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Letter

Extending Vacuum Trapping to Absorbing Objects with Hybrid Paul-Optical Traps

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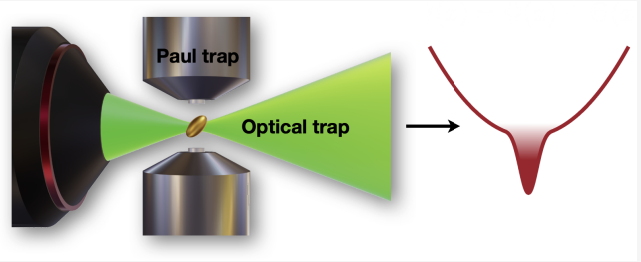
ABSTRACT: The levitation of condensed matter in vacuum allows the study of its physical properties under extreme isolation from the environment. It also offers a venue to investigate quantum mechanics with large systems, at the transition between the quantum and classical worlds. In this work, we study a novel hybrid levitation platform that combines a Paul trap with a weak but highly focused laser beam, a configuration that integrates a deep potential with excellent confinement and motion detection. We combine simulations and experiments to demonstrate the potential of this approach to extend vacuum trapping and interrogation to a broader range of nanomaterials, such as absorbing particles. We study the stability and dynamics of different specimens, such as fluorescent dielectric crystals and gold nanorods, and demonstrate stable trapping down to pressures of 1 mbar.



KEYWORDS: *optical trap, hybrid Paul-optical trap, vacuum trapping, levitated nanoparticles*

INTRODUCTION

The study of micro- and nanosized systems often requires good isolation from the environment. This can be challenging, because they usually either lie on a substrate or are surrounded by a liquid,[1] which can significantly alter their intrinsic physical and chemical properties. Moreover, their small dimensions are often associated with weak signals, which are difficult to separate from environmental noise. A possible solution is to hold the specimen of interest in a trap, preferentially at low pressures.[2−5]

Particle traps can be realized in a number of ways. Paul traps use a combination of AC and DC electric fields for the confinement of charged particles in air and vacuum.[6] Their use for the study of micro- and nanoparticles is nowadays widespread,[5,7−12] for instance, to investigate the optical properties of atmospheric aerosol droplets.[13,14] Optical tweezers—or, more generally, optical dipole traps—allow one to trap dielectric particles near the maximum of a light intensity distribution, like the focus of a strongly focused laser beam.[15−17] Although mostly used to trap dielectric microparticles suspended in a liquid,[18] optical tweezers can also levitate micro- and nanoparticles in air or vacuum.[19,20]

Nevertheless, both approaches have limitations that restrict their use to specific nanoparticle types and interrogation schemes. Optical tweezers, on the one hand, require high powers to trap in vacuum, typically ∼100 mW.[20,21] Such powers are responsible for substantial heating,[22] leading to particle photodamage already at pressures of a few tens of millibars.[23] Hence, low damping regimes—often the most interesting for fundamental studies—can only be accessed with low absorbing materials like silica.[24] On the other hand, Paul traps have low spatial confinement,[25,26] hence limiting the ability to interact with the trapped specimen and detect its position accurately.

Hybrid traps present a possible workaround beyond Paul traps and optical tweezers, which adds further flexibility by combining two types of fields. For instance, trapping of micro- and nanoparticles was reported in magneto-gravitational traps,[27−29] as well as in a Paul trap interfaced to a Fabry−Perot cavity.[30,31]

Here, we implement and characterize a hybrid platform combining Paul and optical traps (see Figure 1). By superimposing a weak but tightly focused laser beam to the potential created by a Paul trap, we form a *dimple* trap,[32] combining high particle confinement with reduced bulk heating. We demonstrate that our platform is versatile both for levitating and optically interrogating in vacuum particles with high optical absorption. In particular, we validate the system with gold nanoparticles, nanodiamonds, and crystals hosting rare earth ions (erbium-doped particles). We also study numerically and experimentally the particle dynamics in terms of its position distribution at equilibrium, which we use to reconstruct the trapping potential.

