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GIANT NEGATIVE MAGNETORESISTANCE IN A NONMAGNETIC SEMICONDUCTOR

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Studies of a classical A^3B^5 semiconductor (InSb) doped with $3d$ magnetic ions (Mn^{2+}, having a localized spin $S = 5/2$) reveal some unexpected transport properties. It is found that the transition from the metallic to the low-temperature insulator phase occurs at an impurity concentration $N_{Mn} \sim N_{cr} = 2 \cdot 10^{17}$ cm^{-3} and a temperature $T < T_{cr} \sim$ $\sim$ 1 K. Under these conditions a giant negative magnetoresistance arises. The experimental results can be explained in terms of the onset of a hard Mott – Hubbard gap Δ in the impurity band formed by the shallow manganese acceptor in InSb at $N_{Mn} \sim N_{cr}$. A model describing the gap formation is proposed.

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Colossal magnetoresistance and metal-insulator transitions in solid-state materials have recently come to the close attention of physicists. Such effects are typical for impurity manganites [1], degenerate magnetic oxide semiconductors [2], and the spin-Peierls magnetic insulator $CuGeO_3$ [3]. For all the above-mentioned materials the influence of the exchange interaction upon the crystal band structure has been found to be of great importance. The aim of the present paper is to demonstrate that similar exchange interaction effects can also be manifested in some classical nonmagnetic semiconductors doped with transition elements (in our case InSb doped with the $3d$ magnetic ions Mn^{2+}).

Our experiments were carried out on p-InSb(Mn) single crystals grown by the Czochralski method. A crystal 4 cm long and 2.5 cm in diameter had a small manganese concentration gradient from $N_{Mn} = 10^{17}$ cm^{-3} to $3 \cdot 10^{17}$ cm^{-3} along the length of the crystal. Hall bars with dimensions of $(7 \times 1 \times 1)$ mm were cut from the crystal transversely to its length. This approach made it possible to study the manganese concentration dependence of the dc conductivity, magnetoresistance, and Hall effect at manganese concentrations close to N_{cr}. Temperature control was effected by means of Lake Shore Cryotronics GaAs diodes and thermistors together with He^4 and He^3 pumped pressure measurements. Magnetic fields of up to 7 T were induced by a superconducting magnet.

The results of the measurements (Figs.1a and 2) demonstrate an exponential temperature dependence of the resistivity for all of the p-InSb(Mn) samples (except for the sample with the maximum manganese concentration (Fig.2b)) at temperatures $T < 1$ K in the absence of magnetic field:

$$\rho = \rho_0 \cdot \exp(\Delta/kT), \tag{1}$$

where Δ is a temperature-independent parameter which is a function of the manganese concentration and reaches its maximum value ($\Delta \simeq 0.6$ meV) at $N_{Mn} \sim 1.5 \cdot 10^{17}$ cm^{-3}.

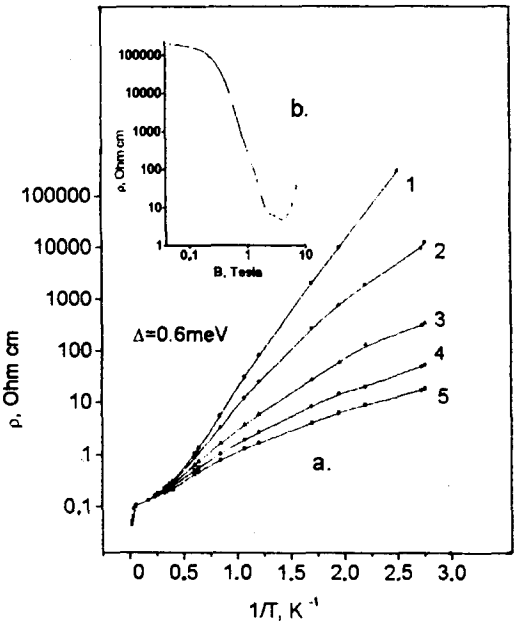

Fig.1. a) Temperature dependence of the resistivity ρ of p-InSb(Mn) for various magnetic fields: *1* - $H = 0$ ($x = 0$); *2* - $H = 0.5$ T ($x = 0.85$); *3* - $H = 1.5$ T ($x = 0.75$); *4* - $H = 0.5$ T ($x = 0.33$). b) $\rho-H$-dependence at $T = 0.4$ K. $N_{Mn} = 1.5 \cdot 10^{17}$ cm^{-3}

