Inhomogeneous spin decoherence and dephasing in room temperature CdSe nanocrystal quantum dots

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Introduction

We use Faraday rotation (FR) time resolved measurements and transmission electron microscopy (TEM) to understand the underlying connection between the decay of room-temperature coherent spin dynamics and the shape and size distribution of an ensemble of CdSe nanocrystal quantum dots (NCQDs). There are two confounding factors in the spin dynamics of these systems:

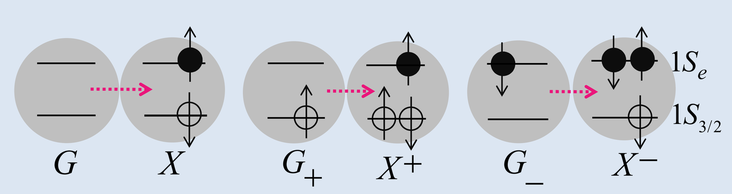
- Magnetic field dependent spin dephasing and decoherence.
- Presence of two distinct components in spin signal with different dynamics and decay characteristics.

We address these two main points by developing a model of dephasing and decoherence arising from g-factor distribution and exciton fine-structure inhomogeneity.

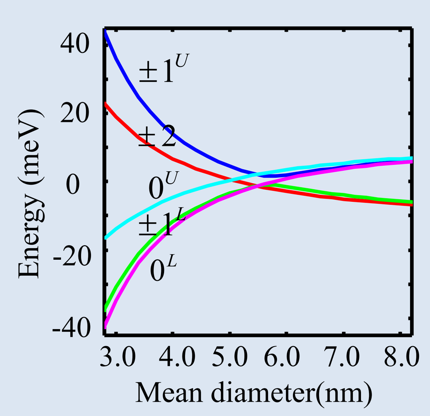
To ensure that the fitting procedure yields meaningful results we examine and minimize the cross-correlation between parameters.

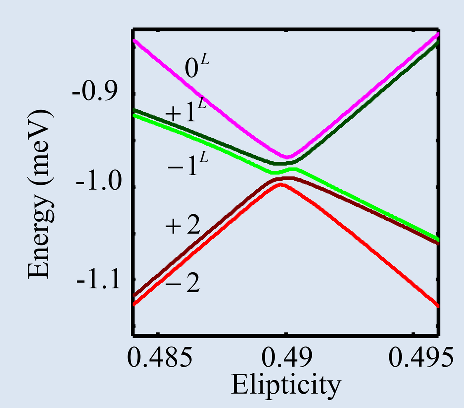
Excitons in NCQDs, Optical excitation, and Faraday rotation

- Optical excitation creates neutral exciton, positive or negative trion depending on the initial charging of the NCQD.

