Anomalous Hall Effect in the (In,Mn)Sb Dilute Magnetic Semiconductor

G. Mihály,[1] M. Csontos,[1] S. Bordács,[1] I. Kézsmárki,[1] T. Wojtowicz,[2] X. Liu,[3] B. Jankó,[3] and J. K. Furdyna[3]

[1]*Department of Physics, Budapest University of Technology and Economics, and Condensed Matter Research Group of the Hungarian Academy of Sciences, 1111 Budapest, Hungary*

[2]*Institute of Physics, Polish Academy of Sciences, PL-02668 Warsaw, Poland*

[3]*Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556, USA*

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High magnetic field study of Hall resistivity in the ferromagnetic phase of (In,Mn)Sb allows one to separate its normal and anomalous components. We show that the anomalous Hall term is not proportional to the magnetization, and that it even changes sign as a function of magnetic field. We also show that the application of pressure modifies the scattering process, but does not influence the Hall effect. These observations suggest that the anomalous Hall effect in (In,Mn)Sb is an intrinsic property and supports the application of the Berry phase theory for (III,Mn)V semiconductors. We propose a phenomenological description of the anomalous Hall conductivity, based on a field-dependent relative shift of the heavy- and light-hole valence bands and the split-off band.

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As promising candidates for spintronic applications, (III,Mn)V dilute magnetic semiconductors have attracted considerable attention during the past few years [1]. In these alloys, the Mn^{2+} ions provide localized magnetic moments and valence band holes at the same time. The presence of charge and spin degrees of freedom and the carrier mediated nature of the ferromagnetic coupling open a new way to the electrical control of ferromagnetism [2]. Devices based on magnetic semiconductors represent a novel generation of information technology where magnetoresistive random access memory functionalities arise simply from bulk properties [3,4]. Moreover, the magnetic state of diluted magnetic semiconductors (DMSs) can be quickly and conveniently characterized by transport measurements by taking advantage of the unusually large anomalous Hall effect (AHE). While AHE is one of the prominent key properties of ferromagnetic semiconductors, it represents a situation of general interest but has not garnered a firm knowledge base.

In general, AHE may arise from scattering processes involving spin-orbit coupling. In the skew scattering and side-jump models [5–7], the Fermi-surface properties of the charge carriers are important. We test this possibility experimentally by modifying the scattering process via application of hydrostatic pressure.

An alternative class of descriptions relates AHE to the Berry phase acquired while the electrons propagate in spin-orbit coupled Bloch bands. In this picture, the AHE arises from near degeneracy points of the bands [6–11] where interband processes become relevant. Spin-orbit coupling and the lack of inversion symmetry may result in this type of AHE even for collinear ferromagnets [10]. Our high magnetic field experiments aim to reveal an AHE contribution arising from the relative displacement of the various spin-up and spin-down bands.

InSb has the largest spin-orbit coupling among the III–V semiconductors and becomes a ferromagnetic metal when a few percent of Mn^{2+} ions are inserted into the In^{+} sites [12–16]. The band structure is well known [17], including the parameters of the partially filled heavy-hole (hh) and light-hole (lh) bands. (In,Mn)Sb is an ideal system to study AHE, and our purpose is to distinguish between the possible AHE mechanisms experimentally.

The $In_{0.98}Mn_{0.02}Sb$ sample was grown by low temperature molecular beam epitaxy (MBE) in a Riber 32 R&D MBE system on closely lattice matched hybrid (001) CdTe/GaAs substrates to a thickness of 230 nm (for further growth details and structural characterization, see Refs. [13,14]). The magnetic properties were investigated by magneto-optical Kerr-effect (MOKE) measurements. The magnetotransport measurements were performed in a six-probe arrangement, where the four side contacts allow the simultaneous measurement of the longitudinal and the transverse voltages. The magnetic field was applied perpendicular to the layer plane. For high-pressure measurements, the samples were mounted in a self-clamping cell, with kerosene as the pressure medium.