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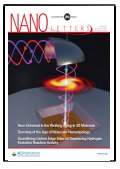

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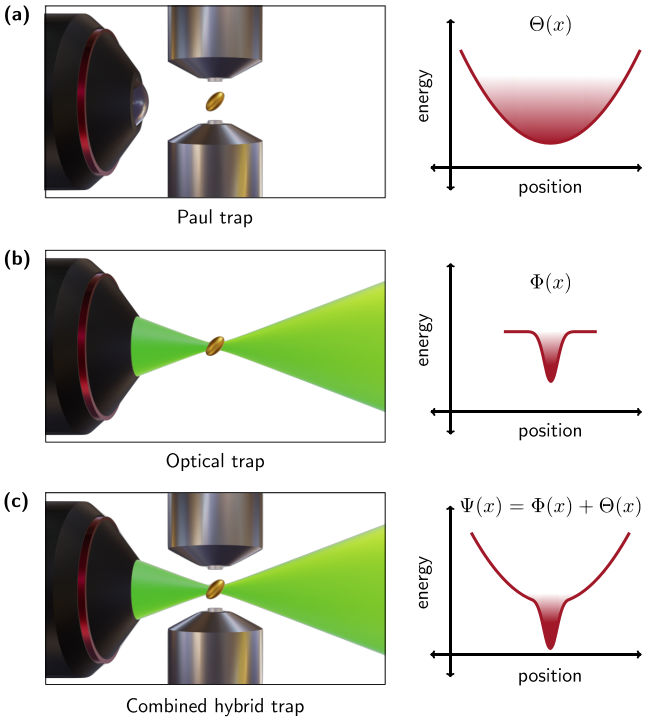

Figure 1. Sketch of the hybrid Paul-optical trap. (a) A charged nanoparticle (here a gold nanorod) is trapped in a deep quadratic potential $\Theta(x)$, created by the Paul trap. The particle may explore a large volume because the potential has a small gradient. (b) The particle is now optically trapped in potential $\Phi(x)$. The particle is tightly confined, but the potential depth is low. (c) When both the Paul trap and the optical trap are activated, the particle is stored in a dimple potential $\Psi(x) = \Theta(x) + \Phi(x)$.

EXPERIMENTAL SETUP

The hybrid Paul-optical platform is sketched in Figure 1 (c). It combines an end-cap Paul trap made of two steel electrodes with rotational symmetry with a 0.8 NA objective lens focusing a 532 nm laser beam (power, $P \leq 20$ mW). The electrodes were designed to provide a linear electric field in a large volume around the trapping region while maintaining high optical access to the particle. Typical parameter ranges for the frequency and amplitude of the driving AC field were 1 kHz−30 kHz and 0.6 kV_{pp} − 2 kV_{pp}, respectively. A piezoelectric stage was used to adjust the position of the electrodes with respect to the laser focus with submicrometer accuracy. The trap, focusing objective, and lens used to collect the forward scattered light are mounted inside a vacuum chamber.

The particles were prepared in ethanol suspensions to load them into the trap. Loading was realized at ambient pressure with a custom-made electrospray, illuminating the trapping volume with a weakly focused 980 nm laser to detect incoming particles. Once a single one was trapped in the Paul trap, we turned on the 532 nm laser, responsible for the optical potential of the hybrid trap. Together, the Paul trap's electric field and the optical gradient force from the focused laser beam produce a dimple-like effective potential, as illustrated in Figure 1 (c). In this situation, the particle is confined to a region that is significantly smaller than what would be achieved with only the Paul trap. Notice that the laser beam alone would not be able to keep the particle trapped at these low powers.

The relative position between the Paul and optical traps must be accurately adjusted to ensure that the potential minima are very close to each other. Otherwise, the particle crosses the optical potential barrier (which is much shallower than the Paul trap potential) and its dynamics are solely governed by the Paul trap electric field, as shown in Figure 2.

