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Adsorption-desorption noise influence on mass sensitivity and dynamic range of nanoresonators with rough surfaces

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In this work we investigate the influence of adsorption-desorption noise on nanoresonators with random rough surfaces. Indeed, surface roughening leads to an increased number of adsorption sites and thus to an increased limit to mass sensitivity and decreased dynamic range leading to increased nonlinear behavior. Extensive analysis of the surface morphology is necessary because it is not only the roughness amplitude w that contributes to adsorption-desorption noise but also the lateral roughness correlation length ξ , and the roughness exponent H that characterizes short wavelength roughness ($< \xi$). The latter is shown to have a comparable effect on adsorption-desorption noise as the roughness ratio w/ξ . © 2007 American Institute of Physics. [DOI: 10.1063/1.2714792]

The exploration of nanoelectromechanical systems (NEMS) is inevitably inspired by the advancement of microelectronics technology into the submicron range.^{1–14} NEMS can attain extremely high fundamental frequencies in the gigahertz range,^{4,15} preserving very high mechanical responsivity,^{4,16} having active masses in femtograms,⁴ and heat capacities far below a yoctocalorie.¹⁷ Although this combination of attributes translates into high force sensitivity and operability at ultralow power, as the resonator size is reduced down to nanoscales the surface to volume ratio increases making nanoresonators susceptible to a wide variety of noise mechanisms.

Following the fluctuation-dissipation theorem systems that dissipate energy are necessarily sources of noise, while the converse is often true. Energy losses in NEMS arise from gas molecules impinging the resonator surface, losses due to bulk defects and impurities, losses due to the thermoelastic effects and other phonon-phonon scattering phenomena,^{18,19} and losses due to surface effects. Studies for SiC/Si NEMS have shown that devices operational in the UHF (ultra high frequency)/microwave regime were made from films that had a low surface roughness, while devices made from rougher films were operational not higher than the VHF range.²⁰ Other studies of Si nanowires have shown the quality factor to decrease by an increment of the surface area to volume ratio.⁹ Recently it was also shown that random surface roughness affects the quality factor and the limit to mass sensitivity of nanoresonators due to momentum exchange noise by impinging gas molecules.²¹ These results indicated that surface effects play a dominant role in NEMS.

Furthermore, as the system size is reduced the number of adsorption sites on the resonator surface grows in proportion to the number of total number of resonator atoms. Hence, nanoresonators are also more susceptible to adsorption-desorption noise generated by impinging gas molecules than bulk mechanical resonators. Indeed, molecular adsorption on resonator surfaces leads to mass loading and thereby changes

to its resonance frequency.^{4,5,22} As the molecules adsorb and desorb due to their finite binding energy and nonzero temperature, the changes in frequency lead to the so-called phase or fractional frequency noise. However, the adsorption-desorption cycle is not intrinsically dissipative since the arrival and departure of the atoms are random and they do not on average change the energy of the resonator (resonator quality factor remains unchanged).⁵ At any rate, the associated noise effects depend on the surface morphology, and they will be further investigated in the present work.

The spectral density of frequency fluctuations arising from adsorption-desorption processes is given by⁵

$$S_{\omega,f}(\omega) = \frac{2\pi\omega_0^2\sigma_{\text{occ}}^2\tau_r}{1 + (\omega - \omega_0)^2\tau_r^2} \left(\frac{m}{M_{\text{eff}}} \right)^2 N_a. \quad (1)$$

Here, the surface is modeled as having N_a adsorption sites, with σ_{occ}^2 representing the variance in the occupation probability of an adsorption site. τ_r is the correlation time for an adsorption-desorption cycle. σ_{occ}^2 and τ_r can be expressed in terms of the adsorption and desorption probabilities, respectively, r_a and r_d : $\sigma_{\text{occ}}^2 = r_a r_d / (r_a + r_d)^2$ and $\tau_r = 1 / (r_a + r_d)$. The adsorption and desorption probabilities are given, respectively, by $r_a = 2Ps / (5\sqrt{mk_B T})$ and $r_d = v_d \exp(-E_b/k_B T)$ with v_d desorption attempt frequency ($\sim 10^{13}$ Hz).^{4,5} P and T are the gas pressure and temperature, respectively, E_b is the binding energy of an adsorbate atom, and s is the sticking coefficient ($0 < s < 1$). In general, both r_a and r_d depend upon the temperature, the nature and preparation of the surface, and the adsorbing species.

