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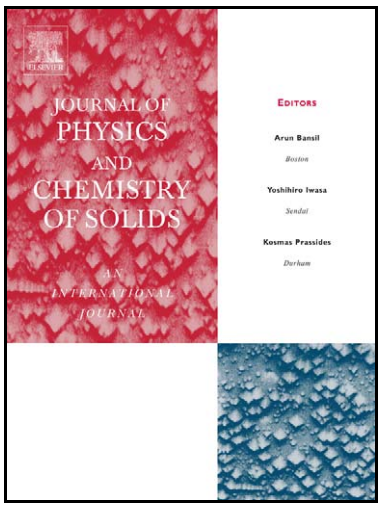

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Hopping conductivity of Ni-doped p-CdSb in strong magnetic fields

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Magnetoresistance (MR) of oriented single crystals of the anisotropic semiconductor *p*-CdSb doped with 2 at % of Ni is investigated between $T = 1.5 - 300$ K in transversal pulsed magnetic fields up to $B = 30$ T. In fields $B \sim 4 - 15$ T at T below 4.2 K the resistivity obeys the law $\ln \rho \sim \eta [B \varphi (B)]^{1/2}$ with $\varphi (B) = a(0)/a(B)$, where a is the carrier localization radius and parameter η depends on $a(0)$, on the acceptor concentration N_A and on the direction of the magnetic field with respect to the crystallographic axes, but does not depend on T. Such behavior gives evidence for MR realized by hopping charge transfer over the nearest-neighbor sites in strong magnetic field. The analysis of the experimental data yields the values of η, agreeing with calculated ones within an error of 10 %, taking into account the effects of the anisotropy of the acceptor states and of the explicit dependence of $a(B)$ due to increase of the activation energy of shallow acceptors in magnetic field and the sensitivity of the metal-insulator transition to B.

I. INTRODUCTION

Interest to semiconductors doped with transition-metal elements is connected to the spin-polarized electron transport, making them attractive e. g. for applications in spintronics [1]. Progress in this rapidly developing area suggests search of new diluted magnetic semiconductor systems. Recently a family of such materials has been enriched by cadmium antimonide doped with Ni (p-CdSb:Ni) [2]. Special interest to this system is connected to strongly anisotropic transport in the group II-V semiconductor p-CdSb [3]. In undoped material the activated conductivity is governed by acceptor bands with energies ~ 3 meV and 6 meV. Fe and Ni substituting for Sb in the lattice act as acceptors, whereas doping with Ag induces the metal-insulator transition (MIT) [3]. The anisotropic quantum transport revealed by Shubnikov-de Haas effect [4] and negative magnetoresistance [5] are observed on the metallic side of the MIT, whereas anisotropic hopping conductivity is realized on the insulating side of the MIT [6].

A new important feature of CdSb doped with Ni is formation of nanosize Ni-rich $Ni_{1-x}Sb_x$ clusters with ferromagnetic (FM) ordering of internal Ni spins [2]. The clusters have a broad size distribution, large non-sphericity and orientations distributed along a preferred direction. The presence of these clusters leads to considerably anisotropic magnetic properties of p-CdSb:Ni, as well as to frustrated ground state and spin-freezing starting at the room temperature [2], making p-CdSb:Ni a perspective spintronics material.

Investigations of hopping transport in p-CdSb:Ni in zero and weak magnetic fields ($B < 6$ T) reveal interesting behavior, uncommon of conventional (non-magnetic) semiconductors. Namely the variable-range hopping (VRH) conductivity at $B = 0$ transforms into the nearest-neighbor hopping (NNH) conductivity in nonzero field. This transition is accompanied with expansion of the temperature interval of the hopping conductivity already in weak magnetic fields, in line with presence of magnetic disorder or strongly inhomogeneous magnetization due to Ni-rich nanoclusters [7], which is damped when B is increased.

In this work detailed investigation of the hopping conductivity in p-CdSb:Ni in strong magnetic field is reported to complete the picture of hopping charge transfer in this material.