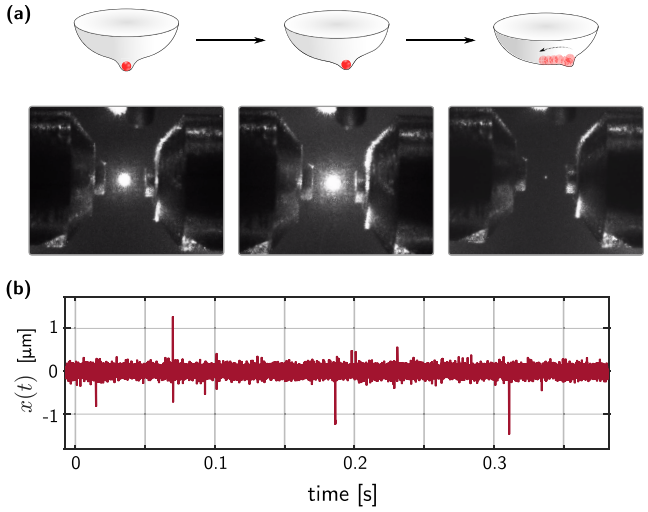

Figure 2. Changing the relative position between Paul and optical traps. (a) The Paul trap is displaced with respect to the optical field with a piezoelectric nanopositioner. If this displacement is small, the trapped particle stays in the potential minimum (i.e., at the focus of the beam). However, if the displacement is increased further, the Paul trap eventually pushes the particle back to its center and its brightness decreases suddenly. The photographs portray the Paul trap moving toward the incoming beam (from top to the bottom of the picture). In this case, the maximum of the particle brightness takes place when the Paul trap and the optical trap are slightly displaced, and the Paul trap exactly compensates the optical scattering force. (b) Time trace of a particle in the hybrid trap (upper-left picture situation) at low optical power (1 mW of 532 nm light). The spikes indicate that the particle hops in and out of the focus.

Position detection is achieved by interferometric measurements of the particle scattered light. When the particle is located in the optical dimple, the 532 nm forward-scattered light is collected and used to illuminate a quadrant photodiode, which returns three electric signals that are proportional to the particle motion along the $x(t)$ (trap axis), $y(t)$ (gravity), and $z(t)$ (beam) directions.

RESULTS AND ANALYSIS

We tested the hybrid trap with both dielectric and metallic particles. Gold nanoparticles are particularly hard to trap in air with optical tweezers, even at ambient pressure.[33] They cannot dissipate efficiently the energy absorbed from the laser beam, and their bulk temperature increases considerably. To assess their survival and stability, we loaded gold nanorods of 33 nm × 63 nm into the Paul trap at ambient pressure, trapped them in the dimple trap with 532 nm light, and brought the system to vacuum. As can be seen in Figure 3, with the hybrid trap the gold particles could be maintained in the trap down to 10 mbar. Below this pressure, and depending on the power of the optical trap, the probability of the nanorods disappearing from the trap increased dramatically. At ∼1 mbar most of the studied particles vanished, except for low optical powers (below ∼3 mW). In the latter case, it was not possible to maintain a stably trapped particle, i.e., it hopped in and out of the focus intermittently and thus received a considerably lower average optical power. We

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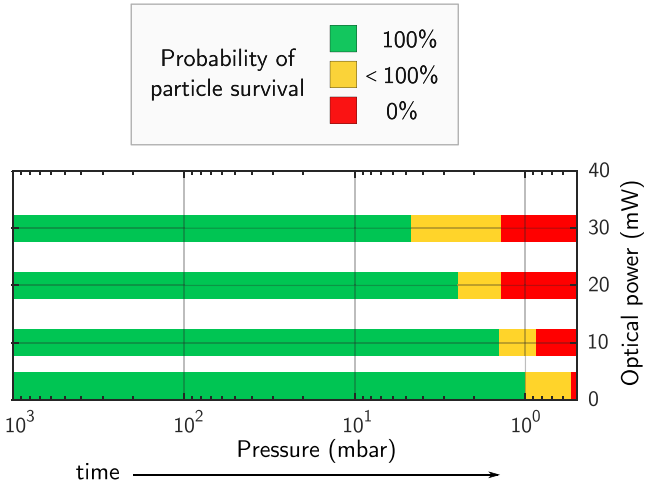

Figure 3. Experimental probability of survival of a levitated gold nanorod in the hybrid trap when the pressure is progressively reduced (using 532 nm light). Green color marks that all particles achieved said pressure, yellow marks the survival of some (but not all) of the trapped particles, and red marks that no particle could be brought to that pressure. Most of the gold nanoparticles are lost around 1 mbar. We repeated the experiment with 1064 nm light, obtaining similar results.

