NANOPHYSICS

Tunable light-induced dipole-dipole interaction between optically levitated nanoparticles

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Arrays of optically trapped nanoparticles have emerged as a platform for the study of complex nonequilibrium phenomena. Analogous to atomic many-body systems, one of the crucial ingredients is the ability to precisely control the interactions between particles. However, the optical interactions studied thus far only provide conservative optical binding forces of limited tunability. In this work, we exploit the phase coherence between the optical fields that drive the light-induced dipole-dipole interaction to couple two nanoparticles. In addition, we effectively switch off the optical interaction and observe electrostatic coupling between charged particles. Our results provide a route to developing fully programmable many-body systems of interacting nanoparticles with tunable nonreciprocal interactions, which are instrumental for exploring entanglement and topological phases in arrays of levitated nanoparticles.

When a dielectric subwavelength particle is illuminated by laser light, the particle is polarized in phase with the incoming electromagnetic wave. The induced dipole makes the particle a high-field seeker, which enables optical trapping in the intensity maximum of focused lasers (1). Additionally, the dipole radiation field acquires the optical phase of the trapping field. This process, called coherent scattering, has been used in combination with an optical cavity to cool the motion of atoms and polarizable nanoparticles in far-detuned traps (2–5). More recently, it has been applied to achieve motional ground-state cooling of single silica nanoparticles in an optical cavity (6) and with real-time feedback (7, 8).

Simultaneous trapping of more than one particle in a single optical potential allows for the creation of a self-organized structure of particles that interact through the scattered light (9). The particles assume steady-state positions at locations where the constructive interference of scattered fields is maximized, thereby minimizing the total energy. This optical interaction is fundamentally conservative and reciprocal, giving rise to a spring-type interaction called optical binding (9, 10). Optical binding between dielectric objects has been realized for microparticles (where the radius is comparable to or larger than the wavelength) in many experiments (10–18). In this regime, the effect is well described by Mie scattering theory (9), in which the light scattered from one of these particles is not spatially coherent over the extension of a neighboring particle. For the case of nanoscale objects, optical binding has been demonstrated with metal particles in liquid, where plasmon resonances enhance the interaction (19–21). However, making full use of the opportunities provided by optically trapped nanoparticle arrays for investigating complex nonequilibrium phenomena requires controllable interactions beyond the current framework of optical binding (22–24). The tools presented in this article pave the way to using the technology of atomic physics (25, 26) for the generation and observation of quantum correlations and topological phases in a fully programmable mechanical array (27, 28).

In contrast with previous experiments, our work shows fully tunable and nonreciprocal optical interactions between two silica nanoparticles—with radius \( r = 105 \pm 3 \) nm appreciably smaller than the wavelength \( \lambda = 1064 \) nm—that are levitated in two distinct, phase-coherent optical traps at a variable trap separation \( d_0 \). Each particle behaves as an induced dipole driven by the total optical field, which is a sum of the trapping field, and the coherently scattered light from the other particle. The interference between these two fields gives rise to the interaction between the particles and affects their motion in all three dimensions. The total light-induced interaction is a combination of a conservative gradient force and a nonconservative radiation pressure force, in analogy to the forces acting on a single nanoparticle in an optical trap (1). The relevant contributions to the optical interparticle forces oscillate periodically and decay with the interparticle distance \( d \) as (29)

\[ F_{1,2} \propto \sin(kd + \Delta \phi) [2\kappa n + (k - 1/n_k) e_1]/kd \]

in the far field \((kd \gg 1, k = 2\pi/\lambda)\). Here, \( \Delta \phi \) denotes the optical phase difference between the trapping lasers at the particle position, \( n \) is