The $\rho-T$-dependence at $N_{Mn} < 10^{17}$ cm^{-3} and $N_{Mn} > 3 \cdot 10^{17}$ cm^{-3} can be described by the variable-range hopping (VRH) conductivity equation [4]

$$\rho = \rho_0 \cdot \exp(T_0/T^x), \tag{2}$$

where $x < 1$, and T_0 is a temperature-independent parameter. In a magnetic field $H < 3$ T and at $T < 4.2$ K the resistivity decreases throughout the whole range of manganese concentration (Figs.1, 2). Investigation of the $\rho-T$-dependence in magnetic fields shows that increasing the magnetic field brings forth a transition from the type of conductivity described by Eq.(1) to one described by Eq.(2). Thus colossal negative magnetoresistance (ρ/ρ_0) should be expected in the millikelvin temperature range if it assumed that the value of x (in Eq.(2)) decreases twofold in a magnetic field $H \sim 2$ T. It should be pointed out that the $\rho-H$-dependence has a "shoulder" that increases as the temperature goes down (Fig.1b). In magnetic fields $H > 3$ T the resistance increases, as is typical for the VRH conductivity in semiconductors.

In a weak magnetic field the Hall constant R_H decreases with decreasing temperature, changes sign (Fig.3a), and increases exponentially at $T < 1$ K (Fig.3b). The R_H-T-dependence is described by

$$-R_H = -R_0 \exp(\Delta/kT), \tag{3}$$

where the value of Δ is the same as that of the parameter Δ obtained in the $\rho-T$-measurements (from Eq.(1)). The high-field Hall constant ($H > 3$ T) increases slightly, remaining positive as the temperature decreases.

We can understand the experimental results if it is assumed that the magnetic-impurity band in InSb(Mn) splits into two bands separated by a Mott - Hubbard gap Δ which is about 10% of the binding energy of the hole on a manganese acceptor [5]. The upper

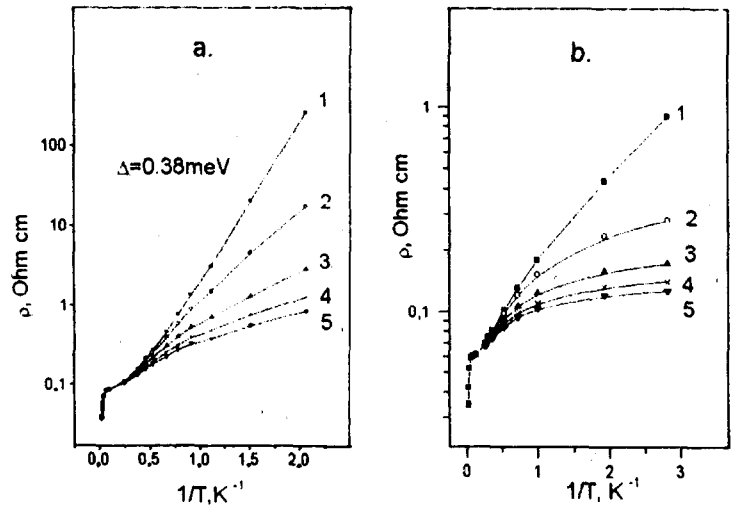

Fig. 2. Temperature dependence of the resistivity ρ of p-InSb(Mn) for various magnetic fields. a) $N_{Mn} = 1.24 \cdot 10^{17}$ cm^{-3}; *1* – $H = 0$ ($x = 1$); *2* – $H = 0.5$ T ($x = 0.75$); *3* – $H = 1.0$ T ($x = 0.5$); *4* – $H = 1.5$ T ($x = 0.33$); *5* – $H = 2.0$ T ($x = 0.25$). b) $N_{Mn} = 2.4 \cdot 10^{17}$ cm^{-3}; *1* – $H = 0$ ($x = 0.75$); *2* – $H = 0.5$ T ($x = 0.1$); *3* – $H = 1.0$ T ($x = 0.01$); *4* – $H = 1.5$ T; *5* – $H = 2.0$ T

band is the electron band (negative sign of the Hall constant) and the lower band is the hole band (positive Hall constant).