Since the number of adsorption sites N_a is proportional to the surface area, extension to the case of a rough surface is as follows. We assume for the roughness profile a single valued random fluctuation $h(r)$ of the in-plane position $r = (x, y)$. For a Gaussian height distribution the rough surface area A_{rough} is given by^{23,24} $A_{\text{rough}}/A_{\text{flat}} = \int_0^{+\infty} (1 + \rho^2 u)^{1/2} e^{-u} du$,^{23,24} with $\rho = \sqrt{\langle (\nabla h)^2 \rangle} = [\int_0^{Q_c} q^2 C(q) dq]^2$ the average local surface slope,²⁴ and A_{flat} the average macroscopic flat contact area, and $Q_c = \pi/a_0$ with a_0 a lower roughness cutoff of the order of atomic dimensions. $C(q) = \langle |h(q)|^2 \rangle$ with $h(q)$ the Fourier transform of the surface

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FIG. 1. Calculations of the limit to mass sensitivity due to adsorption-desorption of gas molecules as a function of the long wavelength roughness ratio w/ξ , for $w=10$ nm, and various roughness exponents H . The inset shows similar calculations as a function of the correlation length ξ , for $H=0.7$, and various roughness amplitudes w (rou: rough and f: flat).

height. Therefore, the extension of Eq. (1) to rough surfaces implies the substitution $N_a \rightarrow N_a(A_{\text{rough}}/A_{\text{flat}})$. From the frequency fluctuations we obtain a frequency shift $\delta\omega_{\text{rough}} = [\int_{\omega_0-\pi\Delta f}^{\omega_0+\pi\Delta f} S_{\omega,f}(\omega)(A_{\text{rough}}/A_{\text{flat}})d\omega]^{1/2}$ with Δf the measurement bandwidth, and the corresponding limit to mass sensitivity (or minimum detectable mass) by $\delta M_{a-d,rough} \approx (2M_{\text{eff}}/\omega_0)\delta\omega_{\text{rough}}$. Integration yields for $\delta M_{a-d,rough}$,

$$\delta M_{a-d,rough} = \delta M_{a-d,flat} \left[\int_0^{+\infty} (1 + \rho^2 u)^{1/2} e^{-u} du \right]^{1/2}. \quad (2)$$

$\delta M_{a-d,flat} \approx (4\sqrt{\pi m \sigma_{oc}}) \sqrt{N_a \arctan(\pi \Delta f \tau_r)}$ is the limit to mass sensitivity for flat surfaces.⁴ Calculations of the limit to mass sensitivity by Eq. (2) require the knowledge of $\langle |h(q)|^2 \rangle$.

A wide variety of surfaces possess the so-called self-affine roughness,^{25,26} with a roughness spectrum that scales $\langle |h(q)|^2 \rangle \propto q^{-2-2H}$ if $q\xi \gg 1$ and $\langle |h(q)|^2 \rangle \propto \text{const}$ if $q\xi \ll 1$.^{25,26} This is satisfied by the analytic model²⁶ $\langle |h(q)|^2 \rangle = (2\pi w^2 \xi^2)/(1 + a q^2 \xi^2)^{(1+H)}$ with $a = (1/2H)$ [$1 - (1 + a Q_c^2 \xi^2)^{-H}$] ($0 < H < 1$), $a = 1/2 \ln(1 + a Q_c^2 \xi^2)$ ($H=0$). Small values of H (~ 0) characterize jagged or irregular surfaces, while large values of H (~ 1) refer to surfaces with smooth hills and valleys.^{25,26} For other models, see also Ref. 27

Moreover, the previous model for $\langle |h(q)|^2 \rangle$ yields for the average local slope ρ the analytic form²⁸ $\rho = (w/\sqrt{2}\xi a) \sqrt{(1-H)^{-1}[(1 + a Q_c^2 \xi^2)^{1-H} - 1] - 2a}$. For $H=0$ and 1 we obtain the correct limiting forms (by taking into account the identity $\ln(T) = \lim_{a \rightarrow 0} (1/u)(T^u - 1)$): $\rho_{(H=0)} = (w/\xi a \sqrt{2}) [a Q_c^2 \xi^2 + \ln(1 + a \xi^2 Q_c^2)]^{1/2}$ if $H=0$ and $\rho_{(H=1)} = (w/\xi \sqrt{2} a) [\ln(1 + a \xi^2 Q_c^2) - a Q_c^2 \xi^2 (1 + a \xi^2 Q_c^2)^{-1}]^{1/2}$ if $H=1$. Furthermore, in the limit of weak roughness ($\rho \ll 1$), we obtain from Eq. (2) the simplified form $\delta M_{a-d,rough} \approx \delta M_{a-d,flat}(1 + \rho^2/4)$, which yields a simple dependence on the roughness amplitude w , namely, $\delta M_{a-d,rough} \sim w^2$ since $\rho \sim w$.

