

Quantum persistent tennis racket dynamics of nanorotors

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Classical rotations of asymmetric rigid bodies are unstable around the axis of intermediate moment of inertia, causing a flipping of rotor orientation. This effect, known as the tennis racket effect, quickly averages to zero in classical ensembles since the flipping period varies significantly upon approaching the separatrix. Here, we explore the quantum rotations of rapidly spinning thermal asymmetric nanorotors and show that classically forbidden tunnelling gives rise to persistent tennis racket dynamics, in stark contrast to the classical expectation. We characterise this effect, demonstrating that quantum coherent flipping dynamics can persist even in the regime where millions of angular momentum states are occupied. This persistent flipping offers a promising route for observing and exploiting quantum effects in rotational degrees of freedom for molecules and nanoparticles.

Quantum control of nanomechanical motion is a challenging task with great potential for future quantum technologies and fundamental tests of physics [1]. Optically levitating nanoparticles in ultrahigh vacuum achieves an unrivalled degree of environmental isolation [2], rendering these systems ideally suited for high-precision sensing [3–5] and for high-mass tests of quantum physics [6–9]. In addition, freely suspending nanoscale dielectrics introduces rigid-body rotations as novel nanomechanical degrees of freedom.

The non-linearity and anharmonicity of nanoparticle rotations induces quantum interference effects which have no analogues in their free centre-of-mass motion. The rotations of levitated objects can be controlled and exploited by using aspherical or anisotropic objects rather than isotropic spheres [10, 11]. Amongst the most recent experimental achievements are frequency-locking of nanorods [5], GHz rotations of anisotropic spheres [12] and nanodumbbells [13, 14], and rotational precession of dielectrics [15]. Together with the breakthroughs of centre-of-mass ground state cooling of spherical particles [16] and rotational cooling of aspherical particles [17, 18], these developments pave the way for cooling the rotational state into the quantum regime [19, 20] and can be used for probing quantum physics [9, 21, 22] with nano- to microscale rigid rotors.

The instability of classical rigid body rotations around the axis of intermediate moment of inertia results in an exponential growth of initial deviations from perfect mid-axis revolutions [23]. The resulting periodic flipping of the orientation of asymmetric rigid bodies, which was first studied by Poinsot in the 19th century [24], is now known as the tennis racket (or Dzhanibekov) effect. Analogies to this classical flipping effect have been exploited in several fields, for instance, qubit control [25], non-equilibrium quantum many-body simulation [26] and Josephson junction dynamics [27]. The frequency of the tennis racket flips crucially depends on the initial rotation

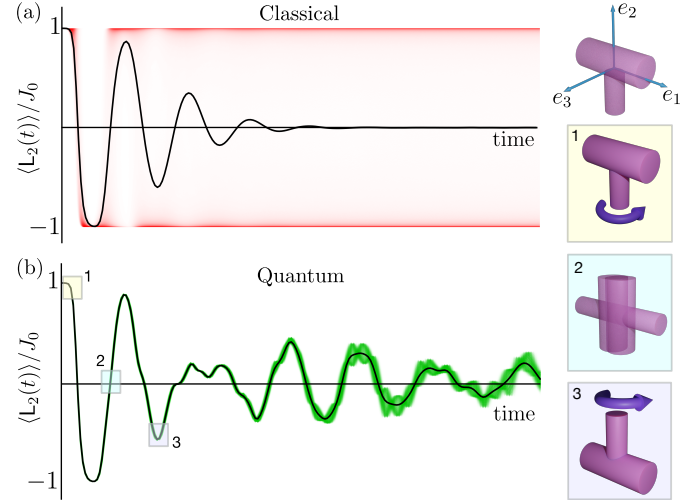


FIG. 1. An asymmetric rigid rotor initially rotating with angular momentum L_2 around its mid-axis e_2 , exhibits periodic flipping of its orientation, known as the tennis racket effect. The flipping of its orientation is manifested by a periodic change of sign of the mid-axis angular momentum, $\langle L_2(t) \rangle / J_0$. The red and green shaded regions show the probability density of mid-axis angular momentum values. (a) Gaussian decay of mix-axis flipping classically. (b) Persistent mid-axis flipping quantum mechanically. The insets (1-3) show the quantum mid-axis orientation at the times highlighted in (b).

state, implying that they quickly vanish when averaged over repetitions of the experiment (or in an ensemble of particles), see Fig. 1(a). However, in this paper we show that the mid-axis rotations become persistent in the quantum regime, see Fig. 1(b).

