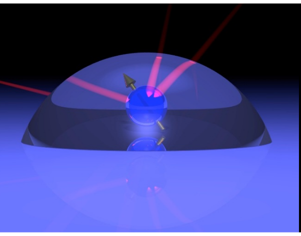

Frequency-domain optical probing of coherent spin dynamics in nanocrystal quantum dots

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Introduction

- Fourier Transform Spin Resonance (FTSR): a technique for measuring GHz-scale coherent spin dynamics in semiconductors using narrow linewidth modulated lasers.
- FTSR measures the Fourier transform of spin dynamics.
- Proof-of-concept: measuring Faraday rotation in CdSe nanocrystal quantum dots.

Motivation

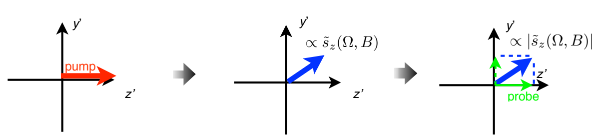
- Single spin spectroscopy
  - Sharp spectral features of spins in single QDs require a narrow linewidth probe.
  - High peak intensity of ultrafast pulses causes undesired shift of QD levels.
- Inhomogeneous QD ensemble
  - Narrow linewidth pump and probe can excite and measure spins in a narrower distribution of QDs.
- Frequency domain technique can be naturally combined with other resonance techniques (e.g. ESR).

FTSR theory

Pump photons initialize spins → Average evolution of spin projection: $s_z(t, B)$ → FTSR measures Fourier transform: $\tilde{s}_z(\Omega, B)$

Modulate pump: $I_{pump} = I_0\,(1+\cos(\Omega t))$ → Build up steady state spin polarization $\propto \tilde{s}_z(\Omega, B)$ → Probe in-phase or out-of-phase: $I_{probe} = I_1\,(1+\cos(\Omega t))$, $I_{probe} = I_1\,(1+\sin(\Omega t))$

Reference frame rotating at Ω:

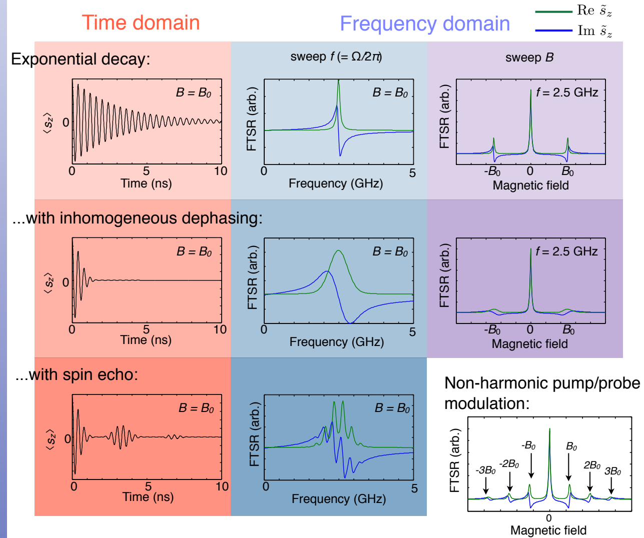


Measure DC component of signal with low bandwidth detector:

$$\Theta_{FR} \propto 2\tilde{s}_z(0, B) + \mathrm{Re}\,\tilde{s}_z(\Omega, B)$$ ← Probe in-phase

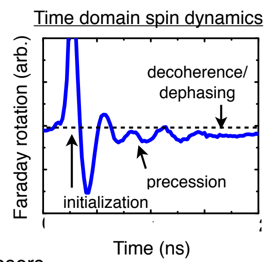
$$\Theta_{FR} \propto 2\tilde{s}_z(0, B) + \mathrm{Im}\,\tilde{s}_z(\Omega, B)$$ ← Probe out-of-phase

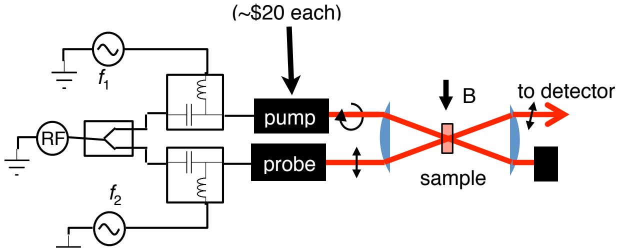
Some simulated FTSR spectra



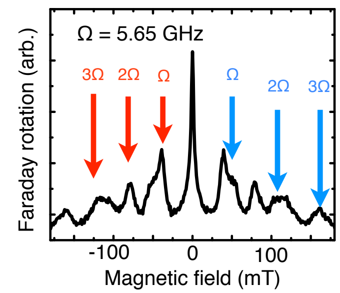
Experimental samples and setup

- Sample: CdSe nanocrystals in solution
- ~ns spin coherence time at T=300K
- Multiple g-factors, lifetimes, inhomogeneous dephasing rates



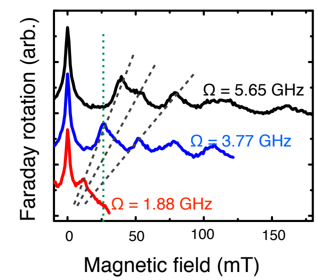


Experimental demonstration

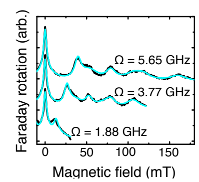


- Multiple sets of peaks → non-harmonic pump/probe
- Position of peaks → g-factors, g_1, g_2
- Width of peaks → spin lifetime
- Broadening with |B| → dephasing

- Peaks shift linearly with Ω.
- Linecut at constant B yields Fourier transform of spin dynamics.



Fits:



Fit model:

$$s_z(B,t) = A\left[\exp\left(-\frac{\Delta g_1 \mu_B B t}{\hbar}\right)\exp\left(-t/\tau_1\right)\cos\left(\frac{g_1 \mu_B B t}{\hbar}\right) + \beta\exp\left(-\frac{\Delta g_2 \mu_B B t}{\hbar}\right)\exp\left(-t/\tau_2\right)\cos\left(\frac{g_2 \mu_B B t}{\hbar}\right)\right]$$

(inhomogeneous dephasing; decoherence; precession; bimodal distribution)

Fit results:

τ_1 (ns)	τ_2 (ns)	g_1	g_2	Δg_1	Δg_2
0.77±0.07	3.02±0.14	1.203±0.007	1.619±0.005	0.00±0.015	0.091±0.008

agrees with time domain results!

Conclusions

- FTSR: a technique for probing spin dynamics with narrow linewidth lasers.
- Optimal trade-off between time resolution and spectral width, low peak intensity.

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