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Electric feedback cooling of single charged nanoparticles in an optical trap

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We demonstrate feedback cooling of the center-of-mass motion of single charged nanoparticles to millikelvin temperatures in three dimensions via applying oscillating electric fields synchronized to their optically observed motion. The observed motional temperatures at weak feedback agree with a simple model and allow us to estimate the charge number of trapped nanoparticles. The agreement between our model and experiments is confirmed by independent measurements of the charge numbers based on a shift in the oscillation frequency induced by a constant electric field. The demonstrated temperature of below 10 mK at 4 × 10⁻³ Pa is lower than that with the conventional optical cooling approach at this pressure by one to two orders of magnitude. Our results form the basis of manipulating cold charged nanoparticles and paves the way to quantum mechanical studies with trapped nanoparticles near their ground state.

Manipulating the motion of objects near their quantum ground state has been a crucial subject in diverse fields from quantum simulations [1–3] and quantum information processing [4] to precision measurements [5, 6]. Cooling atomic ions and ensembles of neutral atoms to their motional ground state has been successful [2, 7]. Specific vibrational modes of nanomaterials and micromechanical oscillators have been brought to their quantum ground state [8, 9]. However, cooling the motion of nanoparticles including more than a few atoms to their motional ground state has been an elusive goal. The main difficulty lies in the absence of an efficient mechanism for cooling.

Cold nanoparticles are expected to possess various applications such as testing quantum mechanics for macroscopic objects [10, 11], ultrasensitive force and mass sensing [12–18], and the laboratory test of the collisional dynamics of interstellar materials [19]. Up to now, cooling the motion of nanoparticles to millikelvin temperatures has been demonstrated via all-optical approaches, where trapping, observing, and cooling them are all based on light scattering [20–25]. The lowest temperature achieved with all-optical approaches is finally limited by random photon recoils [26]. To overcome the limitation from photon recoils, an all-electrical approach for highly charged particles has been proposed [27].

Here, we show that the motional temperature of single charged nanoparticles in an optical trap is efficiently lowered via the optical measurement of the nanoparticle’s position and the application of oscillating electric fields synchronized to their motion. The observed motional temperatures with the electric feedback agree with a simple model only when the mass of the nanoparticle is properly estimated through the time scale of the rethermalization of the motion after it is cooled. The agreement between our model and experimental results is confirmed by independent measurements of the charge number based on the electric-field-induced shift in the oscillation frequency.

Compared to the conventional all-optical cooling method, parametric feedback cooling (PFC) [21, 24], our method, electric feedback cooling (EFC), has two important advantages. First, while in PFC the cooling rate is proportional to $T_{eff}$ [21], EFC has a high cooling rate determined by the magnitude of applied electric fields. Second, in EFC, the feedback signal is purely electrical, and thus does not perturb the optical position measurement, whereas, in PFC, the modulation on the trapping potential for cooling can affect the position measurement. In the present work, we demonstrate $T_{eff}$ below 10 mK at 4 × 10⁻³ Pa, which is about one to two orders of magnitude lower than the values obtained with PFC at this pressure, manifesting the efficiency of our approach. We estimate that, due to the high cooling rate of our approach, photon recoil heating will not be a major obstacle to cooling to near the quantum ground state at lower pressures.

In our experiments, we trap silica nanoparticles with radii of about 100 nm in a one-dimensional optical lattice formed with a fiber laser at $\lambda = 1550$ nm [Fig. 1(a)] [28–30]. We observe the three-dimensional motion of nanoparticles with a photodetector measuring the spatio-temporal variation of the infrared light scattered by them [24]. The area of the power spectral density (PSD) calculated from the photodetector signal is proportional to $T_{eff}$ [21, 24]. In most cases, trapped nanoparticles are positively ionized when the chamber is evacuated and an ion pressure gauge is turned on [31, 32]. If charging does not occur spontaneously, we induce charging with
a corona discharge by applying about 400 V to an electrode placed near the trap region.

