Fluctuation and dissipation of a stochastic micro-oscillator under delayed feedback

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We investigate the dynamics of a microcantilever subjected to the combined forcing from Brownian motion and delayed self-feedback. Specifically, the excitation of the fundamental mode of the cantilever by thermomechanical agitation is utilized as delayed external forcing and the resulting dynamical response is studied as a function of the delay and the coupling strength. A fluctuation-dissipation theorem is derived from the delay Langevin-like equation and its validity is discussed. The relaxation time scale associated with the adsorption processes is established and an experiment to determine the oscillator's effective temperature is proposed. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2365378]

I. INTRODUCTION

Due to widespread progress in miniaturization, stochastic fluctuations of thermodynamic quantities have become an increasingly important issue in many micro- and nanoscale systems. The Brownian motion of a microfabricated silicon cantilever, for example, has recently drawn significant attention in connection to sensing and scanning probe microscopy.[1,2] The present work was originally motivated by the need for increased sensitivity in frequency shift measurements of the resonant oscillations of a microcantilever for biological and chemical sensing,[3] in particular when operated in a fluid, where the quality factor Q is severely deteriorated. Adsorption events occurring at the surfaces of a microcantilever sensor, which are typically of the order of 10^{-9} m^2, significantly modify the surface free energy. In view of Shuttleworth's equation,[4] this modification results in a differential surface tension at the bounding surfaces of the cantilever. Typically, one side of the microcantilever is coated with gold and/or other material such as a self-assembled monolayer of a molecular group, single stranded DNA, hydrophilic/hydrophobic, antigen-antibody, aptamers, etc., so as to functionalize that surface, targeting the desired adsorbate/analyte. This in turn results (for example, by molecular binding) in a variation in one of the dynamic attributes of the cantilever, such as bending, for which a radius of curvature can be defined that is proportional to the induced tension as approximated by Stoney's equation.[5] An important issue herein has been the presence of a thermomechanically driven amplitude of the sensor. By taking advantage of such fluctuations in a continuously tuning feedback system, one can achieve a higher frequency measurement resolution.[6–8] Regardless of the specific character of the thermal bath, when adsorption (reaction) events occur on the surface of the cantilever, such that a response may be obtained by the frequency or deflection measuring devices, the desorption (relaxation) process may be of the order of minutes.[9] During this time, nonequilibrium conditions may prevail, making the very definition of temperature a questionable proposition. Furthermore, measurements of extremely small thermal variations due to biomolecular binding pose a challenging technological problem, due to the minute dimensions of the system and the small values of the Q factor. The feedback approach to attaining narrow spectral peaks as an effort to facilitate precise frequency shift measurements further complicates the extraction of dynamic information from the measurements. In particular, in light of the fluctuation-dissipation theorem (FDT), the measurement of the temperature of the cantilever in the presence of molecular binding due to adsorption or diffusion in a liquid may pose a certain ambiguity due to a possible violation of the FDT.

To overcome these challenges, we propose the following approach. Within the assumptions of the FDT, we begin by theoretically investigating the dynamics of a microcantilever capable of self-sustained oscillations in the feedback mode. The external excitation is thermal in origin, e.g., due to the Brownian motion of the molecules in the surrounding medium. Another source of self-excitation may be internal phonon fluctuations that eventually drive the cantilever into one of its resonance modes. The response of the system is used as a delayed feedback to provide further enhancement of the Q factor such that the Fourier amplitude of the signal could be measured with a spectrum analyzer. We then proceed by discussing the analytical results such that they may be utilized in an experimental investigation.

II. ANALYTICAL RESULTS AND DISCUSSIONS

Under the above assumptions, in a first-order approximation, the dynamics of the cantilever is described by a second-order stochastic delay differential equation. For this system we derive a FDT, and discuss its validity. The FDT was originally established by Callen and Welton[10] as a generalization of the Nyquist relation using a quantum perturbation theory. Further elaborations and developments were subsequently reported by Weber.[11] In general, the FDT is violated in nonequilibrium systems, even in those where the relaxation time of a process is much larger than the observation

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time as, for example, in glassy systems.[12] For an adsorptively modified microcantilever used as a sensor, the relaxation time scale can be in the range of minutes.[9] However, one may stabilize the cantilever in a stationary situation and perform measurements after possible transient effects have died out. For the dynamics considered in this article, the FDT is not violated.

Using the FDT, we investigate the parameter domain defined by the delay and coupling strength and determine those regions where the Q factor is enhanced. Using the microcantilever in those regimes would provide a more efficient sensor. Based on these results, we propose an adsorption experiment from which the temperature of the cantilever may be obtained as a measure of the effect of desorption.