II. EXPERIMENTAL RESULTS

Single crystals of CdSb doped with 2 at % of Ni were prepared by the modified Bridgman method [2, 3]. As observed by X-ray diffraction, the ingots of volume ~ 1 cm^3 were of single phase material with orthorhombic structure (space group D_{2h}^{15}) and had the same lattice parameters as undoped CdSb [3]. For investigations rectangular prisms with the longest edge along the [100] (# 1), [010] (# 2) and [001] (# 3) axes, respectively, were cut from the ingots. The measurements of ρ (T) were made by recording the signal from two different pairs of potential contacts on the samples placed in a He exchange gas dewar, where their temperature could be varied between 1.5 – 300 K to an accuracy of 0.5 %. MR measurements were made in transversal field configurations with **j** || [100] and **B** || [001] (# 1), **j** || [010] and **B** || [100] (# 2), **j** || [001] and **B** || [010] (# 3) at temperatures between T = 1.5 – 300 K in pulsed magnetic fields up to B = 30 T. The magnetic pulse length was 8 ms, the error in the strength of the field was not larger than 5 % and its inhomogeneity did not exceed 0.3 %.

As can be seen from Fig. 1 the values of ρ (T) at B = 0 decrease smoothly below 300 K until a minimum is attained at ~ 30 K followed by strong increase of ρ with further decreasing of T [7]. As shown in Fig. 2 the data of ρ (B) exhibit large positive MR, increasing with B and decreasing with increasing T at liquid He temperatures [approximately above the break in the ρ (B) axis], whereas at higher T (below this break) the dependence of ρ on B is weakened. In addition, MR decreases with increasing T up to ~ 30 K and then starts to increase, in agreement with the behavior of ρ (T) at B = 0 in Fig. 1 [7].

III. ANALYSIS OF THE RESULTS AND DISCUSSION

The plots of ln ρ vs. T^{-1} in the inset to Fig. 1 exhibit two intervals of activated behavior characterized by different slopes between ~ 5 – 20 K and below ~ 5 K, corresponding to the conductivity determined mainly by activation of holes from shallow acceptor states to the valence band and to the hopping conductivity over the states of the impurity band, respectively [7]. In semiconductors transversal MR, connected to non-generate carriers in conduction or valence band (VB), depends on B as follows: $\rho(B) = \rho_0 [1 + C(\mu B)^2]$ for $\mu B << 1$ and $\rho(B) = \rho_0 [1 + D]$ for $\mu B >> 1$, where ρ_0 and μ are zero-field resistivity and carrier mobility, respectively, C and D are constants depending on a scattering mechanism [8].

Generally, the dependence of MR on B in the region of conductivity over the VB states, given by these equations, is always much weaker than the exponential magnetic-field dependence of MR in the region of hopping conductivity (see below), which takes place in p-CdSb:Ni below T ~ 4.2 K [7]. This explains the difference in the behavior of MR in Fig. 2 in the two temperature regions, including a strong increase with B at $T \leq 4.2$ K followed by a weaker one above T ~ 10 K. On the other hand, the equations above mean that temperature dependence of MR in the interval of the conductivity over the VB states is determined presumably by ρ_0 and μ, both parameters being functions of T. In more detail this interval is presented for # 3 (the bottom panel of Fig. 2). Namely, one can see two main features of the MR curves at $T > 4.2$ K: (i) rapid decrease of MR with decreasing T between 4.2 – 30 K and slow increase in the interval of 30 – 300 K; (ii) the weakening of the dependence of MR on B with increasing T. Both these features agrees with the behavior prevised by the equations above, the feature (i) being connected to the factor $\rho(0)$ in both equations [cf. the behavior of $\rho(T)$ at $B = 0$ in Fig. 1] and the feature (ii) due to decreasing $\mu(T)$ with increasing T, which takes place in undoped or weakly doped p-CdSb in the corresponding temperature interval [3, 9].

Below we investigate MR in the low-temperature interval of hopping conductivity in more detail.