In ferromagnetic systems, the Hall-resistivity is often described as a sum of two terms,

$$\rho_H = \rho_{xy} = R_0 B + R_S M, \tag{1}$$

a normal Hall contribution due to the Lorentz force, plus an anomalous Hall term (ρ_{AH}) that is proportional to the magnetization M. Alternatively, a similar separation can be made in the Hall-conductivity,

$$\sigma_H = \sigma_0 B + \chi_S M. \tag{2}$$

Here, the first term corresponds to the normal Hall effect related to the carrier concentration ($\sigma_0 = R_0/\rho^2$), while the second term defines the anomalous Hall conductivity, σ_{AH}.

The analysis of the AHE in terms of ρ_{AH} or σ_{AH} is not a simple technical question. The description by ρ_{AH} implicitly assumes that the Hall signal arising from different processes is additive in the scattering rate, $1/\tau$. In contrast, in the Berry phase picture, the transverse current due to the AHE is additive and does not depend on the electron scattering that determines the longitudinal current.

In most materials—including (In,Mn)Sb—the Hall-resistivity ($\rho_H = \rho_{xy}$) is at least by 1 order of magnitude smaller than longitudinal resistivity ($\rho_{xx} = \rho$). Consequently, the Hall conductivity defined by Eq. (2) cannot be distinguished from the off-diagonal conductivity derived by matrix inversion from Eq. (1), as $\rho_H/(\rho_{xx}^2 + \rho_H^2) \approx \rho_H/\rho_{xx}^2 = \sigma_H$.

In magnetic semiconductors at low magnetic fields, the Lorentz term usually gives a negligible contribution compared to the AHE, and the Hall resistivity seems to be simply proportional to the magnetization [14]. This situation is exemplified in Fig. 1 by the temperature dependence of the Hall signal in (In,Mn)Sb: The development of a large nonlinear contribution to $\rho_H(B)$ and the onset of hysteresis loops signify the crossing of the paramagnetic-ferromagnetic phase boundary [15].

The Hall phenomenon at high magnetic fields, however, is more complex: the anomalous Hall effect is not simply proportional to the magnetization, i.e., the AHE coefficient—either R_S or χ_S—is not a constant, but strongly field dependent. The upper panel in Fig. 2 displays the Hall-resistivity up to $B = 14$ T, at various pressures. Qualitatively, the two terms of Eq. (1) play different roles in the different field ranges. At low fields, the negative contribution of the AHE overcomes the positive normal Hall term, while at high fields, the linear contribution due to the Lorentz force is dominating (the slope of this linear term is temperature independent and corresponds to the concentration of the magnetically active Mn ions [16]). Subtracting the linear contribution from the total Hall signal, the resulting anomalous Hall term has a nonmonotonic dependence on the magnetic field: following a sharp peak, it slowly decays, changes sign, then saturates at high fields. As the magnetization gradually increases with increasing magnetic field, it is clear that the $\rho_{AH} \propto M$ relation is not valid.

Before proceeding, we note that the separation of the normal Hall effect by subtracting a linear term—which is often applied as a first step of the analysis—assumes that the linear field dependence of the normal Hall resistivity is not affected by the multiband nature of the electrical conductivity. In materials where various bands contribute to the conductivity, however, a deviation from the linear variation is expected, since the contribution of the different types of carriers is not simply additive [18]. While this is true, the characteristic field above which this effect may influence the normal Hall term is quite high, given by ρ/R_0, which is ~100 T for (In,Mn)Sb. Another possibility is that the subband Hall contributions to the normal Hall effect themselves are field dependent due to the change in the population of the spin-split bands. As both the band parameters [17] and the value of the exchange coupling is known [19], it is easy to show that in the magnetic field range of the experiments, this leads to less than 1% correction to R_0.

