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Correlations between the thermal vibrations of two cantilevers: Validation of deterministic analysis via the fluctuation-dissipation theorem

Christopher D. F. Honig, Milad Radiom, Brian A. Robbins, John Y. Walz, Mark R. Paul, and William A. Ducker^{a)}

Department of Chemical Engineering and Department of Mechanical Engineering, Virginia Tech, Blacksburg, Virginia 24060, USA

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We validate a theoretical approach for analyzing correlations in the fluctuations of two cantilevers in terms of a deterministic model, using the fluctuation-dissipation theorem [M. R. Paul and M. C. Cross, Phys. Rev. Lett. **92**, 235501 (2004)]. The validation has been made possible through measurement of the correlations between the thermally stimulated vibrations of two closely spaced micrometer-scale cantilevers in fluid. Validation of the theory enables development of a method for characterizing fluids, which we call correlation force spectrometry. © 2012 American Institute of Physics. [doi:10.1063/1.3681141]

The stochastic motion of micron and nanoscale cantilevers in fluid is important to a number of technologies such as atomic force microscopy¹ and to the development of biomolecule detectors.² Thus, there has been a concerted effort to understand the dynamics of cantilevers in fluid,^{3,4} which has been extended to use of microcantilevers as viscometers.^{5,6} In the literature, there are a number of examples illustrating an excellent agreement between experiments and theory for the noise spectrum of a single cantilever in fluid.⁷⁻⁹ The high frequency motion of silicon-based cantilevers is in a regime where inertial effects are important, thus making the analysis difficult due to the subtle interactions of potential and non-potential contributions in the flow field. This is in contrast to the well understood case of oscillating micron scale beads that are held in optical traps in liquid. In this case, inertial effects are negligible and current work on two-particle microrheology continues to make significant progress.¹⁰

A useful development for the field would be to move from the analysis of single cantilevers to the analysis of the correlations between cantilevers. This is because (a) correlation measurements have lower noise: this noise includes both instrumental noise and thermal noise^{8,11} and (b) correlations between cantilevers depend on the spatial variation of the properties of the immersion fluid. Thus, the micron-scale features of the immersion fluid can be examined. However, the implementation of correlation measurements requires a method of analysis.

Using the fluctuation-dissipation theorem, Paul and Cross showed that the *stochastic* dynamics of pairs of small cantilevers in fluid could be analyzed in terms of *deterministic* modeling of the a pair of cantilevers when cantilever 1 is subject to the removal of a force, F_1 ¹²

$$\langle x_1(0)x_1(t) \rangle = \frac{k_B T}{F_1} X_1(t), \quad (1)$$

$$\langle x_1(0)x_2(t) \rangle = \frac{k_B T}{F_1} X_2(t), \quad (2)$$

Equation (1) relates the autocorrelation of stochastic fluctuations in deflection of a single cantilever, $\langle x_1(0)x_1(t) \rangle$, to the deterministic ring down of the same cantilever, $X_1(t)$, after the removal of a step force, F_1 , at time zero. t is the time, x is the deflection, k_B is the Boltzmann constant, and T is the temperature. Equation (2) relates the cross-correlation of stochastic fluctuations between two cantilevers, $\langle x_1(0)x_2(t) \rangle$, to the deterministic time series of deflections of cantilever 2, $X_2(t)$, after the removal of a step force from cantilever 1, F_1 , at time zero.

In this paper, we present a direct experimental measurement of the stochastic cross-correlations of a cantilever pair in fluid, which provides the experimental validation of Eq. (2). These results pave the way for the development of new technologies using the correlated motion of pairs of cantilevers and other objects. For example, we demonstrate that an instrument using the theory can resolve differences in the viscosity of Newtonian fluids, validating the use of the instrument as a rheometer. This effort runs in parallel to the work of others on oscillating micron scale beads that are held in optical traps in liquid.¹³

