

Hall effect in the new diluted magnetic semiconductor p-CdSb:Ni

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The Hall effect is investigated in oriented single crystals of a new diluted group II-V semiconductor p-CdSb doped with 2 at % of Ni in the temperature interval of $T = 1.5\text{--}300$ K and pulsed magnetic fields up to $B = 25$ T. A non-linear dependence of the Hall resistivity, ρ_H , on B is observed, which is strongly pronounced below ~ 10 K and still exists even up to 300 K. From the analysis of $\rho_H(B)$ a positive normal, $\rho_N = R_0 B$, and a negative anomalous, ρ_A , contributions are obtained. The normal Hall coefficient, R_0 , is determined pre-

sumably by activation of holes into the valence band, accompanied with a minor contribution of itinerant holes from the acceptor band at lowest T , whereas ρ_A below ~ 77 K scales with resistivity approximately as ρ^n ($n = 1.6 \pm 0.1$). The anomalous contribution to the Hall effect is attributable to presence of magnetic $\text{Ni}_{1-x}\text{Sb}_x$ nanoclusters, which have been found previously from investigation of magnetic properties of p-CdSb:Ni.

1 Introduction A group II-V diluted magnetic semiconductor (DMS) p-CdSb:Ni has been prepared and investigated recently, demonstrating interesting magnetic properties [1]. Magnetization of this material saturates at 300 K already in the magnetic fields B above ~ 2 T, exhibiting large anisotropy of the saturation magnetization, $M_s = 11.6, 14$ and 30 (in units of 10^{-4} emu/g) along the axes [100], [010] and [001], respectively. In addition, magnetic irreversibility or deviation of the zero-field cooled susceptibility, $\chi_{ZFC}(T)$, from the field-cooled susceptibility, $\chi_{FC}(T)$, is observed already below the room temperature, accompanied with a broad peak of $\chi_{ZFC}(T)$ at temperature $T_b \sim 100$ K [1]. Magnetic properties of p-CdSb:Ni have been explained by presence of nanosize Ni-rich $\text{Ni}_{1-x}\text{Sb}_x$ precipitates (clusters), which are formed in CdSb matrix at small doping levels and have a ferromagnetic (FM) ordering of internal Ni spins at $x < 7.5$ %, a high aspect ratio and preferential orientation of the major axes, as well as large size distribution [1]. The volume fraction of the $\text{Ni}_{1-x}\text{Sb}_x$ phase,

$\eta' \sim 0.01$ % being well below the percolation level, makes the clusters completely independent (isolated).

Analysis of the resistivity, ρ , of p-CdSb:Ni, which is anisotropic (as well as in undoped p-CdSb [2]), demonstrates that it is governed above $T_{cr} \sim 3$ K by activation of holes from the acceptor band (AB) to the valence band (VB), whereas below T_{cr} by a hopping charge transfer over the localized states of the AB, yielding the values of the activation energy, $E_A = 2.45, 2.50$ and 2.85 meV ($T > T_{cr}$), and the acceptor concentration, $N_A = 3.61, 3.37$ and 2.52×10^{16} cm⁻³ in samples # 1, # 2 and # 3, oriented along the axes [100], [010] and [001], respectively [3].

In this work investigations of the Hall effect in p-CdSb:Ni are reported. As observed in various DMS [4–6], presence of magnetic nanoclusters in our material suggests existence of the anomalous, ρ_A , contribution to the net Hall resistivity, ρ_H , which in our case of small η' is expected to be observed along with the normal contribution, ρ_N , providing valuable information on both ρ_A and ρ_N .

2 Results and discussion Single-crystalline material of p-CdSb doped with 2 at % of Ni was obtained with the modified Bridgman method (for details see [1]). For investigations rectangular prisms with the longest edge along [100] (# 1), [010] (# 2) and [001] (# 3) axes, respectively, were cut from the ingots. The same samples have been used previously in the investigations of ρ and magnetoresistance [3]. The Hall resistivity was measured in pulsed magnetic fields up to $B = 25$ T in configurations of $\mathbf{j} \parallel [100]$ and $\mathbf{B} \parallel [001]$ (# 1), $\mathbf{j} \parallel [010]$ and $\mathbf{B} \parallel [100]$ (# 2), $\mathbf{j} \parallel [001]$ and $\mathbf{B} \parallel [010]$ (# 3).

