

Nanoparticle Stored with an Atomic Ion in a Linear Paul Trap

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Radiofrequency (RF) traps enable highly controlled interactions between charged particles, including reactions between cold molecular ions, sympathetic cooling of one ion species with another, and quantum logic spectroscopy. However, the charge-to-mass (Q/m) selectivity of RF traps limits the range of objects that can be confined simultaneously in the same trap. Here, we confine two particles—a nanoparticle and an atomic ion—in the same radiofrequency trap although their charge-to-mass ratios differ by six orders of magnitude. The confinement is enabled by a dual-frequency voltage applied to the trap electrodes. We introduce a robust loading procedure under ultrahigh vacuum and characterize the stability of both particles. It is observed that slow-field micromotion, an effect specific to the dual-field setting, plays a crucial role for ion localization. Our results lay the groundwork for controlled interactions between diverse charged particles, regardless of the difference in their charge or mass, with applications from antimatter synthesis to the generation of macroscopic quantum states of motion.

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The confinement of charged particles in radiofrequency (RF) traps is a fundamental technique for mass spectrometry [1], cold molecular chemistry [2], and quantum information processing [3–5], and has wide-ranging applications from the study of condensed-matter material properties [6] to the detection of millicharged dark matter [7]. Typically, a trap is configured to levitate a specific object with a predefined charge-to-mass (Q/m) ratio; because of the trap's stability properties, only objects with similar Q/m ratios will be stably confined [8]. While this selectivity is advantageous for certain applications, such as mass spectrometry, it limits the range of objects that can be confined simultaneously in the same trap.

To overcome this limitation and trap two species with very different Q/m ratios, a Paul trap driven by two voltage sources has been proposed [9–11], targeting antihydrogen synthesis [9,11] and sympathetic cooling of megadalton particles, including nanoparticles or viruses [10]. In this scenario, the frequency and the amplitude of the first voltage source are tuned to provide confinement for the first species, the frequency and amplitude of the second source are tuned for the second species, and because of a careful choice of parameters, the first source does not

compromise the stability of the second species and vice versa. Here, we experimentally demonstrate such a dual-frequency linear Paul trap, which is driven by 17.5-MHz and 7-kHz voltage sources. This approach allows us to confine vastly different objects, a silica nanoparticle and atomic calcium ions, in the same RF trap, thus creating a hybrid system combining an ultrahigh-quality-factor mechanical oscillator [12] and one of the leading platforms for encoding qubits [5]. The trapped objects differ by six orders of magnitude in their Q/m ratio and by eight orders of magnitude in mass.

Let us examine the conditions for stable trapping of a particle—either a nanoparticle or an atomic ion—in the quadrupole electric potential Φ of a linear Paul trap driven by two voltage sources [9–11] with frequencies Ω_{slow} , Ω_{fast} and amplitudes V_{slow} , V_{fast} . For the nanoparticle in our system, the main confinement is provided by the slow field, and the influence of the fast field can be neglected. One sees this by comparing the frequencies of the nanoparticle's center-of-mass motion in the separate potentials of the two fields (see Supplemental Material [13]):

$$\frac{\omega_{(n)}|_{\Omega_{\text{fast}}}}{\omega_{(n)}|_{\Omega_{\text{slow}}}} = \frac{\Omega_{\text{slow}}}{\Omega_{\text{fast}}} \frac{V_{\text{fast}}}{V_{\text{slow}}} \approx 10^{-2}, \quad (1)$$

where the approximation in the final step is based on typical voltages used in the experiment (Table I). For the ion, on the other hand, under the pseudopotential approximation and the condition $\Omega_{\text{slow}} \ll \Omega_{\text{fast}}$, the slow field acts as a slowly varying dc offset to the fast field, and the stability condition for the ion motion can be written as (see Supplemental Material [13])

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TABLE I. Typical experimental parameters used for co-trapping an ion and a nanoparticle (NP). Q is the charge, m the mass, f_{drive} the trap-drive frequency, V_{drive} the trap-drive amplitude, and ω the frequency of center-of-mass motion in the xy plane of Fig. 1.

