Sympathetic cooling and squeezing of two colevitated nanoparticles

T. W. Penny, A. Pontin, and P. F. Barker

Department of Physics and Astronomy, University College London, Gower St., London WC1E 6BT, United Kingdom

(Received 13 January 2022; revised 8 November 2022; accepted 20 December 2022; published 31 January 2023)

Levitated particles are an ideal tool for measuring weak forces and investigating quantum mechanics in macroscopic objects. Arrays of two or more of these particles have been suggested for improving force sensitivity and entangling macroscopic objects. In this article, two charged, silica nanoparticles, that are coupled through their mutual Coulomb repulsion, are trapped in a Paul trap, and the individual masses and charges of both particles are characterized. We demonstrate sympathetic cooling of one nanoparticle coupled via the Coulomb interaction to the second nanoparticle to which feedback cooling is directly applied. We also implement sympathetic squeezing through a similar process showing nonthermal motional states can be transferred by the Coulomb interaction. This work establishes protocols to cool and manipulate arrays of nanoparticles for sensing and minimizing the effect of optical heating in future experiments.

DOI: 10.1103/PhysRevResearch.5.013070

1. INTRODUCTION

The ability to cool and control the center-of-mass (CoM) motion of levitated nanoparticles and microparticles, coupled with their extreme isolation from the environment, makes them ideal candidates for measuring weak forces. They have been proposed as detectors in the search for dark-matter candidates [1–3], for investigating the macroscopic limits of quantum mechanics [4–8], and for measuring short-range forces [7–11]. To date, only a few investigations have focused on cooling and controlling more than a single particle levitated in vacuum [12,13]. Arrays of levitated nanoparticles are of interest, as they can be used to enhance the detection of dark-matter candidates [2,3], measure vacuum friction [14], and for evidencing the quantumness of gravity via entanglement [15,16]. Even arrays as small as two particles can be useful for increasing the isolation from external noise sources [17]. A first step towards utilizing arrays of levitated particles is the development of tools to control the motion of colevitated particles.

Sympathetic cooling has been used extensively in ion trapping experiments to cool atomic and molecular species where no favorable internal transitions for laser cooling are available [18–22]. It is made possible by the coupling from the Coulomb interaction between colevitated ions. Coupling between two levitated nanoparticles in vacuum has been demonstrated with optical binding [12] and via the Coulomb interaction [13]. The ability to cool and control all particles via a single particle using light, while not illuminating the other cotrapped particles, can be used to minimize heating [23,24], particularly when they contain internal atomic-like systems such as nitrogen vacancy centers, whose internal state manipulation is highly temperature dependent.

The coupling between the particles in the array allows, in principle, to transfer other more complex motional states as well. Among these, squeezed states are considered the simplest, more easily accessible nonclassical states [25,26]. In the quantum regime and in the presence of a multimode system, as is considered here, squeezed states are an important resource which can allow the generation and observation of entanglement between different mechanical degrees of freedom [27,28]. Even far from the quantum domain, squeezed states have important applications for enhanced force sensing [29,30].

In this paper we cotrap a pair of silica nanoparticles in a linear Paul trap that are coupled through their mutual Coulomb repulsion. By implementing a velocity damping scheme [31–39] on just one particle, we sympathetically cool the motion of the second particle to achieve sub-Kelvin normal-mode temperatures. This differs from previous work [13] where both particles were cooled simultaneously. Importantly, we also show that the Coulomb interaction can transfer other states between cotrapped particles by squeezing the normal modes of the system with a parametric drive [27,28,40–45] on just one particle.