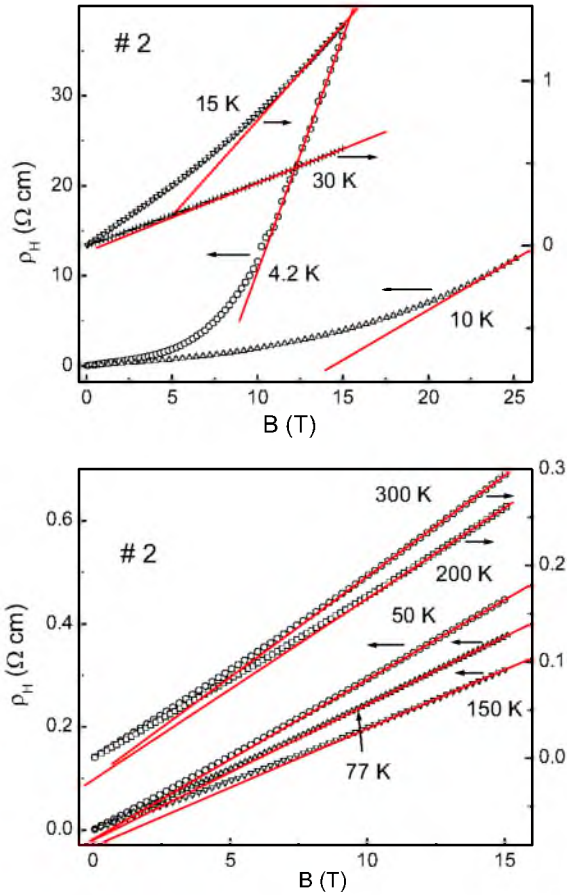


Figure 1 The plots of ρ_H vs. B for # 2. The lines are linear fits.

The dependence of ρ_H on B (an example for # 2 is shown in Fig. 1) exhibits a clear non-linearity or strengthening of $\rho_H(B)$ with increasing field, being most pronounced below ~ 10 K but still persisting even up to 300 K. It can be shown that such non-linear behavior of $\rho_H(B)$ cannot be attributed to the reasons similar with those acting in non-magnetic 3D semiconductors, including presence of the two groups of the charge carriers like the VB holes and itinerant holes of the AB in undoped p-CdSb [7], the magnetic freeze-out of the VB charge carriers or complex

magnetic-field behavior of the resonant bands in n-CdSb:In [8]. Therefore, non-linearity of $\rho_H(B)$ should be associated with the magnetic subsystem existing in p-CdSb:Ni (see Introduction) due to an anomalous contribution to the Hall effect superimposed on the normal one.

In FM compounds

$$\rho_H = \rho_N + \rho_A, \quad (1)$$

where $\rho_N = R_0 B$, $\rho_A = R_s M$. R_0 and R_s are the normal and the anomalous (spontaneous) Hall coefficients, respectively, and M is the magnetization of the material [9]. Applicability of Eq. (1) to p-CdSb:Ni, which, strictly speaking, does not belong to FM materials, can be partially substantiated by saturation of the magnetization when B is increased (see Introduction) at any T , making the second term in Eq. (1) to be independent of B and tending the plots of ρ_H vs. B to linear functions. As can be seen in Fig. 1 this takes place in strong magnetic fields, permitting us to obtain R_0 and ρ_A as the slope and the interception point of the linear parts with the vertical axis, respectively [9]. The dependences of $R_0(T)$ and $\rho_A(T)$ are shown in Fig. 2.