As mentioned in the Introduction, hopping conductivity of p-CdSb:Ni in zero and weak (B < 6 T) magnetic fields has been already investigated, yielding the data of microscopic parameters such as the mean localization radius, $a \equiv a$ (0), the acceptor concentration, N_A, and the anisotropy coefficients $p_j = [m_j^2 / (m_k m_l)]^{1/6}$, where m_j, m_k and m_l are the components of the hole effective mass. The subscripts j, k, l = 1, 2 , 3 ($j \neq k \neq l$) and j = 3, 1, 2 for # 1, # 2 and # 3, respectively, correspond to the direction of the *magnetic field* along the [001], [100] and [010] axes, respectively (see Table I) [7]. As follows from Table I, the relation $a > a_0$, corresponds to proximity to MIT of the acceptor system in all investigated samples (where a_0 = 83.5 Å is the value of a far from the MIT) and the values of N_A are relatively close to the critical MIT acceptor concentration $N_C = 6.28\times10^{16}$ cm^{-3}. In Table I are displayed also the values of the activation energy of the shallow acceptors, E_A, obtained from the linear fit of the plots of ln ρ vs. T^{-1} at B = 0 in the inset to Fig. 1 [7]. In all the samples the weak-field limit corresponds to the condition of $\lambda >> a_0$, where λ is the magnetic length. Under this condition the quadratic dependence of MR, ln ρ ~ B^2, has been observed in p-CdSb:Ni [7], whereas its violation (for $\lambda \leqslant a_0$) is related to the onset of the strong-field limit of MR [10], which will be analyzed below.

As follows from the percolation model of the hopping charge transfer over the nearest sites (or NNH conductivity) [10], the positive MR is connected to shrinkage of the impurity wave functions in the direction perpendicular to ***B*** and is given in strong magnetic field by the expression [6, 10]

$$\ln [\rho (B) / \rho^*]_j = \eta_j [B \varphi (B)]^{1/2}, \quad (1)$$

where ρ^* is a constant,

$$\eta_j = q\, p_j^{1/2} [e / (N_A\, a\, \hbar)]^{1/2}, \quad (2)$$

q = 0.92 and

$$\varphi (B) = a / a (B). \quad (3)$$

Presence of the factors p_j in Eq. (2) reflects the anisotropy of the acceptor states in p-CdSb and the different elasticity of the anisotropic acceptor wave functions to magnetic shrinkage at

different directions of $\boldsymbol{B}$ in p-CdSb [6, 10], which makes MR to be anisotropic and sensitive to the direction of the magnetic field via the coefficient η_j in Eq. (1). On the other hand, it should be underlined that η_j does not depend on T.

Another important point of the analysis of the MR data in strong fields with Eqs. (1 – 3) deals with origin and consecutive account of the magnetic field dependence of the localization radius, which sets in already far from the MIT, when λ becomes comparable with $a_0(0) \equiv a_0$, and is connected to the dependence of E_A on B [10]. Introducing the field $B_0 \equiv \hbar/(ea_0^2)$ with the condition of $\lambda(B_0) = a_0$, this dependence can be written as $E_A(B) = E_A(0) \equiv E_A$ for $B << B_0$ and $E_A(B) = E_A \ln^2(B/B_0)$ for $B >> B_0$ [10]. In our case of $B_0 = 9.5$ T the interval of high fields (see below) corresponds to $B \sim B_0$ or $\lambda \sim a_0$, where $E_A(B)$ cannot be obtained analytically [10]. On the other hand, this function can be approximated well with the relation $E_A(B) = \beta B^{1/3}$, where β is independent of B [10]. Although an analytical expression for β cannot be found, it is reasonable to suppose that β is anisotropic in *p*-CdSb. The dependence $E_A(B)$ can be found approximately from the plots of $\ln \rho(B, T)$ vs. T^{-1} in the temperature interval of the acceptor freeze-out (examples are shown for # 1 in Fig. 3). Then β is obtained from the plots of E_A vs. $B^{1/3}$ which are close to linear between $B = 10 - 15$ T (see the inset to Fig. 3) yielding the values collected in Table I and supporting the expected anisotropy of β. Taking into account the magnetic field dependence of $E_A(B)$ above and using the relation $a_0 \sim E_A^{-1/2}$ [10] one can find $\varphi(B) = \varphi_0(B)$, where $\varphi_0(B) \equiv a_0 / a_0(B)$ is given by the expression

$$\varphi_0(B) = (\beta B^{1/3} / E_A)^{1/2}. \quad (4)$$

On the other hand, as has been mentioned above the acceptor system in all our samples is close to the MIT. The sensitivity of MIT to B brings an additional dependence of a on B, because shrinking of the impurity wave functions by magnetic field diminishes their overlap taking the system away from the MIT at $B = 0$. This process decreases a when B is increased.