We argue that quantum-persistent tennis racket flips are realistically observable with levitated nanoparticles at moderate motional temperatures. Such an experiment would probe quantum rotations in an unprecedented mass and complexity regime. Furthermore, we argue that the quantum tennis racket flipping can be understood by a tunnelling-induced splitting of degenerate eigenenergies close to the classical phase space instability. We discuss the quantum and classical rotor dynamics

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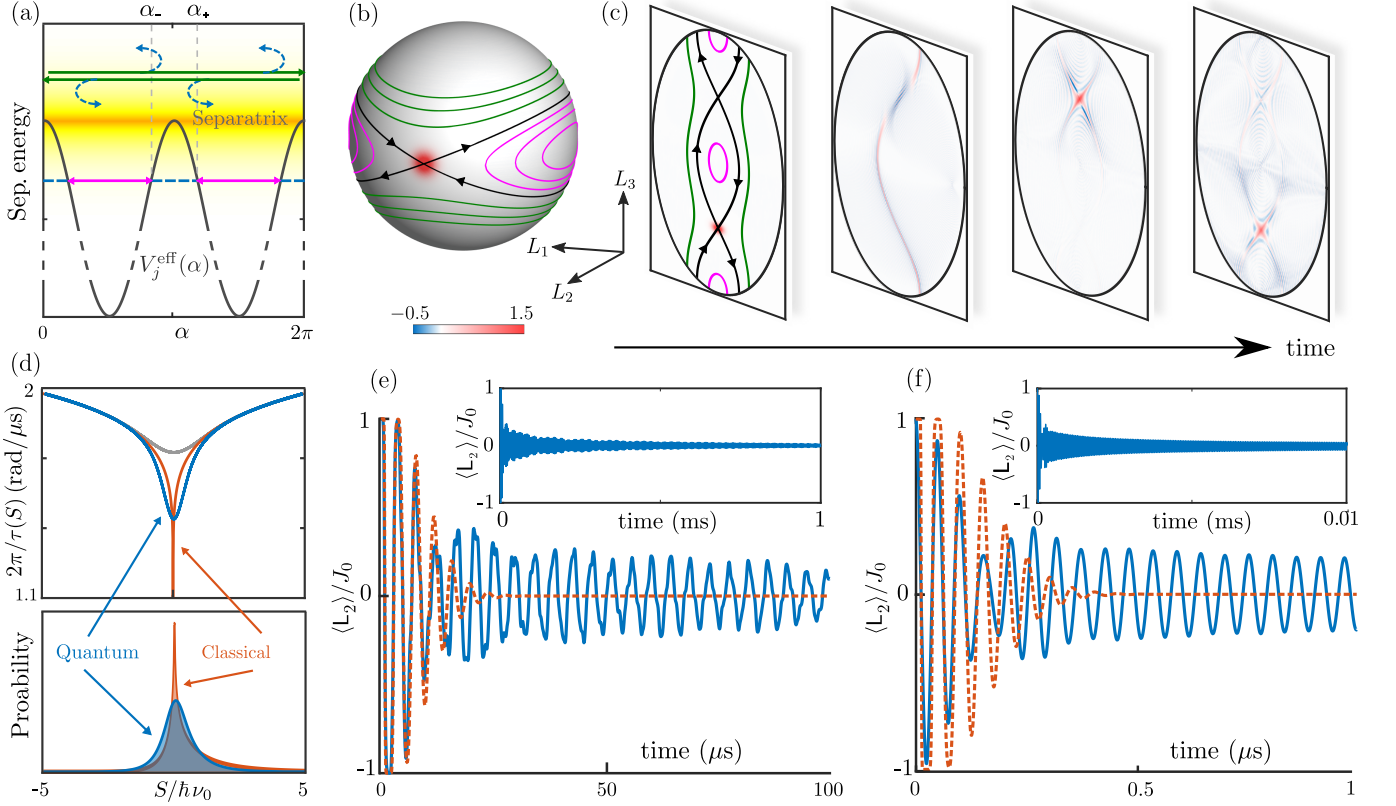


