Anharmonic magnetic response of magnetic nanoparticles detected by atomic rf magnetometry

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Harmonically-excited magnetic nanoparticles (MNP) produce an anharmonic magnetic field $B_{MNP}(t)$, consisting of fundamental (ω_{exc}) and higher ($n\omega_{exc}$) harmonics of the excitation field. We explore the recording of these harmonics with a laser-pumped atomic rf magnetometer (ARFM), operated in two different modes, and report on sensitivity, bandwidth, and constraints thereof.

In the first mode, the ARFM is operated as an M_x-magnetometer [1] based on the optical detection of magnetic resonance in a spin-polarized atomic Cs vapor in a paraffin-coated glass cell. The Cs vapor is exposed to a DC magnetic field B_0 which splits the Zeeman sublevels by $\hbar\omega_L$ (Larmor frequency) and to an rf-magnetic field which leads to a spin-depolarization when the rf frequency matches ω_L. A circularly-polarized laser beam resonant with the Cs $D_1(4 \rightarrow 3)$ hyperfine transition pumps the atoms into non-absorbing dark states. The rf-induced depolarization increases the absorption of the Cs vapor, thereby changing the light transmission in a resonant manner. In this operation mode we tune B_0 such that its Larmor frequency matches one of the harmonics $n\omega_{exc}$ of the $B_{MNP}(t)$ oscillation. The amplitude of the corresponding resonance is a direct measure of the respective harmonic's amplitude. However, the magnetometer readout at higher harmonics is perturbed by multiphoton rf-transitions induced by the excitation field. The corresponding signals cannot be distinguished from the MNP signals proper. We have studied this multiphoton depolarization by supplying a purely harmonic rf-field. The detected multiphoton amplitudes A_n (Fig. **??**, left) increase with a $A_n \propto B^n_{\omega_{exc}}$ power law of the driving field amplitude [2,3], and the corresponding cross-sections (Fig. **??**, center) obey the anticipated $\sigma_n \propto \omega_L^{-(n-1)}$ law.

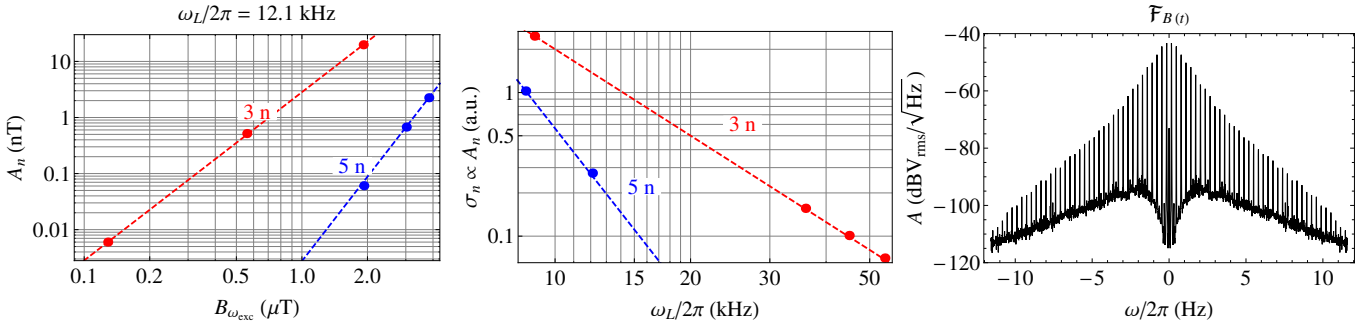

Figure 1: *Left and central graphs show the $A_n \propto B^n_{\omega_{exc}}$ and $\sigma_n \propto \omega_L^{-(n-1)}$ dependencies of the multiphoton process, respectively. The right graph shows the Fourier spectrum of a 157 Hz square-wave modulated $B(t)$ field measured by a self-oscillating magnetometer.*

In order to avoid the multi-photon perturbations we deployed the ARFM in its self-oscillating mode of operation [4]. We orient the magnetometer such that $\vec{B}_{MNP}(t)$ has a large projection onto $\vec{B}_0$, which leads to a frequency modulation of the Larmor frequency. This translates into a transmitted light intensity spectrum given by $\omega_L \pm n\omega_{exc}$. The bandwidth of self-oscillating magnetometers is known to be limited only by the photodetection bandwidth. As anticipated, this mode of operation allows an unperturbed detection of large number of harmonics as evidenced by our recording of a square wave-modulated magnetic field (157 Hz) with a bandwidth exceeding 10 kHz.

References

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