II. THEORY

The equations of motion (EoM) for two harmonic oscillators coupled via the electrostatic force are given by

\[
\ddot{z}_1 + \gamma_1 \dot{z}_1 + \omega_1^2 z_1 = \frac{F_{\text{fluct},1}}{m_1} + \frac{Q_1 Q_2}{m_1 4\pi \varepsilon_0 (z_2 - z_1)^2},
\]

\[
\ddot{z}_2 + \gamma_2 \dot{z}_2 + \omega_2^2 z_2 = \frac{F_{\text{fluct},2}}{m_2} - \frac{Q_1 Q_2}{m_2 4\pi \varepsilon_0 (z_2 - z_1)^2},
\]
where $z_\i$, are the positions of the particles ($i = \{1, 2\}$ denote the particle), $\gamma_i$ are the damping constants, $\omega_i$ are the natural frequencies, $m_i$ are the masses, $F_{\text{fact,}i}$ are the thermal noise forces defined by $\langle F_{\text{fact,}i}(t)F_{\text{fact,}j}(t') \rangle = 2m_i\gamma_i k_B T_0 \delta(t-t') \delta_{ij}$, and $T_0$ is the temperature of the surrounding thermal bath, $Q_i$ are the charges of the particles, and $\epsilon_0$ is the permittivity of free space. Several techniques for levitating multiple nanoparticles exist, but here we will focus on the case of particles cotrapped in a single linear Paul trap. Paul traps confine charged particles using a combination of static and oscillating electric fields.

Considering the axial direction of a linear Paul trap where the trap is formed by only static fields, the uncoupled secular frequencies of the two particles are given by

$$\omega_i = \frac{\epsilon_0}{m_i} \sqrt{z_i^2 - z_1^2},$$

where $u_i = \frac{200k_B T_0}{\epsilon_0}$, $z_i$, are the equilibrium positions of each particle, $z_i^\pm$, can be calculated by setting $\frac{\partial V}{\partial \bar{z}_i} = 0$ and solving for $z_i$. From these, an equilibrium separation of

$$z_{\text{sep}}^\pm = \frac{Q_1 Q_2}{2} = \left( \frac{Q_1 Q_2}{u_2 4\pi \epsilon_0} \right)^{\frac{1}{2}}$$

$$\Delta z_i = z_i - z_i^\pm$$

can be calculated which is mass dependent. Moving to a coordinate system given by the particles’ deviations about their equilibrium positions, $s_i = z_i - z_i^\pm$, and assuming $s_i \ll z_{\text{sep}}^\pm$, the interaction term in Eq. (3) can be expanded to second order about the equilibrium positions. By ignoring damping and external forces, the Euler-Lagrange EoM are found to be

$$\begin{bmatrix} \ddot{s}_1 \\
\ddot{s}_2 \end{bmatrix} = - \begin{bmatrix} V_{11} & V_{12} \\
V_{21} & V_{22} \end{bmatrix} \begin{bmatrix} s_1 \\
\dot{s}_2 \end{bmatrix} = - \begin{bmatrix} 0 \\
\dot{s}_1 \end{bmatrix},$$

where $V_{ij} = \frac{1}{m_i m_j} \epsilon_0^2 \Delta z_i \Delta z_j V(z_1, z_2)$. Assuming the oscillator motion takes the form $s_i = s_{0i} e^{-i \omega t}$, then the problem is reduced to finding the eigenvalues and eigenvectors of matrix $V$, which describe the normal modes of the system. The eigenvalues are given by

$$\omega_\pm = \left( \frac{\omega_i^2 + \Delta \omega_0}{2} \right) \left( 1 \pm \sqrt{1 + \frac{\Delta \omega_0}{\omega_i^2}} \right),$$

where

$$A = \frac{Q_1}{m_1} + \frac{Q_2}{m_2},$$

$$B = \frac{2Q_2}{1 + \frac{Q_2}{Q_1}} \left( \frac{1}{m_1} + \frac{1}{m_2} \right),$$

$$C = \frac{Q_1}{m_1} - \frac{Q_2}{m_2},$$

and

$$D = \frac{4Q_2}{(1 + \frac{Q_2}{Q_1})} \left( \frac{Q_1}{m_1} + \frac{Q_2}{m_2} \right) \left( \frac{1}{m_1} + \frac{1}{m_2} - 2 \right).$$