also observed that, when the particles disappeared from the trap, the event happened rather suddenly, without clear prior changes in particle brightness. We speculate that this is due to the evaporation of the outer layer of the gold particle, which leads to a sudden change in charge-to-mass ratio and to unstable trapping conditions.[6]

To study the dynamics of the particles in the hybrid trap, we analyze the dominant forces in the absence of absorption effects. The center of mass (COM) Langevin equation of motion of a particle in an electric Paul-optical trap can be obtained with Newton's second law:

$$m\ddot{x} + m\Gamma\dot{x} - \frac{QV}{d^2}\cos\omega_d t\cdot x + \alpha\frac{1}{2}\nabla|\mathbf{E}|^2 = \sigma\eta(t) \tag{1}$$

Here, $x(t)$ is the COM motion, m is the particle mass,[34] and Γ is the damping rate due to the interaction with residual gas molecules. From the Paul trap, ω_d is the trap driving frequency, Q is the particle charge, V the trap voltage amplitude, and d is the trap's characteristic size, determined by the geometry of the electrodes. Regarding the optical trap, α is the real part of the polarizability of the particle and $\mathbf{E} \triangleq \mathbf{E}(x, y, z)$ is the optical field, which we approximate as a focused Gaussian beam (see the Supporting Information) to perform numerical simulations. Finally, $\sigma\eta(t)$ is a stochastic force that accounts for the thermal coupling with the environment, with $\eta(t)$ being a unit intensity Gaussian white noise and $\sigma = \sqrt{2k_B T m\Gamma}$.[35,36]

In eq 1 we do not consider gravity, which is small compared to the electric force (from the Paul trap) and the gradient force (from the optical tweezers field), nor the scattering force, which can be neglected for nanoparticles in tightly focused beams—as is our case when the particle is in the dimple of the hybrid trap [Note: When the particle is *pushed* away from the center by the Paul trap, as shown in Figure 1, the scattering force might become apparent]. Since both relevant deterministic forces are gradients, the total potential will be the addition of the two individual trap potentials. However, notice that at low laser powers the levitated particle explores large regions of the optical trap and the dipole force cannot be approximated as a linear restoring force, as is common practice in optical tweezers experiments (i.e., there will be significant nonlinearities in the dynamics). The Paul trap potential, being much larger, can still be safely approximated as a quadratic (time-varying) potential.

If we consider the adiabatic approximation for the Paul trap, i.e., we approximate the time-varying potential by an effective constant potential,[36] then at equilibrium $x(t)$ will follow the Gibbs probability density function

$$\rho_\infty(x) = \frac{1}{Z}\exp(-\beta\Psi(x)) \tag{2}$$

where Z is a normalizing factor, $\beta = 1/k_B T$, and $\Psi(x)$ is the combined hybrid potential, introduced in Figure 1. Hence, by estimating $\rho_\infty(x)$ and inverting eq 2, we can recover the effective potential of the hybrid trap.

This process is shown in Figure 4 with simulated time traces. Here, we studied theoretically the effects of a decrease in the optical field's intensity. The simulations reveal that when the laser power is below 8 mW, the position histograms start to deviate noticeably from a normal distribution. This is due to a reduced particle confinement that lets the particle explore the nonlinearities away from the optical trap center. Eventually, at sufficiently low powers, the particle starts leaving and re-entering the beam focus.