We model the cantilever by an effective point mass, and denote the delays by $\tau_i \in [0,\infty[$, the dimensionless feedback coupling strength by $g_i \in]-\infty,\infty[$, $i=1,2,\ldots$, the effective mass by m, the viscous damping constant by $\gamma \geq 0$, and the elastic constant by $k>0$. The inclusion of thermal white noise $\Xi(t)$ with $\langle \Xi(t)\rangle = 0$ and $\langle \Xi(t)\Xi(t')\rangle \propto \delta(t-t')$, and memory effects, gives a general formulation of the dynamics of any oscillator, since delays are inherent to any experimental arrangements, albeit here introduced deliberately. For finite constant delays, defining the linear operator $\mathcal{L}=\partial_t^2 + a_1\partial_t + a_2$, with $a_1=\gamma/m$, $a_2=k/m$ we can formulate the general dynamical equation as

$$\mathcal{L}z(t) = b_0\Xi(t) + b_1 z(t-\tau_1) + b_2\partial_t z(t-\tau_2), \tag{1}$$

with the average

$$\mathcal{L}\langle z(t)\rangle = b_1\langle z(t-\tau_1)\rangle + b_2\partial_t\langle z(t-\tau_2)\rangle, \tag{2}$$

where $b_1=g_1/m$ and $b_2=g_2/m$ are real constants, and $b_0 = \mu/m$ with the dimensionless parameter μ used to regulate the noise level. In the absence of the delays, Eq. (1) reduces to the ordinary Langevin equation

$$\mathcal{L}z(t) = b_0\Xi(t) \tag{3}$$

for the dynamics of the system. The response z of the linear dissipative system (3) to a time dependent perturbation $\Xi(t)$, is obtained from the Fourier transform of Eq. (3), $z(\omega) = \chi(\omega)\Xi(\omega)$ at frequency ω, where χ is the complex susceptibility. When a microcantilever in thermodynamic equilibrium with its surrounding medium at temperature T, oscillates with a mean energy E under the condition of a generalized resistance R, the FDT yields a generalized expression for the mean square value of the spontaneously fluctuating force Ξ:

$$\langle \Xi^2\rangle = \frac{2}{\pi}\int R(\omega)E(\omega,T)d\omega \tag{4}$$

(this is in the absence of any other externally applied forces[10]). The application of the FDT to the dynamics of the microcantilever at room temperature T at the relatively low frequencies involved, such that $E(\omega)=E\approx k_BT$ $(\hbar\omega \ll k_BT)$, and for a viscous drag force of $-\gamma\dot{z}$, reduces Eq. (4) to $\langle \Xi^2\rangle \approx (2\gamma k_BT/\pi)\int_{\omega_0}^{\omega} d\omega$, where k_B is Boltzmann's constant, and $\hbar$ is Plank's constant. Taking ω_0 to be the dc limit, and by taking the Fourier transform of the Langevin Eq. (3), one can calculate the spectral (power) density

$$\langle |z|^2\rangle(\omega) = \frac{k_BT}{\pi}\frac{\Im\chi(\omega)}{\omega} = \frac{b_0^2\langle \Xi^2\rangle}{(a_2-\omega^2)^2+(a_1\omega)^2} \tag{5}$$

of the Brownian motion around the equilibrium position.

When the delays are nonzero, taking the Fourier transform of Eq. (1), we obtain for the susceptibility

$$\chi(\omega) = b_0[a_2 - i\omega a_1 - \omega^2 - b_1 e^{i\omega\tau_1} + i\omega b_2 e^{i\omega\tau_2}]^{-1}. \tag{6}$$

Under the open-loop conditions the coefficients a_1, and a_2 can be determined experimentally, by impulse excitation, for example.[7,8] Under position feedback, the coefficient b_1, and under velocity feedback the coefficient b_2 may similarly be determined. However, if the feedback coefficients and delays are not known *a priori*, then one may numerically fit Eq. (6) to the measured frequency response of the system, and thus extract these parameters, typically by approximating the exponentials using a Taylor or Padè expansions. Here, these coefficients are assumed known, and we will use experimentally determined values[7,8] for the simulation of the results. Thus, using Eq. (6), we proceed by writing the spectral density for the delayed system as

$$\langle |z|^2\rangle(\omega) = \left(\frac{2\gamma k_BT\mu^2}{\pi m^2}\right)\frac{\delta\omega}{A^2+B^2}, \tag{7}$$

