Photoluminescence and Electron Paramagnetic Resonance studies of bulk GaN doped with gadolinium

Z. Lipińska[1], A. Wysmołek[1], M. Pawłowski[2], M. Palczewska[2], M. Kamińska[1], A. Twardowski[1], M. Boćkowski[3], I. Grzegory[3]
[1] *Institute of Experimental Physics, Warsaw University, Hoża 69, 00-681 Warsaw, Poland*
[2] *Institute of Electronic Materials Technology, Wolczynska 133, 01-919 Warsaw, Poland*
[3] *Institute of High Pressure Physics, Polish Academy of Sciences, Warsaw, Poland*
e-mail: Andrzej Wysmolek@fuw.edu.pl

Theoretical predictions of possible room temperature ferromagnetism in GaN diluted with manganese strongly motivated studies of nitrides based diluted magnetic semiconductors (DMS) with transition metals or rare-earths. Over the last years, above room temperature ferromagnetism for MBE grown GaN layers weakly doped with Gd has been reported by different researchers [for example see ref.1]. The estimated average value of magnetic moment per Gd ion has been obtained as high as 500 times that of Gd atomic value. It has been argued that the observed enhancement of the average magnetic moment resulted from strong contribution of GaN host lattice to macroscopic magnetization of the whole material [1]. The origin of such behavior remains still puzzling and more studies of GaN:Gd are required to clarify this issue.

In this paper we present photoluminescence (PL) and electron paramagnetic resonance (EPR) experiments on strain free GaN bulk crystals of wurtzite structure doped with gadolinium. The samples were grown from the solution of nitrogen in liquid gallium under high (1.5GPa) pressure (HP) of N_2 and at elevated temperatures of about 1500^0C. Doping with Gd was obtained by adding this element to the solution. Both PL and EPR techniques are well known as a powerful experimental tools to study electronic and magnetic properties of transition metals and rare-earth in semiconductors, and they have not been exploited up to now for GaN:Gd system.

In EPR experiment sharp absorption lines were detected, which could be attributed to $Gd^{3+}(4f^7)$ state. Identification of the lines was possible by comparison with other semiconductors doped with Gd. The observed Dysonian shape of the lines originated from presence of high concentration of free electrons in measured GaN. However, the estimated value of free electron concentration was definitively lower than in undoped HP GaN, what indicated purification role of Gd from unintentional donors (presumably oxygen).

PL studies revealed intensive spectra in two energy range: close to GaN band gap (3.1-3.6 eV) and in the "red" range of 1.6-1.8 eV. Band-to band transitions, typical for GaN doped with free electron concentration at a high 10^{18}cm^{-3} level, were clearly seen. They were followed by several sharp peaks in direction of lower energy. In the "red" range sharp structure with main line at 1.77eV energy was detected. The FWHM of this line was as small as 5meV. This structure was related to $Gd^{3+}(4f^7)$ state.

Judging from EPR and PL results Gd dopant was present in the studied samples. However, no ferromagnetic behavior of the samples was observed by means of EPR. This creates some doubts if ferromagnetism observed by others was really related to GaGdN system.

[1] S. Dhar, O. Brandt, M. Ramsteiner, V. F. Sapega, and K. H. Ploog, Phys. Rev. Lett. 94, 037205 (2005)