Evading surface and detector frequency noise in harmonic oscillator measurements of force gradients

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We introduce and demonstrate a method of measuring small force gradients acting on a harmonic oscillator in which the force-gradient signal of interest is used to parametrically up-convert a forced oscillation below resonance into an amplitude signal at the oscillator's resonance frequency. The approach, which we demonstrate in a mechanically detected electron spin resonance experiment, allows the force-gradient signal to evade detector frequency noise by converting a slowly modulated frequency signal into an amplitude signal. © 2010 American Institute of Physics.

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Many precision measurements rely on registering a signal of interest as a change in the amplitude or frequency of an oscillator. In theory, the ultimate precision of such measurements is limited by quantum-mechanical measurement noise; in practice, the precision achievable in an oscillator measurement is often limited by thermomechanical position fluctuations,^{1,2} detector noise,¹ or environmental fluctuations.³⁻⁵ If one is using the oscillator to detect a time-varying *force*, then the requirements for achieving thermally limited or quantum-limited sensitivity can be relaxed by using parametric amplification⁶⁻⁸ to raise both the displacement signal and the oscillator's position-fluctuation noise above the detector's noise floor. Here we propose and demonstrate using parametric amplification to evade both detector and surface frequency noise when using a cantilever to detect a *force-gradient* signal.

Forces acting on microcantilevers are routinely measured at the thermomechanical limit, where the smallest detectable force is set by the force fluctuations, $P_F = 4k_b T \Gamma$, giving rise to the friction Γ experienced by the oscillator. Given the finite sensitivity of displacement sensors, however, achieving thermally limited sensitivity in a force measurement usually requires modulating the signal force at or near the cantilever frequency, f_c , where the resulting displacement is amplified by the mechanical quality factor of the cantilever. In many cases, such modulation is inconvenient or impossible for $f_c \approx 1$ kHz. In magnetic resonance force microscopy (MRFM), for example, force modulation at f_c is often impossible because of the sample's unfavorable spin relaxation times.^{9,10} In electric force microscopy, force detection is inconvenient because of the undesirably long natural response time of the cantilever near resonance^{1,11} and the finite charging time of the sample. These shortcomings are obviated by detecting the signal as a (slowly modulated) force gradient,^{1,9-11} δk . In a force-gradient experiment, the cantilever is driven into self-oscillation via positive feedback¹ and the (modulated) force-gradient signal shifts the instantaneous frequency of the cantilever, $\delta f_c \approx f_c \delta k / 2k_c$, where k_c is the cantilever spring constant.

Achieving thermally limited sensitivity in a force-gradient experiment remains challenging, however. This is illustrated in Fig. 1, in which we plot the power spectrum of cantilever frequency fluctuations seen in three representative magnetic resonance force microscope experiments (detailed below). In the first experiment (solid black line), the cantilever was brought to a height $h=30$ nm above a gold-coated surface, driven to a root-mean-square amplitude of $x_{\text{rms}}=73$ nm, and the tip-sample potential adjusted to minimize tip charge and therefore frequency noise. The observed frequency noise (solid black line) is a sum of following three contributions: (1) thermomechanical position fluctuations (dotted line) with power spectrum $P_{\delta f_c}^{\text{therm}} = 4k_b T \Gamma (f_c / 2k_c x_{\text{rms}})^2$, (2) detector noise (dashed line) having a power spectrum $P_{\delta f_c}^{\text{det}} = P_{\delta x}^{\text{det}} f^2 / x_{\text{rms}}^2$, where $P_{\delta x}^{\text{det}}$ is the detector noise written as an equivalent cantilever displacement fluctuation, and (3) surface noise (dotted-dashed line) $P_{\delta f_c}^{\text{surf}}(f) \propto f^{-1}$ arising from uncompensated tip charge coupling to fluctuating electric field gradients produced by the sample.⁵

In the second and third experiments, the magnetization of unpaired electron spins in the sample was modulated to create the force-gradient signal at 6.28 Hz (blue line) and 100 Hz (green line), respectively, in Fig. 1. We can see in the figure that even though the modulation frequency was cho-

