Quantum Optical Binding of Nanoscale Particles

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(Received 10 May 2024; accepted 21 October 2024; published 3 December 2024)

Optical binding refers to the light-induced interaction between two or more objects illuminated by laser fields. The high tunability of the strength, sign, and reciprocity of this interaction renders it highly attractive for controlling nanoscale mechanical motion. Here, we discuss the quantum theory of optical binding and identify unique signatures of this interaction in the quantum regime. We show that these signatures are observable in near-future experiments with levitated nanoparticles. In addition, we prove the impossibility of entanglement induced by far-field optical binding in free space and identify strategies to circumvent this no-go theorem.

DOI: 10.1103/PhysRevLett.133.233603

Optical fields provide unique opportunities for controlling the mechanical motion of dielectric objects in the quantum regime [1]. Optical tweezers can stably suspend nanoscale to microscale particles [2,3], where their center-of-mass and rotational dynamics can be monitored at the standard quantum limit by homodyning the scattered light [4]. Trapped particles can be controlled by a combination of conservative dipole forces and nonconservative radiation pressure [5], and their rotations can be propelled via conservative and nonconservative optical torques [6]. Combining the exquisite control with precise optical readout enables cooling the mechanical motion of submicron particles into the quantum regime with feedback techniques [7,8] or cavity cooling [9–11].

If two or more dielectric particles are trapped close to each other they interact through the field scattered between them. Such *optical binding* interactions give rise to strong and tunable forces [12] that have been observed in numerous setups, ranging from atomic clouds [13] and nanoscale particles [14–18] to chains of microscale particles in fluids [19]. The dominant mechanism of optical binding is that the interference between the scattered light and the trapping light dynamically changes the optical forces acting on the particles. Importantly, this interaction can be strong, tunable, and nonreciprocal [15], meaning that they seemingly violate Newton's law of action equals reaction. This implies that the linearized particle dynamics close to the trap center are described by a non-Hermitian dynamical matrix, exhibiting exceptional points, unidirectional transport, and spontaneous symmetry breaking [16], with great potential for precision sensing [20–22].

To illustrate the richness of optical binding, let us consider the time-averaged force acting on a subwavelength sphere (dipole approximation) with polarizability α at position $\mathbf{r}_1$ in the laser field $\mathbf{E}(\mathbf{r})e^{-ickt}$, with k the wave number, due to the presence of an identical sphere at position $\mathbf{r}_2$ [12],

$$\mathbf{F}_{12} = \frac{\alpha^2}{2\varepsilon_0}\frac{\partial}{\partial \mathbf{r}_1}\mathrm{Re}[\mathbf{E}^*(\mathbf{r}_1)\cdot \mathrm{G}(\mathbf{r}_1-\mathbf{r}_2)\mathbf{E}(\mathbf{r}_2)]. \quad (1)$$

This force is nonreciprocal, $\mathbf{F}_{12} \neq -\mathbf{F}_{21}$, given that the free-space dipole Green tensor for $r > 0$,

$$\mathrm{G}(\mathbf{r}) = \frac{e^{ikr}}{4\pi}\left[(1-ikr)\frac{3\mathbf{r}\otimes\mathbf{r} - r^2\mathbb{1}}{r^5} + k^2\frac{r^2\mathbb{1} - \mathbf{r}\otimes\mathbf{r}}{r^3}\right],$$

is complex-valued. The force being nonreciprocal, the resulting particle dynamics are nonconservative so that both momentum and energy are not conserved, but constantly supplied and extracted by light scattering (see Fig. 1). The recent progress in quantum cooling of levitated nanoparticles [7,8,10,11,23] and the prospects of exploiting nanoparticle arrays in the deep quantum regime for superior force and torque sensing [15,16,18,24–26] raises the need for a full quantum mechanical description of optical binding interactions between cotrapped particles.

