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SCATTERING INTENSITY FROM BROWNIAN DYNAMICS: APPLICATION TO TOTAL INTERNAL REFLECTION MICROSCOPY

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ABSTRACT. Total internal reflection microscopy (TIRM) measures the position of a Brownian particle above an interface by using its scattering of an evanescent wave. From the knowledge of the trajectory it is possible to reconstruct the interaction potential between the Brownian particle and the wall with nanometer and femtonewton resolution. TIRM relies on the a priori knowledge of the relation $I(z)$ between the particle position and the scattering intensity. We introduced a method to determine experimentally $I(z)$. Such method largely extends the conditions accessible with TIRM.

1. Introduction

Total internal reflection microscopy (TIRM, Fig. 1) [1, 2] permits one to measure the trajectory of a Brownian particle diffusing near a wall by monitoring the intensity I scattered by the Brownian particle under evanescent illumination. From the timeseries of the particle-wall separation distance z it is possible to sample the particle equilibrium distribution and from this to determine the distance resolved interaction potential and corresponding forces with femtonewton and nanometer resolution. TIRM has successfully been applied to the study of particle-wall interactions such as electrostatic [3, 4], van derWaals [5], depletion [6, 7, 8, 9], magnetic [10] and critical Casimir [11] forces.

Notwithstanding, TIRM presents some limits. The central point of the data analysis is the *a priori* knowledge of the relation between the measured scattering intensity I and the corresponding particle distance z. For short penetration depths of the evanescent field and in front of a dielectric surface, it has been demonstrated that $I(z) \propto \exp(-z/\beta)$, where β is the evanescent field penetration depth [1, 2]. This, however, poses considerable constraints to the experimental conditions and the range of forces where TIRM can be applied. In particular, this prevents the possibility of using TIRM with large penetration depths (hundreds of nanometers), in front of reflecting surfaces (e.g., gold which is of interest for biological application because it can be functionalized), and with reflecting particles (e.g., gold). Under these conditions multiple optical reflections between the particle and the wall typically lead to a non-exponential $I(z)$ [12, 13, 14].

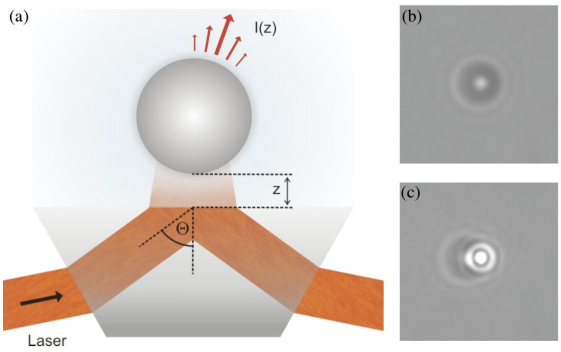

Figure 1. (a) Schematic representation of the TIRM working principle. The incoming laser is totally internally reflected at the glass-water interface generating an evanescent wave. A nearby colloidal particle scatters the light from the evanescent wave. The scattering intensity $I(z)$ depends on the distance z from the colloid to the surface. (b) $4.3\,\mu\mathrm{m}$ diameter polystyrene spherical particle illuminated in bright-field and (b) scattering of an evanescent wave.

Since the scattering of an evanescent field by a particle is a well-posed mathematical problem and can be solved numerically, the distance-intensity relation can in principle be calculated for each of the aforementioned cases. In Ref. [14], for example, the numerically calculated relation was successfully used to correct the experimental data. However, the exact form of the scattering very sensitively depends on parameters –such as the radius and shape of the particle, the refractive indexes of the particle, surface, and liquid medium, the roughness of the surface– that are often prone to signicant uncertainties.

2. Results

We introduced a method to experimentally determine $I(z)$ by making solely use of the distance-dependent hydrodynamic interactions between the particle and the wall [15]. We demonstrated this method by experiments with large penetration depth, gold surfaces and reflecting particles (Fig. 2). When an exponential $I(z)$ cannot be assumed, it can be reconstructed from the acquired intensity time series $I(t)$. The correct $I(z)$ satisfies

$$\begin{cases} MSD(I) &= \left(\frac{dI}{dz}\right)^2 2D(z) \\ S(I) &= \frac{9}{2}\frac{d^2I}{dz^2}\left\|\frac{dz}{dI}\right\|\sqrt{2D(z)} \end{cases} \tag{1}$$

where $D(z)$ is the position-dependent diffusion coefficient of the particle, $MSD(I)$ is the mean square displacement and $S(I)$ is the skweness calculated on $I(t)$.

3. Discussion

Our method largely extends the range of conditions accessible with TIRM, and even allows measurements on highly reflecting gold surfaces where multiple reflections lead to large deviations from an exponential $I(z)$ relationship. This technique will particularly be beneficial for the extension of TIRM to new domains. We have demonstrated TIRM

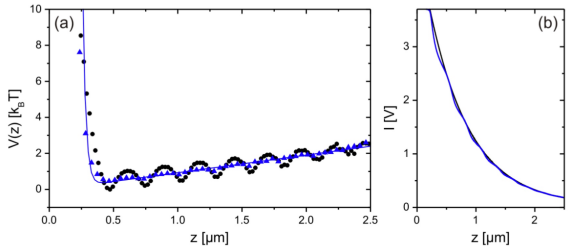

Figure 2. (a) TIRM with a reflecting particle ($400\,\mathrm{nm}$ radius titanium-oxide particle [16]) and large penetration depth ($\beta = 720\,\mathrm{nm}$). Under such conditions an exponential cannot be assumed and, indeed, the potential obtained the exponential $I(z)$ is faulty (black circles). Using the fitted $I(z)$ the correct shape of the potential is retrieved, with a short-range electrostatic repulsion and a gravitational slope far from the surface. (b) Exponential (black) and fitted (blue) $I(z)$.

with a very large penetration depth, which allows one to bridge the gap between surface measurements and bulk measurements, and TIRM in front of a reflecting (gold-coated) surface, which allows plasmonic and biological applications.

4. Outlook

This new technique only assumes the knowledge of the particle radius, which is usually known within an high accuracy and can also be measured in situ, and the monotonicity of $I(z)$. This permits, in principle, to completely automatize the analysis of TIRM data, possibly providing the missing link for a widespread application of TIRM to fields, such as biology, where automated analysis techniques are highly appreciated. This new calibration technique will particularly be benecial for the extension of TIRM to new domains: biological applications; surface plasmon applications; reflecting surface and/or particle; non-spherical particles e.g., cells or organelles; particles whose optical properties are at least partially unknown, e.g., gels and cells.

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