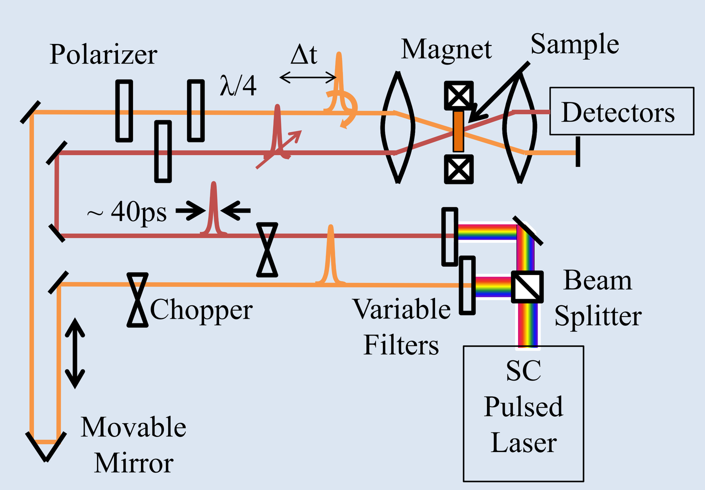


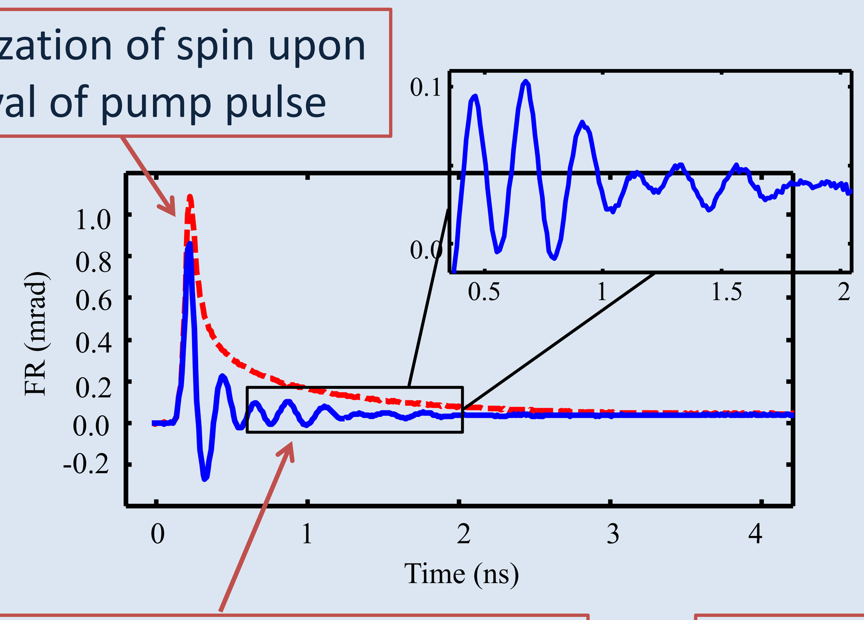
- Exciton fine-structure arising from electron-hole exchange interaction and crystal field splitting is size and shape dependent.

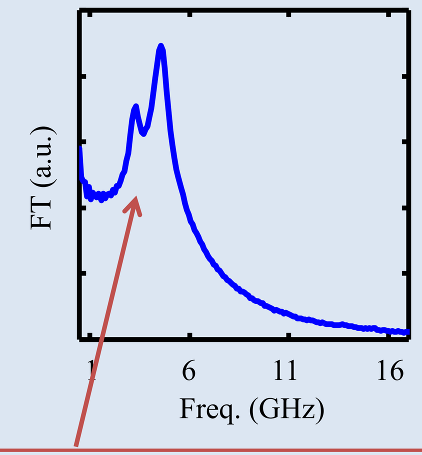




- Reading the Spin: Rotation in polarization of the probe beam is a measure of the projection of spin onto the beam direction.



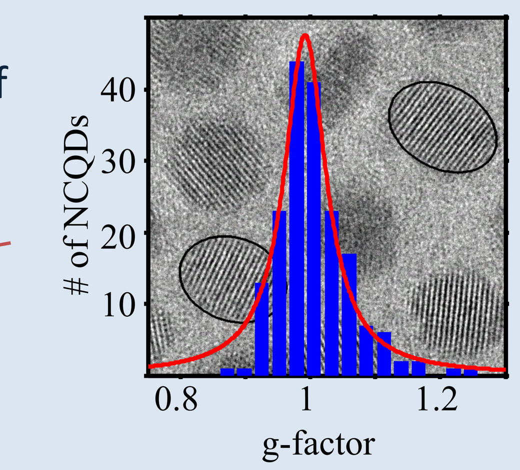




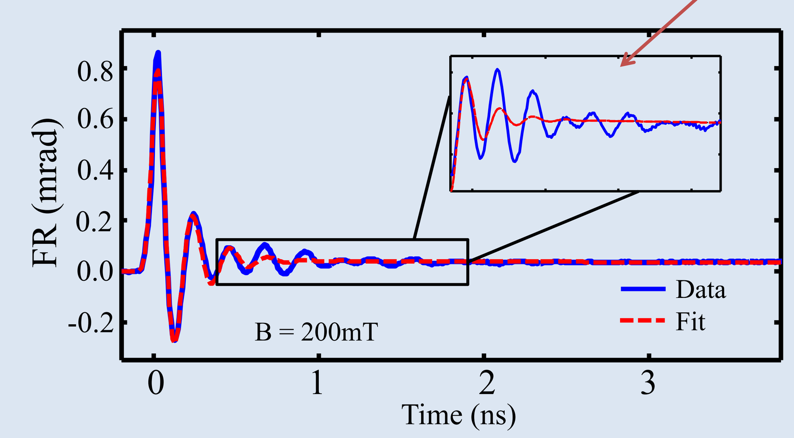
Spin dynamics is size-dependent; g-factor inhomogeneity induced dephasing (gID)

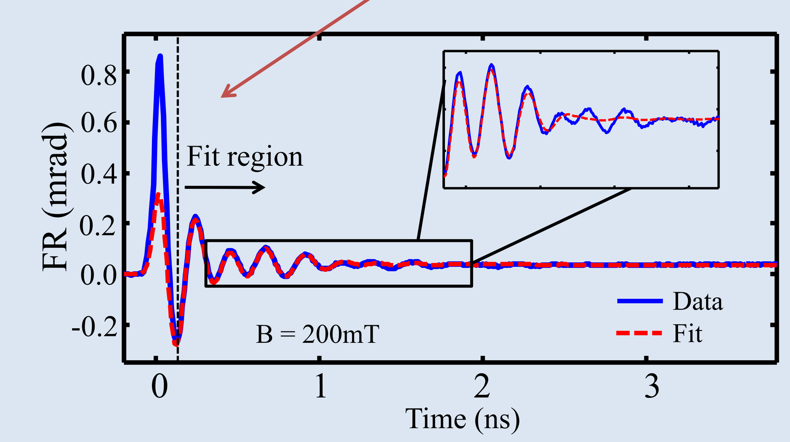
- The size distribution within the ensemble leads to dephasing of the spin signal.
- Measuring the size distribution by TEM imaging we calculate dephasing:

$$\langle S_z \rangle \propto e^{-t/\tau} \times \int_{-\infty}^{\infty} dg P(g-g_0) \cos((g-g_0)\mu_B BT/\hbar)$$

