Thermal nonlinearities in a nanomechanical oscillator

Jan Gieseler¹, Lukas Novotny²* and Romain Quidant¹,³*

Nano- and micromechanical oscillators with high quality (Q)-factors have gained much attention for their potential application as ultrasensitive detectors. In contrast to micro-fabricated devices, optically trapped nanoparticles in vacuum do not suffer from clamping losses, hence leading to much larger Q-factors. We find that for a levitated nanoparticle the thermal energy suffices to drive the motion of the nanoparticle into the nonlinear regime. First, we experimentally measure and fully characterize the frequency fluctuations originating from thermal motion and nonlinearities. Second, we demonstrate that feedback cooling can be used to mitigate these fluctuations. The high level of control allows us to fully exploit the force-sensing capabilities of the nanoresonator. Our approach offers a force sensitivity of 20 zN Hz⁻¹/², which is the highest value reported so far at room temperature, sufficient to sense ultraweak interactions, such as non-Newtonian gravity-like forces.

Recent developments in optomechanics have evolved towards smaller and lighter resonators featuring high Q-factors, which are important for the sensing of tiny masses¹⁻³, charges⁴, magnetic fields⁵ and weak forces⁶⁻⁹. The presence of a force field or the adhesion of a small mass induces a change in the mechanical response and can be monitored by tracking either the oscillation frequency, phase or its amplitude. Ultimately, dissipation losses as well as thermomechanical noise and temperature fluctuations limit the Q-factors of clamped oscillators and consequently their sensing performance⁷⁻⁹. This can be circumvented by using an optically trapped nanoparticle in high vacuum. Indeed, the Q-factor of a levitated particle is limited only by collisions with residual air molecules and can potentially reach 10¹² for small particles in ultrahigh vacuum¹⁰⁻¹⁴. Here, we first show that an optically trapped nanoparticle is sufficiently sensitive that thermal forces drive it out of its linear regime. In addition, we demonstrate that feedback cooling can be used to mitigate frequency fluctuations associated with the thermal nonlinearities, thereby recovering the force-sensing capabilities of the oscillator.

In our experiment, a silica nanoparticle with a diameter of ~75 nm is trapped at room temperature in the focal region of a tightly focused near-infrared laser beam (λ = 1.064 nm, polarized along the x axis). The intensity near the focus of the objective (NA = 0.8) can be well approximated by Gaussian functions (Fig. 1), defining a focal volume of w_x × w_y × w_z ≈ 0.69 μm × 0.54 μm × 1.36 μm (Supplementary Information). For large displacements, the optical potential becomes anharmonic featuring a Duffing nonlinearity. For a Gaussian field distribution, the nonlinear coefficients are given by

\[ \xi_i = -2/w_i^2 \]

where w_i is the beam waist radius (i = x, y) or Rayleigh range (i = z). For small displacements \[ |x| < |\xi_i|^{-1/2} \], the nonlinearity is negligible and the three motional degrees of freedom decouple. Owing to the asymmetry of the optical focus, the oscillation frequencies \[ \Omega_i = (k_i/m)^{1/2} \] along the three main axes are different (\[ \Omega_x/2\pi = 125 \text{ kHz}, \Omega_y/2\pi = 135 \text{ kHz}, \Omega_z/2\pi = 37 \text{ kHz} \]). Here m is the mass of the particle and the linear trap stiffness is given by \[ k_i = \alpha E_0^2/w_i^4 \], where \[ E_0 \] is the electric field intensity at the focus. The gradient of the optical intensity distribution exerts a restoring force \[ F_i^{\text{opt}} = -k_i(1 + \sum_{j=x,y,z} \xi_j x_j) x_i \] on a dipolar particle with polarizability \[ \alpha \] that is displaced from the trap centre by \[ x_i \]. For a sphere of radius \[ a \] and dielectric constant \[ \epsilon_r \], the polarizability is \[ \alpha = 4\pi a^3 \epsilon_0 (\epsilon_r - 1)/(\epsilon_r + 2) \], with \[ \epsilon_0 \] being the vacuum permittivity.

