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First-order longitudinal Hall effect

M. Möllendorf and W. Bauhofer
Max-Planck-Institut für Festkörperforschung,
D-7000 Stuttgart 80, Federal Republic of Germany
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We report the observation of first-order Hall effects determined by independent tensor elements and measured in an unconventional longitudinal geometry: magnetic field parallel to either the current or the Hall field. Monoclinic semimetallic $SrAs_3$ is shown to be a well-suited system for the investigation of this effect.

The Hall effect is a frequently used tool for the investigation of electrical transport properties of solids.[1] The origin of the Hall effect is the Lorentz force deflecting charged particles moving in a magnetic field. The Lorentz force acts perpendicular to the velocity of the charge carriers and to the applied magnetic field. Thus, in the usual Hall-geometry electrical current, magnetic field and Hall electrical field are mutually perpendicular. This Rapid Communication reports on the observation of a Hall effect in a longitudinal geometry with the magnetic field parallel to either the current or the Hall electric field.

Generally, the Hall effect is described by the odd terms of a power-series expansion of the resistivity $\underline{\rho}(\vec{B})$ connecting the electric field $\vec{E}$ with the electrical current density $\vec{j}$:

$$\vec{E}=\underline{\rho}(\vec{B})\cdot\vec{j}\ , \tag{1}$$

$$\rho_{ij}(\vec{B})=\rho_{ij}+\rho_{ijk}B_k+\rho_{ijkl}B_kB_l+\cdots\ . \tag{2}$$

In the following we identify only the linear terms ρ_{ijk} as the Hall effect, neglecting higher-order contributions.

The Onsager relations[2] as well as the crystal symmetry restrict the number of independent components of the Hall tensor. The Onsager relations force the Hall field $\vec{E}_H$ to be always perpendicular to the current $\vec{j}$. First-order longitudinal Hall effects (LHE's) with $\vec{E}_H \parallel \vec{B}$ or $\vec{B} \parallel \vec{j}$ are not allowed in cubic systems by symmetry arguments. However, with $\vec{E}_H \parallel \vec{B}$, terms of third order in $\vec{B}$ have been observed by Grabner[3] in n-type germanium. Longitudinal components of $\vec{E}_H$ are generally allowed in first order for all symmetries lower than cubic if current and magnetic field are *not* applied along the crystallographic axes. This is merely due to an inappropriate choice of the laboratory frame and does not represent a new phenomenon, since those longitudinal components can be expressed as linear combinations of the transverse components. In contrast, monoclinic symmetries entail both aforementioned LHE's described[4] by *independent* tensor elements ρ_{131} and ρ_{232}:

$$\underline{\rho}(\vec{B})=\begin{pmatrix}\rho_{11} & \rho_{12} & 0\\ \rho_{12} & \rho_{22} & 0\\ 0 & 0 & \rho_{33}\end{pmatrix}+\begin{pmatrix}0 & 0 & \rho_{131} & 0 & 0 & \rho_{132} & 0 & \rho_{123} & 0\\ 0 & 0 & \rho_{231} & 0 & 0 & \rho_{232} & -\rho_{123} & 0 & 0\\ -\rho_{131} & -\rho_{231} & 0 & -\rho_{132} & -\rho_{232} & 0 & 0 & 0 & 0\end{pmatrix}\vec{B}+\cdots\ . \tag{3}$$

The orthogonal reference frame 1, 2, 3 is adjusted to the crystallographic directions c, a, and b with 2 parallel to a and 3 parallel to b. In this notation, b is the twofold axis and the monoclinic angle β is between a and c. As far as we know our observation of the LHE in $SrAs_3$ is the first experimental verification of this effect.

