Control of sp-d Exchange Interactions in Pseudo-type II Mn:ZnSe/CdSe Core-Shell Nanocrystal Quantum Dots

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Dilute magnetic semiconductors (DMSs) have been the focus of considerable research due to their potential usability in spin-based electronic devices1. Unpaired electrons of dopant atoms, such as Mn$^{2+}$, can couple strongly to electrons of the semiconductor (sp-d exchange interaction), which should allow for the manipulation of the spin degree of freedom using traditional microelectronic circuitry. We have developed a novel approach for manipulating sp-d interactions between the dopant and the semiconductor wherein Mn ions are incorporated into cores of ZnSe/CdSe core-shell semiconductor nanocrystal quantum dots (NQDs). These NCs represent quasi-type II hetero-structures that allow one to tune both the band edge transition energy and dopant-carrier wavefunction overlap by changing the size of the core and/or shell thickness2. The following is a summary of recent results from a set of doped heterostructures for which we demonstrate tunability of the sp-d exchange interaction energy as a function of hetero-NQD geometry.

Nanocrystals were synthesized using standard wet chemical techniques. A ZnSe core was grown and doped with Mn$^{2+}$ ions followed by growth of an outer CdSe shell. NC thin films were prepared on glass substrates, mounted on a sample stick, and inserted into 8T split coil superconducting magnet with optical access (SpectroMag, Oxford Instruments). Magnetic circular dichroism (MCD) experiments were performed in the Faraday geometry using a standard optical layout consisting of a frequency tunable light source whose polarization is modulated using a PEM (Hinds, Inc.). Light transmitted through the sample was focused onto a high gain APD and signal was measured using standard lock-in techniques.

Figure 1 displays the Zeeman splitting energy for a series of hetero-NCs as a function of magnetic field. MCD is used to measure field induced splitting of the lowest energy (1S) excitonic absorption feature. Shown in the inset is an example MCD spectrum, as expected this is related to the derivative of the absorption spectrum. The red trace in Fig. 1 is derived from a Mn:ZnSe NC (core only) which shows typical evidence of sp-d exchange as inversion of the g-factor sign along with signal saturation at high fields. When the shell thickness increases, the sign inversion (with respect to undoped NC) and saturation behavior are maintained although with decreased magnitude. We assign this decrease in amplitude to a shift in the electron (hole)/Mn overlap integrals. This results from the pseudo-type II band alignment of these heterostructure NCs which causes the carrier wavefunctions to shift their center-of-mass from the core to the shell with increased shell thickness. Thus, we are able to produce doped NCs where the strength of the interaction with Mn spins is controlled by engineering the dimensions of the hetero-NCs.

Acknowledgements

This work was supported by Los Alamos LDRD Funds and the Chemical Sciences, Biosciences, and Geosciences Division of the Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy.

References