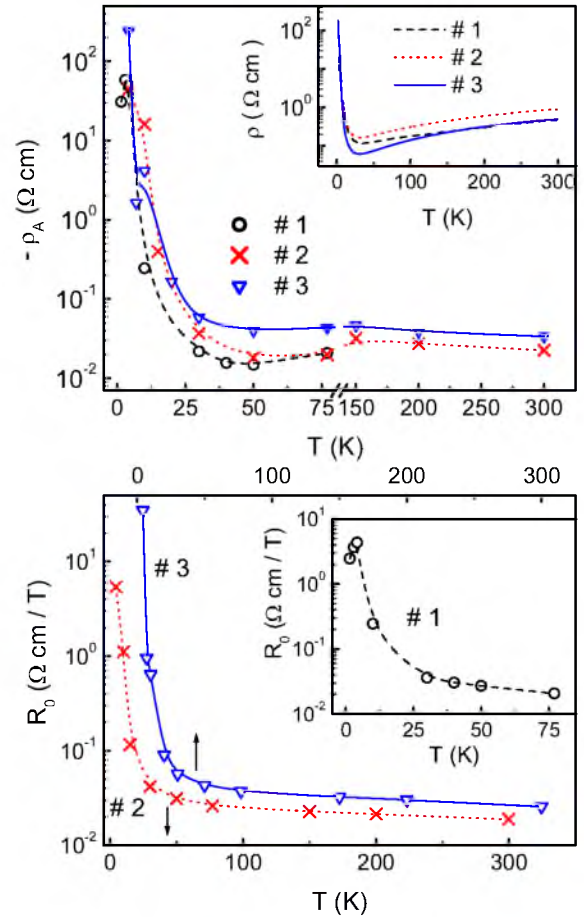


Figure 2 Temperature dependence of ρ_A (top panel) and R_0 (bottom panel) in the investigated samples. The lines are to guide the eye. Inset to the top panel: the plots of resistivity, ρ , vs. T [3].

The Hall concentration, $p_H \approx 1/(eR_0)$, in the interval of extrinsic conductivity $\sim 4.2\text{--}200$ K is determined by the holes activated from acceptors to the VB with concentration $p_V(T)$, and by the holes in the extended states of the AB, having the fraction α , with the concentration $\alpha p_A(T)$:

$$p_H(T) = p_V(T) + \alpha p_A(T), \quad (2)$$

where $p_A(T)$ is the total concentration of the holes in the AB. The interval above is limited by $T > 4.2$ K due to transition to the hopping conduction at lower T [3], whereas temperatures below ~ 200 K guarantee absence of the intrinsic contribution to $p_H(T)$ [2]. Due to non-degeneracy of the holes in our samples [3] we can write:

$$p_V(T) = N_V(T) \exp[-\xi/(kT)] \quad (3)$$

and

$$p_A(T) = 2 N_A / \{1 + \exp[-(E_A - \xi)/(kT)]\}, \quad (4)$$

where ξ is the chemical potential (here the energy is measured from the edge towards the depth of the VB), $N_V(T) = 2 (2mkT)^{3/2} / (4 \pi^{3/2} h^3)$ and $m = 0.2 m_0$ is the mean effective mass of the holes in CdSb (m_0 is the free electron mass) [2, 9]. Above we took into account existence of the two equivalent maxima of the VB of CdSb [2] and consider one unsplit by spin AB (neglecting the Hubbard correlations). Taking into account conservation of the holes,

$$p_A(T) + p_V(T) = N_A (1 - K), \quad (5)$$

where $K = N_D / N_A$ is the degree of compensation and N_D is the concentration of compensating donors, we obtain a complete system of equations for p_V and p_A , given by Eqs. (3)–(5), which is solved numerically by excluding ξ . Finally, $p_H(T)$ is calculated with Eq. (2) and is shown by solid lines in Fig. 3. In calculations the values of E_A and N_A are taken from [3] (see Introduction), the values of $K = 5.5\%$, 13.4% and 17.7% for # 1, # 2 and # 3, respectively, are obtained by fitting the high-temperature parts of $p_H(T)$, whereas those of $\alpha = 4 \times 10^{-3}$, 3.5×10^{-3} and 0.8×10^{-3} for # 1, # 2 and # 3, respectively, by fitting $p_H(T)$ on the low-temperature interval. One can see a good agreement of the calculated curves with the experimental data of $p_H(T)$ in the whole interval between $4.2\text{--}200$ K, where the model given by Eqs. (2)–(5) is applicable as discussed above (the deviations in # 1 taking place below 4.2 K are connected with transition to the hopping conduction [3], where the model above becomes inapplicable). It can be seen also that calculations with $\alpha = 0$ (the dotted lines in Fig. 3), deviate from those with $\alpha \neq 0$ only below ~ 10 K. Together with smallness of values of $\alpha \sim 10^{-3} \ll 1$ this means existence of only a quite narrow interval of the extended states in the AB and negligible role of itinerant holes of the AB

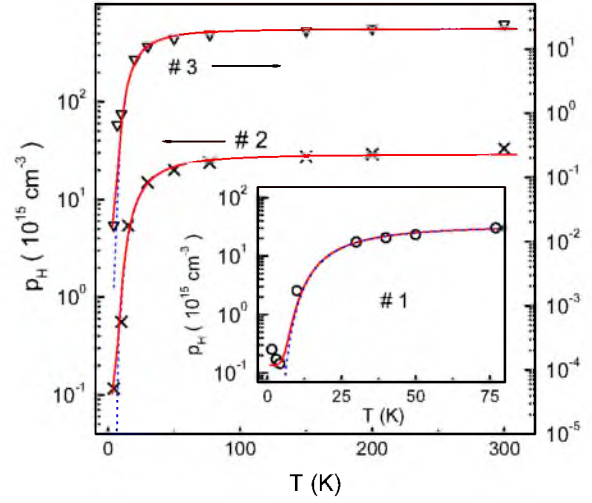


Figure 3 Temperature dependence of p_H in the investigated samples. The lines are calculated as described in the text.

in formation of the normal contribution to the Hall effect, excluding only the lowest temperatures.

