Temperature evolution study of free hole concentration in (Ga,Mn)As

N. Tataryn[1,2], O. Yastrubchak[2], R. Kuna[3], L. Gluba[4,5], L. Borkovska[2], O. Kolomys[2], J.Z. Domagała[3], T.Wosinski[3], J. Żuk[5], M. Sawicki[3] and J. Sadowski[3,6,7]

[1]National Technical University of Ukraine "Igor Sikorsky Kyiv Polytechnic Institute", Kyiv 03056, Ukraine,
[2]V. Lashkaryov Institute of Semiconductor Physics of National Academy of Sciences of Ukraine, 41, Pr. Nauky, Kyiv, 03028, Ukraine
[3] Institute of Physics, Polish Academy of Sciences, Aleja Lotników 32/46, PL-02668 Warsaw
[4]Institute of Agrophysics, Polish Academy of Sciences, Doświadczalna 4, 20-290 Lublin
[5] Institute of Physics, Maria Curie-Skłodowska University in Lublin, Pl. M. Curie-Skłodowskiej 1, 20-031 Lublin, Poland
[6]MAX-IV laboratory, Lund University, P.O. Box.118, 22100 Lund, Sweden
[7]Department of Physics and Electrical Engineering, Linnaeus University, SE-391 82 Kalmar, Sweden

The high-spectral-resolution optical studies of the energy gap evolution, supplemented with electronic, magnetic and structural characterization, have shown that the modification of the GaAs valence band caused by Mn incorporation occurs already for a very low Mn content, much lower than that required to support ferromagnetic spin–spin coupling in (Ga,Mn)As [1]. The combined low-temperature magnetic and optical studies indicate that the paramagnetic↔ferromagnetic transformation in *p*-type (Ga,Mn)As takes place without imposing changes of the unitary character of the valence band with the Fermi level located therein. The whole process is rooted in the nanoscale fluctuations of the local (hole) density of states and the formation of a superparamagnetic-like state.

In the present study the temperature evolution of free hole concentration in (Ga,Mn)As have been investigated the with Raman scattering spectroscopy through the analysis of hole-plasmon related mode in the spectra. A set of 100 nm thick (Ga,Mn)As layers has been prepared by the low-temperature molecular-beam epitaxy (MBE) growth technique, with the Mn contents *x* ranging from 0 (LT-GaAs reference layer) to 1.6%, at approximately 230°C on GaAs (001) semi-insulating substrates. Micro-Raman measurements were performed using 532 nm, 514 nm and 488 nm laser lines in the backscattering configuration in a wide range of temperatures (4-300K) with the resolution of 1 cm^{-1}.

In *p*-type GaAs, as well as in (Ga,Mn)As, longitudinal-optical (LO) phonon mode couples with the hole-gas-related plasmon forming so-called coupled plasmon–LO phonon mode (CPPM). Under the low-temperature MBE growth conditions an excess of arsenic builds into GaAs, mainly in the form of arsenic antisites, As_{Ga}, which act as deep double donors and lead to an *n*-type hopping conductivity in LT-GaAs and ultra-low Mn doped (Ga,Mn)As. In the case of the present set of samples about 0.3% of Mn is needed to form the CPPM band in the Raman spectra, indicating that (Ga,Mn)As turns to *p*-type at around this concentration of Mn. With further increase in *x* the CPPM mode starts to dominate the spectra, simultaneously with a shift of its energy towards the TO-phonon-line wavenumber. This is the direct indication of an increasing hole density with *x*, which depends on temperature and Mn concentration and can be quantified from the full line-shape fitting.

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[1] L. Gluba, O. Yastrubchak, J. Z. Domagala, R. Jakiela, T. Andrearczyk, J. Żuk, T. Wosinski, J. Sadowski, and M. Sawicki, *Phys. Rev. B* (accepted 16 Feb. 2018).