Fluid viscoelasticity triggers fast transitions of a Brownian particle in a double well optical potential

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Thermally activated transitions are ubiquitous in nature, occurring in complex environments which are typically conceived as ideal viscous fluids. We report the first direct observations of a Brownian bead transiting between the wells of a bistable optical potential in a viscoelastic fluid with long relaxation time. We precisely characterize both the potential and the fluid, enabling a neat comparison between our experimental results and a theoretical model based on the generalized Langevin equation. Our findings reveal a drastic amplification of the transition rates compared to those in a Newtonian fluid, stemming from the relaxation of the fluid during the particle crossing events.

Understanding the role of fluctuations in the dynamics of nonlinear systems with multiwell energy landscapes is of paramount importance in many disciplines of both fundamental and applied science [1, 2]. For instance, it has been recognized that thermal noise is responsible for the activation of transitions in a wide variety of processes at mesoscopic scale, such as the magnetization reversals in thin films [3], molecular reactions [4], protein folding [5], adsorption of colloids at fluid-fluid interfaces [6], drug binding [7], photochemical isomerization [8], to name but a few. The transition rates in such situations are well described within the framework of Kramers escape rate theory [9], which is based on the dynamics of a Brownian particle in a metastable state, coupled to its environment through a constant drag coefficient, γ0, and thermal white noise. In particular, in the overdamped limit and in one dimension, the mean time to cross a potential barrier of height \( U \), is given by

\[
\tau_K = \frac{2\pi\gamma_0}{\sqrt{\kappa_S\kappa_W}} \exp\left(\frac{U}{k_B T}\right),
\]

where \( k_B \) is the Boltzmann constant, \( T \) is the environment temperature, and \( \kappa_W > 0 \) and \( \kappa_S < 0 \) are the local curvatures or stiffnesses of the potential well where the particle initially equilibrates, and of the barrier, respectively. Eq. (1) has been experimentally verified by directly visualizing the motion of colloidal particles in water in bistable optical potentials [10, 11]. Optical trapping experiments have quantitatively elucidated further aspects predicted by numerous noise-activated escape theories [1, 2, 12, 13], such as Maxwell-like relations [14], stochastic transitions in periodic potentials [15], Kramers turnover in the intermediate underdamped regime [16, 17], escape-rate optimization by energy-landscape shaping [18], and very recently, the accurate characterization of the transition path dynamics [19].

An important issue that arises when measuring barrier-crossing rates in multidimensional systems, e.g., conformational changes of biomolecules [5, 22, 23], is the emergence of memory due to a coarse-grained description of their dynamics [20, 21]. Such non-Markovian effects were pointed out in a seminal theoretical work by Grote and Hynes in 1981 in the realm of condensed phase reactions [13], where a frequency-dependent friction was introduced. It predicts an enhancement of the reaction rate with respect to Eq. (1), later explored in the context of chemical kinetics [4, 26, 30, 31]. More recently, viscoelastic fluids, widespread in many soft matter systems of biological and technological importance [32–34], have drawn the attention of many researchers, since they give rise to intriguing phenomena at the mesoscale due to their frequency-dependent flow properties [35, 36].

The timescales of Brownian particles embedded in viscoelastic fluids lack a clear-cut separation from timescales of the surroundings due to their complex microstructure. This contrasts with what happens in Newtonian viscous liquids and results in memory friction with large relaxation times. Their motion is commonly described by the generalized Langevin equation [40] with nonequilibrium transient effects that markedly manifest themselves in presence of driving forces [41, 42]. Although generalizations of Kramers rate theory to systems with long-memory friction have been developed in the past [43, 44], apart from a few numerical works [28, 29, 50, 51], their predictions remain largely unexplored in experiments.

In this work, we use optical micromanipulation [53, 54] to show the first experimental realization of thermally-activated transitions of a bead in a bistable potential, in a viscoelastic fluid. We find a significant increase in the crossing rates over the barrier separating the two local minima, as compared to those in a purely viscous environment. This is in quantitative agreement with a theoretical description based on the generalized Langevin equation, which unveils the mechanism underlying the amplification of the barrier crossing rates under viscoelastic conditions.

In our experiments, a spherical silica bead of diameter \( d_p = 0.99 \mu \text{m} \) is trapped by a double-well optical potential in a viscoelastic fluid. Such a potential, \( U(x) \), is generated by two orthogonally polarized optical tweezer-
I. Introduction

Viscoelastic fluids are known to exhibit complex behavior, including shear thinning, shear thickening, and viscoelastic relaxation. In this work, we explore the effect of viscoelasticity on the thermally activated transitions of a colloidal bead in a double-well potential. The bead is confined by two traps, A and B, which are separated by a saddle point, S. The transitions between these wells are thermally activated, and the friction experienced by the particle is dependent on the viscoelastic properties of the fluid.