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**Fig. 1. Experimental setup.** (A) Two laser beams are diffracted by the SLM and focused with the microscope objective to create two distinct optical tweezers. The optical traps are in the vacuum chamber at a pressure of \(-1\) mbar. The light is collected after the focus and used for detection of mechanical modes. We drive the electrodes with a voltage \( V \) to calibrate the number of charges. (B) Camera image of two nanoparticles (radius \( r = 105 \) nm) trapped in two optical traps at a distance \( d_0 = 10 \) μm. (C) Side view (above) and top view (below) of the trap foci. Two parallel laser beams are used to trap two nanoparticles at a distance \( d_0 \). Intrinsic mechanical frequencies along the \( z \) axis \( \Omega_z \propto \sqrt{1 + \eta} \) and \( \Omega_1 \propto \sqrt{1 - \eta} \) are controlled by a single parameter \( \eta \). Polarization is set along the \( y \) axis to maximize dipole radiation along the \( x \) axis. We set the optical phases \( \phi_1 \) and \( \phi_2 \) with the SLM. (D) Our system can be simplified to two harmonic oscillators with frequencies \( \Omega_{1,2} \), which are coupled through dipole-dipole interaction with nonreciprocal coupling rates \( g \pm \bar{g} \). (E) In case of reciprocal coupling \((\bar{g} = 0)\), the normal modes of the system, the COM mode \( z \), and the breathing mode \( z \), are nondegenerate with frequencies \( \Omega_z = \Omega \) and \( \Omega_1 = \Omega + 2g \), respectively. We observe the normal modes at frequencies \( \Omega_z /2\pi = 51 \) kHz and \( \Omega_1 /2\pi = 63 \) kHz in the power spectral density (PSD) of the detector signal, which we fit to determine the mechanical frequencies. a.u., arbitrary units.
the optical phase differences is always at the frequency \( h \).

The breathing mode \( \Omega \) has a higher or lower frequency by \( 2g/2\pi = 8 \text{ kHz} \) for \( \Delta h_0 = 0 \) or \( \Delta h_0 = \pi \), respectively. Black dashed lines are fits to experimental data. The avoided crossing is shifted from \( \eta = 0 \) for \( \Delta h_0 = \pi \) because of the interference of the trapping lasers in the focal plane. The amplitude of the detected motion is not constant owing to variations in detection sensitivity during the measurement.

The linearized dynamics with particle center-of-mass (COM) positions \( z_{1,2} \) follow as (29)

\[
\begin{align*}
\dot{m}_1 z_1 + m_1 \ddot{z}_1 &= -(m \Omega_1^2 + k_1 + k_2)z_1 + (k_1 + k_2)z_2 + (k_1 - k_2)z_0, \\
\dot{m}_2 z_2 + m_2 \ddot{z}_2 &= -(m \Omega_2^2 + k_1 - k_2)z_2 + (k_1 - k_2)z_0
\end{align*}
\]

where \( m \) is the mass of each particle, and the displacement resulting from the homogeneous part of the forces has been absorbed in \( z_{0,2} \). The spring constant \( k_1 = K \cos(kd_0) \cos(h_{\text{tot}})/kd_0 \) describes the conservative part of the optical forces (tunable optical binding), whereas \( k_2 = G \sin(kd_0) \sin(h_{\text{tot}})/kd_0 \) describes a nonconservative interaction, as indicated by a change of sign between the equations. The non-reciprocity of the interaction is maximized by \( \Delta h_0 = \pi/2 + n\pi \), where \( n \in \mathbb{Z} \). The constant \( G = \omega^2 \sqrt{P_1 P_2} \) is a positive function of the trap powers \( P_1,2 \) and the particle polarizability \( \alpha \). The scaling of \( G \) with \( \alpha^2 \) reflects the nature of the dipole-dipole interaction.