We experimentally determine the nonlinear coefficients by parametric excitation through modulation of the trapping laser at a frequency close to the parametric resonance \[ \Omega_{\text{mod}} \approx 2\Omega_i \] (Supplementary Information) and find \[ (\xi_x, \xi_y, \xi_z) = (−7.95, −10.41, −0.98) \mu m^{-2} \], in good agreement with the values estimated from the size of the focus (equation (1)).

The equation of motion for each spatial degree of freedom (\[ i = x, y, z \]) is given by

\[ \ddot{x}_i + \Omega_i^2 x_i + \sum_{j=x,y,z} \xi_j x_j = F_{\text{focu}}/m \]

¹ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, 08860 Castelldefels (Barcelona), Spain, ²Photonics Laboratory, ETH Zürich, 8093 Zürich, Switzerland, ³ICREA-Institució Catalana de Recerca i Estudis Avançats, 08010 Barcelona, Spain. *e-mail: lnovotny@ethz.ch; romain.quidant@icfo.es

Figure 1 | Experimental configuration. A silica nanoparticle is trapped by a tightly focused laser beam. Random collisions with residual air molecules drive the particle into the nonlinear regime of the potential. Inset: the focal intensity distribution forms a trap that can be approximated by a Gaussian potential (white). The deviation from a harmonic potential (red) is described by a Duffing nonlinearity (Supplementary Information).
is the average velocity $Dm$ bar). These pressures correspond $Q h$ allows us to continuously $D m$ to the equipartition principle $D m = D m T$. The thermal amplitude eventually becomes comparable to the particle size. Consequently, a proper description of the particle motion requires the inclusion of nonlinearities. These give rise to a frequency shift $\Delta \Omega_{NL} = \frac{3}{8} \xi \Omega_0 r_{th}^2$.

In contrast to linear thermal frequency fluctuations, nonlinear frequency fluctuations add frequency noise but do not affect the damping.

To resolve the nonlinear frequency shift originating from thermal motion, the nonlinear contribution must be larger than the linear one, that is

$$R = \frac{\Delta \Omega_{NL}}{\Omega_0} = \frac{3\xi Q k_B T}{4 \Omega_0^2 m} \gg 1$$  \hspace{1cm} (3)$$

where $T$ is the temperature of the residual gas and $k_B$ is Boltzmann’s constant. To fulfill the condition in equation (3) a light and high-$Q$ mechanical resonator is required. In our experiment, $m = 3 \times 10^{-18}$ kg and $Q = 10^6$, as determined in a ring-down measurement at a pressure of $P = 0.5 \times 10^{-6}$ mbar. These parameters place us well into the nonlinear regime. Importantly, the dependence of the $Q$-factor on pressure $P$ allows us to continuously tune the system between the linear and nonlinear regimes.

To demonstrate the differences between a thermally driven harmonic oscillator ($R \ll 1$) and an anharmonic oscillator ($R \gg 1$), we compare the particle’s motion at high pressure (6 mbar) and at low pressure (1.2 $\times 10^{-2}$ mbar). These pressures correspond to $Q$-factors of 25 and 12 $\times 10^4$, respectively. At high pressures (low $Q$) the dominant source of frequency fluctuations is linear damping $\Delta \Omega_h = \Gamma_0$ and the power spectral density (PSD) of the particle motion features a single symmetric Lorentzian peak, whose width is equal to the linear damping coefficient $\Gamma_0$ (Fig. 2e). In contrast, at low pressure (high $Q$) nonlinear frequency fluctuations $\Delta \Omega_{NL} = 3\xi \Omega_0/8r_{th}^2$ dominate and we observe an asymmetric