II. Experimental Setup

The experiment is performed in a sample cell containing a micellar viscoelastic fluid. The sample is imaged using high-resolution microscopy, and the particle position is tracked over time. The viscoelastic properties of the fluid are characterized using rheological measurements.

III. Results

A. Thermally Activated Transitions

The thermally activated transitions between wells A and B are studied. The mean crossing times for these transitions are calculated using a maximum likelihood estimation method. The results are compared with theoretical predictions for a Newtonian fluid.

B. Role of Viscoelasticity

The role of viscoelasticity on the thermally activated transitions is investigated. The zero-shear viscosity and relaxation time of the fluid are measured, and the effects of these properties on the mean crossing times are assessed.

IV. Conclusion

In conclusion, the role of viscoelasticity on the thermally activated transitions of a colloidal bead is explored. The results show that the viscoelastic properties of the fluid significantly affect the mean crossing times between wells A and B.

**Figure 1**: Thermally activated transitions of a colloidal bead ($d_p = 0.99 \, \mu m$), embedded in a micellar viscoelastic fluid across an optical double-well potential for Exp. I (see main text): (a) time evolution of the trajectory along $x$, (b) normalized histogram of the crossing times from A to B, and (c) from B to A, respectively. The solid red lines depict the maximum likelihood estimation using an exponential model. (d) Experimental 2D potential, and (e) 1D potential across the critical points A, S, B. The dots represent the experimental data while the solid line corresponds to the fitting to the double-Gaussian potential.

**Table 1**: The resulting values of the potential stiffness around the critical points, $k_{(A,B,S)} = \frac{\mu}{\tau_0} U(x_{A,B,S})$, and the energy barriers, $U_{(A,B)}$, for the whole set of experiments are listed in Table 1. In this table we also report the values of the experimentally measured mean-crossing times, $\tau_{exp}$. To better understand the nontrivial dependence of $k_{(A,B,S)}$ on the parameters that characterize the potential, we first compare them with the mean crossing times estimated by means of Eq. (1) for a particle in a Newtonian fluid of constant viscosity $\eta_0 = 0.042 \pm 0.0052 \, \text{Pas}$ under identical energetic conditions. The values of such estimates, presented in Table 1 as $\tau_K$, and compared with $\tau_{exp}$ in Fig. 2a, reveal a systematic reduction of the mean transition times under viscoelastic conditions, which we attribute to the nonstationary nature of the particle mobility.

In order to assess the role of the viscoelasticity of the environment in the reduction of the mean barrier-crossing times, we measure the zero-shear viscosity and relaxation time of the fluid, $\eta_0 = 0.042 \pm 0.0052 \, \text{Pas}$, and a Newtonian fluid of constant viscosity $\eta_0 = 0.042 \pm 0.0052 \, \text{Pas}$, respectively. The particle is confined within the double-well potential, it exhibits thermally-activated transitions between wells A and B through the saddle point S, as exemplified by the intermittent jumps of a typical trajectory plotted in Fig. 2a. Note that escape events from A to B must be counted separately from those taking place from B to A. Thus, the barrier-crossing time, $\tau$, is defined as the total time that the particle spends in metastable equilibrium within a given well plus the time to spontaneously jump over the barrier to finally reach the neighborhood of the contiguous energy minimum. In Figs. 2b and 2c we show the normalized histograms of $\tau$ for the transitions in Exp. I from A to B and from B to A, respectively. By means of a maximum likelihood estimation using an exponential model, we find that the distribution of $\tau$ is well described by $\rho(\tau) = [\tau_{exp}^{(ve)}]^{-1} \exp[-\tau/\tau_{exp}^{(ve)}]$, with $\tau_{exp}^{(ve)}$ the mean crossing time, as depicted by the red solid lines in Figs. 2b and 2c. This suggests that the activated jumps correspond to a Poisson process.

