I. INTRODUCTION

The measurement of any response function of a physical system is usually based on the same procedure: a small external forcing has to be applied to the system in order to study its response. Small, because we work under the fundamental hypothesis of the linear response theory. The forcing of the system should not drive it to another state where its properties are different. While it is generally easy to fulfill this condition, it may become tricky in some cases. For example, systems that are very fragile or close to a critical point cannot be tested without extreme care. Likewise, systems that are still evolving must be handled with precaution; since they are not in equilibrium, any external forcing may change (delay, accelerate, etc.) their natural evolution.

For systems in equilibrium, the study of intrinsic fluctuations of one property can give an answer to that problem; in Onsager’s hypothesis, these thermally excited fluctuations are governed by the same linear response functions. The fluctuation-dissipation theorem (FDT), resulting from this hypothesis, allows determination of response functions from the study of fluctuations. Let us recall briefly the expression of this theorem. We consider an observable \( x \) of the system and its conjugate variable \( F \). The response function \( \chi_{xF}(f) \), at frequency \( f \), describes the variation \( \delta x(f) \) of \( x \) induced by a perturbation \( \delta F(f) \) of \( F \), that is, \( \chi_{xF}(f) = \delta x(f)/\delta F(f) \). FDT relates the fluctuation spectral density of \( x \) to the response function \( \chi_{xF} \) and the temperature \( T \) of the system:

\[
S_x(f) = \frac{4k_B T}{\omega} \text{Im}[\chi_{xF}(f)],
\]

where \( S_x(f) = \langle |x(f)|^2 \rangle \) is the fluctuation spectral density of \( x \), \( k_B \) is the Boltzmann constant, \( \text{Im}[\chi_{xF}(f)] \) is the imaginary part of \( \chi_{xF}(f) \), and \( \omega = 2\pi f \). Textbook examples of FDT are Nyquist’s formula relating the voltage noise to the electrical resistance and the Einstein’s relation for Brownian motion relating the particle diffusion coefficient to the fluid viscosity. Therefore, the study of fluctuations can give a true zero forcing measurement of a response function. This smart technique has already been used in several experiments, to extract properties of biological, chemical, and physical systems.

The case of out of equilibrium systems is a bit different, since the requirements of FDT are not fulfilled. However, some recent theories for glassy materials point out the interest of the study of fluctuation-dissipation ratio in these slowly aging systems. Experimentally, such a measurement implies an independent determination of fluctuations and response, with a vanishing driving in the second case to avoid any modification of sample aging. Recent experiments, all focused on electrical properties of glasses, have shown the interest of such an approach.

Our experiment was built in this context; in order to test FDT in glassy materials, we developed an original rheometer whose purpose is to measure thermally excited strain in a sample, and compare it to what can be expected from FDT. The experimental setup is based on a classic cylindric Couette rheometer configuration. To measure the angular position of the rotor, we use a differential interferometer (described in Sec. II), which permits detection of thermally excited motion. In Sec. III, we present the forcing method, based on electrostatic interaction, that we use to generate torques comparable to that of thermal noise. Section IV illustrates rheometer operation with a silicon oil, demonstrating its ability to measure response function from thermal fluctuations. We discuss in Sec. V.

II. STRAIN MEASUREMENTS: DIFFERENTIAL INTERFEROMETRY

In order to measure the infinitesimal displacements that are thermally excited in the sample, we use a differential interferometer. Originally proposed by Nomarski, this technique is well suited to measure submicron displacements with better than \( 10^{-13} \text{ m}/\sqrt{\text{Hz}} \) sensitivity.

Figure 1 reports the optical scheme we used in our apparatus. Light emitted by a stabilized He–Ne laser (Melles Griot 05STP903) is brought to the interferometer through a polarization maintaining singlemode optical fiber. The highly...
calcite prism
the fiber is collimated with a convergent lens, and enters a prism by a mirror, and the phase shift interferometer; each beam is reflected back in the calcite two beams cross a region which is the sensitive area of the equal intensities but with orthogonal polarizations. These the beam makes a 45° angle with the neutral axes of the Glo- stain of orthogonal polarization with a Wollaston prism cross polarizations. The light is further split into two beams of orthogonal polarizations. These two beams cross a region which is the sensitive area of the interferometer; it is better than 10^{-12} m/\sqrt{Hz} above 0.6 Hz. Peaks are mechanical resonant frequencies of the apparatus, whereas the base line is due to conditioning electronics and laser phase noise.