FIG. 1. (Color online) Power spectrum of cantilever frequency fluctuations with (peaks S_1 and S_2) and without (solid baseline) a modulated force-gradient signal present. Dashed-dotted black line: surface noise. Dotted black line: thermal noise ($P_{\delta f_c}^{\text{therm}} = 2.8 \times 10^{-7}$ Hz²/Hz). Dashed line: detector noise [$P_{\delta x}^{\text{det}} = (10 \text{ pm})^2/\text{Hz}$]. Noise at frequencies away from the signal peak in the blue and green traces has been removed for clarity.

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sen optimally, the noise in the 6.28 Hz experiment was nevertheless dominated by surface noise; consequently, the observed signal-to-noise ratio S_1/N_S is smaller than the thermally limited signal-to-noise ratio S_1/N_T by a factor of 14. Modulating at 100 Hz does avoid surface noise but the noise is dominated instead by detector noise and the observed signal-to-noise ratio is even worse: S_2/N_D is smaller than S_2/N_T by a factor of nearly 10^3 .

To achieve a thermally limited signal-to-noise ratio in this representative force-gradient experiment would require modulating at a frequency $f_{\text{mod}} \gg 80$ Hz and operating with a detector having a position sensitivity of $P_{\delta x}^{\text{det}} \ll (390 \text{ fm})^2/\text{Hz}$, 26 times smaller than we currently achieve using optical fiber interferometry. Here we introduce a parametric amplification scheme that (1) is compatible with a force-gradient measurement; (2) can be used with a modulated signal, allowing the signal to evade the effect of surface frequency noise; and (3) converts a frequency signal to an amplitude signal at f_c , evading detector frequency noise. In contrast with other applications of parametric amplification in which an externally supplied force gradient amplifies a small force signal, here the signal of interest acts as the amplifier.

As in previous work,¹⁰ a force-gradient signal was generated by interacting unpaired electron spins in a gold-coated film of TEMPAMINE with a high-compliance magnetic tipped cantilever. Experiments were carried out in vacuum ($P=10^{-6}$ mbar) and at cryogenic temperatures ($T=8$ K). The cantilever had $f_c=4829$ Hz, $k_c=7.8 \times 10^{-4}$ N/m, a mechanical quality factor $Q=3.8 \times 10^4$, and a nickel tip of radius $r=2 \mu\text{m}$. A (swept) magnetic field of $B_0=0.50$ T to 0.85 T was applied to polarize the sample spins and a pulsed 17.28 GHz transverse magnetic field from a half-wave microstripline resonator was applied to saturate sample spins. Spin magnetization μ_z in the sample interacted with the magnetic field from the magnetic tip to create a force gradient, Δk_{spin} , which shifted the resonance frequency of the cantilever;^{9,10} $\Delta k_{\text{spin}} = \sum_j \mu_{z,j} \partial^2 B_z^{\text{tip}}(\mathbf{r}_j) / \partial x^2$ where the sum is over all spins in resonance at the given applied field. A small potential, $V_{\text{dc}} \approx 1$ V, was applied between the cantilever and the gold sample coating to control the charge on the cantilever tip. The cantilever was driven below resonance at a frequency $f_d=48f_c/49=4730$ Hz by applying an oscillating voltage $V_{\text{dc}}=33$ V_{rms} from a waveform generator (Agilent 33250A) to a nearby wire. The force applied to the cantilever from the drive wire is

$$F_{\text{wire}}(t) = \frac{1}{2} C' [V_{\text{dc}} + \sqrt{2} V_{\text{ac}} \cos(2\pi f_d t)]^2, \quad (1)$$

