Two-Color Spin Noise Spectroscopy: Using Spin Fluctuation Correlations to reveal Homogeneous Linewidths within Quantum Dot Ensembles

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Introduction: Inhomogeneous broadening is ubiquitous in the physical, chemical and materials sciences, occurring whenever a collection of nominally equivalent constituents differ in size, shape, composition, conformation and/or local environment. Notable examples include ensembles of nanocrystals, quantum dots, nanotubes or molecules. In optically active systems, such broadening typically leads to a spread of the constituents' fundamental absorption or emission energies over an inhomogeneously broadened band of spectral width γ_{inh}, which can be orders of magnitude larger than the underlying homogeneous linewidth γ_h of the individual constituents themselves. Usually, however, γ_h is the essential quantity of interest, since γ_h directly reveals (or at least constrains) the fundamental relaxation rates and coherence times of the system.

Unfortunately, γ_h is generally inaccessible in inhomogenously broadened ensembles using conventional low-power/linear optical spectroscopic techniques, which typically measure time- and ensemble-averaged response functions (such as absorption, photoluminescence, polarization, etc). To circumvent this limitation, various nonlinear optical methods have been developed over the years to extract γ_h from inhomogeneously broadened ensembles; eg spectral-hole burning and four-wave mixing methods, as well as single-particle methods.

Experiment: In this work we develop a novel low-power optical technique— two-color spin noise spectroscopy—and demonstrate that it can be used to reveal the underlying homogeneous linewidth γ_h of the individual constituents that make up an otherwise inhomogeneously broadened ensemble. Specifically we apply these methods to reveal γ_h of singly charged (In,Ga)As quantum dots (QDs) within an inhomogeously broadened QD ensemble. Importantly, this optical technique operates in the linear/low-power regime and does not rely on any excitation or nonlinear response of the system. The key point is that this technique is not based on conventional time-averaged response functions, but rather is based on the intrinsic and random fluctuation properties of the system—in this case, spin fluctuations. In particular, it exploits the fact that spin fluctuations from different QDs in the ensemble are *uncorrelated* in time. By measuring the degree of correlation between two independent noise probes (two probe lasers detuned from each other), we reveal the underlying homogeneous absorption linewidth, γ_h, of positively charged QDs in an ensemble measurement—information that is generally inaccessible to conventional linear spectroscopy.

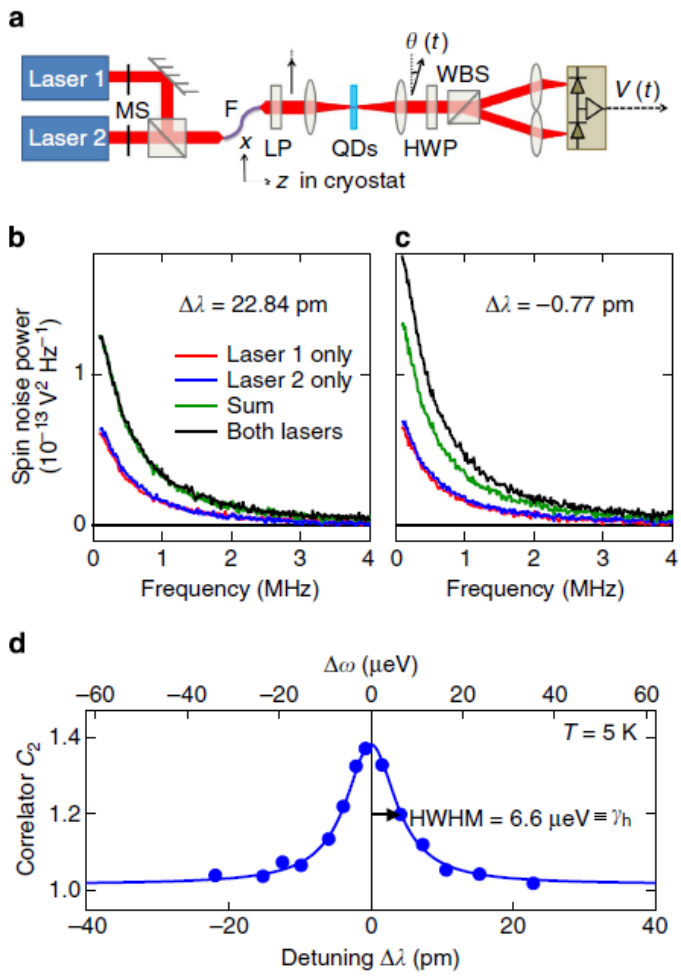

Fig.1 (a) The two-color spin noise experiment. (b,c) The measured spin noise power when the two lasers are far from each other (in which case they probe uncorrelated QD ensembles), or near each other (where the measured spin fluctuations are correlated). (d) the degree of correlation versus relative detuning. The half-width gives γ_h.

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