FIG. 1. Scheme of the differential interferometer. A calcite prism splits input light into two parallel beams with cross polarizations. These two beams are recombined with the same optical component after a reflection on a mirror. The phase shift \( \varphi \) between cross polarizations keeps track of optical path difference in the sensitive area. Analysis of this beam is made with a Wollaston prism, oriented at 45° with respect to the calcite. Contrast between intensities collected by photodiodes is a sinusoidal function of \( \varphi \).

Now that we have a highly efficient tool to measure variations of optical path between two parallel beams, we would like to use it to record the angular position of the rotor in the rheometer. This can be done accurately by interposing a square prism in the light path, and using a single mirror, as shown in Fig. 3(a). When the two beams enter the cube on both sides of a diagonal, the variation of \( \delta L \) is just proportional to that of the angular position \( \theta \) of the prism; as shown in Fig. 3(b), a linear approximation is excellent in all the range accessible in \( \theta \):

\[
\delta L = L_0 \theta. \tag{4}
\]

For our experimental conditions, we compute \( L_0 = 20 \text{ mm/rad} \) with a 30° range available in \( \theta \). Thus using this deflection technique the contrast is proportional to \( \theta \). Specifically, inserting Eq. (4) in Eq. (3):

\[
C = \frac{2\pi}{\lambda} L_0 \theta = A \theta \tag{5}
\]
with $A = 2.09 \times 10^5 \text{rad}^{-1}$. Advantages of this deflection technique are the following: insensitivity to any translation of the prism to rotation around the optical axis, no pointing error due to any displacement, and easy positioning. Moreover, sensitivity to rotations around the second horizontal axis are second order terms in the optical path expression. Therefore, this deflection technique together with the differential interferometer allows $10^{-11} \text{rad/} \sqrt{\text{Hz}}$ sensitivity for the detection of rotation around a vertical axis only.

III. RHEOMETER SETUP AND LOW STRESS GENERATION

Figure 4 illustrates the rheometer setup. A rotor is suspended by two steel wires in a cell of radius $R_r = 7 \text{ mm}$. The rotor, which is made of poly-vinyl chloride (PVC) to reduce its inertial moment $J$, has a radius $R_h = 6 \text{ mm}$ and a height $H_h = 30 \text{ mm}$. The sample fills the 1 mm gap between the rotor and the stainless steel cell. Suspension steel wires have a diameter of 0.2 mm and are tightened to ensure correct centering of the cylindric Couette setup, and to fix the resonant frequency of the torsion pendulum in the middle of the measuring frequency range. In order to remain within the linear range of the interferometer, the displacements due to the forcing technique must remain very small, thus the torque acting on the rotor too. We chose to use electrostatic interaction to achieve this goal, as shown in Fig. 4: two small metallic cylinders (labeled $A_1$ and $A_2$), fixed on the rotor, are set in correspondence with two others (labeled $B_1$ and $B_2$), fixed on the external cell. Each pair of cylinders ($A_i-B_1$ and $A_2-B_2$) constitutes a capacitor. Applying a voltage $\delta v_i = v_B - v_A$ between them creates an attractive force $F_i$ ($i = 1,2$) between corresponding cylinders. Respecting the symmetry around the vertical rotation axis, the total force is zero and we end up with a torque $\Gamma$ acting on the rotor, tending to align all the cylinders in the same plane. Cylinders to apply torque have a 3 mm diameter for a 10 mm height and their distance is about 2 mm. The resulting capacitance is of the order of the pF. Useful driving is achieved when $\delta v_i$ is a white noise with a peak amplitude of about 100 V.

Knowing the value of the torque as a function of the applied voltage and angular position is far too complicated to be trusted, therefore, we cannot directly access the torque value. But we know that $\Gamma$, created by electrostatic interaction, is a quadratic function of the driving voltage $\delta v_i$, that is,

$$\Gamma = B \delta v_i^2,$$

where $B$ is a proportionality constant, which depends on the precise positioning of the cylinders. It can be determined by the inertial calibration procedure described in the next paragraph. This electrostatic stress method allows us to apply extremely tiny torque to the rotor, of order of magnitude $10^{-12} \text{ N m/} \sqrt{\text{Hz}}$.

