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### Giant Diffusion of Nanomechanical Rotors in a Tilted Washboard Potential

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We present an experimental realization of a biased optical periodic potential in the low friction limit. The noise-induced bistability between locked (torsional) and running (spinning) states in the rotational motion of a nanodumbbell is driven by an elliptically polarized light beam tilting the angular potential. By varying the gas pressure around the point of maximum intermittency, the rotational effective diffusion coefficient increases by more than 3 orders of magnitude over free-space diffusion. These experimental results are in agreement with a simple two-state model that is derived from the Langevin equation through using timescale separation. Our work provides a new experimental platform to study the weak thermal noise limit for diffusion in this system.

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Thermal diffusion in a tilted periodic (also named washboard) potential constitutes an archetypal model of transport phenomena in nonequilibrium statistical physics [1,2]. It describes a diverse range of systems, including Josephson junctions [3], molecular motors [4], synchronization phenomena [5], and diffusion on crystal surfaces [6], to name only a few.

Brownian particles moving in the tilted potential at nonzero temperature exhibit two distinct well-characterized limiting behaviors: locked and running states. The first emerges when the potential wells are deeper than the thermal energy scale, the diffusing particle remains in a local minimum. In the latter, a sufficient tilt of the potential, lowering its energy barrier, allows the particle to flow down the potential wells. Depending on the system parameters (friction, temperature, and tilt), both solutions may coexist, and stochastic transitions between locked and running states may occur. It has been shown, both experimentally [4,7,8] and theoretically [9,10], that in this two-state coexistence regime, the effective diffusion coefficient can be enhanced by several orders of magnitude relative to the free diffusion coefficient. However, this has only been experimentally observed in the overdamped regime which is by now a fully understood stochastic process [2].

To our knowledge, an experimental study of giant diffusion in the strongly inertial (underdamped) regime is still missing. The theoretical problem is also more demanding in the underdamped and weak noise limits because the usual expansion of the matrix continued fraction [2] fails to converge well [11]. On these grounds, this nonlinear stochastic system in the low-noise and low-damping limits is still attractive [11].

We propose a simple experimental setup based on the optical trapping of a nanodumbbell in a moderate vacuum. A special feature of our experiment is that the friction coefficient, being linearly proportional to the gas pressure P, can be tuned over several orders of magnitude. Therefore, for a nearly constant temperature, Brownian motion can move from the overdamped to the underdamped regime. This makes the optical trapping setup a suitable platform to study stochastic dynamics in the low-friction regime [12–14].

The main focus of this Letter is the analysis of the rotational diffusivity in the bistability region for an underdamped stochastic mechanical nanorotor traveling in a tilted periodic potential. We provide experimental confirmation of a giant diffusivity by tracking the rotational motion of a single silica nanodimer. Because of the experimentally achieved timescale separation, we show that the effective diffusion is well described by a two-state model for this nearly one-dimensional rotational motion.

The experimental setup consists of a vacuum optical tweezer trapping a dielectric silica dumbbell made of two nanospheres of nominal radius  $68 \pm 7$  nm [15,16]. The continuous wave trapping laser beam (wavelength,  $\lambda =$ 1064 nm and power, 180 mW) passes through an objective lens with a 0.8 numerical aperture, the polarization's ellipticity of which can be changed by a quarter-wave plate. A detailed description of the experimental setup, calibration, and processing procedures is given in the Supplemental Material [17] including Refs. [18–32]. The shape and the size of the dimer are assessed under linear polarization, and correspond to an aspect ratio L/D = 1.8, with two spheres of radius  $R_p = 65.7$  nm, where  $D = 2R_p$ and L is the major axis length of the dimer. Our detection system is able to simultaneously record the particle's center of mass motion and its angular displacement.

The nanodimer experiences both a trapping force and a torque, both of which are induced by elliptically polarized light. In particular, when such an asymmetric Rayleigh scatterer lies in the transverse  $(\hat{x}, \hat{y})$  plane, corresponding to  $\theta = \pi/2$ , the torque reads

$$M = M_C - M_L \sin(2\varphi), \tag{1}$$

where  $\varphi$  is the angle between the dimer main axis and the lab's horizontal  $\hat{x}$  axis. Its time evolution is related to the dimer rotation around the  $\hat{z}$  optical axis.  $M_C$  and  $M_L$  are respectively the *circular* and *linear* contributions of the optical torque which depend on the dimer polarizability tensor and the laser beam polarization [17].

In a linearly polarized optical tweezer, where  $M_C = 0$ , the long axis of the dimer (i.e., in the direction of greatest polarizability) will tend to align with the laser beam polarization. This is because the polarizability of the dimer along its long axis is greater than the polarizability perpendicular to it. As a result, the dimer acts as a torsion balance with a linear restoring torque,  $-2M_L\varphi$ . The libration pulsation  $\Omega_L$  is well approximated in the low friction limit  $(\Gamma_{\varphi} \ll \Omega_L)$  by  $\sqrt{2M_L/I_z}$ , where  $I_{\perp}$  is the moment of inertia. In contrast, for a circularly polarized trap, where  $M_L = 0$ , the asymmetric particle undergoes a constant torque, whatever its orientation  $\varphi$ . As a result of the continuous transfer of angular momentum, the particle will spin about the  $\hat{z}$  axis at constant frequency, as given by  $\Omega_R = M_C / I_\perp \Gamma_{\varphi}$ , where  $\Gamma_{\varphi}$  is the rotation or torsional vibration damping rate about the  $\hat{z}$  optical axis (see Ref. [17] for its determination). Note that the rotation frequency can exceed GHz, which allows us to study material stresses due to centrifugal forces [33,34], measure ultraweak torques [35,36], and examine quantum features of rotation [37,38].

Figure 1(a) illustrates the potential landscape and orientational dynamics for different polarization states. Considering the general case, for elliptical polarization, the potential has a washboard shape:  $U(\varphi) = M_L \sin^2(\varphi) - M_C \varphi$ , and one can observe running and locked states. Below, we only consider elliptical polarization which is held constant at  $\phi_{\lambda/4} = 25^\circ$ , the angle of the quarterwave plate.