To take into account the dependence of MIT on B we treat the acceptor band in p-CdSb:Ni as the Anderson band characterized by two mobility edges, E_c and E_c', separating the energy interval of the delocalized states (E_c, E_c') and the intervals of the localized states outside it (i. e. close to the DOS tails) [11]. If E_a and E_a' are the bottom and the top of this band, respectively, and the energy is measured from E_a towards the valence band, then $E_c' > E_c$, $E_a' > E_a$ and $E_c' < \mu < E_a'$, where μ is the Fermi energy. Such position of μ corresponds to the insulating side of MIT and to the small degree of compensation, $K = N_D / N_A$ (where N_D is the concentration of compensating donors) in our samples [7]. In such case the relation $E_a' - E_c' \approx V_0^2 / (4PI)$ is fulfilled, where V_0 and P are constants describing the degree of disorder (the width of distribution of the hole potential energy) and the number of the nearest neighbors in the acceptor system, respectively, and $I = I(a_0, \gamma_0) = I_0(a_0, \gamma_0)\exp(\gamma_0)$ is the overlap integral [11]. Here $\gamma_0 \equiv R_A/a_0$, $R_A = (4\pi N_A/3)^{-1/3}$ is half of the mean distance between the acceptors and $I_0(\gamma_0, a_0) = (e/a_0)[(3/2)(1 + \gamma_0) + (1/6)\gamma_0^2]$ is the prefactor for hydrogenic impurity wave functions [11]. Near MIT the localization radius can be written as $a = a_0 (1 - E_c'/\mu)^{-\nu}$, where ν is the critical exponent [11]. Taking into account that $\nu = 1$ and the degree of compensation, K is small (μ lies close to E_a') [7] in the last equation we can put $\mu \approx E_a$ and obtain $a \approx a_0 \mu (4PI/V_0^2)$, which gives $a/a(B) \approx a_0(B)I(B)/[a_0 I(0)]$, where $I(B) \equiv I[a_0(B), \gamma_0(B)]$ and $\gamma_0(B) \equiv R_A/a_0(B)$. Finally we get

$$\varphi(B) \approx \frac{(3/2)(1+\gamma_0) + (1/6)\gamma_0^2}{(3/2)[1+\gamma_0(B)] + (1/6)[\gamma_0(B)]^2} \exp\{-\gamma_0[1-\varphi_0(B)]\}. \qquad (5)$$

As can be seen from Fig. 4 the plots of ln ρ vs. $[B\,\varphi(B)]^{1/2}$ exhibit linear behavior in the interval of B between ~ 4 – 15 T, and their slopes do not depend on temperature at $T \leq 4.2$ K in # 1 and # 2 and between 3 – 4.2 K in # 3. The values of the slopes $\eta^{(ex)}$ obtained from the plots in Fig. 4 are collected in Table I. They are compared with $\eta_j^{(cal)}$, calculated with Eq. (2) using the data of N_A, a and p_j in Table I, exhibiting deviations from them of ~ 10 %, 3 % and 5 %, respectively.

Hence, the behavior of MR in p-CdSb:Ni in the interval of B between ~ 4 – 15 T is governed by the NNH conductivity in the limit of strong fields, yielding a complete numerical agreement with predictions of the percolation theory of NNH [10], if the anisotropy of the acceptor states and explicit dependence of a on B are taken into account. It is worth mentioning that the good agreement between $\eta^{(ex)}$ and $\eta_j^{(cal)}$ above supports the values of the parameters N_A, a and p_j obtained from investigations of the resistivity in zero and weak magnetic fields [7].

At this point, the deviations of the plots in Fig. 4 from linearity with increasing of B are attributable to deviation of the function E_A (B) from the $B^{1/3}$ dependence expected for $B \sim B_0$, towards $\ln^2 B$ which sets in for $B >> B_0 \sim 10$ T [see text between Eqs. (3) and (4)], yielding weaker dependence of a (B). On the other hand, the onset of the temperature dependence of η_j in # 3 observed below 3 K (Fig. 4) cannot be attributed to transition to the VRH conductivity (as e. g. in undoped p-CdSb [6]), because for any VRH mechanism positive MR is predicted to strengthen when T is decreased [10, 12], that is the slopes in the bottom part of Fig. 4 are expected to increase with T, but not vise versa. Such behavior cannot be attributed to contribution of the negative MR, connected to damping of the interference of the direct paths in conventional non-magnetic semiconductors in the regime of the VRH conductivity [13, 14]. Indeed, the negative MR of such nature dominates in weak fields but saturates when B is increased [13, 14], whereas no signs of the negative contribution to MR in the weak-field limit have been observed [7]. Although one cannot exclude the appearance of a contribution of the negative MR in strong fields due to presence of the magnetic Ni-rich nanoclusters in p-CdSb:Ni [2]. A final conclusion at this point requires further investigations. Hence, only the NNH conductivity can be identified unambiguously in p-CdSb:Ni in strong magnetic fields.

