

Two-band character of the first-order longitudinal Hall effect

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(Received 8 January 1988; revised manuscript received 12 April 1988)

Measurements on CaAs_3 , EuAs_3 , and AuTe_2 give strong evidence for the assertion that the first-order longitudinal Hall effect (LHE), recently discovered in monoclinic SrAs_3 , is essentially a two-band effect. The observation of significant LHE coefficients in SrAs_3 , CaAs_3 , and AuTe_2 is restricted to limited temperature ranges representing two-band regimes: $T < 150$ K for SrAs_3 , 150 K $< T < 250$ K for CaAs_3 , and $T > 160$ K for AuTe_2 . No LHE could be detected for the predominantly one-band semimetallic compound EuAs_3 .

I. INTRODUCTION

An unusual first-order longitudinal Hall effect (LHE) has recently been observed^{1,2} for the monoclinic, semimetallic compound SrAs_3 . The geometrical arrangement for a LHE is characterized by a magnetic field parallel either to the current or the Hall field. Rotation of the sample with respect to the magnetic field is necessary for an unambiguous determination of the LHE coefficients. The temperature dependence of the LHE coefficients in SrAs_3 suggests that in addition to the requirements on crystal symmetry a significant LHE is restricted to regimes with two-band behavior. This observation is in agreement with the results of a simple model calculation of the LHE coefficients.²

Further strong evidence is here presented for the two-band character of the LHE based on the investigation of the low-symmetry compounds AuTe_2 , CaAs_3 , and EuAs_3 . Nonvanishing LHE coefficients have been found in the two-band regimes of semimetallic AuTe_2 and semiconducting CaAs_3 while no LHE could be detected in the predominantly one-band semimetal EuAs_3 . Our results for AuTe_2 differ from earlier measurements by Sagar *et al.*³

The LHE is defined by Hall coefficients ρ_{ijk} with two equal indices. The separation of transverse (THE) and longitudinal Hall coefficients is achieved by the following method:² The same sample has to be measured in two different rotation schemes, one giving the sum THE + LHE, whereas the other yields a pure THE. The existence of a LHE is then discernible from the following three observations.

(i) THE + LHE curves have larger amplitudes than the corresponding pure THE curves.

(ii) The angle of the zero crossing of the Hall voltage is

shifted from the position for $\mathbf{E}_H \parallel \mathbf{B}$ (or $\mathbf{I} \parallel \mathbf{B}$).

(iii) The angle of the zero crossing shifts with temperature if the ratio of longitudinal and transverse coefficients is temperature dependent; this is generally the case for mutually independent coefficients. A temperature-dependent shift of the zero crossing is a clear indication for a LHE even for a single rotation scheme.

The angular dependence is more sensitive to the existence of a LHE than the amplitude: A ratio $\rho_{\text{LHE}}/\rho_{\text{THE}} = 0.3$ leads to an angular shift of 17° , while the amplitude increases by merely 4.4% with respect to a pure THE. All three above-mentioned features were clearly identified² in SrAs_3 .

II. THEORETICAL ASPECTS

Within the framework of the Boltzmann equation the Hall tensor elements are calculated by solving the appropriate transport integrals. This requires precise knowledge of the Fermi surface, which is not available for the materials under question. However, a formula for a nonvanishing LHE coefficient has been deduced² within a simple two-band model based on the assumption of an energy independent relaxation time τ and the existence of nondiagonal conductivity tensor elements which are symmetry allowed⁴ for the monoclinic point groups $C2/m$ [SrAs_3 , EuAs_3 , and CaAs_3 (Ref. 5)] and C_s [AuTe_2 (Ref. 6)]. Denoting the diagonal conductivity and mobility tensor elements $\sigma_{jj}^{(i)}$ by $\sigma_j^{(i)}$ and $\mu_{jj}^{(i)}$ by $\mu_j^{(i)}$ with $\sigma^{(i)} = n^{(i)} \cdot q \cdot \mu^{(i)}$, we obtained for the longitudinal Hall coefficient ρ_{232}^0 , defined by

$$E_i = \rho_{ij}(B) I_j \tag{1}$$

and

$$\rho_{ij}(B) = \rho_{ij}^0 + \rho_{ijk}^0 \cdot B_k + \rho_{ijkl}^0 \cdot B_k B_l + \dots, \tag{2}$$

$$\rho_{232}^0 = \frac{(\mu_3^{(1)} - \mu_3^{(2)})(\sigma_1^{(2)} \sigma_{12}^{(1)} - \sigma_1^{(1)} \sigma_{12}^{(2)})}{(\sigma_3^{(1)} + \sigma_3^{(2)})[(\sigma_1^{(1)} + \sigma_1^{(2)})(\sigma_2^{(1)} + \sigma_2^{(2)}) - (\sigma_{12}^{(1)} + \sigma_{12}^{(2)})^2]} \tag{3}$$