The dynamics of an asymmetric Brownian particle trapped under elliptically polarized light is six dimensional with translational and rotational degrees of freedom coupled to each other. This generally leads to a highly nonlinear problem [36,39,40]. However, in the following, we show that the giant diffusion phenomenon is well described by one-dimensional rotational dynamics. Comparison between the experiment and numerical simulations is made quantitative provided that an additional term is added to the usual *extinction* optical torque ( $\propto \Re[\mathbf{p} \times \mathbf{E}_{ini}]$ , with **p** the dipole moment and  $\mathbf{E}_{inj}$  the incident electric field). This extra term, called *scattering* torque ( $\propto \Im[\mathbf{p} \times \mathbf{p}^*]$ ), is due to the interference of the fields scattered by the particle [17,41]. This  $\varphi$ angle dynamics reproducing the stochastic jumps between torsion and continuous rotation states is described by the Langevin equation





FIG. 1. (a) Illustration of the energy landscape for the rotation of the Brownian dimer around the optical axis for three different tilts. When the polarization is linear (blue dotted line), the potential is not tilted and the orientation of the dimer remains in the locked state. Conversely, for a circular polarization (solid blue line) the potential wells disappear. For elliptical polarization (dashed line), the rotational motion can change from a running state (spinning) to a locked state (torsional) and vice versa. (b) Surface plot in log-log scales of the angular vibration and translational power spectral densities as a function of the gas pressure for a beam ellipticity of  $\phi_{\lambda/4} = 25^{\circ}$  and an optical power of 180 mW. The solid and dashed curves (red) represent the local maxima of the PSD computed by Langevin simulations of Eq. (2). The three grey horizontal curves at 0.41 mbar, 0.51 mbar, and 0.61 mbar outline the two-state coexistence region where the middle line denotes the maximum of intermittency.

where  $\Gamma_{\varphi}$  is the rotational damping rate and  $M_{\rm th} = \sqrt{2k_BTI_{\perp}\Gamma_{\varphi}}\zeta(t)$  is the thermal torque with  $\zeta(t)$  a Gaussian noise of zero mean value. *M* is given by Eq. (1).

Figure 1(b) shows the position and orientation power spectral densities as a function of pressure, along with the numerical Langevin simulations (solid red curves). For consistency, we also show the frequencies associated with translational motions in the X, Y, and Z directions that are independent of gas pressure. The observed nondegeneracy of the transverse frequencies X, Y is expected for a noncircularly polarized laser beam. In the high friction limit, the torsional dynamics dominate the dimer orientation motion and their resonance frequencies remain constant in relation to pressure. Conversely, the rotational

dynamics is dominant in the low friction regime, i.e., at low pressure, and its angular frequency,  $\Omega_R$ , rises as the inverse of the gas damping.

The red solid curves in Fig. 1(b) show a good agreement between the experiment and one-dimensional simulations of Eq. (2). One can also clearly see that the bistability region is quantitatively well described by the model. The two locked states observed in the spectral densities are also found in the numerical simulations, and correspond to the fundamental peak and its overtone. Although not within the scope of this Letter, we do not preclude contributions with other angles leading to coupling with extra motional degrees of freedom. The experimental determination of the dimer size and shape by the calibration procedure [17] is used for calculating the moments of inertia and polarizabilities. The latter are further supported numerically using COMSOL MULTIPHYSICS. We find that  $I_{\perp} = 2 \times 10^{-32} \text{ kg m}^2$ ,  $M_C$  and  $M_L$  are, respectively, of the order of  $2 \times 10^{-22}$  N m and  $6 \times 10^{-20}$  N m, in good agreement with both experimental and numerical results.

As mentioned above, in the calculation of optical torques the contribution of *scattering* is included in addition to the usual *extinction* contribution [41]. Since the polarization of the trapping beam is not linear, this scattering torque is crucial to reproduce quantitatively the experimental results, particularly when the dimer spins. In many previous works, the scattering torque was ignored because the rotational Langevin equation was only qualitatively discussed. It is worth noting that if the scattering torque is not taken into account, the particle's rotation speed is overestimated by typically an order of magnitude for the actual dimer [17] and much more for a spheroid (data not shown). Some quantitative deviations between theory and experiment appear at low pressure but without greatly affecting the giant diffusion effect.

We now consider the quantitative study of this stochastic nonequilibrium phenomenon. Figure 2 shows the time traces of the angular velocity  $\dot{\phi}$  obtained by short time Fourier transforms in the bistability region. In the locked state, the mean angular velocity is  $\langle \dot{\phi} \rangle_L \equiv 0$ . While the mean rotational speed,  $\langle \dot{\phi} \rangle_R$ , in the running state increases continuously with the pressure drop. The most noticeable feature of the time traces is the two-state noise-induced transitions, where the maximum of intermittency occurs in the middle of the bistability region at about  $P \approx 0.56$  mbar [Fig. 1(b)].

We use the time traces of angular velocities to determine the mean occupation times in the locked and running states. Figure 3(a) shows the distributions  $P(\tau/\langle \tau \rangle)$  of the mean occupation times, recorded for different gas pressures. These distributions decrease exponentially as expected for a Kramers-like problem. Deeper insight into two-state coexistence can be gained if we plot the pressure dependence of the average occupancy times in the locked and running states, as shown in Fig. 3(b), both for experimental



FIG. 2. Time traces of the dimer angular velocity  $\dot{\phi}$  obtained by short-time Fourier transform for the three pressures that are shown as gray horizontal curves in Fig. 1(b).

and numerical data. It can be evidenced that when  $\Gamma_{\varphi} \ll$  $\Omega_L$  (as in this case), the transition rate  $r_L$  out of a locked state (regardless of the final state, locked or running) is independent of the friction coefficient, i.e., of the pressure. This is in agreement with our observations [see Fig. 3(b) red squares and solid line]. Besides, this value is well approximated by the well-known Arrhenius law,  $1/\tau_L =$  $r_L \sim \Omega_L \exp(-E_b/k_B T)$ ,  $E_b$  being the barrier height given by the potential difference between a minimum and the lower neighboring maximum [42]. This offers insight into how a temperature change affects the stability of the torque state. A different behavior is observed for transitions from a running to a locked state for low damping and finite temperatures. This behavior is explained by the fact that the rate  $r_R$  is such that  $1/\tau_R = r_R \sim \Gamma_{\varphi} \propto P$  [43]. Note that some differences occurring at low pressure can be observed



FIG. 3. (a) Probability distribution function of the occupation time from the locked to running states  $P(\tau_L)$ , red symbols, and the running to locked states  $P(\tau_R)$ , blue symbols, for various pressures where the bistability is observed. Exponential behaviors suggest that stochastic transitions between locked and running states are thermally activated, following an Arrhenius-type law. (b) Pressure dependence of the mean occupation times in the locked and running states [symbols: experiment, solid line: Langevin simulations, Eq. (2)].

between the one-dimensional model and the experiment [as seen in Fig. 3(b)]. They are not due to a time windowing effect, but could arise from the coupling between other rotational degrees of freedom, as seen below 0.45 mbar with the appearance of an additional resonance in the spectral densities [Fig. 1(b)].