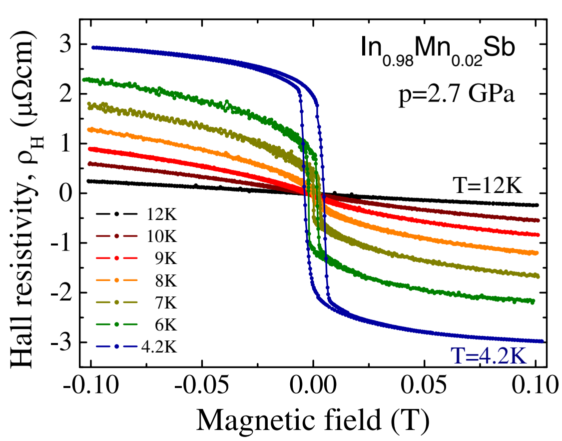

FIG. 1 (color online). Hall resistivity of $In_{0.98}Mn_{0.02}Sb$ at various temperatures, up to magnetic field $B = 0.1$ T.

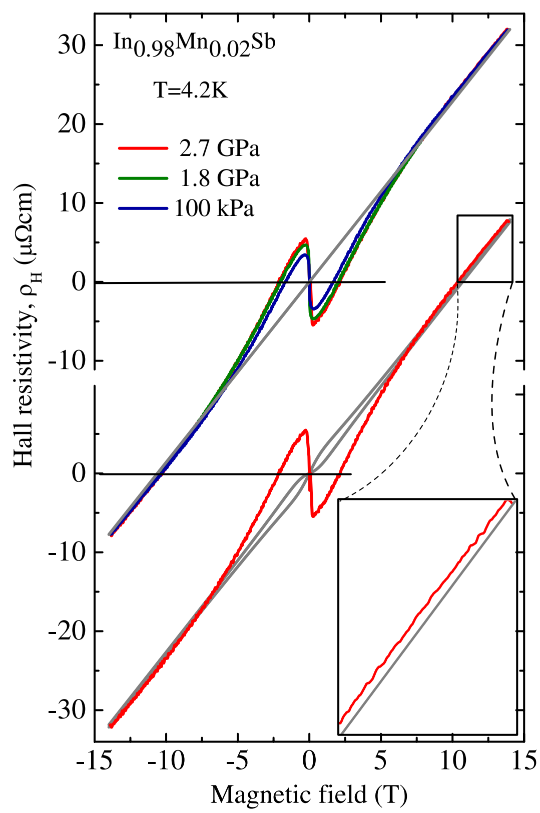

FIG. 2 (color online). Upper panel: Hall resistivity of $In_{0.98}Mn_{0.02}Sb$ at various pressures, up to magnetic field $B = 14$ T. The gray line crossing the origin corresponds to the normal Hall effect. Lower panel: illustration of the possible field dependence of the normal Hall resistivity due to multiband effects; the two gray curves represent the very extreme limits for the field dependence in the normal Hall resistivity. The inset shows the high-field behavior, where ρ_{AH} saturates at a positive value, resulting a small shift compared to the normal Hall term.

The normal Hall effect may however be field dependent for a third reason. In case of different types of carriers, any difference in the magnetic field dependence of the subband resistivities influences the relative weights of the subband Hall contributions [20]. While the contributions of the two subbands to the normal Hall effect cannot be determined separately, the limiting bounds of the field dependence can easily be evaluated. These correspond to the situations when one of the two subbands makes the dominant contribution to the Hall effect, while the magnetoresistance arises solely either from this or from the other subband. These limiting curves can be derived from the experimentally determined magnetoresistance. As a result, the area enclosed by the gray curves in the lower panel of Fig. 2 represents the possible field dependence of the normal Hall resistivity arising from the multiband nature of (In,Mn)Sb. Clearly the observed Hall signal is far beyond even the extreme limits of the normal Hall term. Thus, the strong field dependence has to be attributed to the anomalous Hall effect. Note also that, due to the identical (positive) sign of R_0 in the heavy- and light-hole bands, a negative peak cannot arise from multiband effects under any circumstances.