where $\delta\omega$ is the measurement bandwidth and

$$A = a_2 - \omega^2 - b_1\cos\omega\tau_1 - b_2\omega\sin\omega\tau_2,$$

$$B = a_1\omega + b_1\sin\omega\tau_1 - b_2\omega\cos\omega\tau_2. \tag{8}$$

For $b_1=b_2=0$, this result reduces to that of Eq. (5). The delays may be adjusted such that $\tau_1=\tau_2=\tau$, in which case, using Eq. (7), we can write for the two important cases of position and velocity feedback

$$\frac{\langle |z|^2\rangle(\omega)}{\langle |z|^2\rangle_0(\omega)} = 1 - u(\omega) + u^2(\omega) - \cdots, \tag{9}$$

where u has been defined as $u(\omega)=\Gamma'/(1+\Gamma'')$, and for the off-resonance case, Γ' and Γ'' are given by

$$\Gamma' = \frac{1}{(\omega^2-a_2)^2}\times\begin{cases}\omega^2 b_2^2 - 2a_1b_2\omega^2\cos\omega\tau + b_2\omega(\omega^2-a_2)\sin\omega\tau, & b_1=0,\\ b_1^2 + 2b_1(\omega^2-a_2)\cos\omega\tau + 2a_1b_1\omega\sin\omega\tau, & b_2=0,\end{cases} \tag{10}$$

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and $\Gamma''=a_1^2\omega^2/(\omega^2-a_2)^2$, whereas at resonance, $\Gamma''=0$, and

$$\Gamma' = \begin{cases} b_2^2/a_1^2 - 2b_2\cos\sqrt{a_2}\tau/a_1, & b_1=0, \\ b_1^2/(a_1^2a_2) + 2b_1\sin\sqrt{a_2}\tau/(a_1\sqrt{a_2}), & b_2=0, \end{cases} \tag{11}$$

and $\langle\rangle_0$ denotes the averaging in the absence of reinjection, i.e., Eq. (5). It can be seen that since $0\le\Gamma''<1$, and $|\Gamma'|<1$, then $|u|<1$, and thus the right-hand side (rhs) of Eq. (9) will always be positive as expected. If τ is regulated to meet $\omega\tau\ll 1$, that is, if for the same delay, the frequencies are limited to those with a period T long enough to maintain $\tau/T\ll 1$, then A and B may be expanded such that for $b_2=0$, and weak coupling, small b_1, can be written as

$$\langle|z|^2\rangle(\omega) = \langle|z|^2\rangle_0(\omega)[1-b_1\Delta(\omega)], \tag{12}$$

where

$$\Delta(\omega) = 2\frac{(1+a_1\tau)\omega^2 - a_2}{(a_2-\omega^2)^2+(a_1\omega)^2}, \tag{13}$$

which near resonance takes on the value $\Delta\omega\to\sqrt{a_2}\approx 2m\tau/\gamma$. Thus, for $b_1>0$, cantilevers with a higher mass in less viscous media display less and less Brownian amplitude the longer the delay. To derive the effect of the Brownian forcing upon the cantilever with delayed feedback, we derive first the normal modes of the latter, for the case with $b_2=0$, and $g_1=g$, as a function of (g,τ). We write the formal solution for the case

$$\mathcal{L}z(t) = b_1 z(t-\tau), \quad t\ge t_0, \tag{14}$$

describing the position-delayed damped Hookian oscillations of a cantilever as

$$z(t) = \sum_{n=1}^{\infty} P_n(t)e^{(\alpha_n+i\beta_n)t}, \tag{15}$$

where α_n and β_n compose the nth root of the system

$$\alpha_\pm(\beta) = -\frac{a}{2} - \beta\cot\beta \pm \sqrt{\beta^2\csc^2\beta + \frac{a^2}{4} - c},$$

$$\frac{\sin\beta}{\beta} = -\frac{2}{b}\left(\alpha_\pm + \frac{a}{2}\right)e^{\alpha_\pm}, \tag{16}$$