This Letter identifies unique quantum signatures of nonreciprocal optical-binding interactions and demonstrates how they can be observed in nanoparticle arrays. Specifically, we will see that optical binding in the quantum regime is described by a Markovian quantum master equation, which recovers the conservative and the nonconservative interaction between the nanoparticles and accounts for the ensuing decoherence. It allows us to rigorously derive three results of considerable experimental relevance. First, the quantum fluctuations of the light fields can induce pronounced correlations between the particles

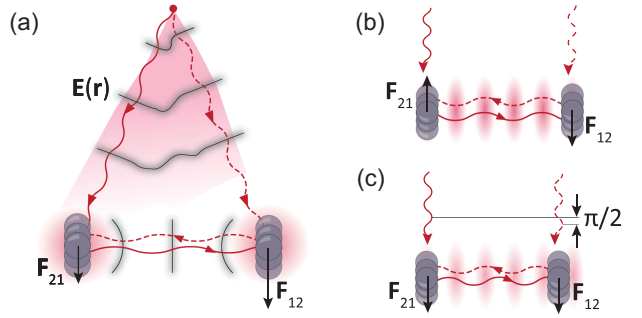

FIG. 1. (a) Two nearby nanoparticles illuminated by a laser $\mathbf{E}(\mathbf{r})$ interact through their scattered light fields. If the laser is phase coherent, the relative local phase can be used to tune the directionality and reciprocity of the mean interaction forces $\langle \mathbf{F}_{12} \rangle$ and $\langle \mathbf{F}_{21} \rangle$. In the quantum regime, this interaction leads to decoherence of the two-particle state and to shot-noise-induced correlations between the particles. (b) If the relative phase vanishes, the mean interaction force is reciprocal $\langle \mathbf{F}_{12} \rangle = -\langle \mathbf{F}_{21} \rangle$, while (c) the interaction becomes antireciprocal $\langle \mathbf{F}_{12} \rangle = \langle \mathbf{F}_{21} \rangle$ for the relative phase of $\pi/2$.

even when the particles would not interact classically. Second, non-Hermitian mode damping practically cannot cool the particle motion to the ground state. Third, free-space optical binding can never lead to entanglement between two cotrapped particles. We will discuss the empirical implications of these findings and present a concrete proposal to observe the quantum signatures of optical binding.

The quantum version of the optical-binding interaction (1) takes the form of a Markovian quantum master equation, which can be derived in five steps: (i) we determine the relation between the incoming light field $\mathbf{E}$ and the polarization field induced inside the two particles. This requires taking into account that the light scattered off one particle contributes to the polarization field inside the other particle. (ii) We calculate the optical forces acting on the two particles and use them to derive the classical Lagrangian describing the combined dynamics of the two particles and the scattered fields. (iii) A mechanical gauge transformation renders the canonical momentum field independent of the particle degrees of freedom. (iv) One derives and canonically quantizes the classical Hamiltonian of the combined particle motion plus scattered fields via a Legendre transformation. (v) The scattered fields are traced out in the weak-coupling Born-Markov approximation. This procedure yields the Lindblad master equation for the two-particle state ρ,

$$\partial_t \rho = -\frac{i}{\hbar}[H + V + V_{\mathrm{opt}}, \rho] + \int d^2\mathbf{n} \sum_s \left(L_{\mathbf{n}s} \rho L_{\mathbf{n}s}^\dagger - \frac{1}{2}\{L_{\mathbf{n}s}^\dagger L_{\mathbf{n}s}, \rho\} \right). \quad (2)$$

Here, H denotes the laser-independent Hamiltonian of the two particles, which may include additional trapping and manipulation fields, and V is the laser-induced time-averaged dipole potential,

$$V = -\frac{\alpha}{4} \sum_{j=1}^{2} |\mathbf{E}(\mathbf{r}_j)|^2. \quad (3)$$

The optical-binding interaction enters at two locations: the Lamb shift gives rise to the conservative optical binding potential

$$V_{\mathrm{opt}} = -\frac{\alpha^2}{4\varepsilon_0} \sum_{\substack{j,j'=1 \\ j \neq j'}}^{2} \mathbf{E}^*(\mathbf{r}_{j'}) \cdot \mathrm{Re}[\mathrm{G}(\mathbf{r}_j - \mathbf{r}_{j'})] \mathbf{E}(\mathbf{r}_j). \quad (4)$$

It is determined by the real part of the Green tensor and can be interpreted as the interaction potential of two induced dipoles. This conservative interaction is always accompanied by the nonconservative optical-binding interaction through the Lindblad operators

$$L_{\mathbf{n}s} = \sqrt{\frac{k^3}{2\varepsilon_0 \hbar}} \frac{\alpha}{4\pi} \sum_{j=1}^{2} \mathbf{t}_{\mathbf{n}s}^* \cdot \mathbf{E}(\mathbf{r}_j) e^{-ik\mathbf{n}\cdot\mathbf{r}_j}. \quad (5)$$