The indices refer to the crystallographic axes a, b, c in the following way: $1 \parallel (a, b)$, $2 \parallel a$, $3 \parallel b$; b is the twofold axis of $C2/m$ and perpendicular to the mirror plane of C_s , a and c include the monoclinic angle β in the $(1,2)$ plane.

Equation (3) can be simplified by assuming (1) a monoclinic hold band, $\sigma_{ij}^{(1)} = \sigma_{ij}^{(h)}$; (2) an ellipsoidal electron band, parallel to the crystallographic axes, $\sigma_{ij}^{(2)} = 0$, $\sigma_1^{(e)} \approx \sigma_2^{(e)}$; and (3) a higher mobility for electrons, $\mu_i^{(e)} \gg \mu_j^{(h)}$. These assumptions are justified by the electrical properties of SrAs_3 .² For ρ_{232}^0 this leads to

$$\rho_{232}^0 = \frac{-ne |\mu_3^{(e)} \cdot \mu_1^{(e)}|^2 \sigma_{12}^{(h)}}{(\sigma_1^{(h)} + \sigma_1^{(e)})[(\sigma_2^{(h)} + \sigma_1^{(e)})(\sigma_3^{(h)} + \sigma_3^{(e)}) - (\sigma_{12}^{(h)})^2]} \quad (4)$$

Equation (4) indicates that within this approach: (i) the existence of a LHE requires the presence of both electrons *and* holes, and (ii) a high mobility of one band (electrons in the case of SrAs_3) enhances the monoclinic contribution of the other band (holes). Expression (4) has been derived for a reference frame parallel to the crystallographic axes a and b . It might be argued that the hole conductivity tensor can be diagonalized by a rotation of the coordinate system. However, such a transformation will now produce a nondiagonal conductivity component for the electron band. In general, the ellipsoids representing different bands of a monoclinic crystal will not have parallel axes. Thus LHE coefficients will exist with differing magnitude in any coordinate system.

III. EXPERIMENTAL RESULTS

The Hall effect measurements have been performed on bar-shaped samples using a fully automatic Hall setup² at a magnetic field strength of 0.8 T which still represents the low-field limit for the transport coefficients of the investigated compounds. Five probes of 100- μm gold wire have been spot-welded directly on the polished surfaces of single crystals grown using the Bridgman method. The crystallographic orientations have been determined with Laue back reflection.

The results for SrAs_3 are in good agreement with the above picture (Fig. 1). At room temperature, SrAs_3 is predominantly n -conducting, and a negligible LHE is found. For $T < 150$ K the conductivity develops a two-band character (indicated by the sign change of ρ_{132} at 110 K) and the LHE increases significantly. SrAs_3 remains a two-band material down to the lowest temperatures, accompanied by a monotonic increase of the LHE coefficients becoming comparable in absolute magnitude with the THE coefficients.

EuAs_3 is a p -type semimetal for $T < 300$ K,⁷ and no LHE is observed. The measurements are complicated at room temperature by the low magnitude of the conventional Hall effect and for $T < 200$ K by the occurrence of a strong extraordinary Hall effect due to the magnetic moments of the Eu^{2+} ions.

Measurements on AuTe_2 by Sagar *et al.*³ claiming the detection of a LHE did not fulfill the requirements for an unambiguous separation of THE and LHE coefficients.