Strontium triarsenide belongs to the large family of triarsenides and triphosphides of divalent metals.[5] Most of these compounds crystallize in the monoclinic system (space group $C2/m$ in the case of $SrAs_3$). The main feature of the triarsenide crystal structures are two-dimensional arsenic layers which can be derived from the black phosphorus structure. The layers are parallel to the (ab) plane which is a plane of easy cleavage. The two s electrons of the metal atoms are transferred to the anion substructure by breaking two arsenic–arsenic bonds per formula unit with respect to black phosphorus. In analogy to α-As, $SrAs_3$ is a semimetal due to an overlap of As nonbonding and antibonding electron states. The combination of several favorable properties makes the Hall effect in $SrAs_3$ easily observable: high conductivity ($\sim 5\times10^2\ \Omega^{-1}\mathrm{cm}^{-1}$) with relatively low carrier concentration ($\sim 6\times10^{17}\ \mathrm{cm}^{-3}$ at 4 K) caused by a high mobility ($\sim 5\times10^3\ \mathrm{cm^2/Vs}$ at 4 K). Large single crystals can be grown by the Bridgman technique. The monoclinic angle β is 112°, markedly different from 90°, proving the pronounced monoclinic structure of $SrAs_3$. Evidence for monoclinic electronic properties is given by the nonellipsoidal Fermi surface which has recently been determined from Shubnikov–de Haas oscillations in the magnetoresistivity.[6]

The electrical measurements are performed with a computerized Hall apparatus typically using a current at 1 mA, a magnetic field of 0.8 T and reversing current and magnetic field for all measurements. The five-probe technique is used with samples cut into bars of $1\times1\times8\text{-mm}^3$ size (voltage probes within the central third).

Figure 1 shows the temperature dependence of resistivity ρ_{22} (a direction) and transverse Hall coefficient ρ_{321} ($\vec{I} \parallel a$, $\vec{E}_H \parallel b$) of $SrAs_3$. The resistivity is rather temperature independent whereas the Hall coefficient changes its sign around 80 K leading to p-type conductivity at low temperatures.

Figure 2 summarizes our observations of the LHE. The two geometrical arrangements (I) and (II) shown in the upper part of Fig. 2 ($\vec{B} \parallel \vec{E}_H$ and $\vec{B} \parallel \vec{J}$) specify the nonvanishing Hall components ρ_{232} and $\rho_{322}(=-\rho_{232})$, respectively. We have deliberately chosen two different geometries allowing for the measurement of the two LHE components

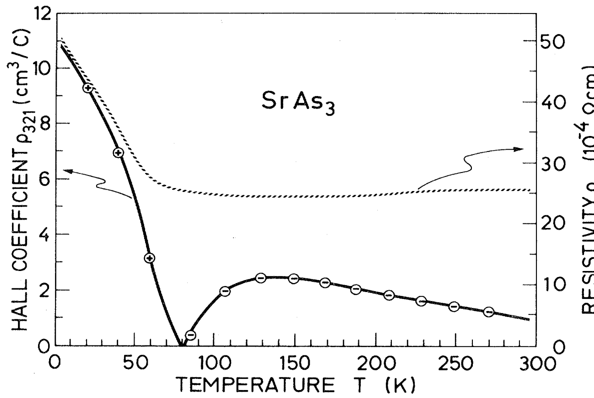

FIG. 1. Resistivity ρ_{22} and transverse Hall coefficient ρ_{321} of $SrAs_3$ as a function of temperature T. The Hall coefficient ρ_{321} changes its sign (symbolized by + and −) around 80 K.