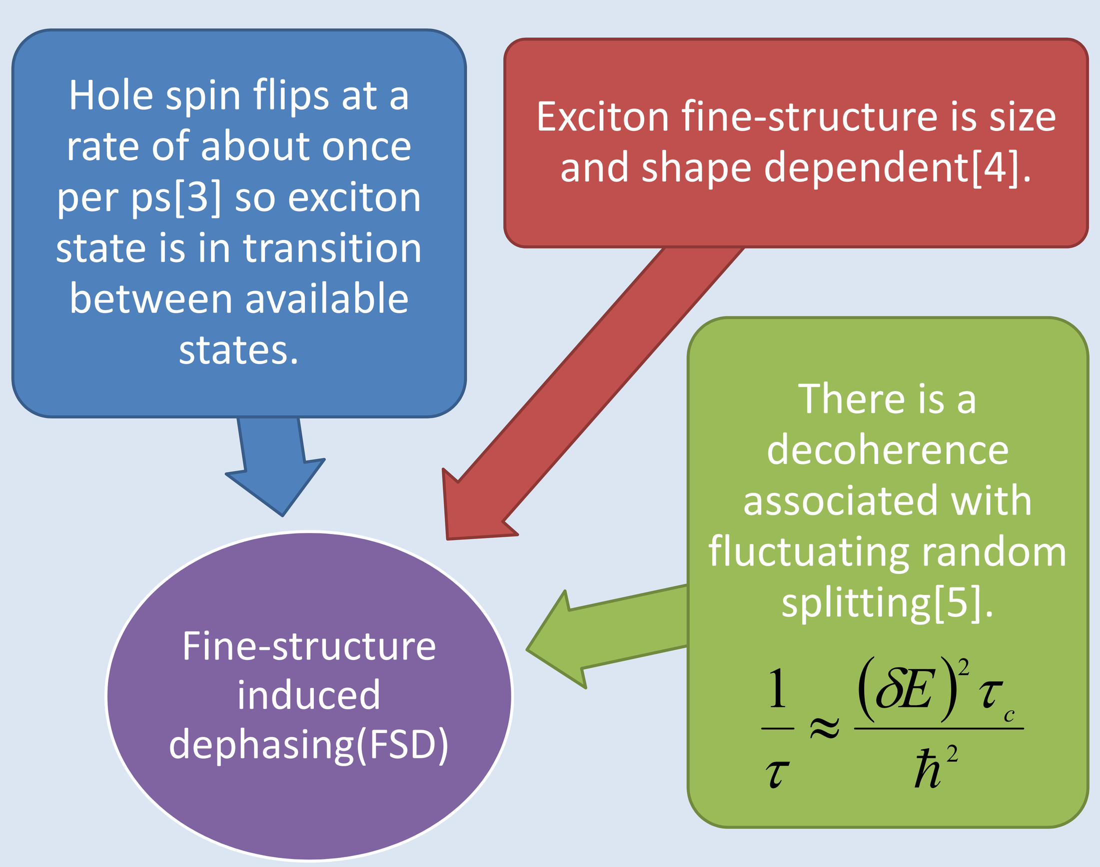


- This approach fails to capture the dephasing/decoherence or the short-timescale behavior of the spin signal.

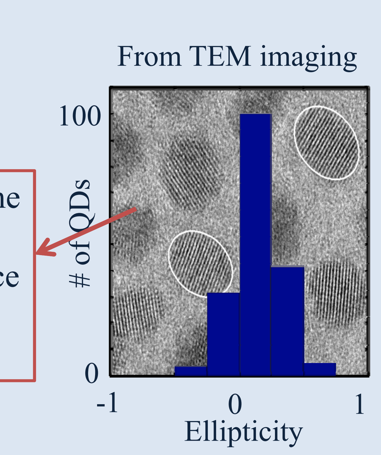




Taking shape anisotropy into account



- The fluctuating splitting given by a combination of exchange interaction and shape-dependent crystal splitting leads to decoherence of exciton spin.
- FSD is large in NCQDs in which the crystal field and shape anisotropy contribution to exciton fine-structure cancel each other (quasi-spherical QDs).