where $C' = \partial C / \partial x$ is the derivative of the wire-cantilever capacitance with respect to the direction of cantilever motion. The resulting amplitude of motion at frequency f_d , $x_d = \chi(f_d) F_d / k_c \approx C' V_{\text{dc}} V_{\text{ac}} \chi(f_d) / k_c$, with $\chi(f_d) \approx (1 - f_d^2 / f_c^2)^{-1}$ the susceptibility and F_d the component of F_{wire} oscillating at f_d , x_d was 99 nm for experiments here. During the experiment, the effective Q of the cantilever was reduced to $Q_{\text{eff}} = 3 \times 10^3$, using negative feedback applied by a piezo at the cantilever base. The sample spin magnetization, μ_z , was modulated at a frequency $f_p = f_c / 49 = f_d / 48 = 98.55$ Hz by pulsing the microwave field in synchrony with the cantilever motion.¹⁰

FIG. 2. Power spectral density of cantilever motion: (a) thermomechanical displacement fluctuations; (b) with negative feedback active while the cantilever was driven at a frequency $f_d = f_c - f_p = 4730$ Hz; and (c) identical condition to (b) but with a microwave field pulsed at $f_p = 98.55$ Hz. Conditions: $B_0 = 0.5925$ T and $h = 200$ nm.

The equation of motion governing the cantilever displacement x is

$$\ddot{x} + \frac{\omega_c}{Q_{\text{eff}}} \dot{x} + \omega_c^2 x + \frac{\omega_c^2 \sqrt{2} \delta k_{\text{spin}} \cos(\omega_p t)}{k_c} x = \frac{\omega_c^2 \sqrt{2} F_d}{k_c} e^{i\omega_d t}, \quad (2)$$

where we have expressed frequencies in angular units, $\omega_p = \omega_c - \omega_d$ and $\delta k_{\text{spin}} = \sqrt{2} \Delta k_{\text{spin}} / \pi$ includes only the first Fourier component of the pulse modulation. We look for a solution of the form $x(t) = \sum_{n=-\infty}^{\infty} a_n e^{i(n\omega_p + \omega_d)t}$ and are particularly interested in two coefficients: $a_0 / \sqrt{2} = x_d$ and $a_1 / \sqrt{2} \equiv \delta x_{\text{spin}}$, the amplitude of the spin-induced oscillation at the cantilever's resonance frequency. We find, for $Q_{\text{eff}} \gg 1$,

$$\delta x_{\text{spin}} = \frac{i \sqrt{2} Q_{\text{eff}} x_d}{2 k_c} \delta k_{\text{spin}}. \quad (3)$$

The central prediction of Eqs. (2) and (3) is that the spin-induced spring constant modulation δk_{spin} at frequency f_p acts to up-convert some of the oscillation at frequency f_d to an oscillation at frequency f_c . The data in Fig. 2 verify this prediction. For reference, Fig. 2(a) shows the power spectrum of thermomechanical motion of the nascent cantilever. In Fig. 2(b) we see the thermomechanical motion peak near 4829 Hz broadened by negative feedback and, in addition, a large peak near 4730 Hz due to the applied drive. In Fig. 2(c) the microwaves have been turned on and an additional narrow peak can be seen, near 4829 Hz, on top of the damped thermomechanical motion. This narrow peak demonstrates the up-conversion of a force-gradient frequency signal at f_p to an amplitude signal at f_c .

We next demonstrated that our parametric up-conversion technique was effective at evading detector noise, allowing us to modulate fast enough to also avoid surface noise. Figure 3(a) is a plot, versus external magnetic field, of the Fourier component at $f_p = 6.28$ Hz of the spin-induced cantilever frequency shift. This modulation rate was chosen to minimize the frequency noise due to the surface and the detector (Fig. 1). The shape of the resulting magnetic resonance signal arises from considering that spins are in resonance with the sum of the tip field and the external field and weighting