FIG. 2. (a) The unstable mid-axis dynamics of an asymmetric top is physically similar to that of a planar rotor in a periodic potential (see the main text for details). The color density shows the probability of different separatrix energies around $S = 0$ (Eq. (4)), with dashed arrows indicating quantum reflections above the potential barrier. (b) Angular momentum sphere shows the separatrix (black line) between classical rotational (green lines) and oscillatory (pink lines) trajectories for fixed $j = 100$. (c) Time evolution of the Wigner function shows periodic dispersion and partial interference (negative value in blue). (d) Upper (Lower) panel: Quantum and classical frequencies (relative probabilities of energy population) close to the separatrix. (e-f): The quantum mid-axis dynamics (blue lines) persist orders of magnitude longer than the corresponding classical dynamics (orange dashed line). (e) Calculated exactly for $J_0/\hbar = 10^6$ by full numerical diagonalisation. (f) Calculated from the approximation (6) for $J_0/\hbar = 10^8$, where a full numerical treatment is impossible.

with analytical and numerical methods, demonstrating that quantum persistent flipping can be observable with GHz nanorotors at milliKelvin temperatures.

Classical dynamics – The rotation dynamics of an asymmetric rigid rotor with moments of inertia $I_1 > I_2 > I_3$ is integrable, conserving the total angular momentum $L^2 = L_1^2 + L_2^2 + L_3^2$ and the energy $H = A_1 L_1^2 + A_2 L_2^2 + A_3 L_3^2$, where $A_i = 1/2I_i$ are the rotation constants ($A_3 > A_2 > A_1$). The body-fixed angular momentum trajectories, $L_i(t)$, follow from Euler's equations of motion [23, 28, 29]. If the initial rotation is closely aligned to the mid axis, $L_2 \gg L_1 \simeq L_3$, the rotation dynamics become unstable, resulting in flipping between the initial angular momentum L_2 and the opposite rotor orientation $-L_2$, [30]. In this limit, the body-fixed mid angular momentum simplifies to $L_2(t) \simeq L_2 \text{cd}(\nu t, q)$, where $\text{cd}(\cdot)$ is the Jacobi CD elliptic function, with $\nu = 2\sqrt{(A_2 - A_1)(A_3 - A_2)}L$ and $q \simeq 1 - 4(A_3 - A_1)|S|/\nu^2$ the elliptic modulus. Here the separatrix energy $S = H - A_2 L^2$ quantifies how close the

dynamics is to the rotor phase space instability at $S = 0$, which defines the separatrix.

The period of the flipping follows from the properties of the Jacobi functions,

$$\tau_{\text{cl}}(S) \simeq \frac{2}{\nu} \ln \left[\frac{4\nu^2}{(A_3 - A_1)|S|} \right], \quad (1)$$

and diverges logarithmically upon approaching the separatrix. This divergence implies that in an ensemble of classical rotation states, each trajectory flips with a vastly different frequency even though their initial angular momenta might be very close. Thus the ensemble-averaged tennis racket flips are expected to quickly decay.

Thermally averaged trajectory – As the practically most relevant scenario, we consider that the particle is initially in a displaced thermal state of mean mid-axis angular momentum J_0 and temperature T , where $A_2 J_0^2 / k_B T \gg 1$. A lengthy and non-trivial calculation for rotational energies clearly exceeding the thermal width of the state yields that the ensemble-averaged rotor dynam-

ics exhibit a Gaussian decay in time [31],

$$\langle L_2(t) \rangle \simeq J_0 \sum_{r=\pm} C_r \sum_{n=0}^{\infty} \frac{(-1)^n}{2n+1} \cos \left[(2n+1) \frac{2\pi t}{\tau_r} \right] \times \exp \left[-(2n+1)^2 \kappa_r^2 t^2 \right]. \quad (2a)$$

where summing over $r = \pm$ covers the contributions for positive and negative separatrix energies. The thermal period and decay rates on both sides of the separatrix are given by

$$\tau_{\pm} \approx \frac{2}{\nu_0} \ln \left(\frac{16}{3} \frac{A_2 J_0^2}{k_B T} \delta_{\pm} \right) + \frac{2\gamma_{\text{EM}}}{\nu_0}, \quad (2b)$$

$$\kappa_{\pm} = \frac{2\pi^2}{\sqrt{3}\nu_0\tau_{\pm}^2} \quad (2c)$$

where $\nu_0 = 2\sqrt{(A_2 - A_1)(A_3 - A_2)}J_0$ is the characteristic frequency and $\gamma_{\text{EM}} \simeq 0.58 \dots$ is the Euler-Mascheroni constant. In addition, we defined the constant