They are the coherent sum of the single-particle photon scattering amplitudes into direction $\mathbf{n}$ with orthogonal polarization directions $\mathbf{t}_{\mathbf{n}s}$, $s = 1, 2$ [27], reminiscent of superradiance [28–33]. The Lindblad dissipator describes nonconservative radiation pressure forces, decoherence, and the nonconservative optical-binding interaction [27]. Together, the conservative and the nonconservative optical-binding contributions in Eq. (2) recover the full optical-binding interaction (1), i.e., $\partial_t \langle \mathbf{p}_1 \rangle = -\langle \partial V / \partial \mathbf{r}_1 \rangle + \langle \mathbf{F}_{12} \rangle$. More details on the derivation and its extension to an arbitrary number of particles of arbitrary size, shape, and optical susceptibility can be found in Ref. [34].

For a physical intuition of quantum optical binding, we consider the case that both particles are deeply trapped in two separate optical tweezers at distance $d \gg 1/k$ (far-field approximation) and with relative optical phase $\varphi = \varphi_1 - \varphi_2$ (see Fig. 2). The motion along the tweezer axes can then be separated from the transverse motion because of a significant mismatch in mechanical frequencies. This simplifies the collective dynamics to two harmonic oscillators with dimensionless quadratures $z_j = \sqrt{m\omega}\mathbf{r}_j \cdot \mathbf{e}_z/\sqrt{\hbar}$ and $p_j = \sqrt{m}\dot{\mathbf{r}}_j \cdot \mathbf{e}_z/\sqrt{\hbar\omega}$, where m is the particle mass and ω is the mean mechanical frequency so that $[z_j, p_{j'}] = i\delta_{jj'}$. Optical binding leads to coupling of the oscillators as characterized by the *reciprocal* and the *antireciprocal* coupling rates

$$g_{\mathrm{r}} = \frac{G}{kd} \cos(kd) \cos\varphi, \quad (6a)$$

$$g_{\mathrm{a}} = \frac{G}{kd} \sin(kd) \sin\varphi. \quad (6b)$$

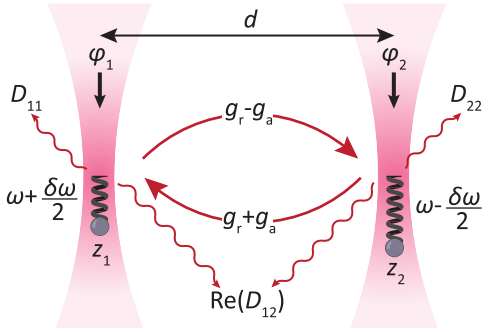

FIG. 2. Two nanoparticles are harmonically trapped with frequencies $\omega \pm \delta\omega/2$ close to the foci of two optical tweezers driven by the same laser with phases φ_1 and φ_2. The light scattered off the particles (i) couples their motion nonreciprocally with coupling rates $g_r + g_a$ and $g_r - g_a$, and (ii) imprints photon shot noise, leading to recoil heating with diffusion constants D_{11} and D_{22}. The photon shot noise is correlated, as described by $\mathrm{Re}(D_{12})$.

They decay as $1/kd$, as expected from far-field dipole radiation, and they oscillate both with the relative tweezer phase φ and with the phase kd as dictated by interference between tweezer and scattered light. The constant G is determined by the tweezer power, tweezer polarization, and particle polarizability [15,16,35]. The two constants (6) enter the linearized master equation together with interaction-independent oscillator dynamics H_0,

$$\partial_t \rho = -\frac{i}{\hbar}[H_0 + \delta H_0, \rho] + 2ig_r[z_1 z_2, \rho] + \sum_{j,j'=1}^{2} 2D_{jj'}\left[z_j \rho z_{j'} - \frac{1}{2}\{z_j z_{j'}, \rho\}\right]. \tag{7}$$