In the rest of this Letter, we focus on the measurement of the effective rotational diffusion coefficient, which is defined by the Kubo relation

$$D_{\rm eff} = \int_0^\infty dt' \langle [\dot{\varphi}(t') - \langle \dot{\varphi} \rangle] [\dot{\varphi}(0) - \langle \dot{\varphi} \rangle] \rangle.$$

This coefficient satisfies the usual Einstein relation  $\lim_{t\to\infty} \langle \varphi^2(t) \rangle - \langle \varphi(t) \rangle^2 = 2D_{\text{eff}}t$ , where  $D_{\text{eff}}$  can in principle be enhanced by orders of magnitude over the free coefficient diffusion  $D_0$  at the crossover from locked to running states. Interestingly, outside the region of bistability, the power spectral density of  $\dot{\varphi}$  allows for a measurement of both  $D_0 = k_B T / \Gamma_{\varphi} I_{\perp}$  and its related timescale  $1/\Gamma_{\varphi}$ . Surprisingly, we can derive a simple two-state stochastic noise model to fit our experimental results in the bistability region (see details in Sec. V.B of the Supplemental Material [17]). Since the parameter range we consider allows large timescale separations  $(1/\Omega_R \ll 1/\Gamma_{\varphi} \ll \tau_L, \tau_R)$ , the spectral density of the rotation speed in the region of bistability reads

$$S_{\dot{\varphi}\,\dot{\varphi}}(\omega) = \frac{D_0}{1 + \frac{\omega^2}{\Gamma_{\varphi}^2}} + \frac{D_{\text{eff}}}{1 + \frac{\omega^2}{\Gamma_r^2}} \tag{3}$$

where  $\Gamma_T = r_R + r_L$ , is the total escape rate, corresponding to the longest timescale of the system. From the timetraces of the angular velocity  $\dot{\varphi}$  (Fig. 2), we compute the power spectral densities of the rotation velocity (defined as the Fourier transform of the rotational velocity autocorrelation function) in the coexistence regime, as shown in Fig. 4(a). Using then Eq. (3), we can determine the effective diffusion  $D_{\text{eff}}$  relative to  $D_0$ , that is displayed in Fig. 4(b) as a function of pressure. Note that  $\Gamma_{\varphi}$  and  $\Gamma_T$  are also measured, allowing us to corroborate the values of the nanodimer size and its aspect ratio. To go further into the two-state model, we use the transition rates  $r_R$  and  $r_L$ deduced from Fig. 3 to calculate  $D_{\text{eff}}$ . In our parameter regime, the two-state model gives an expression that bears much similarities with those developed in [11,44–46]

$$D_{\rm eff} = \langle \dot{\varphi} \rangle_R^2 \frac{r_L r_R}{(r_L + r_R)^3},\tag{4}$$

where  $\langle \dot{\varphi} \rangle_R$  is the average angular velocity taken only over the running states of the whole temporal trace. Comparison of the filled and open circles in Fig. 4(b) indicates that when Eq. (4) is used with the results obtained from occupancy times, good agreement is obtained with the diffusion



FIG. 4. (a) Measured power spectral densities of the dimer angular velocity (dots) and double Lorentzian fit [Eq. (3), solid curves] at the boundaries of the bistable region (P = 0.653 mbar in blue, P = 0.408 mbar in black), and at the pressure around which the maximum of giant diffusion exists (P = 0.560 mbar in red). The dashed line represents the free-space diffusion coefficient  $D_0$  at P = 0.653 mbar, the double arrow displays the excess of diffusion  $D_{\rm eff}/D_0$ , while both the total escape rate  $\Gamma_T$ and the rotational damping rate  $\Gamma_{\varphi}$  are marked by vertical arrows. (b) On the left axis. Pressure dependence of the effective diffusion coefficient  $D_{\text{eff}}$  in units of  $D_0$  estimated by two methods: (i) data from Fig. 4(a) are fitted by using Eq. (3), blue open circles, while (ii) transition rates obtained from Fig. 3 are used to calculate  $D_{\text{eff}}$ from Eq. (4), green filled circles. The black solid line results from the Langevin simulations [Eq. (2)]. On the right axis. Proportion of the mean occupancy in the locked state (red squares: experiment and black dashed line: Langevin simulations).

estimate by spectral densities by using Eq. (3). The occupancy rate in the locked state defined as  $N_{-} = \langle \tau_L \rangle / (\langle \tau_L \rangle + \langle \tau_R \rangle)$  is represented by red squares in Fig. 4(b). We observe that the giant increase in the effective diffusion coefficient is roughly maximal at a pressure where the states are evenly distributed.

In conclusion, we have observed in a very small pressure range a giant increase of the diffusion coefficient in the underdamped and weak noise limit. Carefully calculating the optical torque, a reasonable quantitative agreement has been obtained using a one-dimensional Langevin model. This supports evidence that even though the real system dynamics has six dimensions, it can be reduced to a onedimensional system within at least some parameter regimes. Cooling the translational (and possibly rotational) degrees of freedom would mitigate the rotranslational coupling [15,16], enabling ideally over a wide parameter regime to reach the true one-dimensional Brownian nanorotor as described in Eq. (2). Thus, an optically levitated stator rotator constitutes an outstanding platform to study the issue of jump length distributions [47] and where its dynamics can be controlled down to the quantum ground state [48–50].

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### Giant diffusion of nanomechanical rotors in a tilted washboard potential

PACS numbers:

We describe in detail the experimental setup and how we synthesize the silica nano-dumbbells. We also present the configuration to follow in real time their orientation and the position of their center of mass. In particular, we explain the link between the orientation of the particle and the measured signal. Next, we present the data processing to obtain the angular velocity of the dumbell and how we extract its effective diffusivity. We discuss how to evaluate the size and shape of the dimer using the translational degrees of freedom.

A theoretical part presents some results on the dynamics. First, we calculate the torque for an anisotropic Rayleigh particle, taking into account not only the extinction torque, but also the recoil (or scattering) contribution [I], in order to obtain a good quantitative agreement with the experiments. Finally, we derive the analytical expression for the low frequency behavior observed in the power spectral density (PSD) of the rotational speed. From the Langevin's equation, we get a general expression of the rotational effective diffusion coefficient, and explain why, with our parameters, the latter is identical to the effective diffusion coefficient given by a two-state telegraph noise model [2]-[4].

#### I. EXPERIMENTAL SETUP

A simplified scheme of the experimental setup is shown in Fig. [] An axisymmetric particle (*i.e.*, a dumbbell composed of two silica nanospheres of diameter  $140 \pm 14$  nm,  $\rho = 2200$  kg.m<sup>-3</sup> and refractive index n = 1.45) is trapped in an elliptically polarized laser beam at a wavelength of  $\lambda = 1064$  nm. To do so, the linear polarization at the laser source output is oriented along the x axis by a half waveplate (HWP) plate. The polarization ellipticity is then set by a quater waveplate (QWP), whose angle between the fast axis and x axis in the laboratory frame is  $\phi_{\lambda/4}$ .