The apparatus consists of a pair of commercial AFM cantilevers (here, ORC8-10 B, Bruker, CA) mounted in an antiparallel configuration between two glass slides as shown schematically in Fig. 1, which in the experiments reported here are in a 30 ml beaker of fluid at 23 °C. There are no “moving” parts; the only motion is molecular motion of the fluid and the fluctuating deflection of the cantilevers due to interaction with the fluid at equilibrium. The deflection of each of the cantilevers is measured by the light-lever technique¹⁴ in which a laser (Schäfer + Kirchhoff GmbH, Hamburg, Germany) is reflected by the cantilever onto a position sensitive diode (Phresh Photonics, Reseda, CA). In this case, we use a different wavelength (635 and 680 nm) of laser for each cantilever and (in some experiments) use a wavelength filter over each diode and different light paths to prevent cross-talk between the signals. The signals are recorded synchronously by an Asylum Research Atomic Force Microscope controller (Nyquist frequency, $f_{Ny} = 25$ kHz). The phase lag between detection systems was shown to be negligible in an experiment when both lasers recorded the motion

^{a)}Author to whom correspondence should be addressed. Electronic mail: wducker@vt.edu.

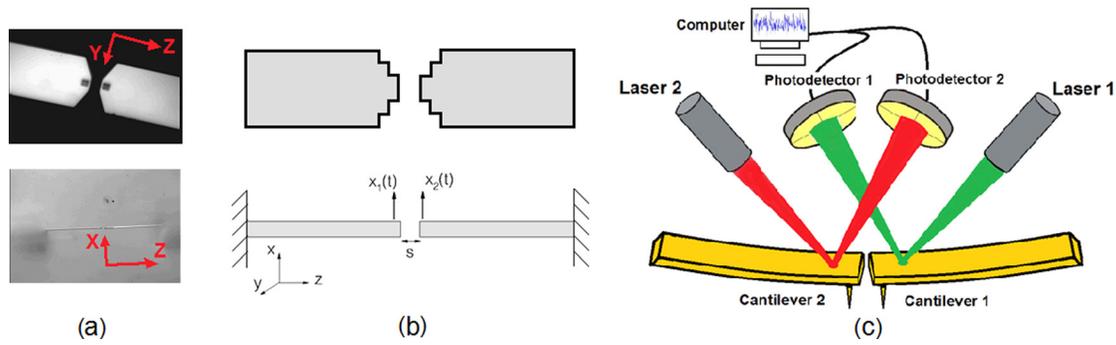


FIG. 1. (Color online) (a) Light microscope image of closely spaced AFM cantilevers seen from above and the side. The cantilevers are $200\ \mu\text{m}$ long and $40\ \mu\text{m}$ wide. (b) Schematic of the cantilevers separated by a distance s , and the shape of the free end of the simulated cantilever. The real cantilever has a flat $10\ \mu\text{m}$ end and then slopes back at about 59° ; this slope is simulated by two equal steps for computational simplicity. (c) Schematic of the detection system.

of a single cantilever. By measurement of the energy spectral density as a function of laser power, we found that the laser had a negligible effect ($<0.5^\circ\text{C}$) on the temperature of the cantilever.

Typically, we measure a time course of 5×10^6 voltage measurements from the diodes at 50 kHz. Subsequent data processing consists of (1) dividing the whole data points into 10 000 bins; (2) subtracting a linear curve fit from each bin to remove drift in the signal; (3) taking a Fourier transform of each bin and calculating the power spectral density (PSD) for each cantilever averaged over all bins; and (4) normalizing the data by multiplying through by $\sqrt{f_{Ny}/A}$, where A is the area under the PSD. By experiment, this gives a value of the autocorrelation of unity at zero time lag. The noise spectra are then $G_{11}(\omega) = \frac{1}{N} \sum_{bins} \hat{x}_1(\omega) \hat{x}_1^*(\omega)$ and $G_{12}(\omega) = \frac{1}{N} \sum_{bins} \hat{x}_1(\omega) \hat{x}_2^*(\omega)$, where $\hat{x}_i(\omega)$ is the Fourier transform of the normalized signal from each cantilever, ω is the frequency, and N is the number of samples used in the average. The cross-correlation is normalized by the geometric mean of the normalization constant for each contributing signal. To remove the normalization on the data, we need to multiply through by $k_b T/k$, which requires measurement of the cantilever spring constant. We calibrated the spring constant in a separate experiment in an Asylum Instruments MFP-3D.¹⁵ In other words, we obtained the voltage-deflection conversion in the correlation force spectroscopy (CFS) by measuring a thermal on the CFS and the MFP-3D and calibrating the voltage-deflection on the MFP-3D.