FIG. 3. (Color online) Cantilever magnetic resonance recorded via modulated force-gradient detection and parametric up-conversion amplitude detection. (a) Optimal frequency-shift measurement ($f_p=6.28$ Hz, RMS amplitude 99 nm and a background of -14.8 mHz subtracted). (b) Surface-noise evading frequency shift measurement [$f_p=100.57$ Hz but otherwise identical to (a)]. (c) Detection of magnetic resonance via parametric up-conversion of a frequency-shift signal (open circles, $f_p=98.55$ Hz and a 0.53 nm background subtracted). The solid line is the signal predicted from Eq. (3) and the frequency-shift signal in (a), scaled to account for the difference in modulation frequencies (Ref. 12) (scale factor=0.70). Conditions: 2.5 mT/pt field step and detection bandwidth $b=1$ Hz.

each spin in resonance by the second derivative of the tip field $\partial^2 B_z^{\text{tip}}(\mathbf{r}_j)/\partial x^2$ at the location $\mathbf{r}_j$ of each spin.¹⁰ Increasing the modulation rate to $f_p=100.57$ Hz, so that surface frequency noise will no longer affect the measurement, does not improve the signal-to-noise [Fig. 3(b)]. This is as expected given the large detector noise apparent at $f=100$ Hz in Fig. 1.

Using a spin modulation rate of $f_p=98.55$ Hz and the parametric up-conversion scheme described above, in contrast, succeeds in evading surface noise. The up-converted amplitude experiment, Fig. 3(c), has a signal-to-noise ratio equivalent to that seen in the optimal frequency-shift experiment, Fig. 3(a). The observed signal (circles) agrees quantitatively with the signal predicted (line) using Eq. (3), calculated using the observed frequency-shift signal in Fig. 3(a), x_d , and Q_{eff} as inputs.¹²

Given the power spectrum of frequency fluctuations observed in Fig. 1, we initially expected the up-conversion experiment to have a signal-to-noise closer to the thermomechanical limit (S_2/N_T in Fig. 1). We found, however, that the signal-to-noise in the up-conversion experiment depended critically on the quality of the sine wave used to drive the cantilever off-resonance at f_d . To explain this finding, we calculated the power spectrum $P_{\delta x_n}(\omega_c)$ of cantilever amplitude noise arising from voltage fluctuations in the drive source by adding a voltage-noise term, $\delta V_n(t)$, to Eq. (1). Voltage fluctuations will be a negligible source of noise when $P_{\delta x_n}(\omega_c) \ll P_{\delta x}^{\text{therm}}(\omega_c)$, the power spectral density of cantilever thermomechanical fluctuations. Meeting this condition requires a source with noise voltage power spectrum that satisfies

$$\frac{4k_b T \Gamma |\chi(f_d)|^2}{k_c^2 x_d^2} \gg \frac{P_{\delta V_n}(\omega_c)}{V_{ac}^2} + \frac{P_{\delta V_n}(\omega_c + \omega_d)}{2V_{dc}^2} + \frac{P_{\delta V_n}(\omega_c - \omega_d)}{2V_{dc}^2}. \quad (4)$$

We conclude that the drive oscillator will contribute negligibly to cantilever position fluctuations at resonance if its voltage amplitude noise is much smaller than -97 dBc/Hz at $f_c=4829$ Hz and much smaller than -125 dBc/Hz at 9559 and 98.55 Hz. Although the amplitude noise for our untuned audiofrequency drive oscillator is unspecified, we note that this level of amplitude noise is challenging to achieve even with a high- Q tuned radiofrequency oscillator.

In conclusion, we have introduced an approach for detecting minute force gradients acting on an harmonic oscillator and have demonstrated that the approach enables the measurement to evade surface and detector frequency noise. We have thus achieved a similar result to Budakian *et al.*,¹³ but without the need to modulate spin magnetization at f_c and in an experiment which is conceptually simpler, easier to implement, and applicable to other forms of scanned probe microscopy beyond MRFM. Calculations indicate that further improvements in the electrical noise in the drive oscillator should enable the technique to better approach thermally limited sensitivity. Although we have chosen to demonstrate this technique using MRFM, we believe the approach is generally applicable to any oscillator force-gradient measurement.

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¹²See supplementary material at <http://dx.doi.org/10.1063/1.3465906> for measurement of the scale factor.

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