The proposed model is based on a dual role of the Mn^{2+} ions.

1. Mn^{2+} ions are shallow acceptor centers ($E_a = 7$ meV), giving rise to free holes in the valence band and to the formation of a narrow impurity band. These Mn^{2+} acceptor centers have a substantial qualitative influece on the characteristics of InSb, causing effects similar to those due to nonmagnetic acceptor centers such as Ge. In InSb(Ge) single crystals free holes having uncompensated spins appear in the valence and impurity bands [6].

We suppose that the exchange interaction between hole spins cause bounded regions with antiferromagnetically correlated spins to appear but that uniform long-range magnetic order is not formed because of the random distribution of the acceptor centers. Such a state can be characterized as a dynamic state with spin fluctuations caused by the production and decay of local antiferromagnetically correlated states. In this situation, splitting of the impurity band takes place only in bounded local regions.

2. In contrast to crystals doped with nonmagnetic impurities, in p-InSb doped with Mn^{2+} ions a magnetic system of localized spins arises. Thus an additional magnetic subsystem appears, and an exchange interaction arises between the localized Mn^{2+} ion spins ($\mathbf{S}^d$) and free-hole spins ($\mathbf{S}^p$). A uniform antiferromagnetic long-range order of the Mn^{2+} ion spins can arise due to the effective exchange interaction between $3d$ spins via spin fluctuations in the subsystem of free-hole spins – order from disorder [7].

Let us write the spin Hamiltonian in the form

$$\mathcal{H} = \sum \mathbf{J}_{ij}^{pp} \mathbf{S}_i^p \mathbf{S}_j^p + \sum \mathbf{J}_{ij}^{pd} \mathbf{S}_i^p \mathbf{S}_j^d, \tag{4}$$

where $\mathbf{J}_{ij}^{pp}$ is the exchange interaction constant taken for free-hole spins, $\mathbf{J}_{ij}^{pd}$ is the exchange constant for the interaction between the spins $\mathbf{S}^d$ and $\mathbf{S}^p$, and i, j are the ion

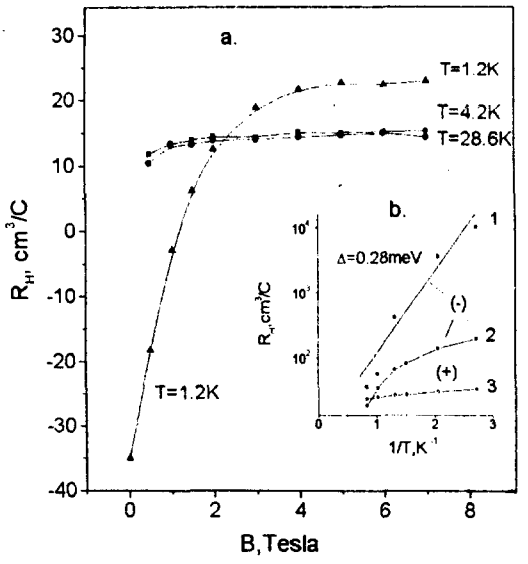

Fig.3. Hall constant R_H as a function of magnetic field (a) and temperature (b). $N_{\mathrm{Mn}} = 2.0 \cdot 10^{17}$ cm^{-3}

positions in the lattice. We assume that $\mathbf{J}_{ij}^{pp} \gg \mathbf{J}_{ij}^{pd}$. As the concentration of $3d$ ions is very small, the direct exchange interaction between the $3d$ spins can be neglected. In the framework of perturbation theory up to second order inclusive, the effective Hamiltonian for the $3d$ ion spins can be written as