Experimental measurements of cantilever fluctuations are compared to finite-element numerical simulations⁹ of the deterministic motion of cantilevers, after removal of the step force, for the precise geometries and conditions of the experiment. We used the nominal values of the cantilever geometry provided by the manufacturer and have fitted the values for the density ($4166\ \text{kg/m}^3$) and Young's modulus (140 GPa) of the cantilever to ensure that the theoretical values of the resonant frequency in air and spring constant of each cantilever matched the corresponding experimentally measured value. This was done because of the large experimental errors in determining precise values of the Young's modulus and the cantilever thickness.

A comparison between the measured auto-correlation, cross-correlation, and noise spectrum for two cantilevers immersed in water and the value determined from Eqs. (1)

and (2) using finite element numerical simulations of deterministic motion are shown in Fig. 2. Clearly, there is an agreement between the direct experiment and the values obtained via Eq. (2): the zero time lag cross correlation between experiment and simulation is 0.98 for the autocorrelation data and 0.94 for the cross correlation data. This

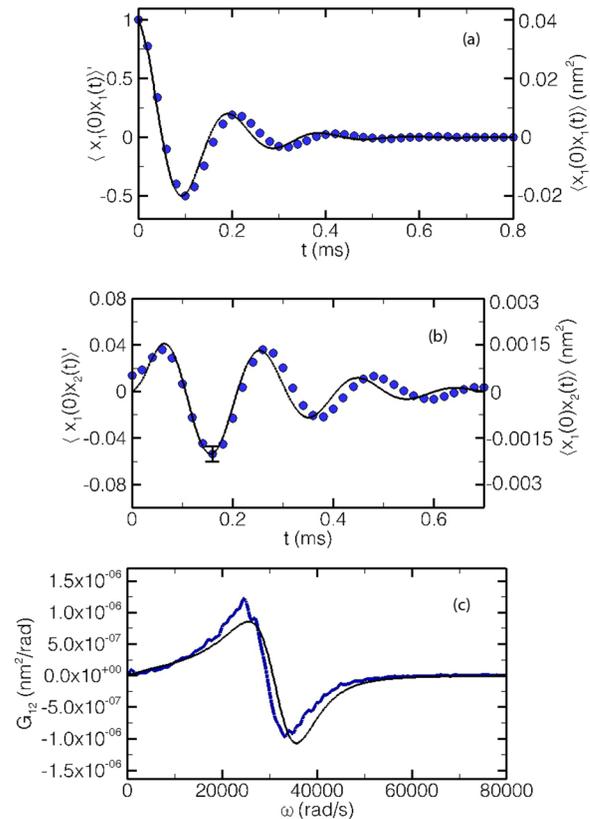


FIG. 2. (Color online) (a) Auto-correlation. (b) Cross-correlation. (c) Noise spectrum, G_{12} . These results are for a pair of commercial AFM cantilevers (ORC8 B: length = $200\ \mu\text{m}$, width = $40\ \mu\text{m}$, and $k \approx 0.1\ \text{Nm}^{-1}$) separated by $8\ \mu\text{m}$ in liquid water at 23°C . In (a) and (b), the experimental measurements are shown using data symbols and the theoretical prediction is given by the solid line. The left axis shows the correlation function normalized by $k_b T/k$ where k_b is Boltzmann's constant, T is the temperature, and the right axis shows the same data in units of nm^2 using the measured value of the spring constant. In (b), the single error bar shows the range of data in three repeat experiments at this time lag. Each repeat experiment has a different pair of cantilevers. In (c), the noisy line is the experimental measurement and the smooth line is the theoretical prediction.