$$C_{+} = \frac{4}{\pi} \left[1 + \frac{3A_3(A_2 - A_1) + A_2(A_3 - A_1)}{3A_1(A_3 - A_2) + A_2(A_3 - A_1)} \right]^{-1} \quad (2d)$$

and the geometry-dependent factor $\delta_{+} = 1 + 3A_3(A_2 - A_1)/A_2(A_3 - A_1)$ for the trajectories above the separatrix. The values below the separatrix, C_{-} and δ_{-} , are obtained by exchanging A_3 and A_1 . The thermally averaged trajectory (2) ensures that $\langle L_2(0) \rangle = J_0$ and provides an accurate approximation to the exact dynamics in a wide range of asymmetric rigid rotor geometries, $10^{-2} < (A_3 - A_2)/(A_2 - A_1) < 10^2$.

The tennis racket dynamics (2) are experimentally observable if the rotor is initially aligned, so that the conserved total angular momentum vector $\mathbf{J}$ always points in approximately the same space-fixed direction in each experimental run. Then the mid-axis angular momentum $\langle L_2(t) \rangle = \langle \mathbf{J} \cdot \mathbf{n}_2(t) \rangle$ is revealed by measuring the mid-axis orientation $\mathbf{n}_2(t)$. On both sides of the separatrix the tennis racket flips decay on the timescale $1/\kappa_{\pm} \sim \ln^2(A_2 J_0^2/k_B T)/\nu_0$ and can thus only be observed if the mean rotational energy exponentially exceeds the rotational temperature. Surprisingly, this is not necessary due to the onset of quantum persistent tennis racket flipping.

Quantum tennis racket tunnelling – The quantum rigid body rotations of an asymmetric top are induced by the free Hamiltonian $H = A_1 L_1^2 + A_2 L_2^2 + A_3 L_3^2$, with the body-fixed angular momentum components L_k , satisfying $[L_k, L_l] = -i\hbar \varepsilon_{klm} L_m$, and ε_{klm} the Levi-Civita symbol. This Hamiltonian commutes with the total angular momentum operator $L^2 = L_1^2 + L_2^2 + L_3^2$ (with eigenvalues $\hbar^2 j(j+1)$), and thus also with the separatrix operator $S = H - A_2 L^2$.

Each separatrix eigenstate $|S_{jn}\rangle$ (with $n = -j, -j+1, \dots, j$), falls into one of four orthogonal subspaces, characterised by the rotational symmetry of the state [32]. The resulting eigenvalues S_{jn} are twofold degenerate for

eigenstates close to the axes L_1 or L_3 , because classical rotations around these axes are stable. However, the degeneracy is lifted close to the classically unstable mid-axis by quantum tunnelling between counter-rotating trajectories of equal energies [33]. We will show that these tunnelling contributions are necessary for persistent quantum tennis racket flipping.

In order to quantify the role of tunnelling for the quantum mid-axis dynamics, it is helpful to transform the Hamiltonian to that of a linear rotor with effective moment of inertia I_{eff} in an effective potential. The Hamiltonian for the asymmetric rotor can be written in action angle variables, $H = (L^2 - L_3^2)[A_1 \cos^2 \varphi + A_2 \sin^2 \varphi] + A_3 L_3^2$ [28], with φ the conjugate coordinate to L_3 . The canonical point transformation via $\alpha = \pi F(\varphi, \Delta)/2K(\Delta)$, with $\Delta = (A_2 - A_1)/(A_3 - A_1)$, $K(\cdot)$ the complete elliptic integral of the first kind, and $F(\cdot)$ the incomplete elliptic integral of the first kind, shows that the dynamics close to the separatrix are that of a quantum planar rotor in a potential, $S_j \simeq \mathbf{p}_{\alpha}^2/2I_{\text{eff}} + V_j^{\text{eff}}(\alpha)$, for $\alpha \in [0, 2\pi)$. Here, $\mathbf{p}_{\alpha}$ denotes the angular momentum operator of the planar rotor (for fixed j), $I_{\text{eff}} = 2\Delta K^2(\Delta)/(A_2 - A_1)\pi^2$, and the j -dependent effective potential is

$$V_j^{\text{eff}}(\alpha) = -\hbar^2 j^2 (A_2 - A_1) \text{cn}^2 \left[\frac{2}{\pi} K(\Delta) \alpha, \Delta \right], \quad (3)$$

with $\text{cn}(\cdot)$ the Jacobi elliptic cosine.