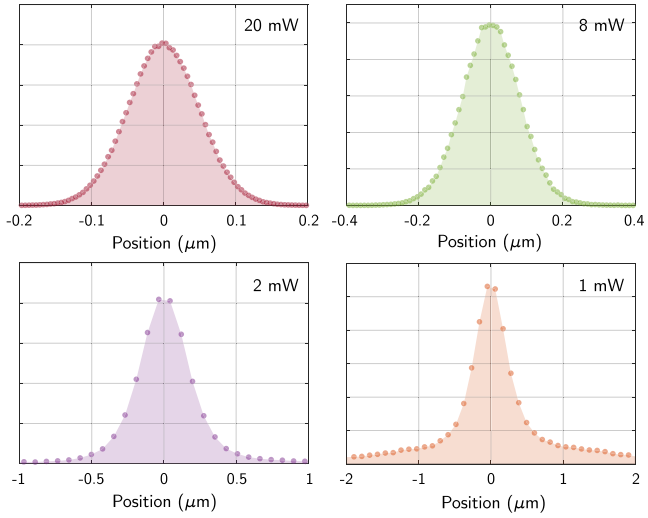

Figure 4. Surge of nonlinearities at low intensities. The plots show numerical simulations of the position density distribution $\rho_\infty(x)$ of 100 nm dielectric nanoparticles (no absorption considered) for decreasing optical powers in the hybrid trap (λ = 1064 nm). Below 20 mW, a reduction in the laser power leads to position distributions that deviate more and more from a normal distribution (quadratic potential case, exemplified with the upper left plot). At 1 mW, the particle leaves and re-enters the beam focus intermittently, resulting in long tails in $\rho_\infty(x)$. Notice that the x-axis scale varies along the different plots.

We observed a similar behavior experimentally. Starting at a high power (e.g., 20 mW of 532 nm laser light), a progressive reduction of the power leads to a blinking of the particle, which we interpret as the particle hopping in and out of the optical trap. Experimentally, the blinking started at higher powers than in the simulations, but this was expected because the two potentials (optical and electric) are never perfectly aligned. Hence, the driving from the Paul trap may push the particle away from the focus before that in the idealized simulation.

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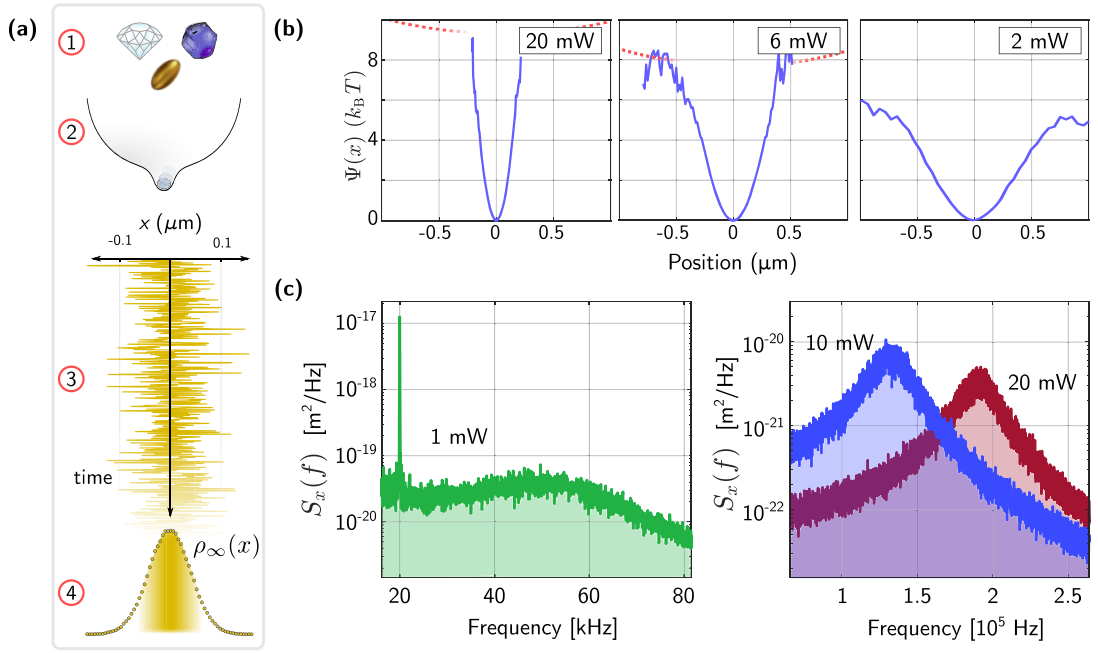