The motion of a trapped charged nanoparticle is continuously attenuated if an applied electric field switches its sign in phase with their motion and provides an electric force opposite to its velocity. Understanding on such a process is achieved by considering the following model. We describe the motion of nanoparticles in a specific direction in the presence of an electric field by the one-dimensional equation of motion:

$$\ddot{q} + (\Gamma_0 + \Gamma_c)\dot{q} + q_n + \Omega_0^2 q = \frac{F_0}{m}$$  \hspace{1cm} (1)

where $q$, $q_n$, $\Gamma_0$, $\Gamma_c$, $\Omega_0$, $m$, and $F_0$ denote the position of nanoparticles, the noise in the feedback signal, the damping rate due to collisions with background gases, the damping rate due to an electric force, the oscillation frequency, the mass of trapped nanoparticles, and a stochastic force from background gases, respectively. The PSD of the particle following eq.(1) is given by [21, 24, 33]

$$S(\Omega) = \frac{2k_BT_0\Gamma_0/m + \Omega^2S_n(\Omega)}{(\Omega_0^2 - \Omega^2)^2 + \Omega^2(\Gamma_0 + \Gamma_c)^2}$$  \hspace{1cm} (2)

where $k_B$, $T_0$, and $S_n(\Omega)$ are the Boltzmann constant, the temperature of background gases, and the PSD of $q_n$, respectively.

According to the kinetic theory for a particle in a gas, $\Gamma_0$ is proportional to the background pressure $P$ at our working pressure range [34]:

$$\Gamma_0 = B \frac{P}{R\rho} \quad B = (4 + \frac{\pi}{2}) \sqrt{\frac{m_{\text{Ar}}}{2\pi k_B T_0}}$$  \hspace{1cm} (3)

where $\rho$, $m_{\text{Ar}}$, and $R$ are the density of the particle, the mass of background gas molecules, and the radius of the particle, respectively. Considering that the electric force is given as an amplitude of $neV/d_{\text{eff}}$ multiplied by a sinusoidal time variation of $\dot{q}/(q_0\Omega_0)$, where $n$ is the charge number, $e$ is the elementary charge, $V$ is the applied voltage amplitude, $d_{\text{eff}} = V_{\text{cal}}/E_{\text{cal}}$ is the effective distance between electrodes calculated with a numerically simulated electric field $E_{\text{cal}}$ for an applied voltage $V_{\text{cal}}$ [Fig. 1(b)], and $q_0$ is the amplitude of the motion at equilibrium, we find $\Gamma_c = neV/(m\Omega_0 q_0 d_{\text{eff}})$.

We first consider the case of weak feedback, where the influence of $q_n$ is negligible. Eq.(2) then indicates that the PSD has the spectral width of $\Gamma_0 + \Gamma_c$, which is broadened with increasing $V$. By integrating eq.(2) to derive the variance of the position $(\langle q^2 \rangle)$, we obtain a self-consistent equation for $q_0$, resulting in the expression for $T_{\text{eff}} = m\Omega_0^2(q^2)/k_B$:

$$T_{\text{eff}} = T_0 \left( \sqrt{1 + \frac{\alpha^2 V^2}{aV}} \right)^2, \quad \alpha = \frac{ne}{2\Gamma_0 d_{\text{eff}} \sqrt{2m k_B T_0}}$$  \hspace{1cm} (4)

Our model suggests that $T_{\text{eff}}$ smoothly decreases from $T_0$ with increasing $V$. We experimentally confirm such a behavior by measuring $T_{\text{eff}}$ for various voltage amplitudes at a fixed pressure [Fig. 2(a)]. We realize cooling trapped nanoparticles in three dimensions by applying electric fields provided by three oscillators independently locked to the photodetector signal via a phase lock loop (PLL) [Fig. 1(a)] [30]. $T_{\text{eff}}/T_0$ is obtained by comparing the areas of the PSDs around the oscillation frequency with and without cooling. In the present study, we assume $T_0 = 300K$, which is expected to be a reasonable assumption at above 10 Pa [35]. For deriving the uncooled area, we use the PSD at around 10 Pa such that $T_0$ is not significantly deviate from 300K. The observed profile is in good agreement with the fitted curve based on our model, showing that our model provides a good understanding on the cooling process at weak feedback.

