Fluorescence Line Narrowing (FLN) Experiments Revisited in the Florida Helix

Rawat, N.; Ngan Hua, K.; Furis, M. (University of Vermont, Materials Science); Bishop, M.; Semenov, D.; McGill, S. (NHMFL-FSU)

Introduction

The recently commissioned HELIX magnet and its split-coil design represent not only a premiere in magnet technology but also an unprecedented opportunity to fully implement the latest ultrafast techniques into the high magnetic field environment towards new discoveries of spin-dependent phenomena in large variety of systems. We revisited Fluorescence Line Narrowing (FLN) in CdSe nanocrystals, a system where the interplay between bright and dark exciton states and exciton spin dynamics [1,2] can now be studied on the sub-nanosecond scale with the new free-space optics setup and the tunable ultrafast OPERA Solo parametric amplifier.

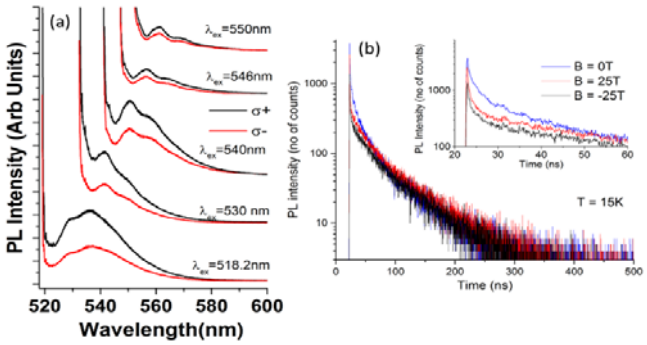

Figure 1. (a) Spin-resolved FLN spectra from five different CdSe nanocrystal sizes within two different ensembles exhibit the expected amount of spin-polarization at B = 25T and T = 15K (b) The sub-nanosecond fast component of exciton spin dynamics (inset) is now revealed for the first time in the HELIX free space optics experiments

Experimental

In an FLN experiment a narrowband laser is carefully tuned to a wavelength resonant to the tail of the ensemble absorption spectrum in order to create excitons only in the largest nanocrystals within the ensemble, thus effectively probing only a subset of nanocrystals sizes. The narrowband ultrafast excitation (~ 1nm) is now obtained from the 30nm broadband output of the 1KHz Coherent OPERA Solo parametric amplifier using a grating and slit setup.

Results and Discussion

Figure 1 (a) shows some of our representative PL spectra and lifetimes. We clearly observe the expected narrow luminescence spectra from different nanocrystals sizes within an ensemble. We also observe a significant amount of polarization that varies as a function of size in agreement with previous CW luminescence experiments performed in the resistive magnets.[3] The T = 15 K long recombination lifetimes are also in agreement with previous measurements. The big news in this experiment is that we can now resolve the fast spin-dependent dynamics component of the decay. (Inset of Fig 1(b)) There are unanswered questions in the fast component of spin-dependent exciton dynamics in virtually every electronic material because of the limitations imposed by fibers experiments. All of those questions can now be answered using state of the art polarization optics and picosecond electronics.

Acknowledgements:

This work was supported by NSF DMR # 1229217 and the NHMFL Visiting Scientist program. A portion of this work was performed at the National High Magnetic Field Laboratory, which is supported by National Science Foundation Cooperative Agreement No. DMR-1157490, the State of Florida, and the U.S. Department of Energy.

References

[1] M. Furis et al, Phys. Rev. B **2006**, 73, 241313.
[2] M. Furis et al. Phys. Rev B 2008, 77, 035328.
[3] M. Furis et al, *J. Phys. Chem. B* **2005**, 109, 15332-15338.