FIG. 1: Simplified top view scheme of the experimental apparatus. The 1064 nm laser light is focused in the vacuum chamber through a high numerical aperture microcscope objective (N.A.= 0.8) and is then divided into two parts: one is sent on the detection system to measure the motion of the particle, the other part is analysed by a polarizer before to reach a balanced photodiode allowing a measurement of the orientation of the particle.

The laser source is an ultra stable and low noise continuous wave laser (AzurLight System) providing up to 7 W of optical power at a wavelength of  $\lambda = 1064$  nm. Its relative intensity noise is of -150 dBc/Hz in the frequency range of interest (10 kHz to 100 MHz). The polarization stability (< 0.1%) and the pointing fluctuations (< 0.5  $\mu$ rad for several hours) are minimized by using a polarizer with a high polarization extinction ratio and including the whole experiment in a box to prevent air fluctuations on the optical path.

The trapping beam is focused in the vacuum chamber by a high numerical aperture objective (Nikon IR Plan-APO : 0.8, W.D : 1mm) and recollected by an aspheric lens of numerical aperture 0.8. The transmitted beam from the trap is splitted in two: one half goes on the center of mass (COM) displacement detection setup, the other half goes on the setup for the detection of the particule orientation.

The COM detection is made by tracking in real time the extinction cross section of the nanoparticle, given by the

spatial integration of the interference pattern between the scattered field and the incident trapping field. For this part, we have used the same setup as in previous work, see  $\boxed{7}$ , and figure 2.



FIG. 2: Detailed top view of the scheme of the COM setup (BS, beam splitter, CM, D-cut mirror and PD, balanced photodiode).

The particle's center of mass displacement and its major axis orientation are recorded simultaneously at sampling rates varying between 10 MS/s to 200 MS/s, what permits to acquire temporal traces from 5 s to 100 s. The change of the gas pressure in the vacuum chamber modifies the damping of the particle's motion.

The nanoparticles (Microparticles GMBH) are loaded inside the vacuum chamber at atmospheric pressure by using a nebulizer (Omron Micro-Air), filled with a suspension of ethanol containing nanospheres. Nanodumbbells are formed by a proper dilution so that in average two nanospheres are contained in a single droplet. Once a particle is trapped, we evacuate the vacuum chamber from other diffusing nanoparticles.

### **II. MEASURED QUANTITIES OF THE ROTATIONAL DEGREES OF FREEDOM**

#### A. Measurement of the dimer orientation and anisotropic polarizability

The light field arriving before the balanced photodiode is described as being the superposition of the field scattered by the particle and the reference field (Fig. 1). A simple analysis makes it possible to relate the differential intensity measured by the detectors to the orientation of the nanoparticle. The trapping beam is defined by

$$\mathbf{E}_{inj}(\mathbf{r}) = E_0(\mathbf{r}) \mathrm{e}^{i\eta(\mathbf{r})} \begin{pmatrix} \cos(\phi_{\lambda/4}) \\ i\sin(\phi_{\lambda/4}) \\ 0 \end{pmatrix}, \tag{1}$$

where  $E_0(\mathbf{r})$  and  $\eta(\mathbf{r})$  are the Gaussian beam amplitude and phase [9, 10],  $\delta = 2\phi_{\lambda/4}$  is the beam ellipticity.

The orientation of the dimer is given by its director axis  $\hat{n} = [\cos \varphi \sin \theta; \sin \varphi \sin \theta; \cos \theta]$ , expressed in the laboratory frame  $(\mathbf{e_x}, \mathbf{e_y}, \mathbf{e_z})$ , with usual polar and azimuthal angles.

The polarizability matrix,  $\underline{\alpha}$ , is diagonal in the dimer's eigenframe, and has two components  $\alpha_{\perp}$ , twice degenerated, and  $\alpha_{\parallel}$ . Their value can be estimated analytically [11-13] or computed numerically. This analysis leads to the real and imaginary parts of the polarizability:  $\alpha'_{\perp,\parallel}$  and  $\alpha''_{\perp,\parallel}$ , respectively. In the laboratory frame, the polarisability matrix and the vectorial electric dipole moment for any orientation is given by

$$\underline{\alpha} = \alpha_{\perp} \mathbb{I} + (\alpha_{\parallel} - \alpha_{\perp}) \hat{n} \hat{n}, \qquad (2)$$

$$\mathbf{p} = \underline{\underline{\alpha}} \mathbf{E}_{\mathbf{inj}},\tag{3}$$

where  $\mathbb I$  is the identity matrix and  $\hat n \hat n$  the dyadic product.

The forward scattered field,  $\mathbf{E}_s$ , in a direction  $\mathbf{r}_L$  towards the collecting lens, depends on the dipole moment of the particle, see Eq. (3) in which one uses the far-field Green tensor [5], Eq. (8.52) and Eq. (8.61)]). By evaluating the modulated intensity received by the photodiode on each polarization channel (along x or along y) as  $E_{inj_{x,y}}.E_{s_{x,y}}^{\star} + E_{inj_{x,y}}^{\star}.E_{s_{x,y}}$ , and integrating on  $\mathbf{r}_L$ , we can write the differential intensity that is measured.

Assuming the dimer is in the (x,y) plane, *i.e.*  $\theta = \pi/2$ , and the half-waveplate is adjusted so that the signal averaged over positions of the dimer is perfectly balanced, we obtain a simple expression for the differential voltage r(t), proportional to the intensity difference :

$$r(t) \propto \frac{E_0^2}{2} \Big( (\alpha_{\parallel}'' - \alpha_{\perp}'') \sin(\delta) \cos\left[2\varphi(t)\right] + (\alpha_{\parallel}' - \alpha_{\perp}') \sin\left[2\varphi(t)\right] \Big). \tag{4}$$

In practice, the measured signal is a nonlinear function of the dimer orientation. To understand the link between the signal spectrum and the characteristic frequencies of the problem, it is interesting to consider a limiting case. In the absence of noise, let us take the following ansatz for the temporal evolution of the orientation of the particle

- $\varphi(t) = \Omega_{\rm R} t$ , when the particle spins,
- $\varphi(t) = \varphi_{eq} + \varphi_0 \cos(\Omega_{\rm L} t)$ , when the particle is in the torsional state around an equilibrium position  $\varphi_{eq}$ .

Therefore, in the rotational state, the signal would peak at the frequency  $2\Omega_{\rm R}$ , while, for the torsional state, the expansion of  $\sin[2\varphi(t)]$  and  $\cos[2\varphi(t)]$  would give a signal frequency at  $\Omega_{\rm L}$ , followed by their harmonics.

In the presence of thermal noise, we observe both experimentally and numerically the presence of the peak of rotation in the spectral density at  $2\Omega_{\rm R}$ , and several peaks associated with the torsional state (both fundamental and harmonics, see fig. 1 of the main text). However, it is difficult to say if the experimental peaks are really harmonics of the 1D problem, or come from the hybridization of the  $\varphi$  motion with the other angles, or even from the coupling with the degrees of freedom in translation.