Upon increasing $V$, however, we observe that $T_{\text{eff}}$ deviates from eq.(4) and increases with $V$. Experimentally we find that the minimum $T_{\text{eff}}$ is obtained at around $\Gamma_0 + \Gamma_c \sim \delta_{B\text{W}}$ with $\delta_{B\text{W}} \approx 2\pi \times 6$ kHz being the bandwidth of PLL. Such a behavior is intuitively understood as follows: when the spectral width of the PSD exceeds $\delta_{B\text{W}}$, the feedback loop starts to oscillate at around $\pm \delta_{B\text{W}}$ and the feedback signal is dominated by undesirable frequency components amplifying the nanoparticle’s motion [Fig. 2(b),(c)]. At strong feedback, the feedback signal includes both cooling and heating components and the ratio between them does not only on the feedback signal amplitude but also on the phase characteristics of the PLL, making it difficult to find the analytical representation of $T_{\text{eff}}$ in the entire amplitude range. For understanding our results, we build a model based on our observation that the amplitude of the cooling component stays nearly constant $\Gamma_0 + \Gamma_c \approx \delta_{B\text{W}}$ at strong feedback. With an assumption that the feedback signal is dominated by the two sideband components with equal amplitudes, we substitute $q_n = neV[\cos(\Omega_0 t + \delta_{B\text{W}} t) + \cos(\Omega_0 t - \delta_{B\text{W}} t)]/(\sqrt{2}m\Omega_0 d_{\text{eff}})$ indicating off-resonant excitations [36]. Considering $\delta_{B\text{W}} \ll \Omega_0$, we find the representation of $T_{\text{eff}}$ at strong feedback:
FIG. 3. (color online). (a) $T_{\text{eff}}$ obtained with EFC as a function of the pressure. The error bars in $T_{\text{eff}}$ are due to the systematic error of $T_0\, (+300 K/0 K)$ estimated from ref. [35]. Our results on PFC for another nanoparticle in the direction of the optical lattice is also presented. (b) The PSDs without EFC at 16 Pa and with EFC at $4 \times 10^{-3}$ Pa.

\begin{equation}
T_{\text{eff}} = T_0 \left[ \frac{\Gamma_0}{\delta_{\text{BW}}} + \frac{(nE)^2}{10m\delta_{\text{BW}}^2 d_{\text{eff}}^2} \right]
\end{equation}

which is in agreement with observed $T_{\text{eff}}$ at large $V$ [Fig. 2(a)]. At an optimum feedback amplitude, the minimum $T_{\text{eff}}$ is approximately given by

\begin{equation}
T_{\text{eff}}^{\text{min}} = T_0 \frac{\Gamma_0}{\delta_{\text{BW}}}
\end{equation}

suggesting that decreasing $P$ (and thus $\Gamma_0$) should result in lower $T_{\text{eff}}$. By measuring $T_{\text{eff}}$ in a wide pressure range between 0.004 Pa and 10 Pa, we find that our simple model is in good agreement with experiments and observe a dramatic decrease in $T_{\text{eff}}$ as $P$ is lowered (Fig. 3). For comparison, we also show data points in previous work with PFC [21] and our results obtained with PFC. We find that within our working pressure range, the temperature obtained with EFC is lower than that with PFC by one to two orders of magnitude. The lowest observed temperature in the present work is between 6 mK and 10 mK at $4 \times 10^{-3}$ Pa for all directions, with the phonon occupation numbers of $2.5 \times 10^3$, $1.3 \times 10^3$, and $6.1 \times 10^2$ in the $x$, $y$, and $z$ directions, respectively.