	Q (e)	m (kg)	f_{drive} (kHz)	V_{drive} (kV _{pp})	$\omega/(2\pi)$ (kHz)
Ion	1	6.6×10^{-26}	17.5×10^3	2.5	4×10^3
NP	800	2×10^{-17}	7	0.15	1.5

$$|a_{\text{eff}}| < q_{(i)}^2/2, \quad (2)$$

where $a_{\text{eff}} = 4\kappa e V_{\text{slow}}/m_{(i)} r_0^2 \Omega_{\text{fast}}^2$, $q_{(i)} = 2\kappa e V_{\text{fast}}/m_{(i)} r_0^2 \Omega_{\text{fast}}^2$, $r_0 = 0.9$ mm is the characteristic distance from the electrodes to the trap axis, $\kappa = 0.93$ is a geometric factor, $m_{(i)}$ is the mass of the ion, and e is the elementary charge. Thus, Eq. (2) imposes conditions on the fast and slow fields under which co-trapping of a nanoparticle and an ion is possible.

We implement the dual-frequency trap by applying a voltage oscillating at $\Omega_{\text{fast}} = 17.5$ MHz to one pair of RF electrodes and a second voltage at $\Omega_{\text{slow}} = 7$ kHz to a second pair. (A similar configuration is described in Ref. [21] for an experiment with $^{40}\text{Ca}^+$ ions.) The trap schematic is shown in Fig. 1(a). The fast voltage signal at Ω_{fast} is provided by a low-noise function generator and preamplified by a high-power amplifier. The schematic shows the function generator and preamplifier as a single ac source. We supply the preamplified signal to a tap of the inductive coil L of a resonant LC circuit in which the trap

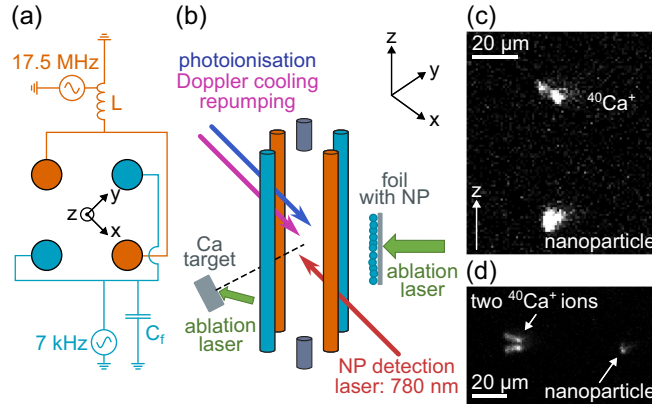


FIG. 1. Experimental setup. (a) Dual-frequency drive of the linear Paul trap. (b) Schematic of the procedure for trapping a nanoparticle (NP) and an ion. Along the z axis, dc end-cap electrodes are indicated in gray. Not shown: two pairs of compensation electrodes for displacement in the xy plane. (c) Camera image of a $^{40}\text{Ca}^+$ ion and a nanoparticle confined in the same Paul trap. (d) A second image in which two ions are confined with a nanoparticle.

electrodes act as a capacitance. In the case of the slow voltage at Ω_{slow} , a different function generator drives a low-noise, high-voltage amplifier. The internal resistance of the slow voltage source and a 4.7-nF capacitor C_f act as a low-pass filter, effectively grounding the fast signal. The slow field sees this capacitor as an open circuit. Similarly, the inductive coil of the resonant circuit acts as a high-pass filter, effectively grounding the slow signal.

As a first step, a nanoparticle is loaded into the trap under ultrahigh vacuum via laser-induced acoustic desorption [22,23] combined with temporal control of the Paul-trap potential [24]. The nanoparticle source is a 300- μm thick aluminum foil with silica nanospheres deposited on the front side; the spheres have a nominal diameter of 300 nm. A pulsed ablation laser (4 mJ, 5 ns) is focused on the back side of the foil [Fig. 1(b)]. The geometry of the trap and the source are as described in Ref. [24], with the exception that here, the distance between dc end-cap electrodes is 3.4 mm. At this stage, only the slow voltage at Ω_{slow} is applied to the trap electrodes, with amplitude $V_{\text{slow}} = 1.4$ kV_{pp}. The end-cap voltage is 56.5 V.

After loading, we cool the nanoparticle's motion using electrical feedback based on optical detection at 780 nm. There are two differences with respect to the protocol of Ref. [25]: first, the nanoparticle's position along the x , y , and z axes is detected with a confocal setup implemented with fiber-coupled avalanche photodiodes [14,23,26,27]. Second, the x - and y -axis feedback signals are combined and sent to a feedback electrode mounted next to the trap, but the z -axis feedback signal is sent to the lower end-cap electrode through a high-pass filter. Feedback cooling operates continuously during the subsequent steps.