Next, we briefly discuss the influence of hydrostatic pressure on the Hall effect. The pressure effects on the magnetotransport have been investigated in details previously [15,16]. Here, we only recall that while the number of charge carriers is not influenced by pressure—as demonstrated by the high-field behavior of the Hall resistivity shown in Fig. 2— the longitudinal resistivity is enhanced. The experimental observations suggest that $\rho(p, T, B)$ has the form of $f(p)\rho(T, B)$, which indicates that the resistivity change is due to the pressure-induced enhancement of the effective mass. (The relative variation of the resistivity is similar to that of InSb pressure gauges used in low temperature experiments [21]). We found that the diagonal conductivity decreases by more than 20% for the applied pressure of 2.7 GPa, as shown in Fig. 3(b). In contrast, the anomalous part of the off-diagonal conductivity is independent of pressure [see Figs. 3(c) and 4(a)], indicating a dissipationless anomalous Hall current [22].

The above observation is in disagreement with the extrinsic scattering picture. Simultaneously, the strong field dependence of the AHE is also suggestive of an intrinsic mechanism, which is independent of the scattering processes and is rather determined by the singularities in the band structure. Note also that the anomalous Hall conductivity changes sign as a function of magnetic field [Fig. 4(a)], which is not expected in the scattering models of the AHE.

Berry-phase calculations of the AHE are based on the four band spherical Luttinger model of the (III,V) semiconductors which takes into account parabolic dispersions for heavy-hole and light-hole bands in the presence of spin-orbit coupling [10,11]. In the ferromagnetic case, an additional term has to be introduced into the total Hamiltonian [10]: $H_{ex} \propto J_{pd}sS$. This represents the exchange interac-

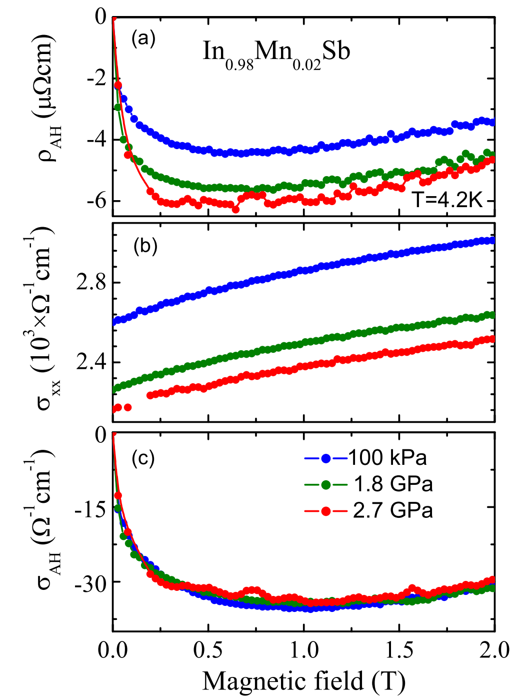

FIG. 3 (color online). (a) Field dependence of the anomalous Hall resistivity at various pressures. (b) and (c) Influence of hydrostatic pressure on the diagonal and off-diagonal terms of the conductivity tensor. In spite of the pressure-induced enhancement of the scattering process, the anomalous Hall conductivity is pressure-independent.

tion between the localized magnetic moments on Mn^{2+} ions (S) and the spins of the charge-carrying holes (s) and results in the spin splitting of the valence bands. Jungwirth *et al.* have shown that if both spin-orbit and exchange coupling (J_{pd}) are important, then AHE is generally nonlinear in the magnetization, and it may have both positive and negative signs [10]. Their numerical calculations—including the influence of the split-off band—gave good estimates for the magnitude of the AHE in (Ga,Mn)As and (In,Mn)As.