Figure 2 | Nonlinearity-induced frequency fluctuations. a, Time trace of the particle motion along $x$ at $1.2 \times 10^{-2}$ mbar. The oscillation amplitude changes randomly and the positions are normally distributed. b,c. However, on timescales that are short compared with the relaxation time $\tau$, the particle motion is sinusoidal with a constant oscillation amplitude over many cycles and has a position distribution featuring two lobes (a right). The part of the time trace when the particle oscillates with a large (small) amplitude is shown in red (green). d, From the long time trace (black) and the short time traces (red, green) we calculate the PSD. For short observation times we observe a Fourier-limited symmetric PSD with an amplitude-dependent centre frequency. The overall PSD (black) results from a temporal average of the instantaneous PSDs (red, green). In contrast, the PSD of a low-$Q$ oscillator (blue) is described by a Lorentzian peak at all times (red thin line). e, Time trace of particle motion at 6 mbar used to calculate the (blue) Lorentzian PSD in d.
peak that is considerably broader than what is expected for the equivalent linear oscillator. However, if we limit the observation time to time intervals $1/\Delta \Omega_{NL} < \tau < 1/\Delta \Omega_s$, we find a clean oscillation with an almost constant amplitude, corresponding to a narrow and symmetric peak in the frequency domain. For large oscillation amplitudes the peak seems downshifted, consistent with the measured negative Duffing nonlinearity. Consequently, for observation times $\gg 1/\Delta \Omega_{NL}$, the non-Lorenzian peak becomes a weighted average:

$$
S_{NL}(\Omega) = \int \rho(E)S_i(\Omega, E) \, dE
$$

over Lorenzian peaks centred at the shifted frequency $\tilde{\Omega}_0(E) = \Omega_0 + 3\xi/(4m\tilde{\Omega}_0) E$ and weighted by the Gibbs distribution $\rho(E) = Z^{-1} \exp(-E/k_B T)$. Here, $Z = \int \rho(E) \, dE$ is the partition function and

$$
S_i(\Omega, E) = \frac{E}{\pi m \tilde{\Omega}_0^2 (\Omega - \tilde{\Omega}_0(E))^2 + (\Gamma_0/2)^2}
$$

is the PSD of a harmonic oscillator with frequency $\tilde{\Omega}_0(E)$ and energy $E$. Note that equation (4) is valid in the low friction limit ($Q \gg 1$). An expression that holds for arbitrary $R$ can be found in ref. 17.

In what follows we present further evidence that the observed frequency fluctuations are a consequence of thermal motion in the nonlinear optical potential. We analyse the correlations between the instantaneous frequencies and energies and we investigate the pressure dependence of the frequency fluctuations.

To quantify the frequency fluctuations, we continuously measure the instantaneous energy $E_i(t)$ and frequency $\Omega_i(t)$ of the three spatial modes ($i = x, y, z$), given by the area and the position of the maximum of the instantaneous PSDs, respectively (Fig. 2e). The instantaneous PSDs are calculated from position time traces $x_i(t)$ (where $t - \tau/2 < t < t + \tau/2$) of $\tau = 20 \text{ ms}$ duration. Figure 3a,b shows the time traces of the relative frequency fluctuations $\Omega_i/(\Omega_0)$ of the three modes for high Q (low pressure) and low Q (high pressure), respectively. Figure 3c shows the correlations between $E_i$ and $\Omega_i$ as a function of pressure, calculated from 30-min-long time traces. The nonlinearity is conservative and, thus, does not change the particle energy, which is determined only by random molecule collisions. Therefore, the energies of the three degrees of freedom are uncorrelated. In contrast, a change in energy of one mode shifts the frequency of all modes (equation (2)). At low pressure ($R > 1$), the nonlinearities dominate and the frequency fluctuations are highly correlated. In contrast, at high pressure ($R < 1$), linear damping dominates and consequently the frequencies become uncorrelated.