Next, we increase the nanoparticle charge $Q_{(n)}$. Recall from Eq. (2) that the values of V_{slow} compatible with co-trapping an ion are bounded from above; a higher value of $Q_{(n)}$ allows the stiffness of the nanoparticle's trap to be maintained while V_{slow} is reduced (see Supplemental Material [13]). When the ablation laser is directed with a pulse energy of 1 mJ to a pure calcium target near the trap [Fig. 1(b)], it is observed that $Q_{(n)}$ increases and saturates at around 300 e in the absence of a fast field. The presence of a fast field allows even higher values of $Q_{(n)}$ to be obtained during this process. Thus, at this point we introduce the fast field with $V_{\text{fast}} \approx 1.5$ kV_{pp}. One to three pulses of the ablation laser correspond to an increase of a few elementary charges; we periodically interrupt this stepwise process to reduce V_{slow} such that the $q_{(n)}$ parameter remains below 0.9. We stop the charging process at an amplitude $V_{\text{slow}} = 160$ V_{pp}, for which $Q_{(n)} \approx 800$ e is reached. Afterward, both charge and mass are assumed to remain constant: a change in either quantity would shift $\omega_{(n)}$, which previously has allowed the addition and subtraction of single elementary charges to be observed [25,28,29], but here we continuously monitor $\omega_{(n)}$ and find no shift.

Once the nanoparticle has been localized and its charge increased, we load a $^{40}\text{Ca}^+$ ion into the dual-frequency trap. First, all laser beams required for ion loading, cooling, and detection are aligned to the geometric center of the trap. Here, the trapped nanoparticle serves as a scattering target onto which the attenuated beams are focused. Next, the amplitude of the fast voltage is set to $2.5 \text{ kV}_{\text{pp}}$. The voltage on one of the trap's two pairs of compensation electrodes is then increased such that the nanoparticle is displaced in the xy plane, making room for the ion at the trap center. (As the ion's charge is three orders of magnitude smaller than the nanoparticle's charge, its equilibrium position is less affected by the compensation field.) The nanoparticle-detection optics and the feedback-cooling parameters are adjusted to account for this new position. Finally, we operate the ablation laser, still focused on the calcium target, at a pulse energy of 0.5 mJ in conjunction with diode lasers at 375 nm and 423 nm for two-step isotope-selective photoionization [15,30]. The success probability for loading an ion via ablation is similar with and without a nanoparticle (see Supplemental Material [13]). Once an ion is loaded, it is detected and Doppler cooled via fluorescence on the $397\text{-nm } 4^2\text{S}_{1/2} \leftrightarrow 4^2\text{P}_{1/2}$ transition, with repumping on the $866\text{-nm } 4^2\text{P}_{1/2} \leftrightarrow 3^2\text{D}_{3/2}$ and $854\text{-nm } 4^2\text{P}_{3/2} \leftrightarrow 3^2\text{D}_{5/2}$ transitions. (Amplified spontaneous emission of the 397-nm diode near the $393\text{-nm } 4^2\text{S}_{1/2} \leftrightarrow 4^2\text{P}_{3/2}$ transition occasionally populates the $4^2\text{P}_{3/2}$ state.) All three diode-laser frequencies are locked to a wave meter.

To verify that a nanoparticle and an atomic ion are co-trapped, we illuminate them with the 397-nm , 866-nm , and 854-nm laser fields and capture their image using an electron-multiplying charge-coupled device (EM-CCD) camera. An example image is shown in Fig. 1(c): nanoparticle and ion are separated by $55(10) \mu\text{m}$. The laser beams are focused on the ion, and the nanoparticle is in the tail of the Gaussian intensity profile; the camera detects elastically scattered light from the nanoparticle. Only 397-nm light is used for imaging, and an optical bandpass filter in front of the camera removes other wavelengths. An example of the ion-loading process is shown in Supplementary Video 1 [13]. The ion typically escapes after several minutes, with the shortest instance being one minute and the longest almost one hour. These storage times are shorter than those for an ion trapped with a single field, which we attribute to the second field, not to the nanoparticle (see Supplemental Material [13]).