# III. TREATMENT PROCEDURES FOR LOCAL ANGULAR FREQUENCY EVALUATION AND TRANSIT TIME EXTRACTION

In this section, we present our data processing to evaluate the instantaneous angular vibration of the trapped nanodumbbell and how we extract the transit times to calculate the effective diffusion constant.



FIG. 3: a) Measured signal r(t) corresponding to the angular motion  $\varphi(t)$  for a gas pressure P = 0.56 mbar, when the particle is in the locked (torsional) state (red curve) or in the running (spinning) state (blue curve). b) Measured Power Spectral Densities obtained from the temporal trace presented in a).

Changes in the polarization of the transmitted beam are related to changes in the instantaneous orientation of the trapped particle, which are recorded by a well-balanced photodiode. This signal is formed by time traces of 5 to 100 seconds. We divide them into subtraces whose length varies between 50  $\mu$ s to 1 ms in order to evaluate the instantaneous angular velocity. For each subsample, we then calculate the one-sided power spectral density, which reads as

$$S_{rr}(\omega) = \int_0^\infty dt \Big\langle \left[ r(t) - \langle r(t) \rangle \right] \left[ r(0) - \langle r(0) \rangle \right] \Big\rangle e^{i\omega t}$$
(5)

Figure 3 shows the measured signals (left panels) and their corresponding spectra (right panels), when the particle is in the locked or rotating state, both at a pressure of 0.560 mbar. Each PSD was calculated for a time window of 100  $\mu$ s. This permits to reconstruct the time trace of angular frequency.



### IV. DAMPING IN TRANSLATION AND ROTATION AND CHARACTERIZATION OF THE SIZE AND SHAPE OF THE PARTICLE

FIG. 4: a) Measured Power Spectral Density of the translation and torsional motion of the trapped nanodumbbell at a pressure of P = 1.53 mbar averaged over a time period of 20 s. b) Time trace of the particle motion in translation along the x axis from which is extracted the slowly varying envelope (red curves). c) Energy autocorrelation for various pressures ranging from 0.1 to 0.634 mbar. The dashed lines represent a fit by an exponential decay. d) Translational damping rates of the nanodimer along the direction of its main axis ( $\perp$ , in blue) and perpendicular to it ( $\parallel$ , in red), as a function of the gas pressure. Symbols are measured values, solid lines are fitted from Eq. (8). The constant difference between the damping rates proves that the trapped object is an asymmetric particle and that, in the long-time averages, the friction of the translation degrees is not affected by the type of angular motion (torsional or rotational regime).

#### A. Determination of translational damping and particle geometry

The shape and size of the particle are determined under linear polarization of the trapping beam using different standard [6] and nonlinear [7] calibration techniques.

Figure 4 shows the experimental results on the translational degrees of freedom. In the high pressure regime, we extract the widths  $\Gamma_i$  of the PSDs by fitting the Lorenztian lineshape on the three axis, see figure 4(a). At low

5

pressures, nonlinearities smear the narrow Lorentzian shape of the PSDs [7]. To circumvent this issue, we extract both the time-varying local frequency and position from the slowly varying envelope of the position signals along an axis [Fig. [4](b)]. This allows a measurement of the time-resolved energy of the mechanical oscillator. Then, from the correlation function in energy [Fig. [4](c)] decreasing exponentially, we infer the characteristic time which corresponds to the inverse of damping rate. This amounts to consider

$$E_{i}(t) = \frac{m\Omega_{i}^{2}(t)x_{i}^{2}(t)}{2},$$
(6)

the time-resolved energy, whose correlations decreases exponentially as

$$\langle E_i(0)E_i(\tau)\rangle \propto e^{-\Gamma_i\tau}$$

Fitting the results of figure 4(c), we obtain  $\Gamma_i$ .

In practice, the main axis of the nanodimer is oriented in the direction of the linear polarization (in the low temperature limit), lying in the  $(\hat{x}, \hat{y})$  plane. This permits to infer  $\Gamma_{\parallel}$  and  $\Gamma_{\perp}$ , the damping rates corresponding to the dimer motion in the eigenframe, along its main axis, and perpendicular to it. Figure 4(d) shows their values as a function of pressure. Given the dimer geometrical cross sections, we logically recover that  $\Gamma_{\perp} > \Gamma_{\parallel}$ . From these results, we can deduce the particle size and shape.

We first evaluate the parallel and perpendicular hydrodynamic drag coefficients in the continuous regime,  $\gamma_{\perp,\parallel} = m\Gamma_{\perp,\parallel}$ , where *m* is the mass of the dumbbell, with the help of the Kirkwood-Reisemann theory [14] [15], and write

$$\mu_{\perp,\parallel} = \frac{\gamma_{\perp,\parallel}}{\gamma_S},\tag{7}$$

where,  $\mu_{\perp,\parallel}$  are the coefficient of the mobility tensor, and  $\gamma_S = 6\pi\eta R_p$ , where  $\eta$  is the fluid viscosity in the continuous limit (at room pressure) and  $R_p$  the radius of one sphere of the dumbbell. Note that we consider the two spheres to be identical.

Then, to evaluate the sphere radius and the dimer's aspect ratio L/D, L being the total length in  $\parallel$  direction, and  $D = 2R_p$ , the diameter of a sphere, we consider the behaviour of both perpendicular and parallel damping rates as a function of the pressure P. For a single sphere, the damping rate can be expressed in terms of Kundsen number  $Kn = \lambda_{\rm mfp}/R_p$ , where we introduce the mean free path of air molecules  $\lambda_{\rm mfp} = \lambda_0 P_{\rm atm}/P$ , with  $\lambda_0 = 68$  nm. For an aggregate of N spheres (here, N = 2), different effective Knudsen numbers  $Kn_{a,\parallel} = 1.23Kn$  (resp.,  $Kn_{a,\perp} = 1.39Kn$ ) is used along the parallel (resp. perpendicular) directions [16]. Finally, the damping rates are obtained as a function of the gas pressure

$$\Gamma_{\perp,\parallel}(P) = C_a^{-1}(Kn_a)\gamma_{\perp,\parallel}/m,\tag{8}$$

where we have used a slip correction factor corresponding to the modified Millikan's formula [17]

$$C_a(Kn_a) = 1 + Kn_a \left[ A + B \exp\left(-\frac{C}{Kn_a}\right) \right], \tag{9}$$

with A = 1.234, B = 0.404 and C = 0.876 the coefficients that Millikan has obtained for a single sphere in the rarefied gas regime. Asymptotically, considering a dimer such that L/D = 2, one recovers for the damping rates ratio the values 1.13 in the continuous regime  $(Kn_a \rightarrow 0)$  and 1.26 in the rarefied regime  $(Kn_a = 100)$  [19].

