Spin Noise Spectroscopy beyond Thermal Equilibrium and Linear Response

Glasenapp, P. (TU-Dortmund); Yang, L. (NHMFL-LANL); Roy, D. (LANL); Rickel, D.G. (NHMFL-LANL); Greilich, A.; Bayer, M. (TU-Dortmund); Sinitsyn, N.A. (Theory Division-LANL); Crooker, S.A. (NHMFL-LANL)

Introduction: Detection of intrinsic and random spin fluctuations – "spin noise" – has attracted considerable interest in recent years, spurred by significant advances in ultrasensitive magnetic measurement techniques including nitrogen-vacancy magnetometry, force-detected magnetic resonance, optical detection, and conventional nuclear magnetic resonance. These advances have enabled studies of ever-shrinking volumes (and numbers n) of spins, wherein the magnitude of statistical fluctuations ($\sim\sqrt{n}$) becomes increasingly important as n decreases and can even exceed the average thermal spin polarization due to an applied field – a fact exploited in recent demonstrations of nanometer-scale nuclear spin imaging**.**

For electron spins, the method of 'optical spin noise spectroscopy' has emerged as a powerful technique for detecting spin dynamics that is based on passively measuring (typically via optical Faraday rotation) the stochastic spin fluctuations of an unperturbed system in strict thermal equilibrium. Initially demonstrated in atomic vapors, it has also been applied to spins in bulk and low-dimensional semiconductors. In general however, the information obtained in thermal equilibrium (spin relaxation rates, g-factors, etc) is necessarily linked to – and constrained by – the system's linear response functions, as mandated by the fluctuation-dissipation theorem. While very useful for revealing the relevant energy levels and associated spin coherences available to a spin system, equilibrium spin noise spectra alone cannot in general reveal the underlying couplings and correlations between these coherences, nor the system's (usually very interesting) response to resonant driving fields. For these reasons, detection of spin fluctuations under nonequilibrium conditions has long been desirable from both theoretical and experimental view points.

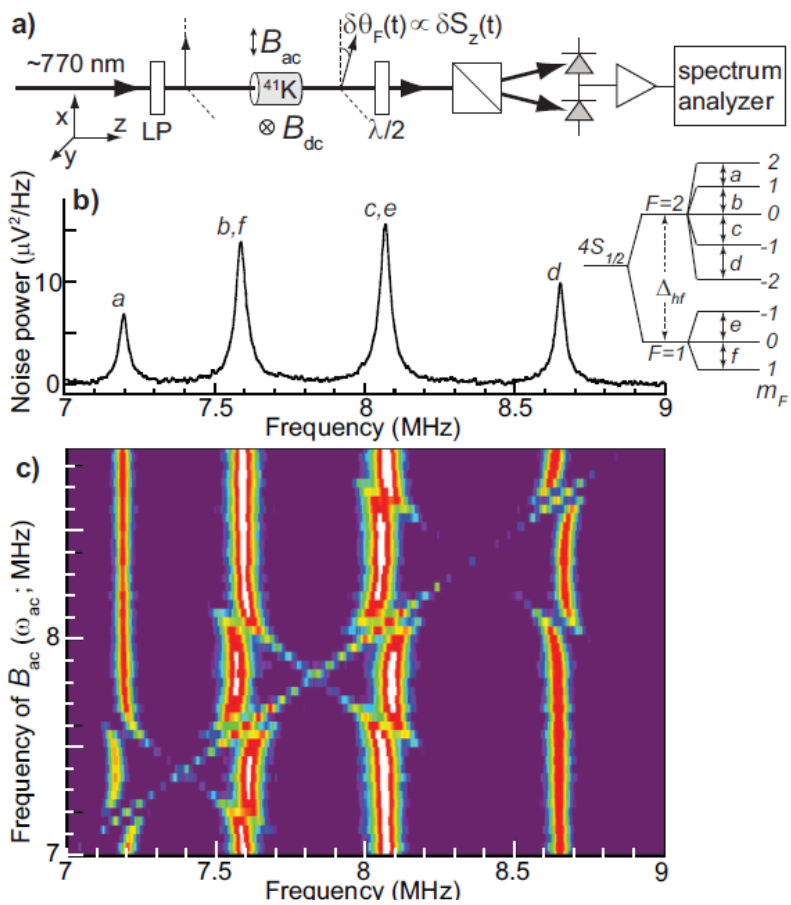

Fig.1 (a) Experimental setup. (b) The spin noise spectrum from ^{41}K in thermal equilibrium. (c) Surface plot of ^{41}K spin noise as B_{ac} is swept through the noise coherences.

Experiment: We demonstrate that, in conjunction with a weak ac magnetic field B_{ac}, spin fluctuations in strictly unpolarized spin ensembles can reveal patterns of correlation and coupling between a spin system's various energy levels, revealing coherent effects beyond thermal equilibrium and linear response [1]. To introduce and benchmark this capability we apply it to a spin system with a non- trivial but very well understood magnetic ground state: a classical vapor of alkali atoms. Specifically, we study ^{41}K atoms, in which the single spin-1/2 valence electron in the 4S state has a multi-level magnetic ground state due to hyperfine coupling with the I = 3/2 nuclear spin. In the presence of B_{ac}, the intrinsic stochastic spin fluctuations alone will clearly and directly reveal the Rabi splittings of driven spin levels, as well as the associated formation of Mollow triplets, Autler-Townes doublets of neighboring spin coherences that share common Zeeman sublevels, the ac Zeeman effect, and even non-linear (multiphoton) coherence effects (See Fig. 1). *This work was supported by the Los Alamos LDRD program.*

[1] P. Glasenapp, Luyi Yang, D. Roy, D. G. Rickel, A. Greilich, M. Bayer, N. A. Sinitsyn & S. A. Crooker, *Physical Review Letters* **113**, 156601 (2014).