To determine whether the theoretical description above is consistent with our demonstration of co-trapping, we characterize the stability of nanoparticle and ion separately in the dual-frequency Paul trap. First, a nanoparticle is confined solely with the slow voltage. The fast voltage is then added with increasing amplitude up to $2.5 \text{ kV}_{\text{pp}}$ while the position and oscillation frequency of the nanoparticle are monitored with a camera and the confocal detection

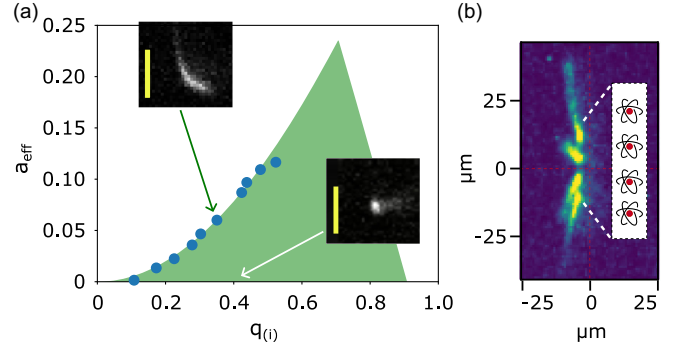


FIG. 2. Influence of the slow field on the ion. (a) Blue dots: instability threshold measured with the ion, plotted in terms of the stability parameters $q_{(i)}$ and a_{eff} . Green area: stability diagram, calculated from experimental parameters. Insets are camera images of the ion for different stability regimes; the scale bar is $10 \mu\text{m}$. (b) Composite image of the ion at four positions along the z axis for $q_{(i)} = 0.4$, $a_{\text{eff}} = 0.06$. The positions along the z axis are set by the end-cap voltage. Inset: schematic of the ion positions along z axis in the absence of micromotion.

setup, respectively. We find that the nanoparticle remains trapped for all amplitudes and that the presence of the fast voltage increases the nanoparticle's 1-kHz frequency in the xy plane by 10 Hz for $V_{\text{fast}} = 2.5 \text{ kV}_{\text{pp}}$. These observations are in agreement with Eq. (1).

Next, the ion is confined for several values of V_{fast} . At each setting, V_{slow} is increased until the ion is expelled from the trap; this threshold amplitude is identified as $V_{\text{slow}}^{\text{max}}$. In Fig. 2(a), the pair of stability parameters $[q_{(i)}, a_{\text{eff}}]$ is plotted for each pair $(V_{\text{fast}}, V_{\text{slow}}^{\text{max}})$. Superimposed on these data points is a stability diagram [4], calculated from the solutions to the Mathieu equation (see Supplemental Material [13]). We see that the experimentally determined thresholds lie at the edge of the stability diagram's stable region, consistent with our expectations. The rightmost data point, at $q_{(i)} = 0.55$, corresponds to $(V_{\text{fast}} = 2.5 \text{ kV}_{\text{pp}}, V_{\text{slow}}^{\text{max}} = 260 \text{ V}_{\text{pp}})$.

Camera images provide further evidence of the slow field's influence on ion confinement: the two insets of Fig. 2(a) show that an ion is well localized when the trap is operated deep in the stable region of the stability diagram, but that its position extends over several tens of micrometers at the instability threshold. This elongation is due to micromotion—not the well-known micromotion at Ω_{fast} in a single-frequency RF trap [16], but at the second frequency Ω_{slow} . Figure 2(b) is a composite of four images of this slow-frequency micromotion in the stable region, each obtained for a different position of the ion along the z axis, set by the end-cap voltage. We see that the micromotion amplitude increases as the ion is displaced further from the origin. This image underscores the importance of positioning the ion at the slow-micromotion minimum. For future experiments based on the controlled interaction of two

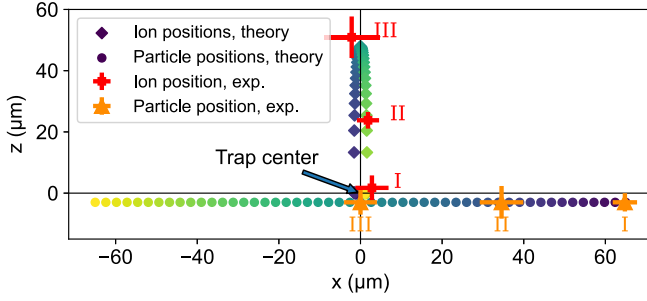


FIG. 3. Ion-particle interaction. Circles: fixed nanoparticle positions in the dual-frequency trap. Diamonds: each ion position is calculated from the corresponding nanoparticle position. Squares: ion positions extracted from Fig. 4. Triangles: nanoparticle position extracted from Fig. 4. The labels next to the data points correspond to the labels in Figs. 4(c) and 4(f). The error bars correspond to the size of the nanoparticle and the ion images in Fig. 4. Ion-nanoparticle pairs are indicated with the same color. Calculation parameters: the nanoparticle's charge is $800 e$, and the ion's secular frequencies along the x and z axes are $\omega_{(i),x} = 4$ MHz, $\omega_{(i),z} = 800$ kHz.

