Change in Spin-Orbit Coupling Upon Addition of Manganese in CdSe Nanocrystals

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Introduction

When impurity ions are added into the lattice of a semiconductor, the electronic properties of the host change depending on the orbital overlap between the host bands and the impurity ions due to changes in exchange interactions. The influence of confinement is also an intriguing question that gives rise to changes in exchange interactions. The origin of these effects has been debated for decades. The classic example of a material that shows exchange interactions that are modified by confinement is Mn doped II-VI semiconductors. Exciton recombination, electron-phonon interactions, and the ways these properties changed with size must be addressed in order to investigate the exchange interactions[2].

Experimental

6.5% MnCdSe (4 nm) is prepared as described previously.[1] CdSe and MnCdSe thin films (100 µm, 10 µM) are prepared by dissolving the sample in pyridine and N-(3-(trimethoxisilyl)-propyl) EthyleneDiamine to form an amorphous organosilane glass. The magneto-photoluminescence was measured at the National High Magnetic Field Laboratory using an Ar+ ion laser (70 µW, 458 nm) circularly polarized using a plastic polarizer and a quarter wave plate coupled through a UV fiber optic into a Janis cryostat (4.2-300 K) in the 30 T resistive magnet. Emission was collected on a separate single mode fiber coupled to a one-meter monochromator (600 g/mm) onto a CCD (512 X 1024 pixels).

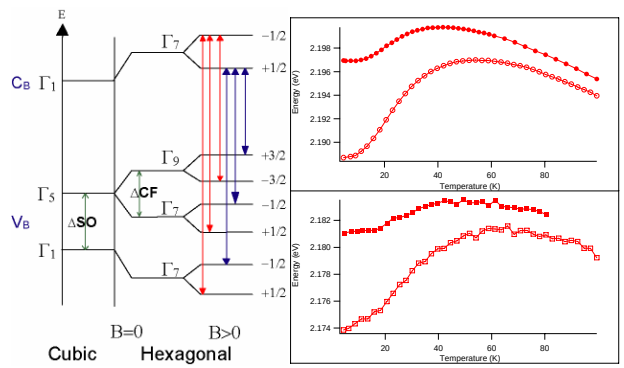

Figure 1. 1A) Allowed transition and splitting of a wurtzite structure. B) 30 T temperature dependent splitting of CdSe (●) and 6.5% MnCdSe (■) under right (●,■) and left (□,○) circularly polarized excitation.

Results and Discussion

Temperature dependent magneto-photoluminescence was measured and fit to a Langevin function $E = E_0 + (E(1) - E(0))\left(\frac{T}{\theta}\right)^3 \int_0^{\theta/T} \frac{x^3}{e^{-x}-1} - A(Cot(\frac{T}{2\theta}) - 1)$ to extract the emitting states. In CdSe, the major transition is between Γ_7 and Γ_9. From the fit of the temperature data, the two emitting states can be extracted. The observed splitting of the crystal field is consistent with the bulk crystal field of a CdSe wurtzite structure. As the phonon density becomes significant (>20 K) both states are indistinguishable due to the thermal energy present.

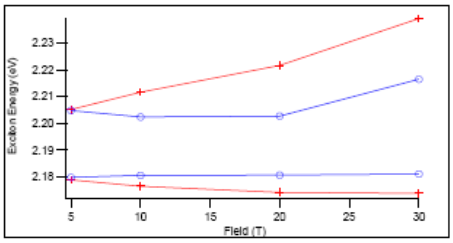

Figure 2. Differentiation of the emitting states upon extraction of the two contributing states as a function of temperature in CdSe resulting in the observed crystal field splitting.

Conclusions

In the case of MnCdSe, the manganese levels influence mixing of the valence levels and therefore depolarization occurs at much lower temperature and the crystal field splitting is decreased. The manganese ground state is in equilibrium with the valence levels of CdSe. This allows all transitions to be populated at zero-field, but the splitting of the 1/2 and 3/2 levels respond to the field. Overall, Mn is responsible for decreasing the spin-orbit energy shown by the lowering of thermal energy to depolarize the circularly polarized electrons.

References

[1] Magana D., et al., JACS, **128(9)**, 2931-2939, (2006).
[2] Magana D., et al., Submitted (2006).