The scaling of \( G \) with \( \alpha^2 \) reflects the nature of the dipole-dipole interaction. At the pressures in our experiment, mechanical damping \( \gamma \) is dominated by the collisions with the surrounding gas. For weak coupling between the \( k_1, k_2 < m \Omega_1^2 \), Eq. 1 yields the eigenfrequencies of the coupled system \( \Omega_\pm (\eta m) = \Omega \mp g^2 \sqrt{\gamma^2 - g^2} \), where we define the conservative and nonconservative coupling rates as \( g = k_1/2m\Omega_1 \) and \( g = k_2/2m\Omega_2 \), respectively. The control parameter \( \eta m = -k_2/m\Omega_2^2 \) defines the value at which the frequency splitting \( \Omega_+ - \Omega_- \) is minimal and \( \Omega \) is the intrinsic mechanical frequency in the absence of interactions at \( \eta = 0 \). Therefore, at the avoided crossing, our system is described as two harmonic oscillators with frequencies \( \Omega_{\pm} \) mutually coupled with non-reciprocal coupling rates \( g \mp \eta \) (Fig. 1D), which...
Fig. 4. Turning off dipole-dipole interaction to detect electrostatic interaction. (A) For arbitrary polarization angle θ, the interference of electric fields is suppressed by \( \cos^2 \theta \). Two special cases of \( \theta = 0^\circ \) and \( \theta = 90^\circ \) are presented in green and orange rectangles, respectively. In the case of \( \theta = 90^\circ \), there is no interference of the trapping and the scattered fields. (B) We measure the coupling rate resulting from the dipole-dipole interaction as a function of the polarization angle θ between particles with an absolute number of \( 1 \pm 1 \) and \( 0 \pm 1 \) charges (circles). The interaction is maximal for the angle \( \theta = 0^\circ \) (green circle). The avoided crossing is unresolved for coupling rates smaller than the mechanical linewidth \( Q \). At the angle \( \theta = 90^\circ \), the dipole-dipole interaction is suppressed (orange circle). We measure coupling resulting from the electrostatic interaction between particles with \( 96 \pm 21 \) and \( 110 \pm 24 \) charges (orange triangle). (C) The avoided crossing is absent for horizontally polarized tweezers (\( \theta = 90^\circ \)) because the far-field dipole-dipole interaction is strongly suppressed. (D) In the absence of optical interactions, we observe the avoided crossing for highly charged particles owing to strong electrostatic interaction.

can be fully controlled by the distance \( d_0 \) and the optical phase difference \( \Delta \phi_0 \). In the case of purely conservative interaction (\( g = 0 \)) and for equal mechanical frequencies, the normal modes of the system become the COM mode \( z_+ = z_1 + z_2 \) and the breathing mode \( z_0 = z_1 - z_2 \). Only the breathing mode is affected by the interaction, such that its eigenfrequency shifts to \( \Omega = \Omega + g \) whereas the COM mode eigenfrequency remains unchanged at \( \Omega = \Omega \) (normal mode splitting). In Fig. 1E, we show the normal modes at \( \Omega / 2 \pi = 51 \text{ kHz} \) and \( \Omega / 2 \pi = 63 \text{ kHz} \) in the power spectral density of the detector signal, where the mechanical frequency is \( \Omega / 2 \pi = 51 \text{ kHz} \) when the interaction is switched off.

To obtain the coupling rate \( g \), we measure the eigenfrequencies as a function of \( \eta = \frac{1}{2} / \pi \) (Fig. 2). We set the trap separation to \( d_0 = 3.15 \text{ μm} \) and the optical phase difference to either \( \Delta \phi_0 = 0 \) (above) or \( \Delta \phi_0 = \pi \) (below), such that the interaction is purely conservative but of either positive (attractive) or negative (repulsive) nature. The spectrogram exhibits an avoided crossing between the normal modes, which typically occurs for equal intrinsic mechanical frequencies (\( \eta = 0 \)), but only if \( |g| > \gamma / 2 \). All measurements are conducted at pressures of \( -1.5 \text{ mbar} \), thus the avoided crossing is observable for coupling rates larger than \( \gamma / 2 \pi = 1.5 \text{ kHz} \). From here on, the coupling rate is expressed in units of the modified mechanical frequency \( \Omega' = \Omega + g \) as the ratio \( g / \Omega' \) is independent of the optical power. We observe a frequency splitting of \( -\approx 8 \text{ kHz} \), which corresponds to a coupling of \( g / \Omega' = 0.2 (0.09 \pm 0.01) \). Other coupling mechanisms can be neglected in our investigations of the light-induced dipole-dipole interaction. First, we select particles with few charges, such that the additional coupling as a result of the electrostatic interaction was smaller than \( |gC / \Omega' = (1.4 \pm 0.6) \times 10^{-3} | \) (29). Furthermore, interparticle coupling arising from the ambient gas or liquid (aero- or hydrodynamic coupling) can be dominant for large objects and small distances; however, in our experiment it is negligible because the ratio of the particle radius to the trap separation is small \( (r / 2d_0 < 0.05) \) (16, 18).