Here, the second term in the first line accounts for the reciprocal interaction between the two particles. While the diagonal elements of the diffusion rate matrix $D_{11} \simeq D_{22}$ account for local shot noise heating, the off-diagonal element

$$D_{12} = \frac{G}{kd}\sin(kd)\cos\varphi + ig_a = D_{21}^{*} \tag{8}$$

gives rise to shot-noise correlations (real part) and antireciprocal coupling (imaginary part). Note that $D_{11}D_{22} > |D_{12}|^2$ so that the time evolution of ρ is completely positive. The interaction also leads to a modification of the local trapping stiffness as described by

$$\frac{\delta H_0}{\hbar} = (g_r + g_a)z_1^2 + (g_r - g_a)z_2^2. \tag{9}$$

Figure 3(a) demonstrates the tunability of optical binding, ranging from predominantly reciprocal coupling (blue), via unidirectional (green) and antireciprocal coupling (orange), to a regime dominated by shot-noise correlations (violet). The points indicate where the coupling becomes purely reciprocal [$g_a = D_{12} = 0$ at (I)], maximally unidirectional [$g_a = g_r$ at (II)], purely antireciprocal [$g_r = \mathrm{Re}(D_{12}) = 0$ at (III)], and where the recoil correlations are maximal [$g_r = g_a = 0$ at (IV)]. In the following, we will discuss three implications of this master equation for upcoming quantum optomechanical experiments with nanoparticles.

First, the master equation (7) predicts that the particles are subject to correlated shot noise as described by the real part of the diffusion rate (8). This implies that the dynamics of the two particles becomes correlated even if both

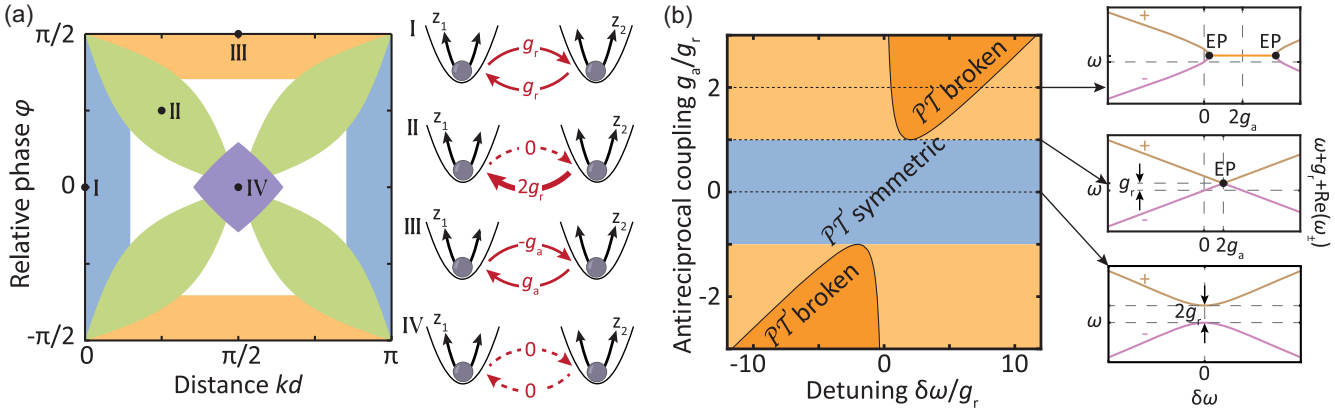

FIG. 3. (a) Different regimes of quantum optical binding between two deeply trapped particles as a function of the relative tweezer phase φ and the tweezer distance kd. The blue region indicates predominant reciprocal coupling $|g_r| > |g_a|$ and $|g_r| > |\mathrm{Re}(D_{12})|$; green indicates predominant directional coupling $|g_r + g_a| > 2|g_r - g_a|$ or $|g_r - g_a| > 2|g_r + g_a|$; orange indicates predominant antireciprocal coupling $|g_a| > |g_r|$ and $|g_a| > |\mathrm{Re}(D_{12})|$; violet indicates predominant recoil-noise correlations $|\mathrm{Re}(D_{12})| > 2\max[|g_r + g_a|, |g_r - g_a|]$. The coupling corresponding to the points (I)–(IV) are depicted on the right. (b) Phases of broken and unbroken generalized $\mathcal{PT}$ symmetry as a function of the mechanical detuning $\delta\omega$ and the antireciprocal coupling g_a. In the blue (orange) area the reciprocal coupling is greater (smaller) than the antireciprocal coupling, $|g_r| > |g_a|$ ($|g_a| > |g_r|$). $\mathcal{PT}$ symmetry is broken in the dark orange region, where the mode frequencies $\mathrm{Re}(\omega_\pm)$ are degenerate. The panels on the right show how the eigenfrequencies vary with mechanical detuning for different g_a as indicated by the dotted lines. In the $\mathcal{PT}$-symmetry broken phase, two exceptional points (EPs) occur.