Provided the eigenvalues are positive ($\omega_\pm > 0$), then the motion is stable and the normal-mode frequencies are given by

$$\omega_\pm. \text{The normalized eigenvectors are given by}
$$

$$e_\pm = \frac{1}{\sqrt{1 + r_\pm^2}} \begin{bmatrix} 1 \\
r_\pm \end{bmatrix}.$$
particles were recorded simultaneously in the same camera and recording the resulting displacement of the image. Both moving the camera a fixed distance with a translation stage modulated by the 637-nm laser (Fig. 1), time traces of the particle from a 637-nm diode laser. Since both particles were illuminated by a 637-nm laser is focused onto each particle individually. The power in each arm is balanced such that the scattering from each particle is approximately equal when measured on the CMOS camera. One of the arms is also focused onto a quadrant photodiode for real-time detection of the motion of one particle. A force is applied to the same particle to either cool or squeeze the normal modes by modulating the power of a 1030-nm laser. The feedback signal is generated from the real-time measurement of the particle position. BS: beam splitter, QP: quadrant photodiode, and SPF: short-pass filter. (b) Two cotrapped particles in the Paul trap. The separation between these particles is $198 \pm 1 \mu m$.

The radial motion of trapped nanoparticles will also couple to form normal modes; however, the coupling scales much more strongly with charge difference than the axial modes. For large charge differences, both particles are almost completely unaffected by the other and oscillate close to their trap frequencies. The radial normal modes can be calculated in a similar manner to the axial normal modes [47].

III. EXPERIMENTAL METHOD

In this experiment we use a linear Paul trap with four parallel rod electrodes for radial trapping and two “endcap” electrodes for axial trapping. The four parallel rods are held in place by two gold-coated printed circuit boards which contain the electrical connections for the rods and have an endcap electrode etched into each [48]. The parabolic coefficients were calculated using finite element modeling and found to be $r_0 = 1.1 \text{ mm}$, $z_0 = 3.5 \text{ mm}$, $\kappa = 0.071$, and $\eta = 0.82$. Typical trap parameters are $V_0 = 100-150 \text{ V}$, $U_0 = 20-50 \text{ V}$, and $\omega_{rf} = 2\pi \times 8 - 12 \text{ kHz}$.

Silica nanoparticles, with charges of up to 6000e, were loaded into the trap at $10^{-1} \text{ mbar}$ using the electrospray technique [48,49]. Two particles were either trapped simultaneously or, after trapping one particle, more nanoparticles were sprayed into the trapping region until a second was caught. Particles were monitored on a complementary metal oxide semiconductor (CMOS) camera using scattered light from a 637-nm diode laser. Since both particles were illuminated by the 637-nm laser (Fig. 1), time traces of the particle motion could also be recorded on the CMOS camera at 1000 frames per second [48,50]. The time traces were calibrated by moving the camera a fixed distance with a translation stage and recording the resulting displacement of the image. Both particles were recorded simultaneously in the same camera image so that the phase difference between the displacement of the particles was known. The camera acted as an out-of-loop detector for measuring the temperature when feedback cooling the particles.

Real-time detection of the particle motion was done using a quadrant photodiode. Individual arms of the 637-nm beam illuminated each particle such that just the motion of one particle was measured on the quadrant photodiode. The signal from the quadrant photodiode was fed to a Red Pitaya FPGA to generate a feedback signal to either cool or squeeze the particle motion. The PYRPL software package was used to filter the position signal of the particle around the appropriate mode, then either delay the signal (to cool the motion) or mix the signal with a sinusoidal wave at twice the central frequency of the mode (to squeeze the motion), followed by amplification. The feedback signal was then used to modulate the power of the 1030-nm diode laser and create a force on the particle. Despite relatively high intensities of $2 \times 10^8 \text{ Wm}^{-2}$ for the 1030-nm laser, the trapping frequencies of particles were shifted by less than 2% due to the additional laser.