particles in a dual-frequency trap, it will be crucial to analyze the impact of slow micromotion. Note that V_{fast} and V_{slow} are applied to different electrode pairs, and capacitive coupling could result in different locations for the fast and slow micromotion minima; one could investigate whether combining the voltages on one electrode pair has a different effect on micromotion.

Having examined the nanoparticle and ion separately, we turn to the question of where the two particles are located when they are trapped together. Recall, for example, that in the loading procedure, it was necessary to position the nanoparticle such that the ion would be loaded at the trap center, to which the lasers had been aligned. In Fig. 3, we calculate the equilibrium position of the ion for different positions of the nanoparticle, using typical experimental parameters. The ion position is determined by the trap potential, Coulomb repulsion from the charged nanoparticle, and dc compensation-electrode voltages V_{c1} and V_{c2} . Here, fixing the nanoparticle position as an input parameter in the calculation is equivalent to fixing V_{c1} and V_{c2} . The nanoparticle positions are chosen to lie on the line $(-65 \mu\text{m} \leq x \leq -65 \mu\text{m}, y = 0 \mu\text{m}, z = -3 \mu\text{m})$, where displacement along the z axis is chosen such that the calculated z -distance between the nanoparticle and the ion for the rightmost nanoparticle position “I” matches the experimental distance in Fig. 4(c), label I. Consider the case of a nanoparticle at $x = \pm 50 \mu\text{m}$: the ion is found near the origin, and we infer that the interaction between nanoparticle and ion is negligible compared to the interaction with the trap. As the nanoparticle is brought closer to the origin, the ion is displaced along the z axis, indicating that the particles' interaction has become significant. Thus, this calculation provides intuition for how the competing forces are balanced. It also highlights that there are two ways to

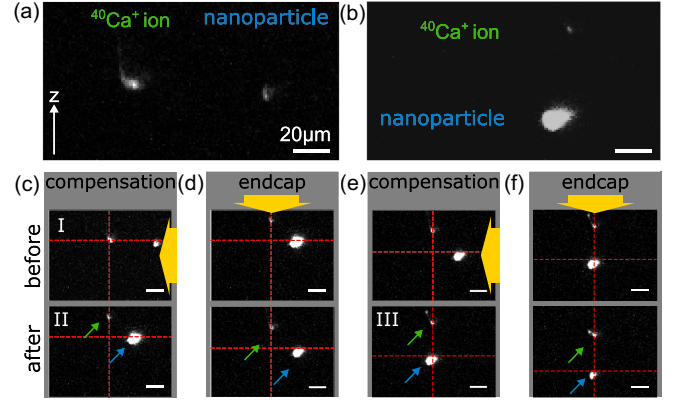


FIG. 4. Ion-nanoparticle configurations. The z axis points in the same direction in all images. (a) EM-CCD images of the xy and (b) z configurations. (c) Starting from an xy pair, we decrease the compensation-electrode voltages that have been used to displace the nanoparticle from the trap center. As a result, the nanoparticle shifts toward the center of the trap, and the ion is repelled along the axis of weakest confinement, which is the z axis here. (d) The voltage on one end cap is increased to compensate for this shift; both particles shift away from that end cap. (e) The compensation-electrode voltages are decreased further until the nanoparticle lies on the z axis. (f) The end-cap voltage is increased further until the ion is at the original position. The red cross in (c)–(f) indicates the potential minimum of the trap for the ion in the absence of the nanoparticle. The labels I–III correspond to the data-point labels in Fig. 3.

trap an ion at the origin, which, as we have just seen, minimizes its micromotion at Ω_{slow} : the nanoparticle must be displaced either in the xy plane or along the z axis.

