Signatures of exciton spin relaxation in photoluminescence spectra of semimagnetic quantum dots

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In this report, we present experimental and theoretical investigations of photoluminescence (PL) spectra of $Cd_{1-x}Mn_xTe$ quantum dots (QDs). We show that reaching thermal equilibrium in the system of the Mn ions and the exciton involves an intermediate stage, in which the exciton spin is relaxed, while the Mn ion spins are not. This stage gives rise to a specific, asymmetric PL lineshape. We develop a theoretical model for the PL spectrum, compare its results with the experiment, and find excellent agreement.

Semimagnetic QDs are an ideal system for investigations of semiconductor magnetism on the nanoscale. In particular, the dot magnetization can be formed without an external magnetic field, purely as a result of the exchange field that the exciton inposes on the Mn ions. From the point of view of potential applications, it is important to establish and tailor the formation dynamics of this magnetization. It has been shown[1, 2] that its development is accompanied by a redshift of the exciton transition energy. Indeed, the driving force for the ordering of the paramagnetic Mn spins is minimization of the exchange interaction between the exciton and the Mn ions. Magnetization formation is thus directly linked to establishing of the thermal equilibrium in this system. Moreover, it was shown [2] that the exciton spin relaxation occurs before the equilibrium is reached.

In this work, we show that the PL line shape is a tool sensitive to the degree of equilibration in the exciton – Mn ion system. In time-resolved PL measurements we directly demonstrate the transition between the relaxation stages: from a totally unrelaxed system, via exciton spin relaxation, to establishing of full equilibrium and forming the magnetization. We show that the equilibration dynamics critically depends on the concentration of Mn ions x. We study a set of samples with x ranging from 1% to 20%. We investigate PL spectra dependence on magnetic field and temperature and pinpoint features related to particular stages of magnetization formation. In particular, we find that exciton spin relaxation leads to a characteristically asymmetric PL spectrum and its narrowing by a factor reaching 2 and dependent on magnetic field and temperature.

The number of Mn ions in the studied dots ranges from about 20 to a few hundred. We thus model the PL lineshape treating the Mn ion spin as a classical variable. It allows us to develop analytical expressions for the PL spectra at each of the relaxation stages. Fitting of the model spectra allows us to extract from the PL measurements the exciton localization volume and the number and temperature of the Mn ions.

[1] J. Seufert *et al.*, Phys. Rev. Lett. **88**, 027402 (2001).
[2] Ł. Kłopotowski *et al.*, Phys. Rev. B **83**, 081306 (2011).