While a classical rigid body exhibits rotational trajectories for energies above the effective potential ($S_j > 0$) and oscillatory motion for energies below the effective potential ($S_j < 0$), a quantum rotor can also be reflected above the potential barrier and tunnel through it [Fig. 2(a) and (b)]. This lifts the degeneracy of energy eigenstates close to the separatrix. The strength of the tunnelling contribution at a fixed energy S is quantified by the transmission amplitude of a single through-barrier tunnelling or above-barrier reflection event,

$$P_j(S) = \exp \left[-\frac{\sqrt{2I_{\text{eff}}}}{\hbar} \int_{\alpha_{-}}^{\alpha_{+}} d\alpha \sqrt{V_j^{\text{eff}}(\alpha) + |S|} \right], \quad (4)$$

where $\alpha_{\pm}$ are the classical turning points (evaluated for $-|S|$). The tunnelling probability approaches unity close to the separatrix $S = 0$, while quickly decaying for non-zero energies [see Fig. 2(a)].

Quantum persistent flipping – For fair comparison with the classical treatment, we consider the dynamics of a displaced thermal state at temperature T initially rotating around its mid-axis with mean angular momentum J_0 ,

$$\rho_0 = \frac{1}{Z} \exp \left(-\frac{H + V_{\text{ext}}}{k_B T} + \frac{2A_2 J_0}{k_B T} L_2 \right). \quad (5)$$

Here V_{ext} is an external potential ensuring that the body-fixed mid-axis points in a well-defined space-fixed direction, and Z is the partition function. The external potential V_{ext} , which is only present initially, serves to align

the body-fixed mid-axis with a well-defined space fixed direction so that the tennis-racket dynamics can be observed by measuring the rotor orientation (see above). Here Z is the partition function. Fig. 2(b) plots the angular momentum sphere for fixed j showing the separatrix (black line) between classical rotational (green lines) and oscillatory (pink lines) trajectories. The Wigner function [34] of a displaced, low temperature thermal state is plotted on the sphere, taking $j = 100$, with its time evolution – showing periodic diffusion and recombination following the separatrix – shown in Fig. 2(c).

Tunnelling and above-barrier reflection become relevant for the rotor dynamics if the initial state (5) has strong support in the region of significant tunnelling probability (4). Using the quarter mean energy, corresponding to approximately one quarter of all states, a straightforward calculation yields the requirement $P_{J_0}(k_B T/4) \simeq \exp(-\pi k_B T/4\hbar\nu_0) \gtrsim 1\%$, or equivalently $\hbar\nu_0/k_B T \gtrsim 0.1$. This requirement depends only on the rotor aspect ratio and angular frequency, and is thus *independent* of its physical size or mass. This is in marked contrast to the logarithmic criterion for observing classical tennis racket flipping discussed above, which is much more restrictive for nanoscopic rotors. We will demonstrate next that strong tunnelling coincides with the occurrence of quantum persistent tennis racket flips.

Describing nanoparticle rotations close to the separatrix for realistic parameters is numerically challenging as semiclassical methods fail *in the deep tunnelling regime* [31, 33], even though macroscopically many states are occupied. Exactly computing the expectation value $\langle L_2(t) \rangle$ requires independently diagonalising the separatrix operator of size $(2j+1)^2$ for each thermally occupied j -value. For nano-scale rotors at GHz rotation frequencies, $J_0/\hbar$ is on the order of tens of millions up to even billions, and since the thermal width includes macroscopically many different j values, this quickly becomes numerically intractable. However, it suffices to determine the eigenvalues for the central $j = J_0/\hbar$ and interpolate to find the possible quantum frequencies $2\pi/\tau_{\text{qu}}(S)$ as a function of S [31].

Close to the separatrix, where tunnelling becomes relevant, the possible quantum frequencies $2\pi/\tau_{\text{qu}}(S) \simeq (S_{j_n} - S_{j_n-2})/\hbar$ split into two branches [blue and gray lines, Fig. 2(d)], from which only the lower (blue) is relevant (at low temperature/fast rotation rates) due to the symmetry of the initial state [31, 33]. Importantly, $\tau_{\text{qu}}(S)$ does not diverge at the separatrix, in contrast to the classical case (orange), *because the tunnelling splitting lifts the degeneracy of the energy levels* [31]. The resulting quantum mid-axis angular momentum dynamics can be approximated by