Finally, it is instructive to compare the analysis above with its simplified variants to discuss their applicability. The first of them is the case, sometimes met in the literature, when the magnetic field dependence of a is neglected, giving φ (B) = 1 and the law $\ln [\rho (B) / \rho^*]_j = \eta_j B^{1/2}$ instead of Eq. (1). It can be shown that the plots of $\ln \rho$ vs. $B^{1/2}$ also contain linear parts approximately in the same field interval of ~ 4 – 15 T with the slopes $\eta^{(ex)}\big|_{\varphi=1}$ independent of T within some temperature

intervals, which are collected in Table I, demonstrating a reasonable agreement with $\eta_j^{(cal)}$ only for # 3. The second variant of simplification is to put $\varphi(B) = \varphi_0(B)$ given by Eq. (4), when the sensitivity of the MIT to B is neglected. It gives in a same way values of the slopes $\eta^{(ex)}\big|_{\varphi=\varphi_0}$ displayed in Table I, which are closer to $\eta_j^{(cal)}$ for # 1 and # 2 than in the previous case, but again an agreement is observed only for # 3. Hence, for two out of the three investigated samples it is important to take into account both effects leading to the dependence of a (B). On the other hand, in # 3 such dependence is weakest due to the smallest value of β (i. e. the weakest dependence of E_A on B – see Table I) and the largest difference of $N_C - N_A$ (the smallest proximity to the MIT – see Table I), where $N_C = 6.28\times10^{16}$ cm^{-3} [7], which makes inclusion of a (B) into the analysis unimportant.

IV. CONCLUSIONS

We have investigated the resistivity and magnetoresistance of p-CdSb with 2 at % of Ni between $T = 1.5$ and 300 K in pulsed magnetic fields up to $B = 30$ T. It is shown that below 4.2 K in the fields between ~ 4 – 15 T the resistivity is governed by the hopping charge transfer over the nearest acceptor sites in conditions of strong magnetic field. A detailed analysis of the magnetoresistance using the microscopic data obtained from investigations of the resistivity of p-CdSb:Ni in zero and weak fields [7], demonstrates numerical agreement of the results with predictions of the percolation theory of the nearest-neighbor hopping conductivity in strong magnetic field [10], if the anisotropy of the acceptor states, as well as the explicit dependence of the localization radius on the magnetic field, are taken into account.

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TABLE I. The values of the parameters N_A, a, p_j and E_A determined from analysis of the resistivity in zero and weak field [7], constant (β) of the high-field dependence of E_A on B, the experimental ($\eta^{(ex)}\big|_{\varphi=1}$, $\eta^{(ex)}\big|_{\varphi=\varphi_0}$ and $\eta^{(ex)}$) and calculated ($\eta_j^{(cal)}$) coefficients of the high-field dependence of MR at different orientations of the magnetic field (j).

Sample No.	N_A (10^{16} cm^{-3})	a (Å)	p_j	E_A (meV)	β (meV T$^{-1/3}$)	$\eta^{(ex)}\big\|_{\varphi=1}$ (T$^{-1/2}$)	$\eta^{(ex)}\big\|_{\varphi=\varphi_0}$ (T$^{-1/2}$)	$\eta^{(ex)}$ (T$^{-1/2}$)	$\eta_j^{(cal)}$ (T$^{-1/2}$)	j
1	3.61	196	0.839	2.45	1.64	2.11	1.65	1.36	1.23	3
2	3.37	180	1.008	2.50	1.53	2.09	1.69	1.50	1.46	1
3	2.51	139	1.182	2.85	1.19	2.20	1.96	2.00	2.09	2

Figure captions

Figure 1. Temperature dependence of the resistivity of p-CdSb:Ni at $B = 0$. Inset: plots of ln ρ vs. T^{-1} at $B = 0$. For convenience the data for # 2 and # 3 are shifted by + 0.5 and + 2.5 units along the vertical axis. The straight lines are linear fits.