The experimental potential is retrieved from the particle trajectories using the equilibrium distribution $\rho(x,y) = \rho_0 \exp[-U(x,y)/(k_B T)]$. As an example, from the data of Exp. I, we obtain the 2D potential $U(x,y)$, plotted in Fig. 2d. Fig. 2e shows the 1D potential across the colinear critical points A, S, and B, $U(x) = U(x,y = 0)$, where the solid line represents the fitting to the double-Gaussian potential, $U(x) = u_1 \exp\left[-(x-\mu_1)^2/\sigma_1^2\right] + u_2 \exp\left[-(x+\mu_2)^2/\sigma_2^2\right] + u_0$, where $u_1,2 > 0$ correspond to the potentials of each individual tweezer, while $\mu_1,2$ and $\sigma_1,2$ are their respective positions and widths, respectively, and $u_0$ is a constant energy value. The values of the fitting parameters for the whole experimental data set can be found in Table S2 in Supplementary Material.

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times, we focus on the description of the particle motion subjected to the double-well potential \( U(x) \), based on the overdamped generalized Langevin equation

\[
\int_{-\infty}^{t} \Gamma(t-s) \dot{x}(s) ds = -U'(x(t)) + \zeta(t),
\]

where the term on the left-hand side represents the history-dependent friction force exerted by the fluid at time \( t \), and \( \zeta(t) \) is a Gaussian stochastic force accounting for thermal fluctuations. We assume that \( \zeta(t) \) has zero mean and its autocorrelation satisfies the fluctuation-dissipation theorem [10], i.e., \( \langle \zeta(t) \rangle = 0 \), and \( \langle \zeta(t) \zeta(s) \rangle = k_B T \Gamma(t-s) \). Moreover, the memory kernel \( \Gamma(t) \) in Eq. (2) is related to the relaxation modulus \( G(t) \) of Eq. (1) via \( \Gamma(t) = 3\pi d_p G(t) \). The specific form of \( G(t) \) leads to a frequency-dependent particle mobility given by

\[
\mu_\omega = \tilde{\Gamma}(\omega)^{-1} = \frac{\gamma_0 + \omega^2 \gamma_0^2 \gamma_\infty}{\gamma_0^2 + \omega^2 \gamma_0^2 \gamma_\infty^2} + \frac{\gamma_0 - \gamma_\infty \omega \tau_0}{\gamma_0^2 + \omega^2 \gamma_0^2 \gamma_\infty^2}.
\]

where \( \tilde{\Gamma}(\omega) \) is the Fourier transform at frequency \( \omega \) of \( \Gamma(t) \), \( \gamma_\infty = 3\pi d_p \eta_S \) and \( \gamma_0 = 3\pi d_p \eta_0 \) are friction coefficients characterizing dissipation at short and long timescales, respectively, whereas elastic effects are quantified by \( \gamma_\infty / \tau_0 \). Based on these assumptions, we solve Eq. (3) numerically using the experimental information of \( U(x) \), \( \gamma_0 \), \( \gamma_\infty \), and \( \tau_0 \). The resulting mean transition times, denoted as \( \tau_{\text{sim}}^{(\text{ve})} \), are reported in Table I. We find a good agreement between \( \tau_{\text{exp}}^{(\text{ve})} \) and their corresponding estimations \( \tau_{\text{sim}}^{(\text{ve})} \), which along with their drastic contrast with \( \tau_K \), hint at the importance of viscoelasticity on thermal activation over the barrier.

From Eq. (3), we also derive an explicit expression for the mean barrier-crossing time in the viscoelastic fluid, \( \tau_{\text{th}}^{(\text{ve})} \), using Kramers rate theory extended to Brownian motion with arbitrarily large memory [16, 67]. By computing the diffusive probability current across the potential barrier, \( j_\text{S} \), and the occupation number in a potential well, \( n_\text{w} \), we find that \( \tau_{\text{th}}^{(\text{ve})} = \frac{n_\text{w}}{j_\text{S}} \), can be expressed as

\[
\tau_{\text{th}}^{(\text{ve})} = \beta \left( \frac{\alpha}{\tau_0} \right) \tau_K,
\]

where \( \tau_K \) is the Kramers’ time with friction coefficient \( \gamma_0 \), given by Eq. (1), and

\[
\beta \left( \frac{\alpha}{\tau_0} \right) = \frac{2\alpha}{1 - \frac{2\alpha}{\tau_0} + \sqrt{\left(1 - \frac{2\alpha}{\tau_0}\right)^2 + 4\alpha \tau_0}}.
\]

is a dimensionless factor which accounts for the coupling with the environment. See Supp. Mat. for more details about the derivation. In Eq. (4), \( \alpha = \frac{\gamma_0}{\tau_0} \) is the ratio between the two friction coefficients, whereas \( \tau_S = \frac{\gamma_0}{\tau_0} \) represents the slowest viscous timescale of the particle when moving in the neighborhood of the barrier unstable point. We can notice that for \( \alpha = 1 \) or \( \tau_0 / \tau_\text{S} \to 0 \), Eq. (4) reduces to the barrier-crossing time of a particle in a Newtonian fluid with constant viscosity \( \eta_0 \), i.e., Eq. (1), for which \( \beta = 1 \). On the other hand, for \( \gamma_0 > \gamma_\infty \) and \( \tau_0 > 0 \), which are the conditions describing viscoelastic behavior, it can be checked that \( \beta < 1 \), hence \( \tau_{\text{th}}^{(\text{ve})} < \tau_K \) for all values of the local curvature \( \eta_S \), thereby quantifying the viscoelasticity-induced reduction in the transition times.

