 1. θ is the angle between external magnetic field and helix axis. Solid line is a fit to a cosine dependence. Bottom panel: Current dependence of the magnetochiral resistance anisotropy of the D helix at $\theta = 0$.

The wires were prepared in similar ways as the helices and subsequently subjected to a torsional deformation. Typical lengths were 1 cm and typical distortions were effected by rotating one wire end around the wire axis over 15°, which resulted in an increase of the 300 K resistance by approximately 5%. Such a treatment introduces screw dislocations into the wire, predominantly of one handedness. These dislocations will act as chiral scattering centers [27]. Figure 3 shows a typical result for the magnetochiral anisotropy of wires subjected to a L and to a D distortion. Clear EMCHA is observed, of opposite sign for the opposite handedness of torsion. Also shown is that the same L wire no longer shows EMCHA after it has been annealed for 24 h at 265 °C, close to its melting point. After this treatment, the zero-field resistance has returned to its value before distortion to within 1%, which proves that most of the screw dislocations have disappeared. Consequently, the EMCHA must vanish, in agreement with our observation. For other wires investigated, the magnitude of the observed effect differed from that in Fig. 3 by up to an order of magnitude. This is to be expected as the number of screw dislocations introduced by our torsional deformation is unknown and may vary strongly between samples. Furthermore, the magnetoscattering of electrons by screw dislocations inside solids has never been studied previously. This makes a quantitative evaluation of the effect impossible at this stage. However, the sign of the magnetoresistance always corresponded to the handedness of the distortion. This proves that scattering of charge carriers by chiral objects in a magnetic field causes EMCHA, in agreement with the predictions for the molecular case by Pospelov [19] and Mussigman *et al.* [20].

Thus far we have considered only diffusive conductors. For ballistic conductors, one can easily show by direct application of time- and parity-reversal symmetry arguments that the carrier transmission probability, and therefore the electrical resistance, may also show EMCHA.

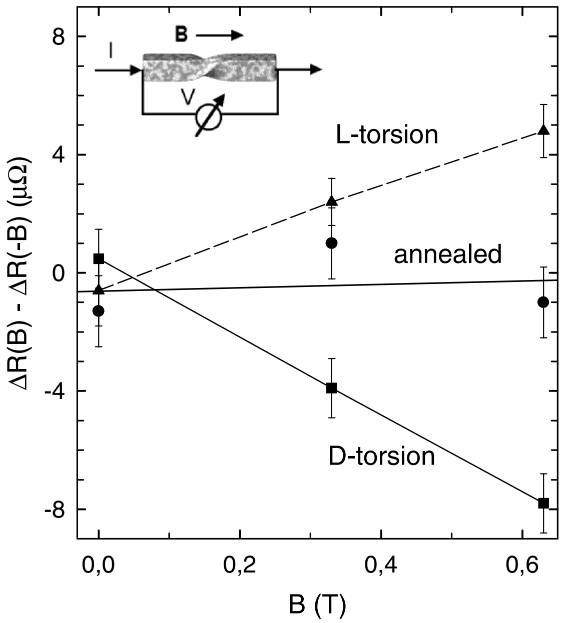

FIG. 3. Two-terminal magnetochiral resistance anisotropy difference $\Delta R(I, B_{\text{ext}}) - \Delta R(I, -B_{\text{ext}})$ of D and L distorted bismuth wires with a length of 10 mm, a diameter of 0.5 mm, and $I = 0.2$ A, at 77 K Also shown is the behavior of the L wire after annealing. Typical zero-field resistance of the wires is 20 mΩ. The inset shows the geometry of our experiment.

Equation (3) will therefore also apply to ballistic conductors. Quantum transport calculations using the simplest possible model, namely, a free electron on a helix in a magnetic field, show that for both ballistic and diffusive conductors EMCHA occurs in their two-terminal resistance [28].

Our experimental results confirm the validity of our conjecture of Eq. (3) and prove the existence of electrical magnetochiral anisotropy in chiral conductors. This effect is fundamental and universal, and should manifest itself in all chiral conductors, ranging from molecules to macroscopic objects. We have identified and demonstrated two mechanisms leading to EMCHA, but these may not be the only ones. Although the effects observed thus far are quite small, they may, in principle, be interesting for spintronics, as they imply that, in chiral conductors, electrical resistance depends not only on the magnitude of spin polarization, but also on its direction. Because of the universal nature of Eq. (2), one may also expect MCHA in other transport phenomena involving the movement of charge in chiral media, such as ion diffusion or heat conduction. In particular, in analogy with the recently reported enantioselective magnetochiral photochemistry [29], one may expect that MCHA in electrochemistry in a magnetic field can lead to enantioselectivity.

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