IV. PARTICLE CHARACTERIZATION

Figure 1(b) shows two particles trapped in the Paul trap. Using the CMOS camera, the equilibrium separation of the particles was measured to be $z_{sep}^2 = 198 \pm 1 \mu m$. This is much larger than the expected amplitude of a single particle in thermal equilibrium with a frequency of $\omega_0 = 2\pi \times 200 \text{ Hz}$ ($\sqrt{q^2} = 6.8 \mu m$), so the Taylor expansion used in Eq. (5) is valid. The power spectral density (PSD) of two coupled particles can be seen in Fig. 2(a), taken at a pressure of $1.3 \times 10^{-2} \text{ mbar}$. Unlike a single uncoupled particle which would display only one mode [48,50], both particles display a mix of the normal modes of a system showing they are axially

FIG. 1. (a) The experimental setup for sympathetic cooling and squeezing. Two 387-nm-diameter, silica particles are trap simultaneously in a Paul trap. One arm of a 637-nm laser is focused onto each particle individually. The power in each arm is balanced such that the scattering from each particle is approximately equal when measured on the CMOS camera. One of the arms is also focused onto a quadrant photodiode for real-time detection of the motion of one particle. A force is applied to the same particle to either cool or squeeze the normal modes by modulating the power of a 1030-nm laser. The feedback signal is generated from the real-time measurement of the particle position. BS: beam splitter, QP: quadrant photodiode, and SPF: short-pass filter. (b) Two cotrapped particles in the Paul trap. The separation between these particles is $198 \pm 1 \mu m$. The radial motion of trapped nanoparticles will also couple to form normal modes; however, the coupling scales much more strongly with charge difference than the axial modes. For large charge differences, both particles are almost completely unaffected by the other and oscillate close to their trap frequencies. The radial normal modes can be calculated in a similar manner to the axial normal modes [47].
of the radial modes, charges of \( Q_1 = 2135 \pm 58 \, e \) and \( Q_2 = 906 \pm 15 \, e \) were calculated using the mass values determined earlier. We can verify the charges by using them to calculate the theoretical values of \( r_{\pm} \) and \( E_{\pm} \) for the axial modes and comparing them to the measured values. We find \( r_- = -1.60 \pm 0.03 \) and \( r_+ = 0.61 \pm 0.04 \), which are close to those used to construct the PSDs in Fig. 2(b), and that \( E_{\mp} = \frac{E_{\pm}}{k_B} = 0.72 \pm 0.03 \), which agrees with values \( E_{\mp} = 0.73 \pm 0.04 \) and \( E_{\pm} = 0.71 \pm 0.04 \) measured from the PSDs. This shows that the radial modes have sufficiently small coupling to be treated as uncoupled.

V. SYMPATHETIC COOLING

By modulating the power of a 1030-nm laser focused onto just one of the particles (particle 1) the normal modes can be cooled. Through the Coulomb interaction, the same modes of the other particle are also cooled. If a force proportional to the velocity of the \( z_+ \) is applied to particle 1, the resulting EoM for the two normal modes are

\[
\ddot{z}_+ + (\gamma_0 + \gamma_{\text{fb}})\dot{z}_+ + \gamma_{\text{fb}} \delta z_+ + \omega_+^2 z_+ = \frac{F_{\text{fluc+}}}{m},
\]

\[
\ddot{z}_- + \gamma_0 \dot{z}_- - \gamma_{\text{fb}} \delta z_- + \omega_-^2 z_- = \frac{F_{\text{fluc-}}}{m},
\]

where \( \gamma_{\text{fb}} \) is the feedback gain and \( \delta z_+ \) is imprecision noise in the detection. Provided \( \omega_- - \omega_+ \gg \gamma_0 + \gamma_{\text{fb}} \), the \( \gamma_{\text{fb}} \delta z_+ \) term in Eq. (21) will have a negligible effect on the \( z_- \) mode and can be ignored. Additionally, in practice the feedback signal is bandpass filtered such that the \( \gamma_{\text{fb}} \delta z_+ \) term does not affect the \( z_- \) mode. Thus the \( z_+ \) mode is cooled while the \( z_- \) mode remains unaffected. Similar equations can be found for the two modes (by interchanging all \( \pm \) subscripts) when cooling the \( z_- \) mode. Since the particles are the same mass, the temperature compression ratio for a mode should be equal in each particle [47]. The cooled mode will have a temperature given by [32,35,37,39]

\[
T_+ = T_{0+} \frac{\gamma_0}{\gamma_0 + \gamma_{\text{fb}}} + \frac{1}{2} \frac{m \omega_+^2}{k_B} \frac{\gamma_{\text{fb}}}{\gamma_0 + \gamma_{\text{fb}}} S_{\text{in}+},
\]

where \( T_{0+} \) is the initial temperature of the CoM mode, and \( S_{\text{in}+} = \int_{-\infty}^{\infty} \delta z_+(t) \delta z_+(0) e^{i\omega t} dt \) is the spectral density of the imprecision noise and is assumed to be white and Gaussian over the linewidth of the oscillator mode. A similar expression is found for the \( z_- \) mode. By increasing the feedback gain, a minimum temperature will be found that is dependent on the frequency of the mode.