In Table I we show the theoretical estimates of the mean crossing times using Eq. (5) for the same set of parameters that characterize the potential of each experiment. We also verify that the measured values of \( \tau_{\text{exp}}^{(\text{ve})} \) and their respective theoretical predictions \( \tau_{\text{th}}^{(\text{ve})} \) are consistent. This is seen in Fig. 2(b), where the experimental mean transition times (blue dots) are plotted against their corresponding theoretical values, while the dashed line represents the ideal prediction followed by Eq. (5).

Furthermore, in Fig. 3 we plot the ratio \( \beta = \tau_{\text{exp}}^{(\text{ve})} / \tau_K \) versus the time-scale ratio \( \tau_0 / \tau_\text{S} \), thus verifying that the coupling of the particle with the viscoelastic surroundings gives rise to a reduction in the mean crossing time in quantitative agreement with Eq. (4). For comparison, the Newtonian case (\( \beta = 1 \)) is represented by the upper

| Exp. | \( \kappa_{A,B} \) (pNnm\(^{-1}\)) | \( \kappa_S \) (pNnm\(^{-1}\)) | \( U_{A,B} \) (kBT) | \( \tau_{\text{exp}}^{(\text{ve})} \) (s) | \( \tau_{\text{sim}}^{(\text{ve})} \) (s) | \( \tau_{\text{th}}^{(\text{ve})} \) (s) | \( \eta_K \) (s) |
|------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| I    | 0.47 (0.42, 0.50) | -0.42 (-0.45, -0.39) | 2.74 [2.35, 3.41] | 20.6 [18.0, 24.0] | 27.8 [11.9, 59.6] | 24.4 [16.7, 42.6] | 87.0 [60.1, 163.3] |
| II   | 0.58 (0.52, 0.63) | -0.45 (-0.49, -0.48) | 2.97 [2.39, 3.69] | 24.7 [21.6, 28.9] | 31.2 [19.4, 69.9] | 26.4 [17.9, 51.6] | 108.1 [71.7, 208.8] |
| III  | 0.67 (0.60, 0.73) | -0.56 (-0.60, -0.51) | 3.14 [2.57, 4.05] | 18.5 [16.0, 21.9] | 24.3 [12.4, 54.0] | 17.0 [11.4, 37.2] | 76.8 [49.2, 167.0] |

\( k \) and \( \tau_0 \) are experimentally characterized with stiffnesses \( \kappa_{A,B} \) at the stable points and \( \kappa_S \) at the unstable saddle point, with energy barriers \( U_{A,B} \). \( \tau_{\text{exp}}^{(\text{ve})} \) is the estimated mean crossing time in the experiments. Using the parameters characterizing the double well and the viscoelastic fluid, the mean crossing times were estimated using Brownian simulations \( \tau_{\text{sim}}^{(\text{ve})} \) and theoretical predictions \( \tau_{\text{th}}^{(\text{ve})} \). For comparison, the last column shows the Kramers predictions \( \tau_K \) with zero-shear viscosity \( \eta_0 = 0.042 \pm 0.0052 \text{ Pa s} \). Values in brackets show 68% confidence intervals of the estimated quantities.
As previously suggested, in contrast to thermally activated transitions, on the mean time that the particle takes to move from one well to the other. Our findings clearly demonstrate that the mean crossing times under viscoelastic conditions are shorter than those expected in a Newtonian fluid of similar zero-shear viscosity. This effect is quantified by a factor \( \beta \) that depends on the fluid properties and on the curvature of the potential barrier, whose values are predicted by a theoretical approach based on the generalized Langevin equation. Our analysis provides direct evidence of the origin of this phenomenon in terms of a non-homogeneous mobility which drastically increases around the energy barrier. Understanding this escaping process should impact our com-

\[
x_S \approx \frac{x_S + \delta}{\gamma_0 - 1} \ln[1 + (\delta/x_S)].
\]

