Efficient injection of spin-polarized excitons and Mn^{2+} ion spin orientation in a CdSe/ZnSe quantum dot

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The ability to control over the spin of excitons injected to a semiconductor quantum dot (QD) is desired in terms of possible QDs applications in quantum information processing. In particular, identification of polarization-conserving optical excitation channels of CdSe/ZnSe QDs is required for spin manipulation of a single Mn^{2+} ion recently incorporated into these dots [1]. So far, such excitation channels have been experimentally recognized in different QD systems. Most of these channels were exploiting resonant excitation of either an excited QD state [2], or a ground state of an adjacent, spontaneously coupled dot [3]. Both of these approaches demand tuning the excitation energy to a particular transition of the studied QD.

Here we show that optical excitation of CdSe/ZnSe QD with the circularly-polarized light of photon energy appropriately close to the QD emission energy provides a significant spin-polarization of excitons captured by the dot. Experimentally, self-assembled CdSe/ZnSe QDs are excited quasi-resonantly at 488 nm by the Ar-ion laser. The efficiency of spin-transfer is monitored in polarization-resolved measurements of neutral exciton (X) photoluminescence (PL) intensity. The detrimental impact of anisotropy-related X fine structure splitting (FSS) is reduced by application of magnetic field in Faraday configuration. At fields exceeding a few Tesla, the Zeeman splitting dominates over FSS and X emission lines become almost fully circularly-polarized. As a result, their intensities enable determination of spin-polarization degree of excitons captured by the dot.

The X PL is studied under circularly- and linearly-polarized excitation. Interestingly, in the latter case, when no spin-polarization of injected excitons is expected, we do observe a non-zero degree of X PL circular-polarization. More specifically, the lower-energy X emission line has larger intensity than the higher-energy one. This effect is related to a phonon-mediated exciton spin-flip process causing its spin-relaxation towards the lower-energy state. Such an interpretation is independently ascertained by an observation of a shorter decay time of the higher-energy X in a time-resolved experiment. The comparison between decay profiles of both X emission lines enabled us to obtain the X spin-flip rate dependence on the magnetic field. Under circularly-polarized excitation, the intensities of X emission lines are governed by an interplay of optically-induced exciton spin-transfer and X phonon-assisted relaxation. Taking into account the independently determined X spin-flip rate we extract the spin-polarization degree of excitons injected to the QD under excitation with σ^+ and σ^- polarizations. It is similar in both cases and reaches almost 50% for magnetic field in the range of 3-10 T.

The robustness of the presented excitation channel is confirmed by similarly high efficiency of the exciton spin-transfer determined for several, randomly selected CdSe/ZnSe QDs. The same effect is demonstrated also for CdSe/ZnSe dots doped with single Mn^{2+} ions, what enabled us to obtain a significant optically-induced Mn^{2+} spin orientation.

[1] J. Kobak, T. Smoleński et al., *Nat. Commun.* **5**, 3191 (2014).
[2] C. Le Gall et al., *Phys. Rev. Lett.* **102**, 127402 (2009).
[3] T. Kazimierczuk et al., *Phys. Rev. B.* **79**, 153301 (2009).