We now discuss the limit of our approach at even lower pressures. From eq. (6), we anticipate that $T_{\text{eff}}$ reaches submillikelvin temperatures at $P < 10^{-3}$ Pa. Recently, photon recoil heating has been identified as the main obstacle in cooling nanoparticles to submillikelvin temperatures with PFC [26].

In the absence of feedback-induced heating, the phonon occupation number at low pressures is given by $v \sim \Gamma_0/\Gamma_c$, where $\Gamma_0 \approx 2 \pi \times 10^9$ Hz is a typical value for photon recoil heating [26]. Hence, with the cooling capability of EFC demonstrated in the present study, photon recoil heating is not expected to be a major obstacle to cooling to near the ground state. Another issue which may prevent cooling at low pressures is the measurement noise affecting the feedback signal [33]. Assuming that the feedback signal includes white noise originating from the noise in the position measurement, we take $q_n = \Gamma_c q_{mn}$ and find the representation of $T_{\text{eff}}$ for $\Gamma_0 \ll \Gamma_c$ as $T_{\text{eff}} = T_0 \Gamma_0/\Gamma_c + m\Omega_c^2 \Gamma_c S_{mn}/2k_B$ with $S_{mn}$ the PSD of $q_{mn}$. Using the typical value of $S_{mn} = 2 \times 10^{-25} \text{m}^2/\text{Hz}$ for the $y$ direction estimated with the noise floor in our system, we find that the minimum value of $T_{\text{eff}} = \sqrt{2m\Omega_c^2 S_{mn} T_0 \Gamma_0/k_B}$ is expected to be around $40 \mu$K at $P = 1 \times 10^{-6}$ Pa. Decreasing the pressure by two orders of magnitude, or placing the vacuum chamber in a cryogenic environment at 4 K, both of which are feasible, will allow us to reach near the ground state ($v \sim 2$). If the detection efficiency, mainly limited by the numerical aperture of the lens, is improved, we expect that cooling to near the ground state will be possible at even higher pressures due to decreased $S_{mn}$. Note that the noise floor for detecting the motion of nanoparticles is close to their ground state (several $\mu K$) in our setup.

In what follows, we show that EFC provides a unique means to measure the mass and the charge number of trapped nanoparticles that are crucial for understanding and predicting their behavior. The mass can be derived from $R$ obtained with eq. (3) if $\Gamma_0$ is correctly measured. We find that the reliable way to determine $\Gamma_0$ is to observe the time evolution of $T_{\text{eff}}$ in rethermalization experiments [35], instead of extracting the spectral width of the PSD that has been often used in previous work [24]. We first prepare trapped nanoparticles cooled via EFC. After EFC is turned off, we observe an exponential growth of $T_{\text{eff}}$ [Fig. 4(a)]. The time constant of the growth is...
the inverse of $\Gamma_0$. The spectral width extracted from the PSD tends to exhibit broadening by fluctuations in the oscillation frequency presumably due to the vibration of optics, the fluctuations in laser intensity, and thermal nonlinearities [28, 37]. We confirm that at our working pressure range the measured values of $\Gamma_0$ are proportional to the pressure as expected from eq.(3) [Fig. 4(b)]. The slope of this plot $g$ is used for deriving $m = 4\pi B^3/(3\rho^2g^3)$.

The value of $\alpha$ obtained by fitting eq.(4) to the cooling curve at weak feedback [Fig. 2(a)] reveals the charge number as $n = 4d_{\text{eff}}\alpha^2P/(2\pi\hbar T_0B^3/3g/pe)$, which we denote as $n_{\text{AC}}$. For the $z$ direction, we have $d_{\text{eff}} = 5.0\text{mm}$. To test whether the obtained values of $n_{\text{AC}}$ are correct, we introduce a new method for determining $n$ independently from the measurement of Fig. 2(a). We apply a dc electric field and observe an induced shift in the oscillation frequency in the $z$ direction. The presence of a gradient shifts the position of the potential minimum, at which the oscillation frequency is lowered due to the anharmonicity of the potential. By measuring the frequency shift in the direction of the optical lattice, which has a well-defined sinusoidal structure with a spacing of $\lambda/2$, we obtain the applied gradient precisely. The oscillation frequency in the $z$ direction in the presence of a dc voltage $V$ is given by [30]