and where $P_n(t)$ is an arbitrary polynomial of degree less than the multiplicity of the nth root,[8] and we have furthermore defined the parameters $a=a_1\tau=\gamma\tau/m$, $c=a_2\tau^2=k\tau^2/m$, $b=-a_2g\tau^2$. These polynomials are determined from the prefunctions $u(t)$ on the interval $[t_0-\tau,t_0]$. If the system is vibrating in its normal mode when the feedback is engaged at $t_0=0$ such that $(g,\tau)\neq(0,0)$, the prefunction can be set to $u(t)=u(0)\cos\omega t$. We have previously described the experimental realization[7,8] of the rhs of Eq. (14). We note that, when $\tau=0$, Eq. (14) only presents a single oscillation frequency, while for $\tau\neq 0$, there is an infinite number of solutions, some oscillatory, the others nonoscillatory (growing or decaying). The condition $\tan\beta=a\beta/(\beta^2-c)$ implies that $\alpha_+=0$, i.e., there are no growing modes. Figure 1 displays the distribution of the n roots $\lambda_n=(\alpha_n+i\beta_n)$, $n=1,2,\ldots$ of Eq. (16), corresponding to the state of the oscillator in (15). Figure 1(a) displays the roots for a fixed delay at various gain settings, while Fig. 1(b) shows the roots for a fixed gain at several delay settings. In the (τ,g) plane, we thus search for the solutions in the vicinity of the nondelayed $(b_1=b_2=0)$ eigenvalue $\lambda_0=(-1.25+76.0i)\times 10^3$. The distribution of the roots around this value (point $1.0+i$) is shown in Fig. 2. Figure 3 shows the behavior of such eigenvalues α, and $\beta=2\pi f$ for each (g,τ). The division of the (τ,g) domain by negative and positive regions of the real part of the roots suggest that for the corresponding oscillations one can expect suppression or amplification. Only (conjugate) solutions near the imaginary axis are stable. In particular, from the frequency distribution $f(g,\tau)$, one may infer that for low coupling strength g, delay controlled frequency tuning occurs primarily around $T_0/4$ and $3T_0/4$, for which, as seen from α distribution, the amplitude of the oscillations remains

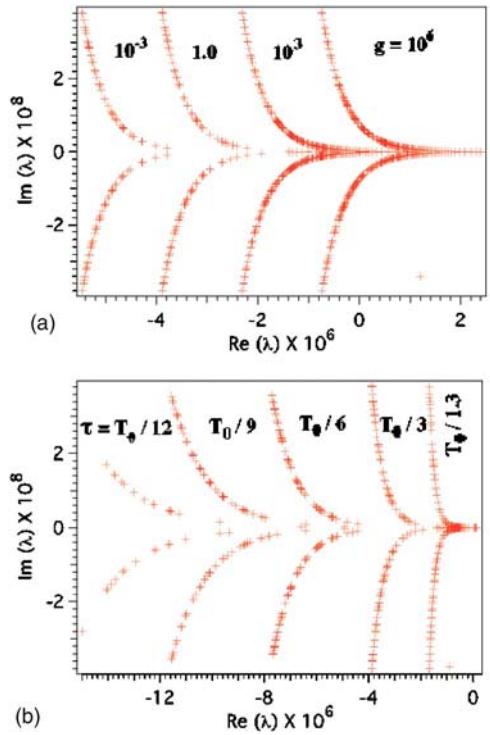

FIG. 1. Infinite complex roots λ of the delayed system of Eq. (2), for some values of (a) g parameter at fixed delay $\tau=T_0/3$, and (b) τ parameter at fixed gain $g=1.0$. The roots migrate from $\mathrm{Re}(\lambda)\to-\infty$ to $\mathrm{Re}(\lambda)\to+\infty$ with increasing gain as seen in (a), or $\mathrm{Re}(\lambda)\to 0$ with increasing delay as in (b). For $g\to 0$ these roots degenerate to the nondelayed value of $\lambda=(-1.25+76.0i)\times 10^3$.

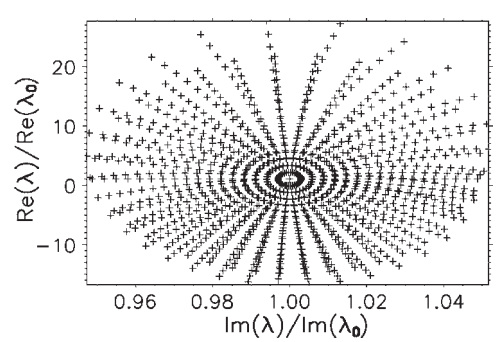

FIG. 2. Distribution of the roots in the vicinity of the nonretarded eigenvalue. Both axes have been scaled with the corresponding nonretarded eigenvalue.