In the experiment, the feedback was implemented by applying a bandpass filter to the position signal of the particle to remove noise and other modes from the feedback signal, then amplifying and adding a \( \pi/2 \) phase shift to estimate the current velocity of the particle [35,39]. Figure 3(a) shows the spectra of the two modes cooled to minimum temperatures of \( T_+ = 200 \pm 10 \) mK and \( T_- = 190 \pm 40 \) mK (calculated from the area under the PSDs and the previously measured masses) at a pressure of \( P = 3.2 \times 10^{-7} \) mbar and spectra of the modes with no cooling (\( T_+ = T_- = 293 \) K) at \( P = 1.3 \times 10^{-2} \) mbar. As expected, the temperature compression ratio of a mode was found to be the same for each particle.
to remain in the underdamped regime where the theory is valid, the oscillator frequency would have to be increased and the background gas damping would have to be reduced. An oscillator at a frequency of ~500 Hz and a background pressure of ~10^{-11} mbar would be sufficient provided other noise sources are kept negligible.

VI. SYMPATHETIC SQUEEZING

Squeezing the motion of a mechanical oscillator has been achieved with several methods including parametric modulation [27,28,40–45], nonadiabatic shifts [54–56], and back-action evading measurements [57–60]. Parametric modulation is usually implemented by modulating the oscillator spring constant at twice the mode resonance frequency. In the Paul trap this would drive both particles. In order to show a sympathetic operation, we use a measurement-based scheme to parametrically drive only one particle. This scheme also avoids acting on either particle with a linear drive that would appear from modulating the trap potential [61,62]. A measurement of the particle position was filtered around the appropriate mode using a Lorentzian bandpass filter with a bandwidth of 152 Hz then mixed with a sinusoidal signal at twice the resonance frequency of that mode to produce a signal to modulate the 1030-nm laser. An additional delay was added to the signal to minimize errors to the phase response of the filter. The laser imparted a force $F \propto z_\pm \sin(2\omega_{01}t)$ onto particle 1. If we were, for example, squeezing the $z_+$ mode, this results in the following EoM for the normal modes:

$$\ddot{z}_+ + \gamma_0 \dot{z}_+ + \omega_0^2 z_+ + (z_+ + \delta z_+)G\sin(2\omega_{01}t) = \frac{F_{\text{fluct},+}}{m},$$

$$\ddot{z}_- + \gamma_0 \dot{z}_- + \omega_0^2 z_- + (z_- + \delta z_+)G\sin(2\omega_{01}t) = \frac{F_{\text{fluct},-}}{m},$$

where $G$ is a gain applied to the signal. Similar to the case for sympathetic cooling, the $z_-$ mode remains unaffected by the applied force provided $\omega_0 - \omega_{01} \gg \gamma_0$. In this instance, $\delta z_+ \ll z_+$ and therefore has a negligible impact on the dynamics of the system. Thus the $z_+$ mode experiences a parametric drive while the $z_-$ mode is unaffected. In the case of squeezing with a parametric drive, the variance of the X and Y quadratures is given by [41]

$$\langle X^2 \rangle = \langle x^2 \rangle \frac{1}{1-g},$$

$$\langle Y^2 \rangle = \langle x^2 \rangle \frac{1}{1+g},$$

where $g = G \times \omega_0/2\gamma_0$. It can be seen that this limits the maximum achievable squeezing to ~3 dB before the onset of parametric instability in the X quadrature [63,64].