$$\Omega_z = \Omega_0 \left[ 1 - \left( \frac{CV + F_0/(\pi m\lambda)}{(\Omega_0/2\pi)^2} \right)^2 \right]^{1/4} = \frac{ne}{\pi m\lambda d_{\text{eff}}}, \quad (7)$$

where $\Omega_0$ is the oscillation frequency in the absence of gradients and $F_0$ is the offset gradient mainly due to the radiation pressure by the trapping laser. We observe that the oscillation frequency varies as a dc electric field is applied, in good agreement with eq.(7) [Fig. 5(a)]. From the curvature of this plot $C$, we obtain the value of $n = 4\pi^2\lambda d_{\text{eff}}CB^3/(3\rho^2g^3e)$, which we denote as $n_{\text{DC}}$. Here we find that the two methods for determining $n$ have different dependences on $g$: $n_{\text{AC}} \propto g^{-1/2}$ and $n_{\text{DC}} \propto g^{-3}$. This fact indicates that unless the value of $g$ is properly measured we encounter an unrealistic result of $n_{\text{AC}} \neq n_{\text{DC}}$. In fact, with the conventional method for determining $g$, i.e., extracting the spectral width from the PSD [21, 24], we found large discrepancies between $n_{\text{AC}}$ and $n_{\text{DC}}$ by up to a factor of 10.

With $g$ obtained with the rethermalization measurements, we arrive at reliable values of $n$ with $n_{\text{AC}}$ and $n_{\text{DC}}$ agreeing within 25%. For the specific case of the presented data in Fig. 4 and Fig. 5(a), the mass and the charge number are $m = 5.3(3) \times 10^{-18}\text{kg}$ and $n_{\text{DC}} = 74(5)$, respectively, where the errors are dominated by the error in determining $g$. The comparison between $n_{\text{AC}}$ and $n_{\text{DC}}$ for multiple experimental runs is shown in Fig. 5(b). In our system, we have not succeeded in observing the variation of $n$ by a single $e$, which was used to determine $n$ in previous work [31]. We infer that this is because the charge variation in our system is much larger than in the previous work. The large charge variation possibly suggests that ionized gas molecules around the trap region are denser than in previous work.

In conclusion, we demonstrate an efficient approach for cooling the center-of-mass motion of single optically trapped nanoparticles via the combination of the optical observation of the particle position and the application of electric fields synchronized to their motion. Our cooling approach provides a unique means to characterize the properties of trapped nanoparticles. The demonstrated temperatures of below 10mK at $4 \times 10^{-3}\text{Pa}$ are about one to two orders of magnitude lower than those obtained with the conventional method at this pressure. The advantages of our approaches lie in the strong cooling force independent from $T_{\text{eff}}$ and the clean observation signals unaffected by feedback signals. We envision that cooling to near the ground state is achieved at a pressure of $1 \times 10^{-8}\text{Pa}$, or at a pressure of $1 \times 10^{-6}\text{Pa}$ in a cryogenic environment at 4 K. Due to the high cooling rate of our approach, photon recoil heating, which has been a severe limitation with the conventional optical cooling, will not be a major obstacle down to around the ground state; rather, the noise in the feedback signal originating from position measurements will be of the largest concern. Our cooling strategy is also applicable to particles trapped in an ion trap, where they can be observed with a weak probe laser that has a minimum heating effect.

Note added: After the submission of this manuscript, two related works by the group of ETH Zurich [38] and by the group of ICFO [39] have appeared on arXiv.

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[32] The chamber is equipped also with a capacitance gage, providing the pressure values between 0.5 and 133 Pa with an accuracy of 0.27 Pa. Outside this pressure range, the pressure values are recorded with an accuracy of 15%.