The Luttiger parameters of InSb [17] indicate that for $In_{0.98}Mn_{0.02}Sb$, where the hole concentration is $n = 3 \cdot 10^{20}$ cm^{-3}, the Fermi level is only $\sim$150 meV away from the lower lying split-off band. Because of the exchange splitting, the majority and minority spin bands move about $\pm$25 meV apart from each other. In the Berry-phase picture, the dominant contributions to AHE arise from the nearly degenerate points of the bands located close to the Fermi energy [6–9]. The vicinity of the split-off band—even without band-crossing—may then have a significant effect on the Berry phase acquired by the heavy- and light-holes, and the large shift of up- and down-spin bands may be responsible to the measured field dependence of the AHE.

It is important to note that as the magnetic field is varied, the relative position of the bands shifts linearly with the absolute value of the magnetization (due to the exchange

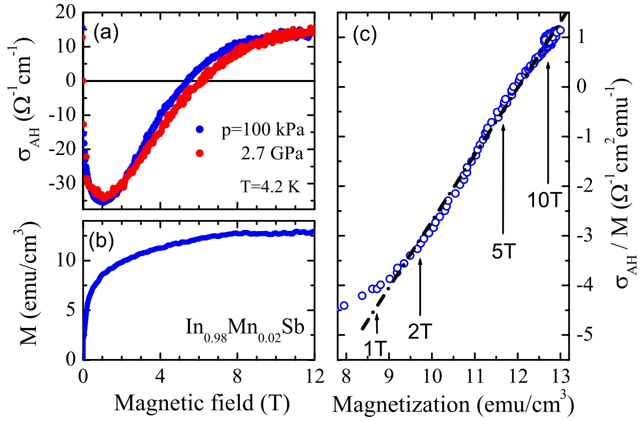

FIG. 4 (color online). Comparison of the field dependence of (a) the high-field anomalous conductivity and (b) the magnetization. (c) $\sigma_{\rm AH}/M$ as a function of M. The variation of $\sigma_{\rm AH}/M$ as a function of M is clearly seen to be linear over a wide field range above about 2 T. The dash-dotted line corresponds to Eq. (3).

origin of the splitting). Assuming that the corresponding correction in the anomalous Hall coefficient is also linear in band shift, i.e., $\chi_S \sim (1 - \alpha|M|)$, one obtains the anomalous Hall conductivity varying as

$$\sigma_{\rm AH} \propto M(1 - \alpha|M|). \tag{3}$$

Such a behavior is demonstrated in Fig. 4, where the experimentally determined $\sigma_{\rm AH}(B)$ and the corresponding $M(B)$ curves are plotted in the from of $\sigma_{\rm AH}(B)/M$ versus M. The observed linear variation above $B \approx 2$ T confirms the above phenomenological picture (using only one fitting parameter, $\alpha = 0.05\ {\rm cm}^3/{\rm emu}$) [23].

Linear magnetization dependence of the high-field AHE has been observed in ferromagnetic Mn_5Ge_3 [24]. This phenomenon has an intrinsic origin, too, but with a different physics behind; it was attributed to Berry phase contributions due to spin orientation fluctuations. By now, it is clear that intrinsic AHE mechanisms of various origin dominate in many systems, but extrinsic processes may prevail in other cases, and a crossover between the two types of mechanisms has also been reported [25].

In conclusion, we showed that—in contrast to the general belief—in (In,Mn)Sb, the AHE is not simply proportional to magnetization. The anomalous Hall signal (either R_S or χ_S) can even reverse sign before saturating at high field. We attribute this behavior of AHE to Berry-phase effects, and we propose a qualitative description of the field-dependent AHE, where exchange splitting leads to a relative shift between the valence bands and the nearby split-off band. The intrinsic nature of the AHE was also confirmed by high-pressure experiments: we demonstrated that the off-diagonal terms of the conductivity tensor are not influenced, while the diagonal terms are reduced due to the pressure-induced enhancement of the scattering process.

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