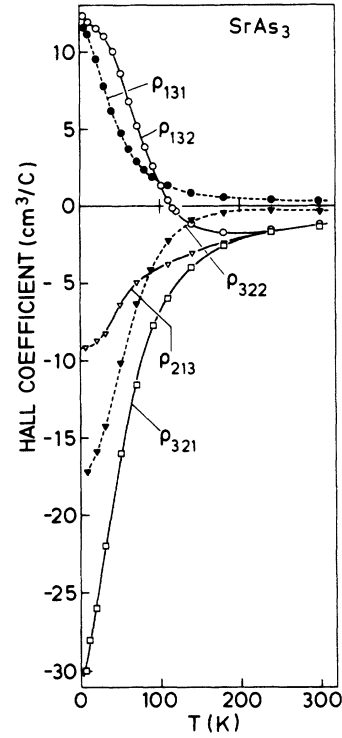


FIG. 1. Temperature dependencies of the transverse Hall coefficients ρ_{213} , ρ_{321} , and ρ_{132} (open symbols) and the longitudinal coefficients ρ_{131} and ρ_{322} (solid symbols) of SrAs_3 .

We have therefore applied our measuring method to AuTe_2 . The data for ρ_{22} , ρ_{33} , and ρ_{231} (corresponding to R_{aa} , R_{bb} , and R_{abc} in the notation of Ref. 3) show good agreement with the values given in Ref. 3, indicating a comparable quality of the samples [Fig. 2(a)]. Figure 2(b) displays angle-dependent Hall effect measurements using the appropriate rotation scheme for the detection of a LHE. The lines represent fits of cosine functions to the data points. No indication of a LHE is found for $T < 160$ K. An increasing contribution of longitudinal Hall coefficients above 180 K manifests itself in the angular shift α_z of the zero crossings of the Hall voltage [Fig. 2(d)]. At 180 K, a LHE is clearly detected by the angle-dependent measurements, whereas the change in amplitude ($< 5\%$) still remains within the order of the absolute experimental accuracy. Note that a LHE in AuTe_2 is only detected when the *decreasing* THE coefficient [Fig. 2(c)] indicates a *growing hole contribution* to the electrical conductivity.

The most striking evidence for the two-band character of the LHE is found for semiconducting CaAs_3 . The crystal structure of CaAs_3 displays a small triclinic distortion of the SrAs_3 structure.⁵ Its physical properties, however, essentially still obey the $C2/m$ selection rules.⁸ The electrical behavior of CaAs_3 changes from n type at room temperature to p type at low temperatures [Fig. 3(b)]. The angle-dependent Hall effect measurements [Fig. 3(a)] clearly demonstrate the occurrence of a LHE in the vicinity of 200 K. The transverse coefficient ρ_{231} changes its sign at 190 K; therefore both electrons *and*

holes contribute to the conductivity in this temperature regime. No significant LHE is observed in the temperature ranges with predominantly one-band conductivity, n type for $T > 250$ K and p type for $T < 150$ K. Note that due to the vanishing transverse coefficient ρ_{231} the maximal Hall voltage occurs close to the longitudinal