coupling rates (6) vanish. This can be seen from the quantum Langevin equations for the mode operators in the corotating frame $a_j = (z_j + ip_j)e^{i(\omega+g_r)t}/\sqrt{2}$,

$$\frac{d}{dt}\begin{pmatrix} a_1 \\ a_2 \end{pmatrix} = -iH_{\mathrm{NH}}\begin{pmatrix} a_1 \\ a_2 \end{pmatrix} + \begin{pmatrix} \xi_1 \\ \xi_2 \end{pmatrix}, \tag{10}$$

where the dynamical matrix is

$$H_{\mathrm{NH}} = \begin{pmatrix} -\frac{\delta\omega}{2} + g_a & -g_r - g_a \\ -g_r + g_a & \frac{\delta\omega}{2} - g_a \end{pmatrix}, \tag{11}$$

with the mechanical detuning $\delta\omega$, which is controlled by the power difference of the tweezer traps, and we performed a rotating-wave approximation. The white-noise operators are correlated $\langle \xi_{j'}^{\dagger}(t')\xi_j(t)\rangle = 2D_{jj'}\delta(t-t')$, implying that the particle motion becomes correlated even when $g_r \simeq 0$ and $g_a \simeq 0$, so that the dynamical matrix mediates no coupling. Intuitively, whether the two particles oscillate in phase (common mode) or in antiphase (differential mode) controls if their scattered fields interfere constructively or destructively, thus increasing or decreasing the recoil heating rates. As a consequence, the two normal modes thermalize to different temperatures in the presence of external damping. For the particles from [15], the resulting difference between the normal modes' recoil heating rates is between 10% and 15%. It could be observed by means of reheating measurements [37] if the noise on the particle is dominated by recoil heating, which occurs at pressures below 10^{-7} mbar in state-of-the-art experiments. It is a direct consequence of the correlation between the quantum noise forces (and thus cannot be obtained by naively adding single-particle recoil heating to the classical equations of motion).

Second, non-Hermitian damping of the quasinormal modes [right eigenvectors of (11)] cannot prepare the particle motion in the ground state. For $g_a \neq 0$, the dynamical matrix (11) is non-Hermitian with eigenfrequencies

$$\omega_{\pm} = \pm\frac{1}{2}\sqrt{\delta\omega^2 + 4g_r^2 - 4g_a\delta\omega}. \tag{12}$$

For $|g_a| < |g_r|$ the normal mode frequencies are real and the dynamical matrix (11) remains in the unbroken phase of a generalized $\mathcal{PT}$ symmetry [38]. However, the system can enter a symmetry-broken regime if $|g_a| > |g_r|$ when passing through an exceptional point at $\delta\omega = 2g_a \pm 2\sqrt{g_a^2 - g_r^2}$. Then, the eigenfrequencies turn into a complex conjugated pair, so that one mode gives rise to an exponential growth in time, while the other leads to an exponential decay. The exponential suppression of mechanical motion is most pronounced for $g_r = 0$ and $\delta\omega = 2g_a$. In this case, the Langevin equation (10) describes the uncoupled dynamics of the two collective modes $(a_1 \pm ia_2)/\sqrt{2}$ of the two oscillators at fixed relative phases $\pm\pi/2$. In the absence of quantum noise, the mode $(a_1 - ia_2)/\sqrt{2}$ would decay exponentially, while $(a_1 + ia_2)/\sqrt{2}$ would increase exponentially. The unavoidable presence of quantum noise due to photon scattering acts as a finite temperature bath, forcing the decaying mode to saturate at the effective occupation $D_{11}kd/2G - 1/2 \geq 0$. In practice, the recoil rate clearly exceeds the coupling rate, $D_{11} \gg G/kd$, so that the stationary state of the decaying mode is far away from the ground state. Observing this saturation of the damped quasinormal mode despite the absence of external heating sources, such as gas collisions or stray fields, presents a signature of quantum optical binding that will become relevant in near-future experiments. The phase diagram Fig. 3(b) demonstrates the occurrence of exceptional points and the time reversal symmetry-broken phase, as observed experimentally in Refs. [16,17].

