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Magnetotransport studies of Ga(Mn,Fe)N bulk crystals

C. Jastrzebski[*, 1], **W. Gebicki**[1], **M. Zdrojek**[1], **M. Bockowski**[2], **B. Strojek**[3], **T. Szyszko**[3], **M. Kaminski**[3], and **S. Podsiadlo**[3]

[1] Faculty of Physics, Warsaw University of Technology, Koszykowa 75, 00-662 Warsaw, Poland
[2] High Pressure Research Center, Polish Academy of Science, Sokolowska 29/37, 01-142 Warsaw, Poland
[3] Faculty of Chemistry, Warsaw University of Technology, Noakowskiego 3, 00-664 Warsaw, Poland

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Magnetoresistance and Hall effect in (Ga,Mn)N, (Ga,Mn,Si)N, and (Ga,Fe)N bulk crystals has been studied in the temperature range from 4.2 K to 300 K. No anomalous Hall effect contribution to the Hall voltage vs. magnetic curve has been found. The negative magnetoresistance has been found only in samples with metallic type of conductivity and is interpreted as resulting from weak localization of current carriers.

1 Introduction

The ferromagnetism in III-V semiconductors heavily doped with transition elements like Mn, Fe, Ni, Co, Cr has been studied since the preliminary reports on ferromagnetism in GaAs:Mn appeared in 1999. Dietl *at al.* [1] predicted on the basis of Zener model of ferromagnetism that some wide band semiconductors like GaN and ZnO heavily doped with manganese are ferromagnets with Curie temperature T_c above room temperature. This paper drove attention of many research teams and stimulated them to verify experimentally the results of Dietl's calculations.

In the paper [1] sine qua non conditions of growth of ferromagnetic material with $T_c > 300$ K have been formulated. The necessary concentration of manganese in GaN has been estimated about 5% with the concentration of holes above $3x10^{20}$ cm^{-3}. Both conditions are difficult to fulfill experimentally. It has been shown that high concentration of manganese could be achieved either by nitridization of pure metallic Ga in supercritical ammonia (bulk crystals) or low temperature MBE growth (thin films). Little is known about the solubility of the other transition metal ions in GaN. Probably the solubility of Fe as well as the other transition elements is much lower than the solubility of Mn and it doesn't exceed 0.5% [2, 3]. That high concentration of Mn ions in the crystals rises questions concerning formation of paramagnetic ions clusters and ferromagnetic inclusions. Dietl model assumed that ferromagnetism in doped III–V semiconductors comes from isolated paramagnetic ions with the exchange interactions transmitted through polarized spins of free current carriers. For the concentration of manganese ions exceeding 5% it is a challenging task to avoid clustering of the manganese ions in GaMnN crystals. In consequence inclusions as a factor responsible for ferromagnetic contribution to the magnetization have been intensively studied but the results are contradictory [3, 4].

The concentration of holes in GaN:Mn system needed to achieve ferromagnetic material with Tc exceeding room temperature is difficult to realize. Bulk crystals are typically n-type with high concentration of electrons (10^{19}/cm^{-3}). Uncontrolled defects like oxygen ions forming shallow donors are difficult

[*] Corresponding author: e-mail: cez_j@if.pw.edu.pl, Phone: +48 22 660 5181, Fax: +48 22 660 5447

to eliminate in bulk crystals technology. Many efforts have been done to compensate and convert the material to p-type through doping it intentionally with shallow acceptors ions. Unfortunately the ionization energy of typical acceptors in GaN is relatively high (ionization energy of Mg acceptor is about 200 meV). This factor makes difficult to achieve the desired concentration of holes. It has been shown in some recent publications that the existence of ferromagnetism in transition elements doped bulk crystals is possible also in n-type materials and compensated materials with more than 5% of manganese the low carrier concentration. The mechanism of ferromagnetism in such materials is not clear [2, 5].

The reports on ferromagnetism in GaN heavily doped with transition elements are contradictory. The possibility of growth GaN:Mn system with more than 5% of manganese has been verified experimentally both in bulk crystals and in MBE grown thin films. Early magnetization measurements showed a paramagnetic behavior of GaN:Mn bulk crystals but in the later papers a ferromagnetic type magnetization dependence on magnetic field has been reported. Several different T_c temperatures have been proposed [6, 7].

Transport measurements and particularly magnetoconductivity and Hall effect are useful tools of magnetic materials characterization and may be helpful in to conclude the controversy. Negative magnetoresistance is a sine qua non condition of ferromagnetism but it is also observed in strongly compensated, paramagnetic semiconductors due to the weak localization and s–d interactions [7, 8]. The coincidence of negative magnetoresistance with anomalous Hall effect is considered a hard test for bulk ferromagnetism.