Figure 2. The dependence of the resistivity of p-CdSb:Ni on magnetic field.

Figure 3. The dependence of ln ρ on T^{-1} for # 1 at B = 11 T, 13 T and 15 T (from down to up). Inset: plots of E_A vs. $B^{1/3}$. The dotted lines are guides for the eye and the solid lines are linear fits.

Figure 4. Plots of ln ρ vs. $(B\,\varphi)^{1/2}$. The lines are linear fits.

Fig. 1

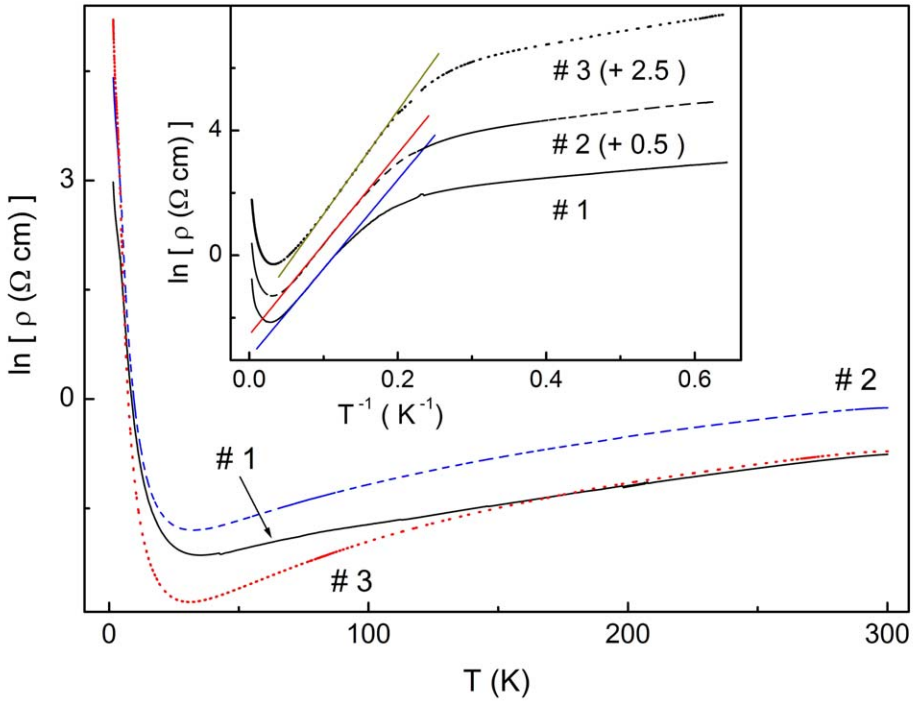

Fig. 2

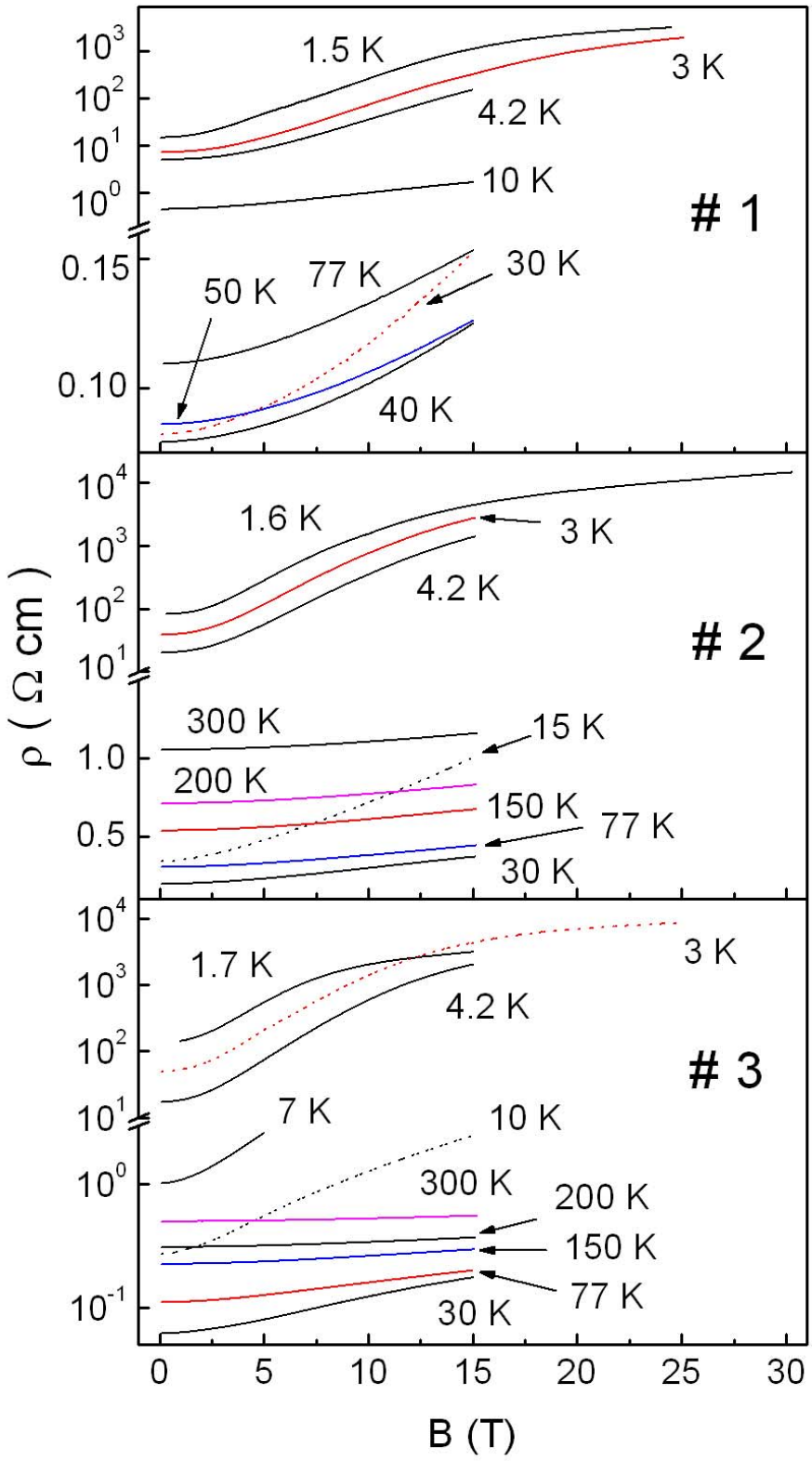

Fig. 3

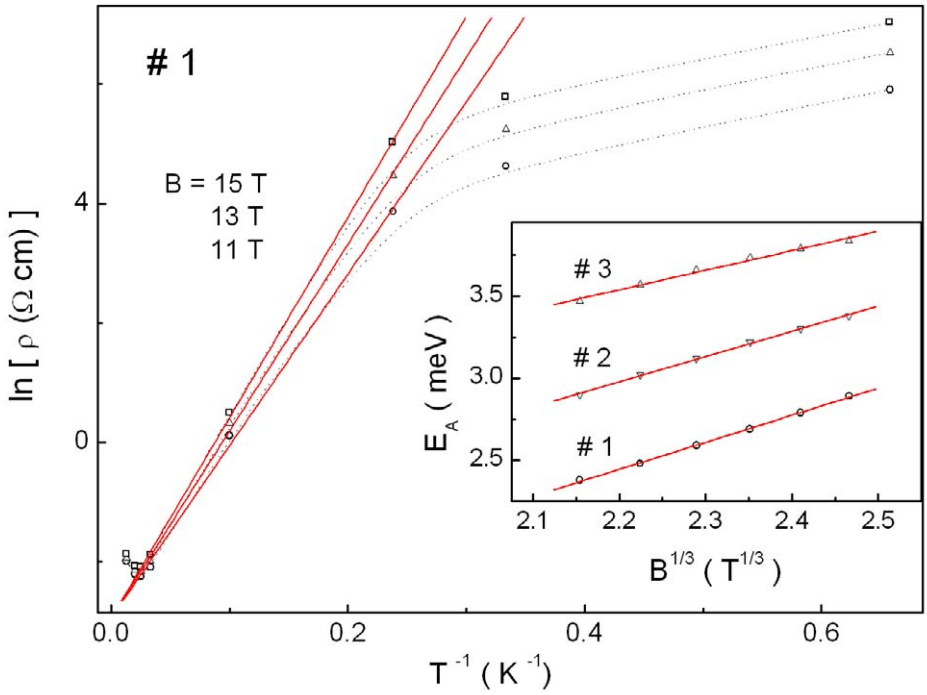

Fig. 4