- We use FSD+gID to describe the exciton spin component of FR data and gID to model the electron contribution to spin signal.

$$\Gamma_{FSD}(t) = \int_{-\infty}^{\infty} P(\mu) \exp(-t/\tau(\mu)) d\mu$$

$$= \frac{1}{\sqrt{1+2t\tau_c k^2 \delta_\mu^2/\hbar^2}} \exp\left[\frac{-(\bar{\mu}-\mu_{qs})^2}{\hbar^2/(t\tau_c k^2)+2\delta_\mu^2}\right] \exp\left[-t\alpha^2 B^2 \tau_c/\hbar^2\right]$$

$$f_e(t) = \cos(g_1 \mu_B Bt/\hbar) \exp(-\Delta g_1 \mu_B Bt/\hbar) \exp(-t/\tau_1)$$

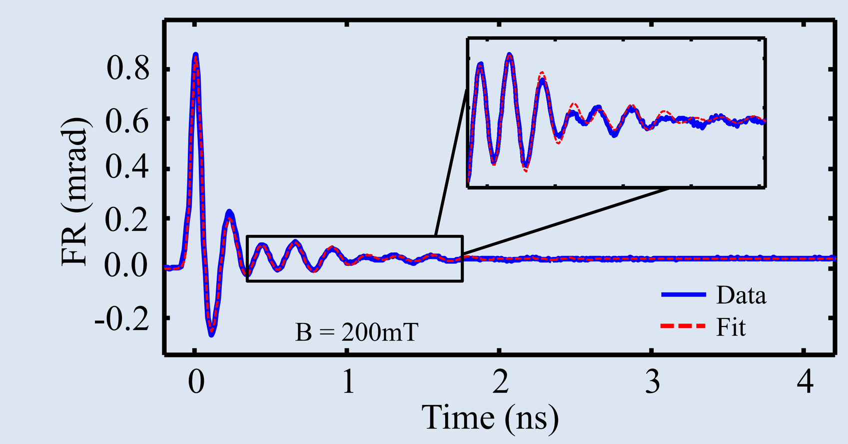
$$f_{exc.}(t) = \cos(g_2 \mu_B Bt/\hbar) \exp(-\Delta g_2 \mu_B Bt/\hbar) \Gamma_{FSD}(t) \exp(-t/\tau_2)$$

Fitting FR data to FSD +gID model

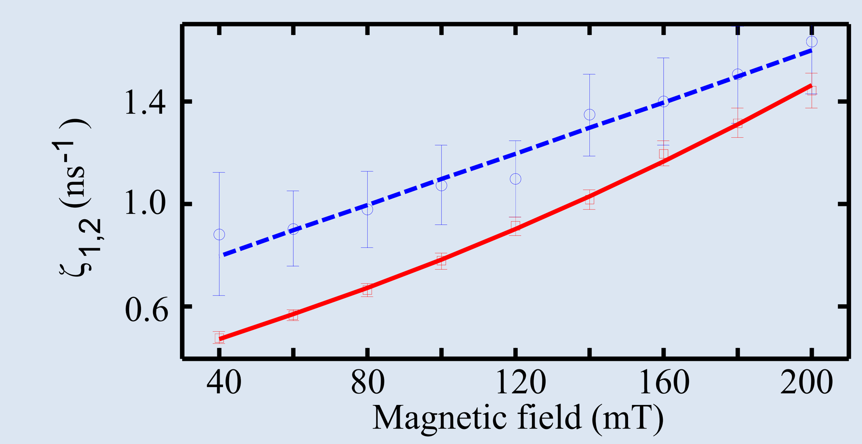
- To reduce cross-correlation between and find reliable fit parameters we combine exponential decoherence and dephasing terms of each spin component into one parameter and fit the data.

$$\varsigma_1 = 1/\tau_1 + \Delta g_1 \mu_B B/\hbar$$

$$\varsigma_2 = 1/\tau_2 + \Delta g_2 \mu_B B/\hbar + \alpha^2 B^2 \tau_c/\hbar^2$$



- We disentangle decoherence and dephasing parameters using the B-dependence of these combined decay rates.



- Spin lifetime is 1.69±0.28 ns for electrons and 3.11±0.47 ns for excitons.

Conclusion

- g-factor inhomogeneity is not enough to model the FR data.
- By including the FSD we are able to model the short-timescale feature.
- Exciton spins in Quasi-spherical NCQDs have longer lifetimes. The rest of the population contributes to the short-timescale spike. This is in agreement with what hypothesized by Gupta *et al.*[2].
- FSD accounts for the unexpectedly strong B-dependence of the decay of the exciton component.

References

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