Valence band mixing and spin pumping efficiency in room-temperature CdSe nanocrystals

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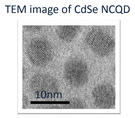
Introduction

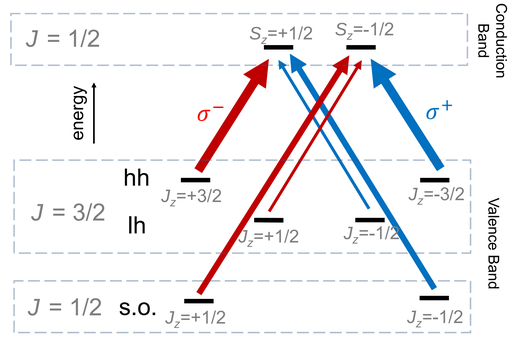
- Optical excitation of spins in semiconductors is a powerful technique to study the initialization of coherent spin states in both bulk materials and nanostructures. We studied the room temperature optical spin pumping process in chemically synthesized colloidal nanocrystal quantum dots (NCQDs).
- With the goal of finding optimum pump energy for optical excitation and understanding the physics behind we measure the efficiency of spin pumping vs. excitation energy in CdSe NCQD ensembles with different mean particle size through time resolved Faraday rotation (TRFR) measurements.

TEM image of CdSe NCQD



Optical spin pumping

- A left-handed circularly polarized photon creates an up spin if its energy is tuned to a hh transition. It creates a down spin if its energy is tuned to lh or so sub-bands.
- Opposite helicities create spins with opposite polarization.
- The relative strength of hh, lh, s.o. transitions are 3:1:2 in bulk semiconductors.



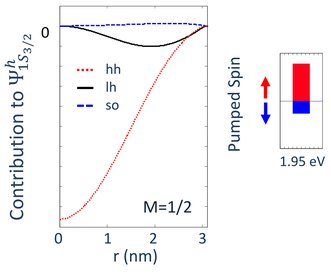
Optical transitions in nanocrystal QDs

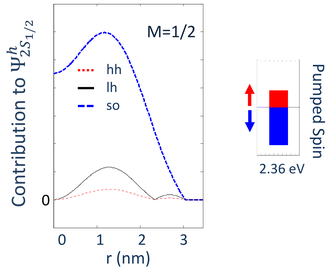
- Hole wave function is a superposition of hh, lh, and so sub-bands; includes all of the Bloch functions u_{J,J_z} .

$$\Psi_{F,M}^{h,\pm}(\boldsymbol{r}) = \sum_{l}\sum_{J,J_z} f_{l,m}^{v,\pm}(\boldsymbol{r}) u_{J,J_z}$$

Hole wave function and electron spin output of an excitation

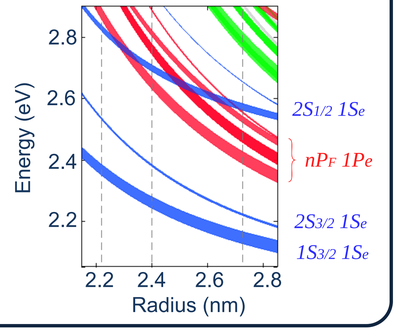
- Lowest energy transition, $1S_{3/2} \rightarrow 1S_e$, is hh dominant.
- $2S_{1/2} \rightarrow 1S_e$ is a split-off sub-band dominant.





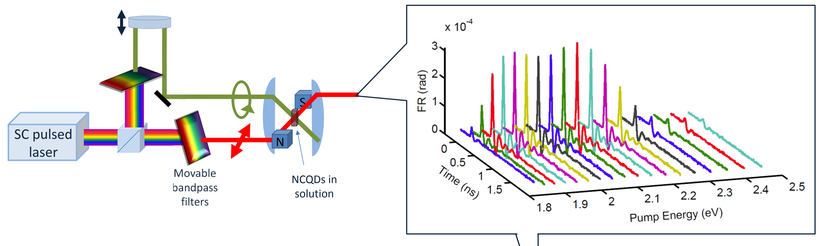
Optical transition energies vs. NCQD size

- The 2S1/21Se has a significant feature. It switches its position in energy with the nPF1Pe group as the size of NC gets bigger.

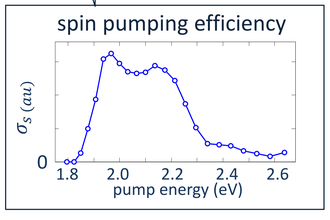


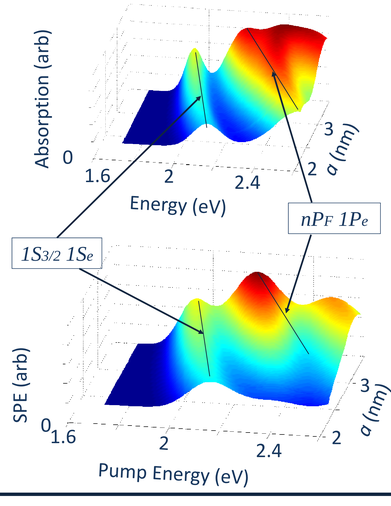
Measuring efficiency of optical spin pumping

- Spins are excited and read through time resolved Faraday rotation measurements.
- By varying the excitation energy the spin signal vs pump energy is studied.



- Measured signal is spin signal plus noise : $\mathcal{S}(t) = s(t) + n(t)$;
- Standard deviation of the signal with noise subtracted is a measure of spin pumping efficiency (SPE) $\sigma_s = \sqrt{\sigma_{\mathcal{S}}^2 - \sigma_n^2}$.

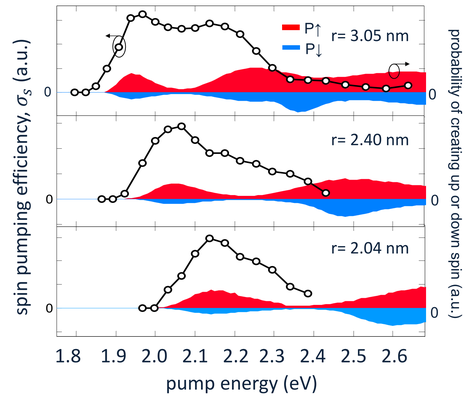




- Spin dependent optical transition strength calculations predict two peaks in SPE for large NCQDs and one for smaller sizes.
- Spin pumping through nPF 1Pe transition is quenched in smaller NCs because of the overlap with 2S1/2 1Se.

Spin pumping efficiency in different sizes of NCQDs

- The calculated spin dependent transition strength from hh(red) and lh+so (blue) sub-bands follow the experimental data and features qualitatively.



- The only peak in smaller NCQD's spin data and the first peak from r = 3.05nm is due to 1S3/21Se transition.
- There is a second peak in the bigger NCQD sample corresponding to the group of nPF1Pe transitions.
- The overlap of s.o.-dominant transition 2S1/21Se and the mainly hh transitions nPF1Pe, quenches a second peak in sample of smaller NCQDs.

Conclusion

- The competition between the transitions from hh, lh, and so sub-bands determine the spin polarization output of optical pumping at different pump photon energies.
- Size dependence of relative position of different energy states in NCQDs is responsible for emergence of one or two peaks in SPE data.

References

[1] A. I. Ekimov, F. Hache, M. C. Schanne-Klein, D. Ricard, C. Flytzanis, I. A. Kudryavtsev, T. V. Yazeva, A. V. Rod-ina, and A. L. Efros, J. Opt. Soc. Am. B 10, 100 (1993).

Acknowledgements

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