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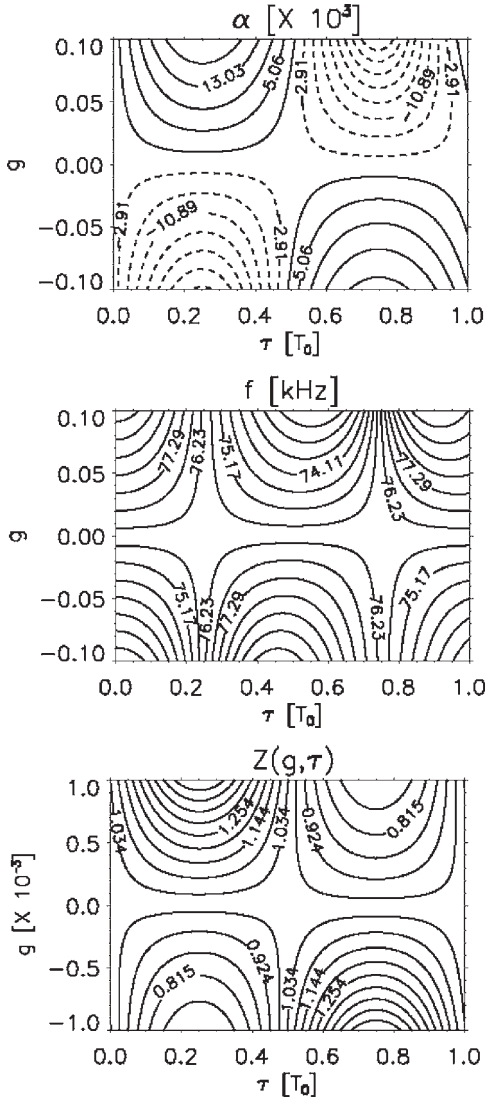

FIG. 3. Eigenvalues (top two) of the delayed system of Eqs. (2), and (bottom) the corresponding system response of (7) in the frequency domain for the case $b_2=0$. Here, $f=\beta/2\pi$, and $Z(g,\tau)=\langle|z|^2\rangle(\omega)/\langle|z|^2\rangle_0(\omega)$, while T_0 is the period of oscillations in the absence of reinjection, i.e., $b_1=0$. The dashed contours depict the negative values, whereas the positive values are shown by solid lines.

relatively unaltered. We also note that a fast transition from amplification to suppression is feasible for delays around $T_0/2$, which for low gain occurs with a minimum frequency shift.

Using the results of this analysis and depending on the available experimental conditions, we can then choose the most convenient delay times and forcing amplitudes in order to enhance the quality factor Q. Furthermore, one may take advantage of these results in the following gedanken experiment. Under the open loop conditions, using a spectrum analyzer, the noise spectrum, i.e., the spontaneously fluctuating position of the cantilever, can be measured at the resonance frequency ω_0 to yield $\langle|z|^2\rangle(\omega_0)=\zeta=k_BT/\pi\gamma\omega_0^2$. Using this experimental information and the functional relationship $T(\gamma)=(\pi\zeta\omega_0^2/k_B)\gamma$, the temperature can be inferred if the dissipation factor is known. However, if the FDT is not valid, then T does not represent the "true" temperature of the cantilever. Thus, denoting the outcome of this measurement S, its deviation $\Delta_e=T-S$ from a parallel measurement of the temperature of the bath T provides a measure of how far the system is from the equilibrium. Now, if equilibrium prevails initially, turning the reinjection on, for example, such that $b_1\neq 0$ and $b_2=0$, one may monitor how Δ_e varies and possibly changes sign as a result of moving the experimental parameters in the (τ,g) plane. Sign changes are expected as a result of suppression ("cooling") or amplification of oscillations. Thus, for a fix τ, and scanning g, the first measurable deviation Δ_e provides a threshold g_{th} for the system's affinity to memory. The depth of this memory may be adjusted in the interval $\tau\in[nT_0,(n+1)T_0]$, with $n=0,1,2,\ldots$. Exposing the system to the environment under investigation, adsorption events may modify this affinity, and thus shift g_{th}. It is therefore plausible to convey dynamic information akin to the adsorptive processes by also tuning τ and trace out a trajectory in the (τ,g) plane. A different analyte concentration or composition may map out different trajectories, which, upon availability, augmented by Δ_e would shed light on nonequilibrium nature of the associated processes.

III. CONCLUSION

In view of FDT, when $b_1=b_2=0$, the temperature of the cantilever may be obtained from spectral density measurements. On the other hand, in the case of feedback, the correct temperature may not be obtained from the spectral density alone. However, in the stationary state, this is not due to a violation of the FDT, and one may account for the temperature deviation by using the calculation of Eq. (7) to account for the contribution of the delay mechanism.

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