As can be seen from the top panel of Fig. 2, the anomalous contribution to the Hall effect in p-CdSb:Ni is negative, comprising of the weak variation of $\rho_A(T)$ above ~ 50 K and a very strong increase with decreasing T from ~ 50 to 4.2 K. One can mention also large scattering of the data of $\rho_A(T)$ between different samples, exceeding considerably the error of their determination from the high-field linear approximation of $\rho_H(B)$ in Fig. 1. The normalization of ρ_A by M_s , shown in Fig. 4, leads to closer values of ρ_A/M_s for different samples, however it cannot decrease the scattering of the data in whole.

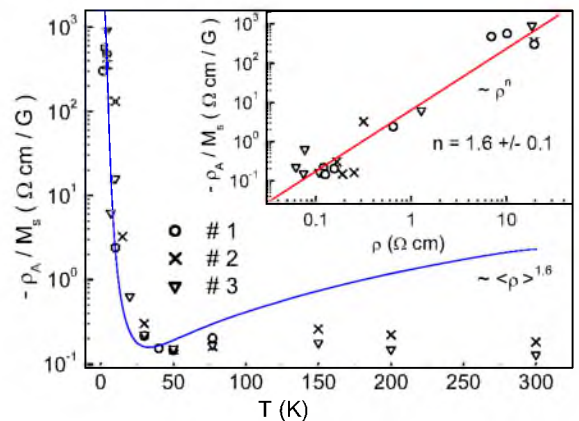


Figure 4 The dependence of ρ_A/M_s in the investigated samples. The line is the low-temperature fit to the experimental data with the function $\langle \rho(T) \rangle^{1.6}$. Inset: ρ_A/M_s vs. ρ in the double-logarithm scale with the line as a linear fit.

Various microscopic models of the anomalous Hall effect (AHE) predict the power-law scaling of ρ_A vs. resistivity ρ , yielding relations like $\rho_A \sim \rho^n$, where n is a constant (see below). Therefore, we compared the dependences of $\rho_A(T)$ and $\rho(T)$, despite of more similar behavior of $\rho_A(T)$ and $R_H(T)$ following from Fig. 2, restricting the interval of analysis to $T < 77$ K where all three parameters, $\rho_A(T)$, $\rho(T)$ and $R_H(T)$ behaves approximately in a similar way, exhibiting a strong upturn with lowering T (Fig. 3). To test a possibility of the power-law scaling above we plotted ρ_A/M_s vs. ρ at $T \leq 77$ K, where $\rho(T)$ for each sample was taken from the inset to the upper panel of Fig. 2, in the double-logarithm scale as shown in the inset to Fig. 4. Indeed, the linear approximation of these data yields $n = 1.6 \pm 0.1$ within four decades of ρ_A / M_s and more than two decades of ρ . In addition, as can be seen from the main part of Fig. 4, ρ_A/M_s behaves between ~ 1.5 –77 K similar to the law $\langle \rho(T) \rangle^{1.6}$, where $\langle \rho(T) \rangle$ is the mean resistivity obtained by averaging the plots in the inset to the upper panel of Fig. 4.

The microscopic models of AHE in FM materials predict the values of $n = 1$ and 2, taking into account the skew scattering and the side jump, respectively, both induced by the spin-orbit coupling [9]. In a theory of multiband FM metals with dilute impurities [10] the power-law dependence $\sigma_A \sim \sigma^n$ of the anomalous Hall conductivity, σ_A , on the conductivity σ , is shown to have values of $n = 1$ or 1.6. On the other hand, for diluted magnetic semiconductors with intrinsic FM behavior the intrinsic Berry phase theory of the AHE predicts the relation of $\rho_A \sim \rho^n$ with $n = 2$ [11].

Experimentally, the values of n between 1–2, as well as outside this interval, were observed in different FM materials (see e. g. [5, 9, 12]), demonstrating non-universality of n in the power-law scaling $\rho_A \sim \rho^n$, which is in line with absence of a universal mechanism of AHE in various FM systems.

At this point, it is important to mention vast analysis of AHE performed using the experimental literature data in 28 different compounds, belonging to five different types of FM materials [13]. This analysis has yielded the value of $n \approx 1.6$, suggesting a broad universality class of this value but in another law, $\sigma_A \sim \sigma^n$, which has been found within over than five decades of σ irrespective of metallic or activated (including hopping) conduction [13].