In Fig. 3d we plot the oscillator amplitude response to parametric modulation (black) and to thermal excitation (scatter plot) at a pressure of $10^{-5} \text{ mbar}$. Independently of the origin of the excitation, there is a linear relationship between the particle’s energy and the particle’s oscillation frequency. However, whereas the response to an excitation at a fixed frequency is sharp, the thermal response is downshifted and much broader. Indeed, according to equation (2), an average thermal excitation of the orthogonal modes ($y$ and $z$) leads to an average shift in the frequency of the mode under consideration (here $x$). The shift of the centre of the frequency distribution ($\approx 2.5 \text{ kHz}$) is in good agreement with the value estimated from the measured nonlinear coefficients and average thermal amplitudes (Supplementary Information). In addition, because the energies and hence the amplitudes of the modes fluctuate independently (Fig. 3c), we get a distribution of frequencies for a given oscillation amplitude of the considered

\[ S_{NL}(\Omega) = \int \rho(E)S_i(\Omega, E) \, dE \]
mode. In contrast, when we drive the system parametrically at a fixed frequency, we simultaneously cool the orthogonal modes. Therefore, the contribution to the frequency shift originating from the orthogonal modes is negligible and we obtain a sharp response. In Fig. 4 we show the PSD of the relative frequencies $\Omega_i/\Omega_{i0}$ (IPSD) as a function of pressure. The IPSDs are calculated from 30-min-long time traces of the instantaneous frequencies (Fig. 3a,b). For low $Q_i$, the IPSD is flat as expected for a harmonic oscillator. In contrast, for high $Q_i$, the nonlinear coupling maps the Lorenzian PSD of the amplitude onto the frequency PSD, which can be described by

$$S_i(\omega) = I \frac{\Omega_i/\pi}{\Omega_i^2 + \omega^2} \quad (5)$$

where $I$ is the total spectral power, which is independent of pressure. The characteristic cutoff frequency $\Omega_c$ has a clear pressure dependence, as shown in Fig. 4c. This further confirms that the fluctuations arise as a combination of nonlinearities and thermal excitations.

The small mass and high $Q$-factor make the levitated nanoparticle an ultrasensitive force sensor with a sensitivity of $S_i = 4k_B T m_0 \Omega_c/Q \approx (20 \pi N^2) Hz^{-1}$ at room temperature, which is comparable to the best values achieved at cryogenic temperatures. There are several ways to couple a force to a levitated nanoparticle, including electric$^{20,21}$ and magnetic$^{20,23}$ coupling or more subtle forces due to fluctuating charges within the particle. In addition, in contrast to trapped ions, the still considerable mass of the particle makes it susceptible to gravitational forces.

In addition to constant forces, a levitated nanoparticle can detect position-dependent forces. A position-dependent force causes a frequency shift. Therefore, following the resonance frequencies of the nanoparticle allows for measuring a force gradient with a precision given by the noise floor of the frequency fluctuations. In practice though, as shown above, nonlinear effects lead to frequency fluctuations in ultrahigh-$Q$ oscillators. It is possible to surpass this limit by operating the oscillator at special points$^{25}$. Furthermore, as we show in the following, nonlinear frequency fluctuations can also be suppressed by feedback cooling$^{11}$. Feedback cooling lowers the oscillation amplitude and therefore reduces the thermal motion of the oscillator. Under the action of feedback, the effective temperature reduces to $T_{ef} = (T_0/I_0) T$, where $I_0$ is the total damping with feedback$^{11,26}$. As shown in Fig. 4d, we manage to reduce the frequency fluctuations to the level of the laser intensity fluctuations, which become the main source of frequency noise. Using active stabilization techniques, relative laser intensity noise can be brought to the level of $10^{-8}/\sqrt{Hz}$ (ref. 27).

We demonstrate the improved sensitivity by mimicking a periodic potential landscape. A modulation at 50 mHz is applied to the trapping laser. The modulation causes a variation of the force gradient, which is measured as a frequency shift. As shown in Fig. 5, without feedback the signal is overwhelmed by noise whereas with feedback, the fluctuations are suppressed down to the level of laser intensity fluctuations and the applied signal is clearly visible. Using feedback cooling, we are able to improve the sensitivity of the oscillator by two orders of magnitude and achieve a relative frequency noise floor $\sqrt{S_i/\Omega_c}$ of $3 \times 10^{-3}/\sqrt{Hz}$ for frequencies below 1 Hz and $1 \times 10^{-4}/\sqrt{Hz}$ for frequencies larger than 10 Hz. In the absence of laser intensity noise the highest sensitivity is obtained when linear and nonlinear fluctuations contribute equally. As feedback cooling reduces both the effective temperature $T_{ef}$ and the effective quality factor $Q_{ef} = \Omega_0/I_{0b}$, there is an optimum feedback gain where the effective $Q$-factor is $Q_{ef}^{opt} = R^{-1/2} Q$ (Supplementary Information). Thus, under the assumption that with cooling the oscillation remains in the linear regime, the frequency spectral density is given by (Supplementary Information)