To study the rheological behavior of the sample, we have to measure the response of the rotor to an external forcing, i.e., the response function $\chi_{\theta} = \delta \theta(f)/\Gamma(f)$ where $\delta \theta(f)$ and $\Gamma(f)$ are, respectively, the amplitudes at frequency $f$ of the angular position variation $\delta \theta$ and of the applied torque $\Gamma$. For small displacements, $\delta \theta$ is simply proportional to the output of the interferometer, the contrast $C$. Being $\Gamma$ linear in $\delta v_i^2$, then in Fourier space $\chi_{\theta}$ is just proportional to the transfer function $\chi_{C\delta v_i^2}$. Specifically, using Eqs. (5) and (6):

$$\chi_{\theta} = \frac{\delta \theta(f)}{\Gamma(f)} = \frac{A}{B} \frac{C(f)}{\Delta(f)} = \frac{A}{B} \chi_{C\delta v_i^2},$$

where $\Delta(f)$ is the amplitude at frequency $f$ of the Fourier transform of $\delta v_i^2$. The proportionality constant $P_c = A/B$ in Eq. (7) can be found using an inertial calibration of the measurement.

To illustrate this method, we consider that the cell is filled with a fluid of viscosity $\eta$ and density $\rho$ and that the rotor is subjected to an oscillating external torque of amplitude $\Gamma(f)$ and pulsation $\omega = 2\pi f$. In such a case the equation of motion of the rotor in Fourier space is

$$-J \omega^2 \delta \theta(f) - i\alpha \omega \delta \theta(f) + k \delta \theta(f) = \Gamma(f),$$

where $J$ is the moment of inertia of the rotor, $\alpha$ is the damping coefficient, and $k$ is the effective spring constant of the torsion pendulum.
where \( \mathcal{J} \) is the rotor inertia moment, \( k \) the steel wires stiffness, and \( \alpha \) is a geometric factor which depends on the size of the rotor and of the gap between the cell and the rotor. For our geometry \( \alpha = 4 \pi \frac{H_R}{(1/R_R^\text{c} - 1/R_C^\text{c})} = (5 \pm 0.5) \times 10^{-5} \text{ m}^3 \), within the approximation that the velocity profile of the fluid inside the gap between the rotor and the cell is linear, that is \((R_C - R_R)^2 < \eta/(2 \pi f \rho)\). Equation (8) directly leads to the expression of the response function of this torsion pendulum

\[
\chi_{\theta} = \frac{\delta \theta(f)}{\Gamma(f)} = \frac{1}{(k - J \omega^2) - i \alpha \eta / \omega}.
\]

(9)

From Eq. (9) we see that \( \text{Re}(1/\chi_{\theta}) = (k - J \omega^2) \) is a parabola whose quadratic coefficient is \( J \). The rotor inertia moment \( J \), being known with good accuracy, can be used to calibrate the measurement. From Eq. (7) we find that

\[
\text{Re} \left( \frac{1}{\chi_{\theta} c^2} \right) = P_c (k - J \omega^2).
\]

(10)

To determine \( P_c \) we have just to fit the real part of the inverse of the transfer function \( 1/\chi_{\theta} \) with a parabola. The coefficient of the quadratic term is just the product of \( P_c J \).

Thus \( P_c \) is determined with good accuracy from this parabolic fit. The inertial calibration technique allows a precise reading of the applied torque, and a complete determination of the response function \( \chi_{\theta} \). After calibration, the response function can of course be generalized for any fluid of complex shear modulus \( G = G_1 - i G_2 \), specifically,

\[
\chi_{\theta} = \frac{\delta \theta(f)}{\Gamma(f)} = \frac{1}{(\alpha G_1 + k - J \omega^2) - i \alpha G_2}.
\]

(11)

IV. RHEOMETER OPERATION

To excite the rheometer we used a white noise source for \( \delta \dot{\omega} \). Simultaneous acquisition of \( \omega \) and \( \delta \dot{\omega} \) is done by a computer via a 16 bits data acquisition system. The computer calculates \( \delta \dot{\omega}^2 \) and the response function \( \chi_{\theta} \).