Third, free-space optical binding cannot mediate entanglement. One could be tempted to expect that optical binding can induce entanglement between the two particles, given that changing the relative laser phase with twice the mechanical frequency, $\varphi = 2\omega t$, transforms the coherent optical-binding interaction in the corotating frame into the two-mode squeezing Hamiltonian

$$H_r(t) \simeq \frac{\hbar G}{2kd}\cos(kd)[a_-^2 + (a_-^{\dagger})^2]. \tag{13}$$

Here, $a_- = (a_2 - a_1)/\sqrt{2}$. However, even if the antireciprocal coupling constant is tuned to zero, $g_a = 0$, the optical-binding interaction can always be written as a local operations and classical communication (LOCC) channel by formulating a stochastic feedback quantum master equation, which models optical binding via a feed-forward loop of independent and local homodyne measurements and which yields Eq. (7) in the ensemble average [35]. If such a LOCC description is possible the interaction cannot mediate entanglement, in agreement with findings for unidirectional quantum transport [39,40]. In free space, the vast majority of scattered photons contributes to decoherence while only a tiny fraction mediates the coherent optical binding interaction. However, if the effective recoil heating rate can be made smaller than the coupling rate, then optical binding would generate entanglement. Likewise, Eq. (13) cannot squeeze below the ground-state width if the coupling rate is smaller than the recoil heating rate [35].

The conditions that preclude entanglement generation can be circumvented in several ways: (i) recoil-scattering decoherence can be reduced by homodyning the position of both particles via detection of the backscattered light. The thus obtained measurement record may be used to determine the conditional quantum state of the system, which evolves according the optical binding master equation (2) together with a stochastic measurement

superoperator given in the Supplemental Material [35]. If the fields are tuned to purely conservative optical-binding interaction, the effective recoil heating rate of the conditional state $D'_{11} = D_{11}(1-\eta_{\mathrm{det}})$ is reduced according to the total detection efficiency η_{det}. By achieving sufficiently large detection efficiencies, one might thus reduce the recoil heating rate below the conservative coupling rate g_{r} and thereby enable the generation of entanglement via optical binding. (ii) The local recoil rates in Eq. (10) can be reduced by squeezing the electromagnetic vacuum state in the quadratures commuting with the local noise [41]. In practice, this requires squeezing a single electromagnetic free-space mode for each particle, ideally with a large spatial overlap ζ with the scattered fields of the respective particles. The effective recoil heating rates are given by $D'_{11} = D_{11}[1-|\zeta|^2(1-e^{-r})]$, where r is the squeezing parameter. (iii) Finally, one may enhance the conservative optical-binding interaction by placing the particles in an optical cavity, realizing coherent scattering of both particles into the same cavity mode [18,42]. Since the cavity output can be detected with high efficiency, such a setup may be utilized for entanglement via single-photon detection and postselection [42]. Note that, for the particles from [15], roughly one out of ten photons contributes to optical binding, while the remaining light scattering leads to decoherence. In the future it might therefore be necessary to combine two or more of the mentioned techniques to enable entanglement via optical binding.

In summary, we presented the quantum theory of optical binding between two polarizable point particles and used it to discuss important implications for near-future experiments with levitated nanoparticles. The possibility for tunable two-mode squashing may well become relevant for measuring small interaction forces between the particles, e.g., due to gravity. We expect that the ability to continuously tune the interaction from fully reciprocal to fully nonreciprocal will render nanoparticle arrays an ideal platform for exploring and exploiting non-Hermitian quantum physics in near-future setups.

Acknowledgments—H. R., K. H., and B. A. S. acknowledge funding by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation)–439339706. B. A. S. acknowledges funding by the DFG–510794108 as well as by the Carl-Zeiss-Foundation through the project QPhoton. U. D. acknowledges support from the Austrian Science Fund (FWF, Project DOI 10.55776/I5111).

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