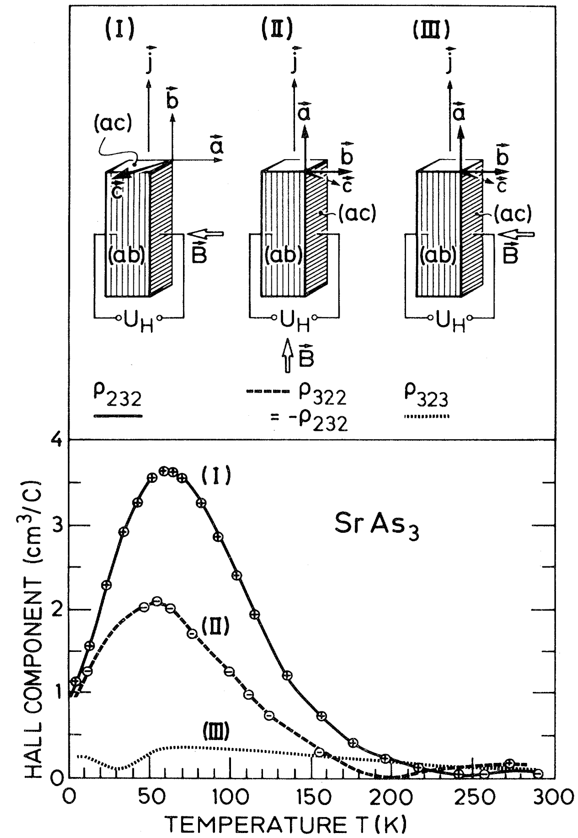

FIG. 2. Upper half: three different geometrical arrangements for observing a longitudinal Hall effect. Crystallographic axes are indicated by arrows, crystallographic planes by letters within brackets. The corresponding Hall components are given below each figure. Lower half: temperature dependence of the Hall components measured in the geometrical arrangements shown above. Note the large ratio ρ_{232}/ρ_{323} of allowed to forbidden components around 70 K. Cases I and II give opposite signs (symbolized by + and −) of the Hall component.

which, according to Onsager's relation, must be equal in magnitude and opposite in sign. Geometry (III) defines the vanishing component ρ_{323}. The corresponding measurements are depicted in the lower half of Fig. 2. The LHE for cases (I) and (II) is unambiguously observed in a limited temperature interval around 70 K. The absolute value of ρ_{232} differ by a factor ~ 2 for the geometries (I) and (II) but both values are much larger [one order of magnitude for case (I)] than the value of ρ_{323} for the forbidden geometry (III). We ascribe the nonzero value of (III) to misalignment effects.

We want to point out that the LHE maximum falls into a temperature region where the transverse Hall component (THE) changes its sign. The ratio of maximal LHE (around 70 K) and THE (at 5 K) amounts to 0.35 which is one order of magnitude larger than Grabner's third-order effect at 0.8 T. We did not yet determine the other independent LHE component ($\rho_{131} = -\rho_{311}$) because of difficulties in preparing samples perpendicular to the easy cleavage *ab* plane.

The possibility of a nonvanishing LHE component in monoclinic crystals is obvious from symmetry arguments; however, an explanation of the LHE within a microscopic picture seems to be quite difficult. Therefore, we want to give some elementary arguments based on the existence of a nonvanishing off-diagonal resistivity component to elucidate the occurrence of a LHE and to account for the differences between geometries (I) and (II).

In case (II) (Fig. 2) a current component perpendicular to $\vec{B}$ is created due to the nonvanishing σ_{12} term. This component vanishes for infinitely long samples due to a transverse electric field arising even without a magnetic field.[7] On the other hand, for short samples the Hall field becomes increasingly shorted by the current electrodes. Thus, for geometry (II) we can never expect to measure the full Hall component which explains the factor of 2 difference of the observed values. Case (I) can be explained in a complementary way. The Lorentz force deflects the charge carriers within the (*ac*) plane, and leads to an electrical field component in the *a* direction due to the nonvanishing ρ_{12} ($\rho_{12} \neq 0$ is equivalent to $\sigma_{12} \neq 0$). In geometry (III) the charge carriers are again deflected by the Lorentz force within the (*ac*) plane but there is no nondiagonal conductivity component carrying the index 3 which could cause an electric field in the *b* direction.

Our model cannot explain the symmetry-allowed LHE components in the eight point groups with diagonal resistivity terms only. This raises the question if the existence of an off-diagonal resistivity term is a necessary condition for the occurrence of a LHE, since symmetry alone cannot guarantee the existence of a physical effect.

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