#### B. Determination of the rotational damping rate

Using the instantaneous rotation frequency (obtained by short-time Fourier transform methods), we calculate the spectral density of the rotational frequency and fit it using Eq. (19) that has been obtained from the Ornstein-Ulhenbeck theory, see Sec. (VB). This procedure allows to deduce the rotational damping rate  $\Gamma_{\varphi}$  at any pressure. Figure 5 shows such an example at 0.225 mbar with a rotational damping rate of  $\Gamma_{\varphi}/2\pi = 143$  Hz.

To confirm the estimated size of the beads and the aspect ratio obtained from the damping rates on the translational degrees of freedom, we plot the measured rotational damping rate as a function of the gas pressure. From the estimated size of the beads and the aspect ratio L/D obtained from the translational damping rate, we take from ref. [19] the rotational damping rate normalized by the rotational damping rate of the unit sphere ( $\Gamma_{\varphi,\text{Sphere}} = 8\pi \eta R_p^3/I_{\text{Sphere}}$ ).



FIG. 5: a) Temporal trace of the rotation frequency at 0.225 mbar. b) PSD of the rotation pulsation for the same experimental parameter as a). The extraction of the cut-off frequency allows a measurement of the rotational damping rate of the particle.



FIG. 6: Evolution of the rotational damping rate  $\Gamma_{\varphi}$  as a function of the gas pressure. The dotted line represents the theoretical damping rate on the rotational degrees of freedom.

In the case of an aggregate of N spheres, the rotational damping rate can be expressed via a correction slip factor and the rotational damping rate of a single sphere as

$$\Gamma_{\varphi,\text{Dumbbell}} = C_n \Gamma_{\varphi,\text{Sphere}}.$$
(10)

From [19], figure 4], we extract the rotational drag torque in units of the rotational drag torque of the elementary sphere for an aspect ratio L/D of a given particle using the fitted formula

$$C_n = 3.62 - 6.97 \frac{L}{D} + 4.37 \left(\frac{L}{D}\right)^2.$$
 (11)

Figure 6 shows the measured damping rates for the rotation and the good agreement with the theoretical formulas given by Eqs. (10) and (11) for an aspect ratio L/D = 1.8 and with radii of 65.7 nm.

### V. DYNAMICS OF THE DIMER ORIENTATION

# A. Expression of the optical torque and equation of the rotational motion for an axisymmetric anisotropic dielectric particle

The torque due to the field-dipole interaction is  $\blacksquare$ 

$$\vec{M} = \left[\frac{1}{2}\operatorname{Re}\left(\mathbf{p} \times \mathbf{E_{inj}}^*\right)\right] + \left[\frac{k^3}{12\pi\varepsilon_0}\operatorname{Im}\left(\mathbf{p} \times \mathbf{p}^*\right)\right],\tag{12}$$

where we take into account both the extinction and scattering contribution to the total torque – respectively corresponding to first and second term in brackets, Eq. (12).

In the particular case where the dimer is perpendicular to the beam axis,  $\theta = \pi/2$ , and  $\vec{M} = M\mathbf{e_z}$ . The optical torque is then given by

$$M = M_C - M_L \sin(2\varphi), \tag{13}$$

$$M_C = \frac{|E_0(\mathbf{r})|^2}{2} \left[ \frac{\alpha_{\parallel}'' + \alpha_{\perp}''}{2} - \frac{k^3}{6\pi\varepsilon_0} \left( \alpha_{\parallel}'' \alpha_{\perp}'' + \alpha_{\parallel}' \alpha_{\perp}' \right) \right] \sin \delta,$$
(14)

$$M_L = \frac{|E_0(\mathbf{r})|^2}{2} \left[ \frac{\alpha'_{\parallel} - \alpha'_{\perp}}{2} + \frac{k^3}{6\pi\varepsilon_0} \left( \alpha''_{\parallel} \alpha'_{\perp} - \alpha'_{\parallel} \alpha''_{\perp} \right) \right] \cos \delta.$$
(15)

In Eq. (13), the first term,  $M_C$ , does not depend on the dimer orientation, and has a maximum value for circularly polarized light. It describes the torque transfer via absorption and the finite-size effect of the scatterer. Note that  $M_C$  is strictly zero for a non-absorbing spherical Rayleigh particle, as extinction and scattering contributions cancel out [1]. It is however slightly non zero for, e.g., a non-absorbing nanodimer, or slightly ellipsoidal nanoparticle.

The second term,  $-M_L \sin(2\varphi)$ , is of maximum amplitude for linearly polarized light, and is also related to the geometric asymmetry of the dimer. Within the limit of small angles, it is reduced to a restoring torque, around the equilibrium position defined by  $M(\varphi_{eq}) = 0$ .

While the scattering torque is generally negligible for lossy Rayleigh particles that have large  $\alpha''$ , it must absolutely be taken into account here, as it induces a drop of roughly an order of magnitude for  $M_C$ . This makes it possible to obtain a numerical value of the rotation frequency at low pressure in agreement with the experiment (see figure 1(b) in the main text).

We have computed numerically the complex polarizabilities  $\alpha_{\parallel}$  and  $\alpha_{\perp}$  for the dimer of the experiment, with a Finite Element Method (FEM), Comsol Multiphysics. To tackle the convergence problems of FEM, we have first solved the Maxwell equations for a dimer made of non-overlaping spheres, for which one can use a very acurate method, based on the decomposition of electric fields on spherical harmonics. The results we obtained are in very good agreement with FEM simulations, when we use improved meshes and boundary conditions for the FEM. We typically obtain a 1 % mismatch between both methods, on the electric dipole moment  $\mathbf{p} \equiv \int_{\text{Dimer}} \varepsilon_0(\varepsilon_r - 1) \mathbf{E} dV$ .

We then used this FEM for a dimer with the same size and aspect ratio as those measured, see Sec. (IV), made of overlaping spheres. **p** has been computed, and the polarizabilities  $\alpha_{\perp,\parallel}$  were inferred from Eq. (3). Then,  $M_C$  and  $M_L$  are computed from Eq. (14, 15).

Note that we allowed slight changes in the dimer geometry (departing not more than 5% from the measured values of D and L/D), so as to obtain a value of  $M_C$  that gives the best agreement with experimental results, (fig. 1(b) in the main text). The torque  $M_C$  is indeed evaluated as the difference of two terms that have a close value, see inside the brackets of Eq. (14), and is therefore very sensitive to numerical inacuracies on  $\alpha''_{\parallel, \perp}$ .

Our approach, using Eq. (14), considerably improves the numerical evaluation of the frequency of rotation in the regime of low pressures.