In our experiment, we achieve full control over the conservative and nonconservative coupling rates. Because the interaction arises from the interference between the trapping and scattered fields, we expect the coupling rate to oscillate with particle distance with a period of \( \lambda \) and decay as \( d_0^{-3} \) owing to the far-field nature of the dipole radiation at distances \( d_0 > \lambda \). We measure the normal mode splitting for trap separations \( d_0 \) in the range of \( 2.2 \) to \( 3.7 \text{ μm} \) and for phase differences \( \Delta \phi_0 = 0 \) (blue points) or \( \Delta \phi_0 = \pi \) (orange points) to maximize the conservative interaction (Fig. 3A). Good agreement is observed with our theoretical model and the measured coupling rates in both cases (29). At a distance of \( d_0 = 2.2 \text{ μm} \) and for \( \Delta \phi_0 = 0 \), we observe the maximum coupling of \( g / \Omega' = 0.186 \pm 0.017 \). The effect of the dominant nonconservative interaction is apparent for the trap separation of \( d_0 = 2.2 \text{ μm} \) and the phase difference of \( \Delta \phi_0 = 0.8 \pi \) (Fig. 3B). The constant pumping of energy into the system as a result of the nonconservative forces increases the particle motional amplitude by an order of magnitude. The eigenfrequencies are degenerate for \( \eta \in [0, 0.07] \), from which we estimate the couplings of \( g / \Omega \approx 0 \) and \( \xi / \Omega \approx -0.017 \). To demonstrate its dependence on the optical phase difference \( \Delta \phi_0 \), we measure the normal mode splitting at a fixed separation of \( d_0 = 3.2 \text{ μm} \) (Fig. 3C). Our theoretical model of linear interactions (blue line) fails to fully predict the observed behavior because of several effects. For example, the actual interparticle distance is different from the trap separation owing to the radiation pressure force of the dipole radiation, which provides a constant displacement force. Moreover, in the absence of an additional cooling mechanism, the particles are able to explore nonlinear terms in the interaction Hamiltonian, which affects the eigenfrequencies and modifies the normal mode splitting. We observe a zero crossing because of the absence of the conservative forces at the phase of \( \Delta \phi_0 = 0.8 \pi \), in agreement with our measurement in Fig. 3B. In future work, feedback cooling can be used to constrain the particle motion within linear dynamics.

Rotating the trapping laser polarization by an angle \( \Theta \) from the \( y \) axis provides for another way to control the dipole-dipole interaction (Fig. 4A). The magnitude of the dipole radiation along the \( x \) axis is smaller by a factor of \( \cos \Theta \) owing to the characteristic spatial profile of the dipole radiation in the far field. The interference of the dipole radiation with the trapping field is suppressed by a factor of \( \cos \Theta \) as a result of the scalar product of the two field components. Altogether, this yields a decrease of the coupling rate by \( \cos^2 \Theta \) in the far-field approximation, which is confirmed in the measurement in Fig. 4B (circles and blue line). For the angle \( \Theta = 90^\circ \), the residual dipole-dipole interaction scales with \( (kd_0)^{-3} \) because of the radial near-field component of the radiated field. We estimate the coupling of \( g / \Omega' \approx 6 \times 10^{-4} \) at \( d_0 \approx 3 \text{ μm} \), which we are unable to detect in the current experiment as \( g / \gamma < 10^{-2} \) (Fig. 4C) (29). Suppression of the dipole-dipole interaction allows us to explore electrostatic interaction between strongly charged particles. We trap particles with absolute charges \( |q_1| / e = 96 \pm 21 \) and \( |q_2| / e = 110 \pm 24 \) and equal signs \( (q_1q_2 = |q_1q_2|) \) and observe an avoided crossing as a result of...
electrostatic coupling (Fig. 4D). The ordering of normal mode frequencies $\Omega_2 < \Omega_1$ reflects the repulsive interaction between the particles. The measured coupling $g_C/\Omega_2 = -0.035 \pm 0.003$ fits well to the expected $g_C/\Omega_2 = -0.047 \pm 0.015$. Because the electrostatic coupling rate scales as $\propto d_0^{-3}$, we infer that electrostatic interaction between particles—each carrying a single charge—can be resolved at a distance of $d_0 \approx 2 \mu$m and at pressures below $10^{-3}$ mbar. Altogether, our platform allows for exploring hybrid schemes with both dipole-dipole and electrostatic interactions.