To test the performance of the rheometer, we study a silicon oil whose viscosity, measured with an industrial rheometer (HAAKE, SR100), is \( G_1 = 0 \) and \( \eta = (3.3 \pm 0.1) \text{ Pa s} \) at \( \omega = 0 \). In Fig. 5, we plot as a function of frequency, the real and imaginary part of the inverse of the response function of our rheometer, \( 1/\chi_{\theta} \). As expected from Eqs. (9) and (10), the real part is a parabola centered on \( f = 0 \text{ Hz} \). The fit of this parabola allows us to determine \( P_c \) and \( k \). Once \( P_c \) is known, from the linear fit of the imaginary part (for \( f \to 0 \)), we can access the value of the viscosity; for a Newtonian liquid, the slope is the viscosity up to the known geometrical factor \( \alpha \). We measure \( \eta = (3.2 \pm 0.3) \text{ Pa s} \), in agreement with the other measurement. The main source of uncertainty in this determination is that for the geometrical factor \( \alpha \), we cannot ensure better than 10% centering of the rotor with our current experimental setup. We notice a deviation to the linear behavior for \( f > 20 \text{ Hz} \); it is due to the non-Newtonian behavior of this silicon oil at high frequencies.

This measurement corresponds to a classic rheological experiment, except for the tiny values of stress and strain applied to the material: the rms value of the shear applied is less than \( 10^{-3} \text{ s}^{-1} \) with a rms displacement of about 4 nm. But we can even perform a true zero applied stress determination of viscosity with this experimental setup. Indeed, let us apply FDT to the rheometer, that is rewrite Eq. (1) for coupled variables \( \delta \theta \) and \( \Gamma \); when no external forcing is applied, the angular position fluctuation spectrum \( S_\theta \) is given by

\[
S_\theta = \frac{4 k_B T}{\omega} \text{Im}(\chi_{\theta}^\ast)
\]

(12)

Thus measuring thermal noise of the rotor angular position gives a determination of the imaginary part of \( \chi_{\theta}^\ast \), with true zero forcing. In Fig. 6 we plot the measured noise spectrum and that computed using Eq. (12) and the previously determined values of \( \chi_{\theta}^\ast \). The agreement with the prediction of FDT is good between 0.2 and 100 Hz (the peaks above 15
external driving, we deduce from the measurement of \( G \) the value of \( \eta \).

Hz are due to mechanical vibrations, which are amplified by the rheometer). This agreement shows that FDT can be used to determine viscosity of a liquid, without applying any shear to it.

Indeed from the best fit of noise spectrum using Eq. (12) one can measure the viscosity which is the only unknown of Eq. (12). From this best fit we get \( \eta = (3.05 \pm 0.3) \text{Pa s} \) which is in good agreement with the value measured directly.

V. DISCUSSION

In this article, we described a rheological experiment based on a classic cylindrical Couette setup. The use of a differential interferometer combined to a deflection technique allows us to measure the angular position with a sensitivity better than \( 10^{-10} \text{rad}/\sqrt{\text{Hz}} \). Extremely low shear can be applied to the sample; using electrostatic interaction in a capacitor, we can apply torques as small as \( 10^{-12} \text{N m} \) to the rotor. An inertial calibration of the measurement allows us complete determination of the response function of the rheometer.

This rheometer can be used in two different ways:

(i) Classical rheological measurement. Applying a small external driving, we deduce from the measurement of \( \chi_{\theta T} \) and Eq. (11) the value of \( G \).

(ii) Noise measurement. Without any external forcing, we record angular position noise \( S_\theta \). If the sample is at equilibrium, FDT is valid and Eqs. (11) and (12) allow us to determine \( G \). [From Eq. (12) only the imaginary part of \( \chi_{\theta T} \) can be determined, but the real part can be deduced via Kramers--Kronig relations.]

This second procedure provides a new tool to investigate rheological properties of a media. When used alone, it provides a true zero stress rheological measurement, that, for example, can be useful for highly fragile materials. When compared to the classical measurements, it tests the validity of linear response hypothesis; if the two measurements disagree, the sample is out of equilibrium. This can be due either to intrinsic process in the material (e.g., aging, chemical reactions, etc.) or to the external forcing applied in the classical measurement.

This rheological experiment was built in order to test FDT in aging materials, preliminary results of this application can be found in Ref. 10. Based on a cylindric Couette setup, this technique can certainly be extended to other materials or configurations, and may find useful applications in various research domains.

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