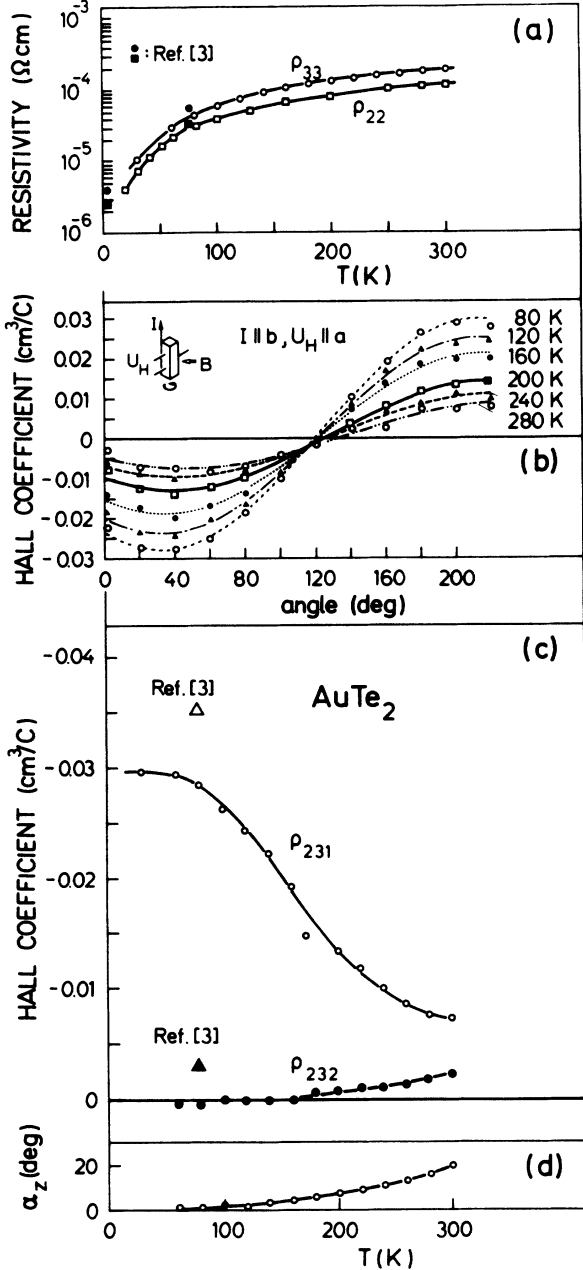


FIG. 2. (a) Electrical conductivities of AuTe₂ along a , ρ_{22} , and along b , ρ_{33} , as a function of temperature. Corresponding values of Ref. 3 are indicated. (b) Angular dependence of the Hall coefficient of AuTe₂ at different temperatures for the indicated rotation scheme. Longitudinal contributions manifest themselves by the shift of the zero crossing for $T \geq 200$ K. (c) Temperature dependencies of the transverse Hall coefficient ρ_{231} (open symbols) and the longitudinal coefficient ρ_{232} (solid symbols). The data were taken at the appropriate angular positions of (b). Triangles mark the values given in Ref. 3. (d) Temperature variation of the angle α_z where the Hall voltage changes its sign (relative to the position at 80 K).

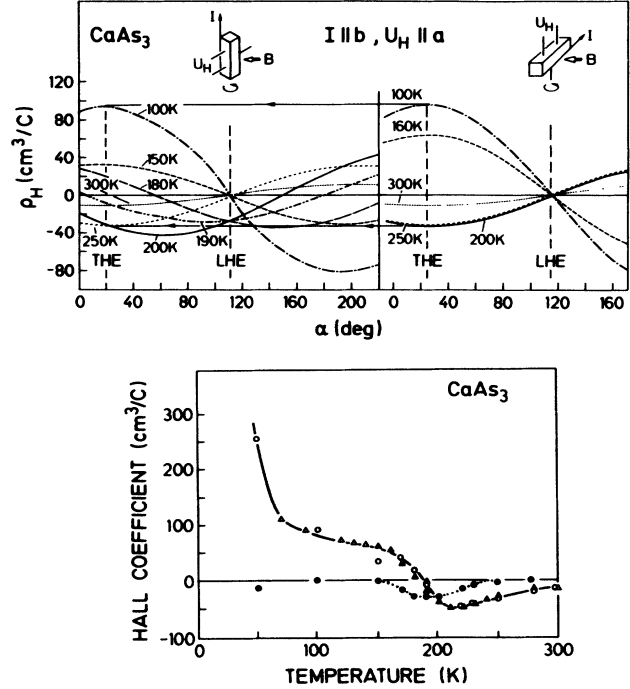


FIG. 3. Upper part: Angular dependence of the Hall coefficient of CaAs₃ for two different rotation schemes. Lines drawn at the maximal values of the THE curves (right-hand side) intersect the corresponding THE + LHE curves (left-hand side) at the positions of a pure THE (shown for 100, 200, and 250 K). Lower part: Temperature dependencies of the transverse Hall coefficient ρ_{231} (open symbols) and the longitudinal coefficient ρ_{232} (solid circles). The data for ρ_{231} are taken from two different samples.

geometry. An equivalent observation² has been made for SrAs₃.

IV. CONCLUSION

The measurements on SrAs₃, EuAs₃, AuTe₂, and CaAs₃ strongly support our model claiming the two-band character of a *significant* first-order LHE. Symmetry-allowed longitudinal Hall coefficients can be strongly enhanced by two-band effects. No LHE could be observed in the predominantly one-band temperature regimes of the investigated compounds.

The experimental results can be summarized as follows. A single monoclinic band yields a marginal, hardly measurable LHE. This represents the case of a small perturbation due to the low symmetry. In contrast, two bands with differently inclined ellipsoids can lead to LHE coefficients whose magnitudes are comparable to the THE coefficients.

ACKNOWLEDGMENTS

I am grateful to M. Wittmann, M. Hartweg, and H. Binder for growing the CaAs₃, EuAs₃, and AuTe₂ single crystals. I would like to thank Monika Latzko and Sonja Sachs for sample preparation and assistance with the measurements.

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