Let us refer to the case of nanoparticle displacement in the xy plane as an xy pair and to the case of displacement along the z axis as a z pair. Examples of the two configurations are shown in Figs. 4(a) and 4(b). The nanoparticle in an xy pair experiences excess micromotion and is more sensitive to voltage noise on the trap electrodes than it would be in the z configuration. Thus, it is desirable to rotate an xy pair into a z pair. A procedure for this transformation is shown in Figs. 4(d)–4(f), in which the voltages on the compensation electrodes and end-cap electrodes are adjusted in four steps. While it is possible to reduce the transformation to two steps, there is a risk that the ion's displacement along the z axis exceeds the $100\text{-}\mu\text{m}$ waist of the Doppler-cooling beams. For the three nanoparticle horizontal positions in Figs. 4(c)–4(f), we have extracted the ion positions relative to the trap center; these were plotted in Fig. 3. The extracted ion coordinates are $\{[x, z]_{\text{I,II,III}}\}_{\text{exp}} = \{[3_{-4}^{+4}, 2_{-4}^{+4}], [-2_{-3}^{+3}, 24_{-3}^{+3}], [-2_{-7}^{+7}, 51_{-7}^{+7}]\}$. The corresponding theoretically predicted ion positions are $\{[x, z]_{\text{I,II,III}}\}_{\text{th}} = \{[-1_{-0.1}^{+0.1}, 1.9_{-1.9}^{+2.6}], [-1.3_{-0.6}^{+0.3}, 31.0_{-9.0}^{+7.0}], [-0.0_{-0.2}^{+0.2}, 45.9_{-2.8}^{+2.5}]\}$, where the uncertainties propagate from the uncertainty of the nanoparticle position. We see good agreement between the theoretically calculated and

experimentally measured positions. Following the rotation to a z pair, we observe that if the ion is lost from the trap, another ion can be reloaded directly in this configuration.

We have observed that only a single ion can be loaded in the z configuration, but in the xy configuration, we can co-trap two ions with a nanoparticle, as shown in Fig. 1(d). To achieve this, the energy of the ablation laser pulse is increased by 10%, to 0.55 mJ; the other co-trapping parameters remain unchanged. We have never observed more than two ions co-trapped with the nanoparticle, but we expect that this should be possible with improvements to the experimental setup. The two-ion limit is not due to the nanoparticle or to the dual-frequency drive: in the absence of the nanoparticle, and with a single frequency Ω_{fast} and amplitude V_{fast} , we trap at most two ions. (For lower amplitudes, we are able to trap several ions.) This points toward problems specific to our trap geometry or RF source: in state-of-the-art traps, it is standard to trap dozens of ions with similar secular frequencies. Co-trapping multiple ions with a nanoparticle is an interesting prospect as one can couple the nanoparticle to different collective motional modes of the trapped ions, which may offer advantages due to noise suppression [31] or due to the mode frequency.

Calcium ions and a charged silica nanoparticle have been simultaneously confined in a dual-frequency Paul trap. We have studied the ion-nanoparticle pair stability as function of the two RF amplitudes, developed a reliable procedure for loading the trap, and demonstrated a method to swap between different geometric configurations. *In situ* charging of the trapped nanoparticle is necessary to reduce the slow-field amplitude to a level that permits ion trapping. The mismatch of six orders of magnitude between the particle's charge-to-mass ratios leads to a differential response to static electric fields, which we found to be beneficial for tuning the ion-nanoparticle separation and maximizing ion-loading efficiencies. As a next step, we plan to analyze the kinetic energies and heating rates of both particles in the trap.

This Letter has broad applications: it opens up the study of controlled reactions between charged particles with vastly different masses, such as between positrons and anti-protons [9,11], or between a wide range of cold molecular ions [10]. Furthermore, our results lay the groundwork for a hybrid system in which a motional state of a nanoparticle is coupled to external or internal degrees of freedom of trapped ions, with prospects for sympathetic cooling (see Supplemental Material [13]). While the center-of-mass motional frequencies of nanoparticles in ion traps lie in the kilohertz regime, rovibrational modes have megahertz frequencies [32] similar to those of the ions' center-of-mass motion, enabling both resonant [33] and dispersive couplings [34]. Such a hybrid system has been achieved by coupling single superconducting qubits to a piezoelectric resonator [35], a surface-acoustic-wave resonator [36], and

a bulk-acoustic-wave resonator [37]; the qubit serves to generate and probe quantum states of motion of the resonator [38]. Building on these pioneering results, levitated particles offer the novel prospect of preparing quantum mechanical superpositions with a spatial extent larger than the mechanical oscillator itself.

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The authors declare no competing interests.

Data availability—Source data are available on Zenodo at [39].

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