$$\mathcal{H}_{\mathrm{eff}}^{d} = \sum \mathbf{J}_{ij}^{pd} \langle \mathbf{S}_j^p \rangle \mathbf{S}_i^d + \sum (\mathbf{J}_{ij}^{pd})^2 / J_{ij}^{pp} \cdot K_{ij}^{pp} \cdot \mathbf{S}_i^d \mathbf{S}_j^d, \tag{5}$$

where $\langle \mathbf{S}_j^p \rangle$ is the spin averaged over configurations of free holes, and $K_{ij}^{pp} = \langle S_i^p S_j^p \rangle - -\langle S_i^p \rangle \langle S_j^p \rangle$ is the spin correlation function of the holes. The first sum in the effective Hamiltonian in Eq.(5) describes the magnetization of the Mn^{2+} ion spins by the mean field $\mathbf{H}_{mf}^{d} = \sum \mathbf{J}_{ij}^{pd} \langle \mathbf{S}_j^p \rangle$. The value differs from zero if uniform long-range magnetic order exists in the free-hole spin subsystem.

As we have said, in the hole spin subsystem there is correlation only at finite sizes, and we have only the second sum in Eq.(5). This sum describes the effective exchange interaction of the spins $\mathbf{S}^d$ through correlations between hole spins. Thus the effective exchange interaction $V_{ij}^{dd} = \sum (\mathbf{J}_{ij}^{pd})^2 / J_{ij}^{pp} \cdot K_{ij}^{pp}$ can bring about the uniform antiferromagnetic long-range ordering of the $\mathbf{S}^d$ spins at a suitable ordering temperature. The internal molecular field $H_{MF} = \sum V_{ij}^{dd} \langle S_j^d \rangle$ arises and splits the impurity band into two bands with spins up and spins down.

Thus a hard gap of an exchange nature appears in the impurity band (see Fig.4b). To put it in another way, a supernarrow semiconductor appears in the band gap of InSb. This hard gap can be closed in an external magnetic field H_0 due to the antiferromagnetic–ferromagnetic (spin-flip) transition when $H_0 \simeq H_{MF}$. And indeed in our case the energy Δ agrees with the value $g\mu_B H_0 \simeq 0.4$ meV at the value of the magnetic field $H_0 \sim 3$ T that provides the gap-closing effect (spin-flip effect).

The effective exchange interaction length for V_{ij}^{dd} is determined by the correlation length ξ_p of the $\mathbf{S}^p$ spin fluctuations. On the assumption that $\xi_p \simeq \eta_p$ (η_p is the mean free path of the holes), the V_{ij}^{dd} interaction is of the long-range type.

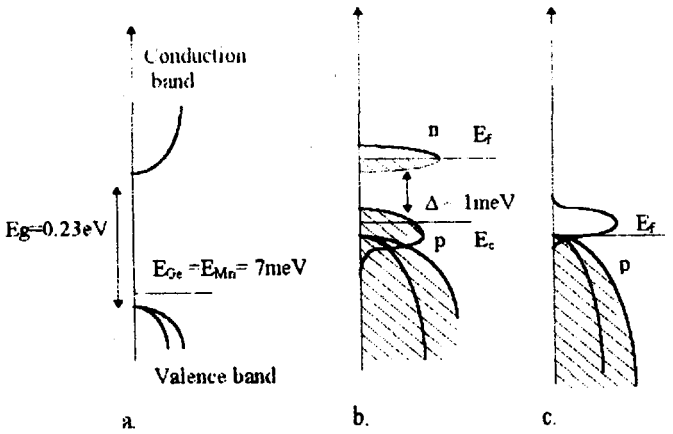

Fig.4. Band structure of p-InSb(Mn). a) $N_{\rm Mn} = N_a \ll N_{cr}$; b) $N_a \simeq N_{cr}$; c) $N_a > N_{cr}$

It is quite clear that uniform long-range ordering of the $\mathbf{S}^d$ spins can exist only at concentrations of the Mn^{2+} ions corresponding to a mean distance $\langle r \rangle \leq (\xi_p \simeq \eta_p)$. The equality of ξ_p and η_p correlates with a critical concentration N_{cr} and a critical temperature T_{cr} corresponding to simultaneous onset of the long-range magnetic order and the gap in the impurity band (metal–insulator transition).

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