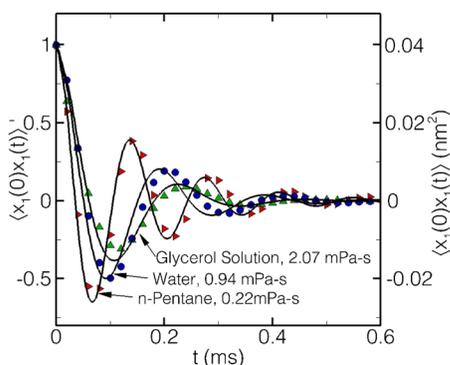


FIG. 3. (Color online) Autocorrelation of equilibrium fluctuations in cantilever displacement for an AFM cantilever in a series of Newtonian fluids. The left axis is normalized by $k_b T/k$ and the right axis is in units of nm^2 .

validates the use of the theory (Eq. (2)). Further validation is provided in Fig. 3 which compares the left and right side of Eq. (1) and Fig. 4 which compares the left and right side of Eq. (2) for fluids of different viscosity (pentane ($\eta = 0.22$ mPa s), water ($\eta = 0.94$ mPa s), and 24 wt. % glycerol in water ($\eta = 2.07$ mPa s)). Again, the close match validates the theory.

The close fit between the experiment and the model using the theory was obtained using the known values of the

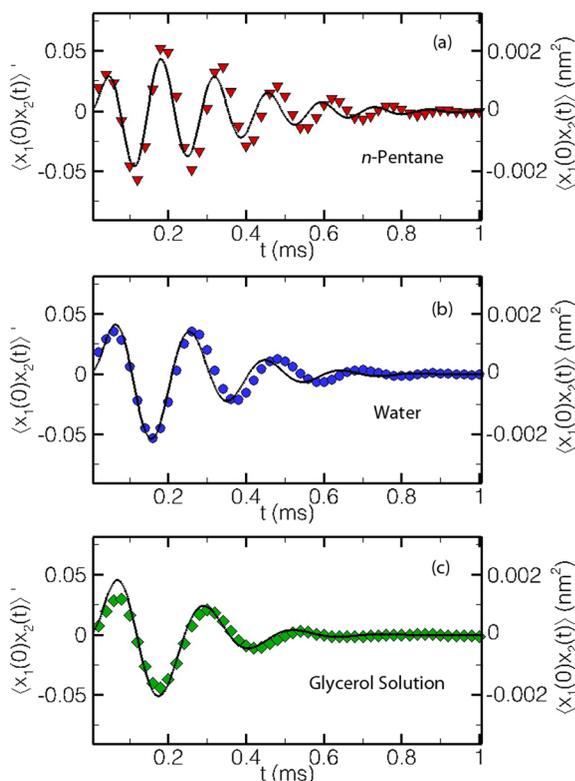


FIG. 4. (Color online) Cross-correlation of equilibrium fluctuations in cantilever displacement for a pair of AFM cantilevers in a series of Newtonian fluids. Experimental measurements are shown as data symbols and theoretical predictions are shown by the solid lines. The left axis is normalized by $k_b T/k$ and the right axis is in units of nm^2 .

viscosity. This suggests that the analysis of the correlated vibrations of two cantilevers can be used as a rheometer to measure the unknown viscosities of solutions. The principal advantages of such a technique are (in common with other cantilever techniques)^{4,5} (1) it is minimally invasive: there are no “moving” parts, we simply monitor the thermally driven oscillations at equilibrium; (2) small sample volume; (3) the device is simple; (4) the cantilevers are fixed in space, so any number of solutions or suspensions can be washed across them for analysis; (5) the dimensions of the wet parts are only millimeters, so the device could be used as a dip probe; and (6) alignment of the cantilevers is not critical (for \sim micrometer changes in cantilever lateral or vertical separation, there was only very small variation in both our experimental and simulation results). Once the rheology of the immersion fluid is understood, the two cantilever method can also be used for analyzing the properties of molecules that straddle the gap between two cantilevers.

In summary, we have validated the theory of Paul and Cross by showing that the experimental values of the autocorrelation and cross-correlation in fluctuations of the deflections of micrometer-sized cantilevers are given by the deterministic ring-down of the same cantilevers. The good agreement using the known viscosity of solution suggests that a pair of cantilevers can be used as a simple rheometer with no moving parts; we name that technique CFS.

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