Higher-order Correlations in the Magnetization Fluctuations of Ultrathin PtCoPt Films

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Introduction

Spontaneous magnetization fluctuations can occur in ferromagnetic materials, even in thermal equilibrium, particularly when the magnetic anisotropy energy becomes comparable to or less than the available thermal energy. These intrinsic thermodynamic fluctuations encode valuable information about the magnetization dynamics of the system itself, because their frequency spectrum $S(\nu)$ is intimately and necessarily related to the dissipative (imaginary) part of the magnetic susceptibility $\chi''(\nu)$, in accord with the fluctuation-dissipation theorem [namely, $\chi''(\nu) \sim \nu S(\nu)/kT$]. Spectroscopy of this intrinsic "magnetization noise" can therefore provide an alternative and entirely passive means of measuring magnetization dynamics that does not require driving, exciting, or perturbing the system away from thermal equilibrium—in contrast with most conventional methods for measuring magnetic resonance or ac susceptibility.

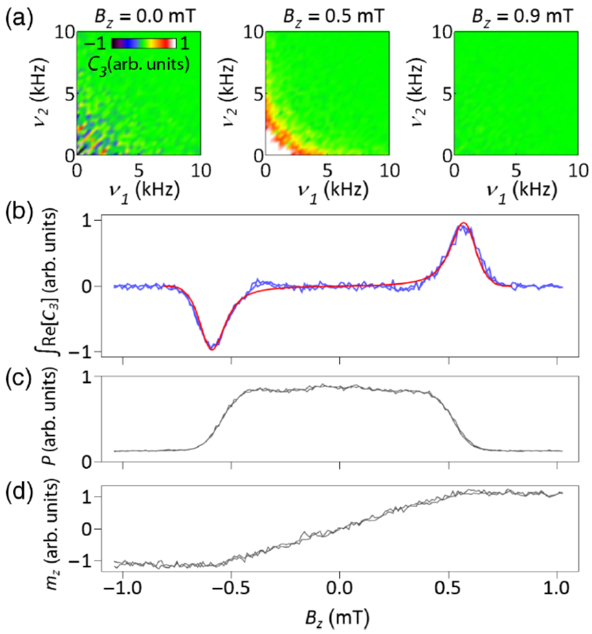

Figure: Measuring higher-order correlations of the magnetization noise at a location on the sample near the SRT that exhibits linear magnetization and large fluctuations. (a) The real part of the third-order correlator, $Re[C_3(\nu_1,\nu_2)]$, acquired at B_z= 0, 0.5, and 0.9 mT. $C_3(\nu_1,\nu_2)=\langle a(\nu_1)a(\nu_2)a^*(\nu_1+\nu_2)\rangle$, where $a(\nu)$ is the Fourier transform of the fluctuation (or 'noise') signal $\delta m_z(t)$. (b) The integral $\int Re[C_3]\, d\nu_1\, d\nu_2$ measured as a continuous function of B_z. The red line shows this dependence calculated from the magnetic free energy. (c) The simultaneously measured total noise power $P=\int S(\nu)d\nu$ versus B_z . (d) The average magnetization $m_z(B_z)$ at this same location, as measured by conventional MOKE, showing a constant susceptibility for $|B_z| < B_{sat}$. The third-order correlator is approximately zero except when $|B_z| \sim B_{sat}$, indicating a skewness in the distribution of $\delta m_z(t)$.

Results and Discussion

We use scanning optical magnetometry to study the broadband frequency spectra of spontaneous magnetization fluctuations, or "magnetization noise," in an archetypal ferromagnetic film that can be smoothly tuned through a spin-reorientation transition (SRT). The SRT is achieved by laterally varying the magnetic anisotropy across an ultrathin Pt/Co/Pt trilayer, from the perpendicular to in-plane direction, via graded Ar+ irradiation. The magnetization noise depends strongly on applied in- and out-of-plane magnetic fields, revealing local anisotropies and also a field-induced emergence of fluctuations in otherwise stable ferromagnetic films. Crucially, we demonstrate that higher-order correlators can be computed from the noise. An important consequence and potential advantage of measuring fluctuations $\delta m_z(t)$ directly in the time domain is that *all* possible time correlators can, in principle, be retrieved and analyzed from the noise signal. Importantly, however, the n^{th}-order time correlator can contain additional information that is not trivially related to $S(\nu)$, particularly in the presence of interactions, inhomogeneous broadening effects, and/or non-Gaussian noise due to (for example) the discrete nature of the system. In general, only the full set of all correlators contains complete information about an interacting system. Higher order correlations have been studied theoretically and experimentally in magnetic systems such as spin glasses and amorphous magnets, but we are not aware of any prior experimental studies of higher-order correlations of fluctuations in magnetic films in thermal equilibrium. Such correlators, however, play an important role in the theory of phase transitions and nonlinear thermodynamics. Therefore, their experimental measurement is highly desirable as a tool for characterization of material phases and for tests of fundamental theoretical predictions, such as higher-order fluctuation relations and for testing the universality of scaling exponents. These results highlight broadband spectroscopy of thermodynamic fluctuations as a powerful tool to characterize the interplay between thermal and magnetic energy scales, and as a means of characterizing phase transitions in ferromagnets.

References

[1] Balk, A. L. *et al.*, Physical Review X **8**, 031078 (2018).