We have demonstrated the controllable (attractive and repulsive) light-induced dipole-dipole interaction between two silica nanoparticles levitated in distinct optical traps with coupling rates up to 20% of the mechanical frequency. These results expand the toolbox of optical binding by trapping in a phase-coherent optical tweezers array, which will enable further studies of optical interactions between Rayleigh particles (9) or atoms (30) at subwavelength distances. Furthermore, we control the nonreciprocal interactions between the particles by tuning conservative and non-conservative interactions. By effectively switching off the optical interaction, we observe electrostatic interaction between two charged particles. The demonstrated strength and level of control of optical and electrostatic interactions in arrays of levitated solid-state objects, in combination with the previously realized quantum state preparation, provides a platform that may open up many research avenues in quantum physics. We foresee that this platform—with a possible addition of an optical cavity—can be used for quantum simulation with mechanical degrees of freedom (31–33), enhanced quantum sensing (34), collective effects (35), and phonon transport and thermalization (24).

REFERENCES AND NOTES
1. A. Ashkin, J. M. Dziedzic, J. E. Bjorkholm, S. Chu, Opt. Lett. 11, 288 (1986).
2. D. R. Leibrandt, J. Lobaszewicz, V. Vuletić, I. L. Chuang, Phys. Rev. Lett. 103, 103001 (2009).
3. M. Hosseini, Y. Duan, K. M. Beck, Y.-T. Chen, V. Vuletić, Phys. Rev. Lett. 118, 133601 (2017).
4. U. Delić et al., Phys. Rev. Lett. 122, 123602 (2019).
5. D. Windley et al., Phys. Rev. Lett. 122, 123601 (2019).
6. U. Delić et al., Science 367, 892–895 (2020).
7. L. Magnin et al., Nature 595, 373–377 (2021).
8. F. Tekbenjhanishi, M. L. Mattana, M. Rossi, M. Frimmer, L. Novotny, Nature 595, 378–382 (2021).
9. K. Dholakia, P. Zemánek, Rev. Mod. Phys. 82, 1767–1791 (2010).
10. M. M. Burns, J.-M. Fournier, J. A. Golovchenko, Phys. Rev. Lett. 63, 1233–1236 (1989).
11. W. Singer, M. Frick, S. Bernet, M. Ritsch-Marte, J. Opt. Soc. Am. B 20, 1568 (2003).
12. S. A. Tatarkova, A. E. Carruthers, K. Dholakia, Phys. Rev. Lett. 89, 283901 (2002).
13. S. Mohanty, J. Andrews, P. Gupta, Opt. Express 12, 2746–2753 (2004).
14. M.-T. Wei, J. Ng, C. T. Chan, H. D. Ou-Yang, Sci. Rep. 6, 38883 (2016).
15. D. S. Bykov et al., Light Sci. Appl. 7, 22 (2018).
16. Y. Arta, E. M. Wright, K. Dholakia, Optica 5, 910 (2018).
17. J. Darraviková et al., Light Sci. Appl. 7, 17135 (2018).
18. V. S. Svak et al., Optica 8, 220 (2021).
19. Y. Zhang, C. Gu, A. M. Schwartzberg, S. Chen, J. Z. Zhang, Phys. Rev. B 73, 165405 (2006).
20. F. Svedberg, Z. Li, H. Hu, M. Käll, Nano Lett. 6, 2639–2641 (2006).
21. V. Demergis, E.-L. Florin, Nano Lett. 12, 5756–5760 (2012).
22. W. Lechner, S. J. M. Habraken, N. Kiesel, M. Aspelmeyer, P. Zoller, Phys. Rev. Lett. 110, 143604 (2013).
23. D. Holzmann, M. Sonnleitner, H. Ritsch, Eur. Phys. J. D 68, 352 (2014).
24. S. Liu, Z. Yin, T. Li, Adv. Quantum Technol. 3, 1900099 (2020).
25. S. Ebadí et al., Nature 595, 227–232 (2021).
26. P. Scholl et al., Nature 595, 233–238 (2021).
27. V. Peano, C. Brendel, M. Schmidt, F. Marquardt, Phys. Rev. X 5, 031015 (2015).
28. N. Goldman, J. C. Budich, P. Zoller, Nat. Phys. 12, 639–645 (2016).
29. See the supplementary materials.