$$\langle L_2(t) \rangle \simeq J_0 \int dS \lambda(S) \cos \left[\frac{2\pi t}{\tau_{\text{qu}}(S)} \right], \quad (6)$$

where the separatrix weight $\lambda(S)$ is well approximated by the classical probability distribution for the separatrix

energy S [31],

$$\lambda(S) \propto \exp \left[\frac{S}{2} \left(\frac{A_1}{(A_2 - A_1)\chi_1^2} - \frac{A_3}{(A_3 - A_2)\chi_3^2} \right) \right] \times K_0 \left[\frac{|S|}{2} \left(\frac{A_1}{(A_2 - A_1)\chi_1^2} + \frac{A_3}{(A_3 - A_2)\chi_3^2} \right) \right] \quad (7)$$

with $\chi_{1/3}^2 = k_B T + \hbar J_0 A_{1/3}$, which includes contributions from both the thermal width and quantum uncertainty, and $K_0(\cdot)$ is the zeroth order modified Bessel function of the second kind.

Fig. 2(e) shows the exact quantum simulation of the tennis racket dynamics for $J_0/\hbar = 10^6$, while (f) shows the approximate quantum tennis racket flipping (6) for $J_0/\hbar = 10^8$, at the temperature $(A_3 - A_1)\hbar J_0/k_B T = 1.25$, where we used the values for moments of inertia $I_1 = 4.4 \times 10^3 \text{ amu } \mu\text{m}^2$, $I_2 = 3.5 \times 10^3 \text{ amu } \mu\text{m}^2$, and $I_3 = 1.7 \times 10^3 \text{ amu } \mu\text{m}^2$, close to state of the art experiments [11–13, 15]. Both plots clearly demonstrate the persistence of quantum tennis racket flips in the tunnelling regime. The simulation for such a high value of J_0 in Fig. 2 is only possible by using the classical approximations for the overlaps Eq. (7), and a continuous approximation of the quantum frequency as a function of separatrix energy for the central J_0 value [31]. The lack of high frequency components in (f) is because (6) only accounts for the lowest energy differences, i.e. only includes low frequency components.

Discussion – Mid-axis rotations of quantum asymmetric rotors exhibit strong non-classical signatures, which can be observed with cutting-edge technology pushing nanoscale rotations towards a quantum regime [5, 12, 13, 15, 35]. For a given rotational temperature, observation of the quantum dynamics requires spinning the particle with characteristic frequency ν_0 so that $\hbar\nu_0/k_B T \gtrsim 0.1$. This relation is independent of the rotor mass but only a function of its aspect ratio. For instance, observing quantum effects at GHz rotation rates requires moderate cooling to a rotational temperature of 100 mK. While this is the same temperature required to see quantum effects in a GHz linear oscillator, we note the rotation rate is given by the initial state, unlike the geometry-dependent frequency of a linear oscillator. Furthermore, for nanomechanical oscillators the resulting amplitude is femtometers, while the nanorotor superposition state extends over the extent of the particle.

State-of-the-art optical techniques enable the preparation of GHz mechanical rotations with nanoscale dielectrics [12, 13], and beyond THz for linear molecules [35]. Rotational cooling is within reach either by cavity or feedback techniques [19, 20], which have been successfully implemented for cooling the centre-of-mass motion of dielectrics [16, 36]. Three-dimensional orientation of the nanoparticles and molecules can in principle be achieved by exploiting a permanent electric or magnetic dipole moment, which aligns with an external static field [37]. Angularly accelerating trapped and aligned particles to high rotation speeds has been implemented by both the

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radiation torque exerted by a circularly polarised laser beam [11–13], and by adiabatic alignment in the field of an optical centrifuge [35, 38].

Once released, the rotor starts dispersing and recurring as shown in Fig. 2. The rotor orientation can be measured from light scattering of a weak optical probe pulse [31]. Orientational decoherence due to scattering of gas atoms [39] requires an experiment with nanorotors to be performed in high vacuum, so that on average less than one collision occurs during the tennis racket flips. **The orientation of nanoscale particles can be optically measured with high precision via light scattering as demonstrated experimentally e.g. [5, 10–15, 31].**

In conclusion, we provided a full theoretical treatment

of the classical and quantum tennis racket effect. The observation of persistent tennis racket flips will probe the quantum nature of rotations in an unparalleled regime of macroscopic moments of inertia, where the objects rotate with billions of angular momentum quanta [5, 11–13, 15].

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