Finally, the equation of motion for the degree of freedom in rotation around the optical axis is described by a Langevin equation

$$I_{\perp}\ddot{\varphi} = -\Gamma_{\varphi}I_{\perp}\dot{\varphi} + M(\varphi) + \xi(t), \tag{16}$$

where  $M(\varphi) = M_C - M_L \sin(2\varphi)$ .  $I_{\perp}$  represents the inertia moment around the optical z axis,  $\Gamma_{\varphi}$  is the rotational damping and  $\xi(t)$  is a centered Gaussian noise, such as  $\langle \xi(t_1)\xi(t_2)\rangle = 2\Gamma_{\varphi}I_{\perp}k_BT \ \delta(t_2 - t_1)$ . We used the Sivak algorithm [23] to compute the time traces.



FIG. 7: Calculated Power Spectral Densities for both quantities  $\dot{\varphi}$ , panel (a) and r(t), panel (b), at the maximum of superdiffusion peak, under elliptical polarisation ( $\phi_{\lambda/4} = 25^{\circ}$ ). Relatively good agreement with experimental results (see panel (a) and Fig. 4(a) of the main text) is obtained. The characteristic frequencies  $\Gamma_T$  and  $\Gamma_{\varphi}$  are shown by arrows on panel (b). The inset, on panel (b) displays the fundamental and harmonics of the torque signal (the three low frequency peaks) as well as the spin peak (the highest frequency bump).

### B. Low-frequency behaviour of the PSD : a superdiffusion

The goal is to calculate the correlation function of the rotaional speed in the bistable regime, to then obtain the power spectral density of the rotational speed and the effective diffusion coefficient of the rotation. We start with [2]:

$$S(t) = \left\langle \left[ \dot{\varphi}(t) - \left\langle \dot{\varphi}(t) \right\rangle \right] \left[ \dot{\varphi}(0) - \left\langle \dot{\varphi}(0) \right\rangle \right] \right\rangle \tag{17}$$

Consider first that  $M_L = 0$ , *i.e.* the trapping beam is circularly polarized. Then, one observes from Eq. (16) that  $\varphi(t) - \frac{M_C}{I_{\perp}\Gamma_{\varphi}}t$  is an Orstein-Ulhenbeck (O.U.) process. In such case, as  $\dot{\varphi}$  is simply shifted by a constant, we find the relation known for the O.U. process:

$$\left\langle \left[ \dot{\varphi}(t_1) - \left\langle \dot{\varphi}(t_1) \right\rangle \right] \left[ \dot{\varphi}(t_2) - \left\langle \dot{\varphi}(t_2) \right\rangle \right] \right\rangle = \frac{k_B T}{I_\perp} \, \mathrm{e}^{-\Gamma_{\varphi} |\mathbf{t}_2 - \mathbf{t}_1|} \tag{18}$$

Using Eq. (5), we obtain

$$S_{\dot{\varphi},\dot{\varphi}}(\omega) = \frac{1}{2} \int_{-\infty}^{\infty} S(t) e^{i\omega t} dt = \frac{k_B T}{I_\perp} \frac{\Gamma_{\varphi}}{\omega^2 + \Gamma_{\varphi}^2}$$
(19)

Eq. (19) has been used to fit the experimental data of figure 5 obtained in the spin state.

At zero frequency, we obtain the diffusion coefficient for angular frequency in a spin state, which is identical to the free space diffusion

$$D_o = \frac{k_B T}{\Gamma_{\varphi} I_{\perp}}.$$
(20)

We now consider the general case for a washboard potential.

Starting from the equation of motion, and integrating between 0 and t. One obtains

$$\dot{\varphi}(t) = \dot{\varphi}(0) \ \mathrm{e}^{-\Gamma_{\varphi} \mathrm{t}} + \int_{0}^{\mathrm{t}} \frac{1}{\mathrm{I}_{\perp}} \mathrm{M}[\varphi(\mathrm{u})] \ \mathrm{e}^{-\Gamma_{\varphi}(\mathrm{t}-\mathrm{u})} \mathrm{d}\mathrm{u} + \int_{0}^{\mathrm{t}} \frac{\xi(\mathrm{u})}{\mathrm{I}_{\perp}} \ \mathrm{e}^{-\Gamma_{\varphi}(\mathrm{t}-\mathrm{u})} \mathrm{d}\mathrm{u}.$$
(21)

As  $M_L \neq 0$ , extra time dependant contributions have to be considered to calculate S(t). For a given time t, we consider all the times  $0 < t_1 < t_2 < \cdots < t_N < t$  that correspond to a change of state (from spin to torque or from torque to spin). Integrals of the following type will now contribute to  $\langle \dot{\varphi}(t)\dot{\varphi}(0) \rangle$ :

$$\left\langle \dot{\varphi}(0) \int_{t_i}^{t_{i+1}} \frac{1}{I_\perp} M[\varphi(u)] \,\mathrm{e}^{-\Gamma_{\varphi}(\mathbf{t}-\mathbf{u})} \mathrm{d}\mathbf{u} \right\rangle,$$
(22)

in which the state of  $\varphi(u)$  is purely torquing or purely spinning. For large times,  $\varphi(u)$  and  $\dot{\varphi}(0)$  are uncorrelated, and upon averaging,  $\langle M[\varphi(u)] \rangle$  appears in the contribution to  $\langle \dot{\varphi}(t)\dot{\varphi}(0) \rangle$ . But, when the particule torques,  $\langle M[\varphi(u)] \rangle = 0$ . Finally, the torque states do not contribute to  $\langle \dot{\varphi}(t)\dot{\varphi}(0) \rangle$ .

Let's now consider only the intervals  $[t_i..t_{i+1}]$  where the particle continuously rotates. Their contribution to  $\langle \dot{\varphi}(t)\dot{\varphi}(0) \rangle$ , can be rewritten as

$$\left\langle \dot{\varphi}(t)\dot{\varphi}(0)\right\rangle = \sum_{i} I_{S_{i}} = \sum_{i} \left\langle \dot{\varphi}(0)\right\rangle \left\langle \int_{0}^{t_{i+1}-t_{i}} \frac{1}{I_{\perp}} M[\varphi(t_{i}+v)] e^{\Gamma_{\varphi} v} e^{-\Gamma_{\varphi}(t-t_{i})} dv \right\rangle,$$
(23)

where the random variable  $t_{i+1} - t_i$  has an exponential law of mean time  $\tau_R$ , independently of the starting time  $t_i$ . Taking the average on  $t_{i+1} - t_i$ , one obtains

$$I_{S_i} = \left\langle \dot{\varphi}(0) \right\rangle \int_0^\infty \frac{\mathrm{e}^{-\tau/\tau_{\mathrm{R}}}}{\tau_R} d\tau \int_0^\tau \frac{1}{I_{\perp}} \left\langle \mathrm{M}(\varphi(\mathbf{t}_i + \mathbf{v})) \right\rangle \,\mathrm{e}^{\Gamma_{\varphi} \mathbf{v}} \mathrm{e}^{-\Gamma_{\varphi}(\mathbf{t} - \mathbf{t}_i)} \mathrm{d}\mathbf{v} \tag{24}$$