2 Experimental

(Ga,Mn)N and (Ga,Fe)N bulk crystals have been grown by nitridization of pure metallic Ga in supercritical ammonia method described elsewhere [9]. To increase the concentration of free carriers samples (Ga,Mn,Si)N crystals have been used, because it is possible to (Ga,Mn,Si)N samples show higher concentration of Mn than (Ga,Mn)N crystals. The crystals were rather small and formed irregular plates or needles. Indium contacts were soldered to the samples to make the classic Hall geometry where possible or the four probe geometry to study only the magetoconductivity. The ohmicity of all the contacts has been carefully verified. Hall effect and longitudinal magnetoresistance have been measured at temperatures between 4.5 K and 300 K in magnetic field varying in the range ±7 T. Oxford Instruments Spectromag 4000 cryostat and standard Kithley DC Hall effect setup has been used. Because the crystals used for the measurements were quite irregular the Hall voltage and longitudinal voltage drop due to the sample magnetoresistance have been separated numerically. All the measured samples exhibit n-type conductivity.

3 Results and discussion

The results of the magnetoresistance measurements are presented at Fig. 1(a,b), (Ga,Mn,Si)N and (Ga,Fe)N respectively, and are correlated with Hall effect measurements (Fig. 2 and Fig. 3). The clear difference between the two systems (positive magetoresistance of (Ga,Mn,Si)N crystals and negative magneoresistance of (Ga,Fe)N system) is seen. At Fig. 2a and Fig. 2b Hall voltage vs. magnetic field and temperature dependence of free electron concentration vs. temperature in (Ga,Fe)N system are presented. The dependence of Hall effect on magnetic field and the electron concentration vs. temperature in (Ga,Mn,Si)N system are presented at Fig. 3a and 3b respectively. For both compounds no correction of the Hall voltage vs. magnetic field curve for anomalous Hall effect has been seen (Fig. 2a and Fig. 3a). Considerable difference between the temperature dependence of electron concentration in (Ga,Fe)N and (Ga,Mn,Si)N is observed. The relatively high concentration of electrons (~$6x10^{19}/cm^3$) weakly depends on temperature exhibiting metallic conduction above Mott transition. The lower concentration of carriers (~$10^{19}/cm^3$) in (Ga,Mn,Si)N depends exponentially on temperature. An important coincidence between the results of magnetoresistance and Hall effect measurements has been observed. The negative magnetoresistance coincides with the metallic conductivity of the material. The positive type of magnetoresistance coincides with temperature dependent type of conductivity characteristic for nondegenarate semiconductors. This coincidence has been observed in all the examined (Ga,Mn)N, (Ga,Mn,Si)N, and (Ga,Fe)N bulk crystals.

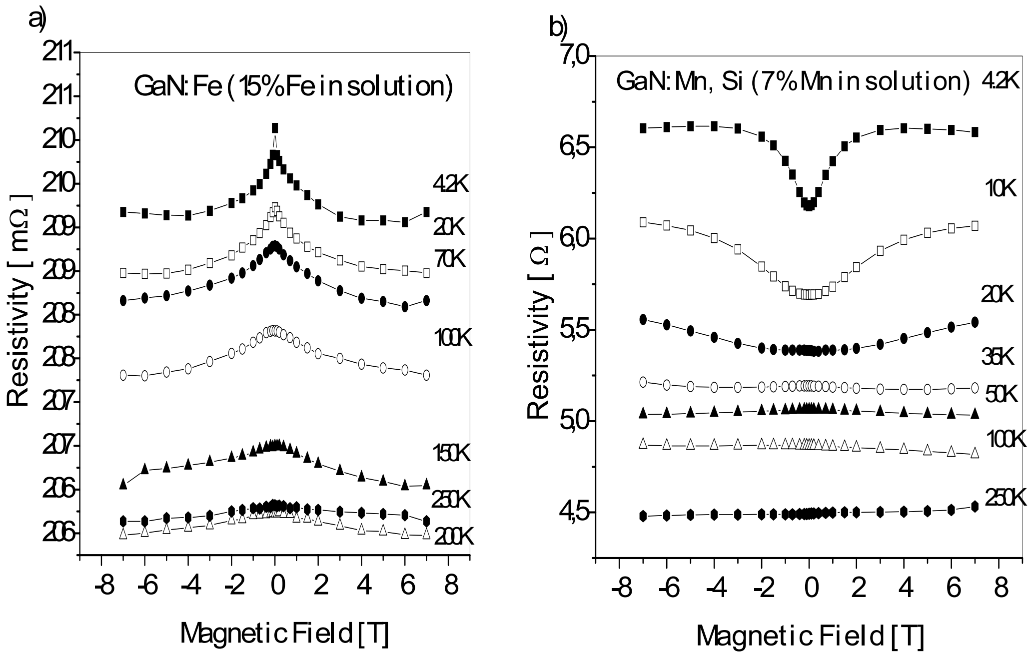

Fig. 1 Temperature dependence of magetoresistance in (Ga,Fe)N (a) and in GaMnSiN (b) system.