$$S_f^{(opt)} = \frac{\Omega_0}{Q_{opt}} = \frac{\Omega_0}{Q} R^{1/2}$$
For the values presented here we obtain $\sqrt{S_{\text{opt}}^0} / Q_0 \sim 10^{-6} / \sqrt{Hz}$, sufficient to sense ultraweak interactions, such as non-Newtonian gravity-like forces\textsuperscript{26}. However, to reach this value, relative intensity fluctuations have to be suppressed down to the same level. The ultimate cooling limit is defined by the zero point motion $r_{zp} = \sqrt{\eta/2mT_0}$. To resolve the nonlinear frequency shift due to the zero point motion $\Delta \Omega_{zp}$, the condition

$$\frac{\Delta \Omega_{zp}}{\Delta \Omega_l} = \frac{3}{8} Q_0 r_{zp} \gg 1$$

has to be satisfied in analogy to equation (3). This requires a $Q$-factor of $Q = \Omega_0 / \Delta \Omega_0 \approx 10^{10}$. In the absence of other noise sources, this regime is reached for pressures below $10^{-9}$ mbar.

In conclusion, we have demonstrated that a laser-trapped nanoscale particle in high vacuum defines an ultrasensitive force sensor. The thermal motion of the residual gas drives the nanoparticle into its nonlinear regime, which gives rise to frequency fluctuations. Using a parametric feedback cooling scheme, we can stabilize the nanoparticle and suppress its nonlinearities, without sacrificing sensitivity. We expect that feedback-controlled nanoparticles will find applications for sensing a wide range of interactions, including van der Waals and Casimir forces\textsuperscript{28}, nuclear spins\textsuperscript{4} and gravitation\textsuperscript{24}.

Received 3 July 2013; accepted 30 September 2013; published online 24 November 2013