Figure 5. Potential reconstruction. (a) Process of potential reconstruction. A nanoparticle (1) oscillates in the hybrid potential (2). A time trace $x(t)$ of the trapped particle is measured for a few seconds (3). From the time trace, we compute the histogram of the position (4) to estimate the Gibbs probability density function $\rho_\infty(x)$ (see eq 2). From this density function, we may obtain $\Psi(x) = \frac{-1}{\beta}\ln(Z\rho_\infty(x))$. (b) Reconstructed trap potentials for three different powers of 532 nm laser light (blue line) with a levitated nanodiamond ($r \sim 70$ nm) at ~100 mbar. The dashed red lines are only a guide to the eye for the Paul trap section of the combined potential. Potentials reconstructed with other particle species can be found in the Supporting Information. (c) PSDs of the motion at different laser powers (pressure inside the vacuum chamber of 50 mbar, $\lambda = 532$ nm). At 1 mW the optical field is not strong enough to keep the particle in the dimple, and the driving ($\omega_d = 20$ kHz) dominates the dynamics. At higher laser powers, the particle is trapped in the dimple of the potential well, where it oscillates driven by Brownian motion. As expected, both the trap frequency and the confinement increase with laser power.

The potential reconstruction from experimentally trapped dielectric particles is shown in Figure 5, which plots the profiles obtained at different powers (similar results are shown for gold and erbium-doped nanoparticles in the Supporting Information). The potential has the shape of a dimple trap, consisting of a large, flatter trapping volume (created by the Paul trap), superimposed to a tighter optical dipole trap, as represented in Figure 5 (b) with data from a levitated nanodiamond. In these time traces, we filtered out the contribution of the driving AC field, which is never completely eliminated through field compensation and perturbs the results. The presence of nonlinearities in the detection system (i.e., in the correspondence between detected signal and position) also posed a problem for such potential reconstructions. To correct for this, we eliminated fractions of the recorded time traces in which it was clear that the particle was away from the trap center and inverted the remaining data with the expression found in Gittes et al.[37]

PSDs of the particle motion are also shown in Figure 5 (c) for different optical powers. At 1 mW, the optical field is too weak to keep the particle in the focus. As a result, the particle hops in and out of the dimple and the dynamics are dominated by the Paul trap driving (peak at $\omega_d = 20$ kHz). This hopping disappears as the laser power is increased to higher values and is almost absent for powers as low as 10 mW. In this situation, the particle oscillates with a frequency in the range of hundreds of kHz, significantly larger than in typical experiments with nanoparticles in Paul traps.[12,26]

CONCLUSIONS

In conclusion, we built a hybrid nanoparticle trap by combining a Paul trap with a weak but highly focused optical beam, and demonstrated its suitability as an experimental platform to trap and store highly absorbing particle species in vacuum.[12,38,39] Even though our experiments were performed with nanoparticles, similar results are expected with larger particles of up to a few micrometers.[13,40]

We validated our platform with gold, diamond, and erbium-doped nanoparticles, trapping and detecting them at optical powers below 10 mW, which are low compared to typical optical tweezer powers (~100 mW). We also verified that the hybrid scheme easily allows a reduction of the vacuum pressure, even below ~5 mbar with gold nanoparticles. This contrasts with previous optical levitation experiments in vacuum, where due to optical absorption of the trapped particles,[22,41] the range of materials was limited to just a few options. In our hybrid scheme there is still room to reduce the heating, since in most experiments the presence of the optical field is only required to perform short measurements. To store the particles the Paul trap suffices, allowing for arbitrarily long levitation times.[12]

Moreover, we used the measured position traces to reconstruct the effective potential with the Boltzmann−Gibbs distribution, comparing the results to numerical simulations of the stochastic equation of motion. As is intuitively expected, the trap potential has a dip in the middle, corresponding to the optical dipole gradient potential.

Due to its versatility, the studied hybrid platform is suited to investigate small objects under experimental constraints that are

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difficult to overcome with other trapping techniques. In aerosol science, hybrid traps could improve the confinement of the levitated particles, boosting the sensitivity of the Raman and infrared signals that are commonly targeted.[13] Another promising direction is the study of isolated particles that strongly interact with light, such as metals[42] or particles with internal degrees of freedom.[12,39] Prime examples of these are diamonds with color centers, quantum dots, or crystals hosting rare earth ions, which live in a domain that is mostly classical but still not completely free of quantum effects.[43] Finally, the ability to both move the optical trap and change its depth, in combination with control over the Paul trap, could be used to experimentally explore complex dynamics in bistable potentials.[44−46]

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.0c02025.

Information regarding the experimental methods, numerical simulations, and particles that have been used in this work (PDF)

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Notes

The authors declare no competing financial interest.

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