Using \( \delta = 0.15 \mu m \), we find the value \( \mu_{\text{eff}} \approx 25.7 \mu m pN^{-1}s^{-1} \) for Exp. I, which is one order of magnitude greater than the zero-frequency mobility \( \mu_0 = \gamma_0^{-1} \approx 2.6 \pm 0.3 \mu m pN^{-1}s^{-1} \), and very close to the high-frequency mobility \( \mu_\infty = \gamma_\infty^{-1} \approx 26.8 \pm 0.7 \mu m pN^{-1}s^{-1} \) associated to the solvent. This is in stark contrast to the characteristic mobilities in the stable points A and B, which for Exp. I are respectively \( \mu_{\text{eff}} \approx 2.7 \mu m pN^{-1}s^{-1} \) and \( \mu_{\text{eff}} \approx 3.1 \mu m pN^{-1}s^{-1} \), i.e., closer to \( \mu_0 \). Therefore, the short-time response of the viscoelastic fluid, which gives rise to a lower friction around the unstable saddle point, is responsible for enhancing the probability of transitions across the barrier.

In summary, we have investigated the effect of viscoelastic memory friction on the transitions of a micronsized bead in a double-well optical potential, and in particular, on the mean time that the particle takes to move from one well to the other. Our findings clearly demonstrate that the mean crossing times under viscoelastic conditions are shorter than those expected in a Newtonian fluid of similar zero-shear viscosity. This effect was quantified by a factor \( \beta \) that depends on the fluid properties and on the curvature of the potential barrier, whose values are predicted by a theoretical approach based on the generalized Langevin equation. Our analysis provides direct evidence of the origin of this phenomenon in terms of a non-homogeneous mobility which drastically increases around the energy barrier. Understanding this escaping process should impact our com-

FIG. 2. (a) Experimental mean crossing time in a viscoelastic fluid vs. the corresponding one in a viscous fluid. The dotted line represents Kramers prediction for a Newtonian fluid with constant viscosity \( \eta_0 \). (b) Experimental mean crossing times vs. theoretical values predicted by means of Eq. (5). The dashed line depicts perfect agreement between \( \tau_{\text{exp}}^{(w)} \) and \( \tau_{\text{th}}^{(w)} \).

As previously suggested, in contrast to thermally activated transitions with constant mobility in a Newtonian environment, the fast transitions under viscoelastic conditions are triggered by the high-frequency components of the particle mobility. To verify this, we estimate the effective mobility, \( \mu_{\text{eff}} \), experienced by the particle along the barrier crossing events. Note that such events take place on a timescale which is comparable or shorter than \( \tau_0 \), and much shorter than the typical time spent in the metastable states A and B, as indicated by the arrow in the inset of Fig. 3. Taking into account that the potential around the unstable point is harmonic with stiffness \( \kappa_S \), the mean time to go from a position \( x_S \) to a neighboring point \( x_S + \delta \) can be approximated by \( \Delta t = (\kappa_S \mu_{\text{eff}})^{-1} \ln[1 + (\delta/x_S)] \). Using \( \delta = 0.15 \mu m \), we find the value \( \mu_{\text{eff}} \approx 25.7 \mu m pN^{-1}s^{-1} \) for Exp. I, which is one order of magnitude greater than the zero-frequency mobility \( \mu_0 = \gamma_0^{-1} \approx 2.6 \pm 0.3 \mu m pN^{-1}s^{-1} \), and very close to the high-frequency mobility \( \mu_\infty = \gamma_\infty^{-1} \approx 26.8 \pm 0.7 \mu m pN^{-1}s^{-1} \) associated to the solvent. This is in stark contrast to the characteristic mobilities in the stable points A and B, which for Exp. I are respectively \( \mu_{\text{eff}} \approx 2.7 \mu m pN^{-1}s^{-1} \) and \( \mu_{\text{eff}} \approx 3.1 \mu m pN^{-1}s^{-1} \), i.e., closer to \( \mu_0 \). Therefore, the short-time response of the viscoelastic fluid, which gives rise to a lower friction around the unstable saddle point, is responsible for enhancing the probability of transitions across the barrier.

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prehension of plenty of transport mechanisms in nature, commonly occurring in non-Newtonian conditions, such as those involving microorganisms and biomolecules [68–71], as well as in activated processes in other types of non-equilibrium systems with intrinsic memory, such as active matter [72] and glassy materials [73]. Finally, this phenomenon can be exploited to envisage new approaches to selectively deliver microscopic assays in artificially generated potential landscapes [74–75].

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