Further computation based on the timescale separation between damping and rotation :  $1/\Gamma_{\varphi}$  and  $1/\Omega_{\text{Rot}}$  shows that averaging on the possible values of  $\varphi(t_i)$  cancels the modulated term in M:

$$\left\langle \mathbf{M}(\varphi(\mathbf{t}_{i} + \mathbf{v})) \right\rangle = M_{C}$$

We notice that the spin contributions at the different  $t_i$  will have the same value, except for the coefficient  $\exp(-\Gamma_{\varphi}(t-t_i))$ . Then, the only non-negligible contribution is the last one, between  $t_N$  and t, when the dynamics ends in spin state. If the particle would end in torque state, the contribution to time correlation function would not be the one from the last state (between  $t_N$  and t) but the former one, between  $t_{N-1}$  and  $t_N$ , whose contribution is weighted by an  $\exp(-\Gamma_{\varphi}\tau_L)$  coefficient, and then negligible, in our problem where  $1/\Gamma_{\varphi} << \tau_L$ . This point was discussed in ref. [4], where no damping was considered, and can now be generalized to a washboard dynamics, Eq. (16), due to the timescale separation between  $1/\Gamma_{\varphi}$  and  $\tau_L$ .

To go on, let us denote  $P_N$  the probability to end in spin state, knowing one starts in spin state. Considering only this main contribution :

$$\left\langle \dot{\varphi}(t)\dot{\varphi}(0)\right\rangle = P_N \left\langle \dot{\varphi}(0)\right\rangle \left\langle \int_0^{t-t_N} \frac{M_C}{I_\perp} e^{\Gamma_{\varphi} \mathbf{v}} e^{-\Gamma_{\varphi}(t-t_N)} d\mathbf{v} \right\rangle.$$
(25)

In the long time limit,  $t - t_N$  has a uniform law between 0 and  $\tau_R$ . We obtain immediately, as  $\tau_R >> 1/\Gamma_{\varphi}$ :

$$\left\langle \dot{\varphi}(t)\dot{\varphi}(0)\right\rangle = P_N \left\langle \dot{\varphi}(0)\right\rangle \frac{M_C}{\Gamma_{\varphi}I_{\perp}}.$$
 (26)

Therefore, the only non negligible contributions to  $\langle \dot{\varphi}(t)\dot{\varphi}(0) \rangle$  occur when the particule starts in spin state and ends in spin state.

The probability to end in spin state, knowing that we start in spin state (so as to have  $\langle \dot{\varphi}(0) \rangle \neq 0$ ) is the probability to make an even number of transitions. Borrowing from [4], Eq. (6)], it reads

$$P_N = \frac{1}{\tau_R + \tau_L} \Big[ \tau_L \mathrm{e}^{-\mathrm{t/T}} + \tau_R \Big]$$
(27)

where  $1/T = 1/\tau_R + 1/\tau_L$ , and considering that S(t) = S(-t), one obtains :

$$\left\langle \dot{\varphi}(t)\dot{\varphi}(0)\right\rangle = \frac{k_b T}{I_{\perp}} e^{-\Gamma_{\varphi}|\mathbf{t}|} + \frac{M_{\rm C}}{\Gamma_{\varphi}I_{\perp}} \left\langle \dot{\varphi}(0)\right\rangle \frac{1}{\tau_{\rm R} + \tau_{\rm L}} \Big[\tau_{\rm L} e^{-|\mathbf{t}|/\mathrm{T}} + \tau_{\rm R}\Big]$$
(28)

By ergodicity :

$$\left\langle \dot{\varphi}(0) \right\rangle = \frac{\tau_R}{\tau_R + \tau_L} \left\langle \dot{\varphi} \right\rangle_{\rm \tiny R}$$

where  $\langle \dot{\varphi} \rangle_{\rm R}$  is the spinning frequency averaged only on spining events of the temporal trace, at the pressure considered. This frequency can be measured experimentally. Besides, the coefficient  $\frac{M_C}{\Gamma_{\varphi}I_{\perp}}$  can be evaluated using the mobility  $\mu$  in the spining state [22]:

$$\left\langle \dot{\varphi} \right\rangle_{\rm R} = \Gamma_{\varphi} \mu \; \frac{M_C}{\Gamma_{\varphi} I_{\perp}}$$

So that the PSD is :

$$S_{\dot{\varphi},\dot{\varphi}}(\omega) = D_o \frac{1}{1 + \left(\frac{\omega}{\Gamma_{\varphi}}\right)^2} + D_{\text{eff}} \frac{1}{1 + \left(\frac{\omega}{\Gamma_T}\right)^2}$$
(29)

with

$$D_{\text{eff}} = \left[\Gamma_{\varphi}\mu\right] \left(\frac{M_C}{\Gamma_{\varphi}I_{\perp}}\right)^2 \frac{r_R r_L}{(r_R + r_L)^3},\tag{30}$$

where  $r_R = 1/\tau_R$  (resp.  $r_L = 1/\tau_L$ ).

The formula Eq. (29) has been used to fit the experimental data in the main text.

Computing the values in the spining state (e.g. at 0.3 mbar), in Risken notation 22, we obtain

$$\Theta/d \equiv 4k_b T/(2M_L) = 0.12$$
$$F_o/\sqrt{d} \equiv \frac{2M_C}{I_\perp \Gamma_\varphi} \sqrt{\frac{I_\perp}{2M_L}} = 7.5$$

Thus, we notice from [22], Fig. (11.30)], that the mobility in the spin state part of the temporal trace is  $\Gamma_{\varphi}\mu \sim 1$ .

Consequently, with our experimental parameters, we recover the expression given by the "two-state" theory toy model [2, 3]. The coefficient  $\frac{M_C}{\Gamma_{\varphi}I_{\perp}}$  is evaluated from the experimental data, as the slope of  $\langle \dot{\varphi} \rangle_{\rm R}$  versus  $1/\Gamma_{\varphi}$ , in the low pressure spin state.

At first sight, the fact that the washboard giant diffusion, on a particle that undergoes forcing, damping and nonlinear oscillations can be described by a toy model with 2 static states (and no dynamics) may seems surprising. Starting from the equation of motion, we have shown that this is due to the strong time scale separation between rotation  $(1/\Omega_{\text{Rot}})$ , damping  $(1/\Gamma_{\varphi})$ , and lifetime of the 2 states,  $\tau_R, \tau_L$ . It could be interesting, in further works, to study the deviations to the 2-state model, when the timescale separation blurs out.

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