Figure 4 shows phase-space plots of the quadratures of the $z_+$ mode of both particles, with and without a parametric drive applied to particle 1 at a pressure of $P = 1.2 \times 10^{-7}$ mbar. Squeezing was performed at a relatively high pressure to reduce the measurement time required to accurately sample the squeezed thermal state. The quadratures were extracted from

![FIG. 3. (a) Spectra for the cooled $z_+$ (left) and $z_-$ (right) modes at a pressure of $P = 3.2 \times 10^{-7}$ mbar (dark lines) shown alongside the spectra of the modes with no cooling at $P = 1.3 \times 10^{-7}$ mbar (light lines) where the modes are expected to be in thermal equilibrium with the surrounding gas. (b) Temperature of the normal modes in the experiment (markers) and theory (lines) vs feedback gain. The disagreement between theory and experiment suggests there is an additional source of heating for the normal modes.](image-url)
equal charges; therefore the mode energies should be equal as they
with only particle 1. The particles used here have approximately
state for the $z$ mode. By a 50-Hz low-pass filter to the
the displacement data by applying a demodulation followed
the demodulation frequency until maximal squeezing was
Anomalous heating of the mode was also seen as the gain of
the squeezing operation was increased. Simulations based on
the elongations of the phase-space distributions in both particles.
Simulation-based on the experimental parameters, implemented with the leapfrog
algorithm, suggest that this is due to using a tightly focused beam to generate the force for squeezing the particle. Since the
beam waist is comparable to the amplitude of particle motion,
the particle feels an additional position-dependent component
to the feedback force. Taking the heating into account, we
were able to measure $-1.70 \pm 0.05$ dB and $-1.69 \pm 0.05$ dB
of squeezing in the $z_{\pm}$ mode of particles 1 and 2, respectively.
The squeezing gain was limited by the nonlinear effect of the
tight beam generating a spiraling in the phase-space plots \[56\].
Both the $z_{\pm}$ mode could be squeezed using this method.

Transferral of the thermal squeezed state between the particles
shows that nonthermal states can also be transmitted via the Coulomb force. For an array of coupled nanoparticles, this property could be used to control and readout the state of the
array from only a single nanoparticle. For large arrays, this
could be technically easier than reading out the state of every
single nanoparticle.

\section{VII. Conclusions}

We have demonstrated and characterized coupling between
two charged nanoparticles that produces two orthogonal normal modes of motion. Both sympathetic cooling and
squeezing of the motion of one particle were shown through
interaction with another coupled particle. This demonstrates
thermal and nonthermal states can be transferred via the
Coulomb interaction. Such techniques could be extended to
a mixed species system with particles of similar masses or
to an array of many nanoparticles. As a result, this work
represents an important tool in implementing the creation of
macroscopic quantum superpositions \[15,65–68\] via coupling of
the normal modes of a cotrapped silica-nanodiamond pair
to the internal quantum state of a nitrogen vacancy center in
the nanodiamond. Such a technique can be applied to experiments which have previously been limited by internal heating
of samples \[24\].

Our results are a first step towards schemes that utilize the
coupling between levitated particles. Large arrays of coupled
particles would have a range of motional frequencies that
scale with the number of particles. This would be ideal for
sensing over a wide range of frequencies simultaneously, such
as in ultralight dark-matter searches \[3\]. Using sympathetic
techniques would allow control and readout of the entire array
from a single particle, simplifying the experimental procedure
compared to an uncoupled array.

\section{Acknowledgments}

The authors would like to acknowledge useful discussions
with Animesh Datta. The authors acknowledge funding from
EPSRC Grant No. EP/N031105/1 and the H2020-EU.1.2.1
TEQ project Grant Agreement No. ID 766900.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4}
\caption{Phase-space diagrams showing a thermal and squeezed state for the $z_{\pm}$ mode of both particles at a pressure of $P = 1.2 \times 10^{-2}$ mbar. Both particles display squeezed states despite interaction with only particle 1. The particles used here have approximately equal charges; therefore the mode energies should be equal as they appear here.}
\end{figure}


[54] M. Asjad, G. S. Agarwal, M. S. Kim, P. Tombesi, G. Di Giuseppe, and D. Vitali, Robust stationary mechanical squeez-


[57] V. B. Braginsky, Y. I. Vorontsov and, K. S. Thorne, Quantum nondemolition measurements, Science 209, 547 (1980).