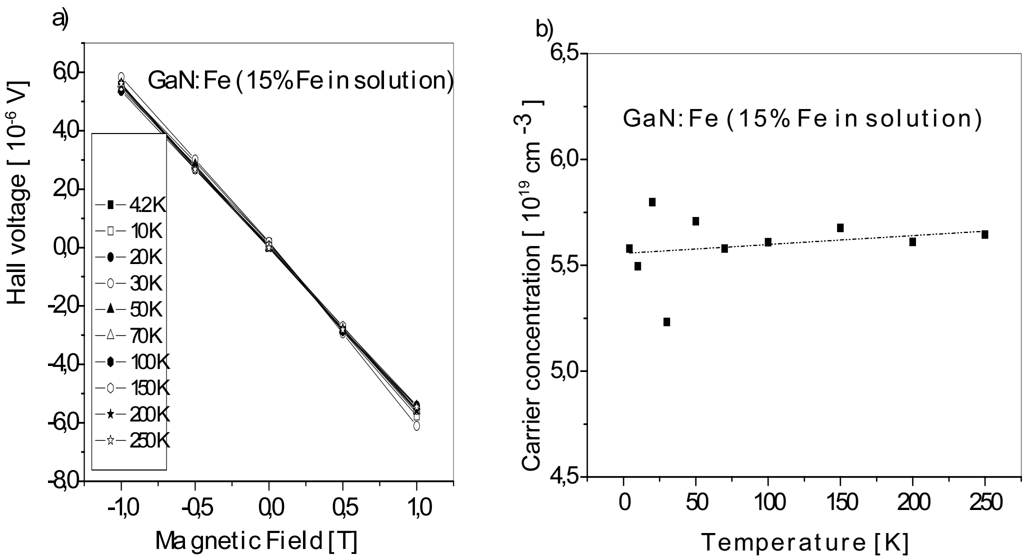

Fig. 2 Hall effect vs. magnetic field at various temperatures (a) and temperature dependence of electron concentration (b) in (Ga,Fe)N bulk crystals.

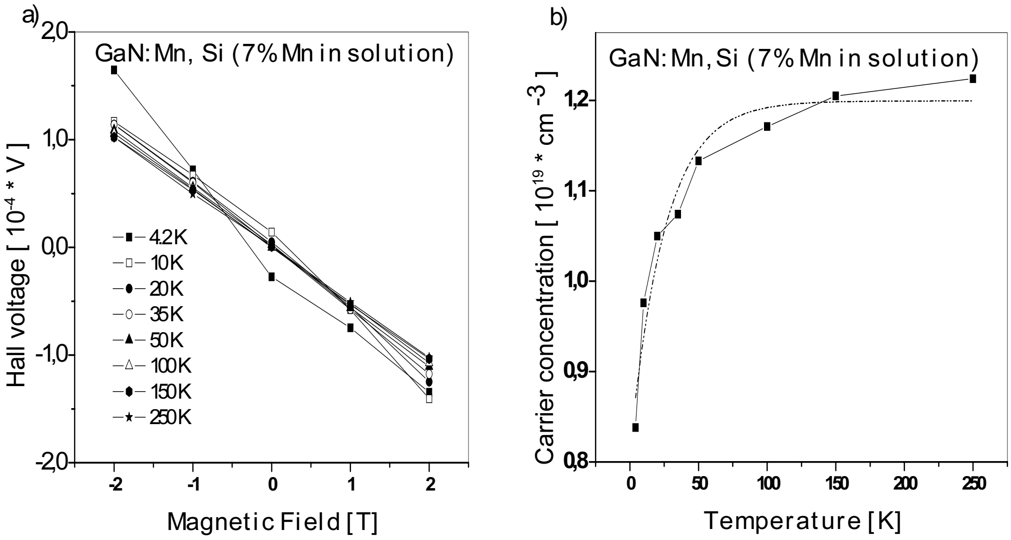

Fig. 3 Hall effect vs. magnetic field at various temperatures (a) and temperature dependence of electron concentration (b) in (Ga,Mn,Si)N bulk crystals.

This result strongly suggests that the negative magnetoresistance of (Ga,Fe)N crystal is related to the weak localization of carriers and to the quantum corrections to the magnetoresistance [10, 11], not to the ferromagnetic ordering of the magnetic moments of paramagnetic ions.

We conclude the paper as follows:

In all the (Ga,Mn,Si)N, (Ga,Mn)N and (Ga,Fe)N samples no anomalous Hall effect has been found in spite of relatively high concentration paramagnetic ions (Mn or Fe) and high concentration of free electrons. The negative magnetoresistance is related to the metallic type of conductivity and is not related to the ferromagnetic ordering of magnetic moments. Probably the results can be explained within the model taking into account weak localization and s–d interaction of free electrons.

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