References
1. Chastel, I. et al. A nanomechanical mass sensor with yoctogram resolution. Nature Nanotech. 7, 301–304 (2012).
2. Yang, Y. T., Callegari, C., Feng, X. L., Ekinci, K. L. & Roukes, M. L. Zeptron-scale nanomechanical mass sensing. Nano Lett. 6, 583–586 (2006).
3. Cleland, A. N. & Roukes, M. L. A nanometre-scale mechanical electrometer. Nature 392, 160–162 (1998).
4. Ruga, D., Budakian, R., Mamin, H. J. & Chui, B. W. Single spin detection by magnetic resonance force microscopy. Nature 430, 329–332 (2004).
5. Stipe, B. C., Mamin, H. J., Stowe, T. D., Kenny, T. W. & Ruga, D. Noncontact friction and force fluctuations between closely spaced bodies. Phys. Rev. Lett. 87, 096801 (2001).
6. Moser, J. et al. Ultrasensitive force detection with a nanotube mechanical resonator. Nature Nanotech. 8, 493–496 (2013).
7. Postma, H. W. C., Koizinsky, I., Husain, A. & Roukes, M. L. Dynamic range of nanotube- and nanowire-based electromechanical systems. Appl. Phys. Lett. 86, 221105 (2005).
8. Cleland, A. N. & Roukes, M. L. Noise processes in nanomechanical resonators. J. Appl. Phys. 92, 2758–2769 (2002).
9. Ekinci, K. L., Yang, Y. T. & Roukes, M. L. Ultimate limits to inertial mass sensing based on nanoelectromechanical systems. J. Appl. Phys. 95, 2682–2689 (2004).
10. Ashkin, A. Optical levitation by radiation pressure. Appl. Phys. Lett. 19, 283–285 (1971).
11. Gieseler, J., Deutsch, B., Quindant, R. & Novotny, L. Subkelvin parametric feedback cooling of a laser-trapped nanoparticle. Phys. Rev. Lett. 109, 103603 (2012).
12. Li, T., Kheifets, S. & Raizen, M. Millikelvin cooling of an optically trapped microsphere in vacuum. Nature Phys. 7, 527–530 (2011).
13. Romero-Isart, O., Juan, M. L., Quindant, R. & Cirac, I. J. Toward quantum superposition of living organisms. New J. Phys. 12, 033015 (2010).
14. Chang, D. et al. Cavity opto-mechanics using an optically levitated nanosphere. Proc. Natl Acad. Sci. USA 107, 1005–1010 (2010).
15. Epstein, P. S. On the resistance experienced by spheres in their motion through gases. Phys. Rev. 22, 710–713 (1933).
16. O’Hanlon, J. F. A User’s Guide to Vacuum Technology 3rd edn (Wiley, 2003).
17. Dykman, M. I. & Keivoglaz, M. A. Theory of nonlinear oscillator interacting with a medium. Phys. Rev. 5, 265–441 (1984).
18. Lifshitz, R. & Cross, M. C. Review of Nonlinear Dynamics and Complexity (Wiley-VCH, 2009).
19. Dykman, M. I., Mannella, R. R., McClintock, P. V. E., Soknin, S. M. & Stocks, N. G. Noise-induced narrowing of peaks in the power spectra of underdamped nonlinear oscillators. Phys. Rev. A 42, 7041–7049 (1990).
20. Neukirch, L. P., Gieseler, J., Quindant, R., Novotny, L. & Nick Vamivakas, A. Observation of nitrogen vacancy photoluminescence from an optically levitated nanodiamond. Opt. Lett. 38, 2976–2979 (2013).
21. Geisselmann, M. et al. Three-dimensional optical manipulation of a single electron spin. Nature Nanotech. 8, 175–179 (2013).
22. Zurita-Sánchez, J., Greifelt, J.-J. & Novotny, L. Friction forces arising from fluctuating thermal fields. Phys. Rev. A 69, 022902 (2004).
23. Knünz, S. et al. Injection locking of a trapped-ion phonon laser. Phys. Rev. Lett. 105, 013004 (2010).
24. Arvanitaki, A. & Geraci, A. A. Detecting high-frequency gravitational waves with optically levitated sensors. Phys. Rev. Lett. 110, 071103 (2013).
25. Villanueva, L. G. et al. Surpassing fundamental limits of oscillators using nonlinear resonators. Phys. Rev. Lett. 110, 177208 (2013).
26. Mertz, J., Marti, O. & Mlynek, J. Regulation of a microcantilever response by force feedback. Appl. Phys. Lett. 62, 2344–2346 (1993).
27. Seifert, F., Kwee, P., Heurs, M., Willke, B. & Danzmann, K. Laser power stabilization for second-generation gravitational wave detectors. Opt. Lett. 31, 2000–2002 (2006).
28. Geraci, A. A., Papp, S. B. & Kitching, J. Short-range force detection using optically cooled levitated microspheres. Phys. Rev. Lett. 105, 101101 (2010).

Acknowledgements
This research was funded by ETH Zurich, Fundació Privada CELLEX, ERC-QMES (No. 338763) and ERC-Plasmolight (No. 259196). We thank A. Bachtold and M. Spasenović for valuable input and help and Inaki Gonzalez for his assistance in preparing Fig. 1.

Author contributions
J.G. and L.N. developed the set-up, J.G. performed the experiments and analysed the data. L.N. and R.Q. supervised the work. All authors contributed to discussing the results and writing the manuscript.

Additional information
Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to L.N. and R.Q.

Competing financial interests
The authors declare no competing financial interests.