It can be seen that the value of $n \approx 1.6$ found in [13] coincides with one of predictions of the model [10], but has more general meaning because the model [10] deals only with the metallic conductivity.

The same value of $n = 1.6 \pm 0.1$ obtained from the analysis of our resistivity data (Fig. 4) may reflect the general universality established in [13]. However, the values of ρ , ρ_A and ρ_N are comparable below ~ 77 K (Fig. 2), where the power-law scaling is observed (inset to Fig. 4). This hinder drawing of σ_A from our data unambiguously, which suggests a strong domination of the anomalous contribution to the Hall effect, and does not permit us to obtain

the exponent in the law $\sigma_A \sim \sigma^n$ being a subject of analysis in [13]. Moreover, as have been mentioned above, the investigated samples of p-CdSb:Ni does not belong directly to FM material. Namely, the array of $Ni_{1-x}Sb_x$ nanoparticles possesses features of a FM system only partially, because due to large size distribution (see Introduction) the unblocked clusters exist even below the blocking temperature T_b [1]. Hence, the large scattering of the data of ρ_A (Figs. 2 and 4) may be connected to existence of such unblocked clusters. One can find another feature of AHE in our material, which is also attributable to its poor similarity with FM systems: namely, a linear dependence of $\rho_H(B)$ takes place above ~ 10 T (Fig. 1), whereas the magnetization saturates already above ~ 2 T [1], meaning that $\rho_A(B)$ does not match the behavior of $M(B)$ in detail.

3 Conclusions Investigations of the Hall effect in the new diluted magnetic semiconductor p-CdSb:Ni have been done between 1.5–300 K in pulsed magnetic fields up to 25 T. Both the normal and the anomalous contributions to the Hall resistivity are observed and analyzed.

References

- [1] R. Laiho, A. V. Lashkul, K. G. Lisunov, E. Lähderanta, I. Ojala, and V. S. Zakhvalinskii, *Semicond. Sci. Technol.* **21**, 228 (2006).
- [2] E. K. Arushanov, *Prog. Cryst. Growth Charact.* **13**, 1 (1986).
- [3] R. Laiho, A. V. Lashkul, K. G. Lisunov, E. Lähderanta, M. A. Shakhov, and V. S. Zakhvalinskii, *Mechanisms of Hopping Conductivity of p-CdSb:Ni in Magnetic Field* (manuscript submitted to ICTMC-16).
- [4] S. R. Shinde, S. B. Ogale, J. S. Higgins, H. Zheng, A. J. Millis, V. N. Kulkarni, R. Ramesh, R. L. Greene, and T. Venkatesan, *Phys. Rev. Lett.* **92**, 166601 (2004).
- [5] A. Oiwa, S. Endo, S. Katsumoto, Y. Iye, H. Ohno, and H. Munekata, *Phys. Rev. B* **59**, 5826 (1999).
- [6] P. Lengsfeld, S. Brehme, G. Ehlers, H. Lange, N. Stusser, Y. Tomm, and W. Fuhs, *Phys. Rev. B* **58**, 16 154 (1998).
- [7] R. Laiho, A. V. Lashkul, K. G. Lisunov, E. Lähderanta, M. O. Safonchik, and M. A. Shakhov, *Semicond. Sci. Technol.* **19**, 602 (2004).
- [8] R. Laiho, A. V. Lashkul, K. G. Lisunov, E. Lähderanta, M. O. Safonchik, and M. A. Shakhov, *Semicond. Sci. Technol.* **21**, 918 (2006).
- [9] S. V. Vonsovskii, *Magnetism* (Nauka, Moscow, 1971), Ch. 25.
- [10] S. Onoda, N. Sugimoto, and N. Nagaoka, *Phys. Rev. Lett.* **97**, 126602 (2006).
- [11] T. Jungwirth, Q. Niu, and A. H. MacDonald, *Phys. Rev. Lett.* **88**, 207208 (2002).
- [12] B. A. Aronzon, D. Yu. Kovalev, A. N. Lagarkov, E. Z. Meilikhov, V. V. Rylkov, M. A. Sedova, N. Negre, M. Goiran, and J. Leotin *JETP Lett.* **70**, 90 (1999).
- [13] T. Fukumura, H. Toyosaki, K. Ueno, M. Nakano, T. Yamasaki, and M. Kawasaki, *Jpn. J. Appl. Phys.* **46**, L642 (2007).