Figure 1 shows calculations of the limit to mass sensitivity $\delta M_{a-d,rough}$ as a function of the roughness ratio w/ξ for

various roughness exponents H . It is evident that $\delta M_{a-d,rough}$ increases with respect to that of a flat surface ($\delta M_{a-d,flat}$),⁵ with increasing surface roughness or equivalently decreasing roughness exponent H and/or increasing the long wavelength roughness ratio w/ξ . Furthermore, the inset depicts the influence of the roughness amplitude w for various correlation lengths ξ , because in many studies only the use of the roughness amplitude is considered to describe roughening. In fact, as the inset of Fig. 1 shows the influence of the roughness amplitude on $\delta M_{a-d,rough}$ is significant over a wide range of correlation lengths ξ . At any rate, comparison of the curves in Fig. 1 indicates that the influence of the exponent H is equally important as that of the ratio w/ξ .

Although the $\delta M_{a-d,rough}$ increases with surface roughening because of the increasing surface sites, the adsorption-desorption on a corrugated surface is far more complex. Indeed, the dominant effect of surface corrugation for molecules impinging under normal incidence on a surface is a reduction of the sticking coefficient.²⁹ However, for non-normal incidence the role of surface corrugation is more complex. The additional parallel momentum can lead to an enhancement or a suppression of the sticking coefficient depending on the type of corrugation and the energy regime.^{29,30} Moreover, variations of the binding energy on a rough surface should also be considered.

Besides the limit to mass sensitivity, adsorption-desorption noise influences also the linear dynamic range (DR). The latter is a widely used concept in amplifier characterization, expressing the window of input power in which the amplifier behaves linearly.^{4,12} The DR for a nanoresonator is defined as the ratio of its maximum vibration amplitude $\langle x_c \rangle$ at the onset of nonlinearity to its rms displacement noise floor within the operation bandwidth Δf .^{4,12} The criterion at the onset of nonlinearity is the level at which the largest term beyond the quadratic (Hooke's law) grows to become 10% the size of the second order term.^{4,12} Therefore, DR is given by $DR_{a-d,rough} = 10 \log[\langle x_c \rangle^2 / \int S_{a-d,rough}(\omega) d\omega]$, where the roughness correction yields

$$DR_{a-d,rough} = DR_{a-d,flat} - 10 \log \left[\int_0^{+\infty} (1 + \rho^2 u)^{1/2} e^{-u} du \right] \quad (3)$$

with $DR_{a-d,flat} = 10 \log[\langle x_c \rangle^2 / \int S_{a-d,flat}(\omega) d\omega]$,^{4,12} the dynamic range for flat surfaces including only the adsorption-desorption noise effects. Although a complete treatment should include thermomechanical noise, momentum exchange, and in more general of any contributing noise effects through additional terms in the spectral density $S(\omega)$ appearing in the denominator of $DR \propto \log[\langle x_c \rangle^2 / \int S(\omega) d\omega]$, at the present work we would like to show solely the influence of the adsorption-desorption noise. For weak roughness ($\rho \ll 1$), Eq. (3) yields the simpler analytic expression $DR_{a-d,rough} \approx DR_{a-d,flat} - 10 \log(1 + \rho^2/2)$. Figure 2 shows detailed calculations of DR as a function of roughness parameters. As Fig. 2 shows increasing surface roughness (decreasing H and/or increasing w/ξ) significantly decreases the dynamic range leading to increased nonlinear behavior. The latter, on the other hand, is not only desirable in certain ap-

FIG. 2. Calculations of the DR as a function of the long wavelength roughness ratio w/ξ , for $w=10$ nm, and various roughness exponents H . The inset shows similar calculations as a function of the correlation length ξ , for $H=0.7$, and various roughness amplitudes w . The plots are shown DR on the natural logarithm scale (rou: rough and f: flat).

plications such as in signal processing,³¹ but also translates into interesting physics.³²

In conclusion, it was shown that surface roughening in NEMS leads to an increasing number of adsorption sites and thus to an increasing limit to mass sensitivity and decreasing dynamic range. The present calculations illustrate that careful analysis of the surface morphology is needed. This is because it is not only the roughness amplitude that contributes to adsorption-desorption noise, but also the lateral correlation length and the roughness exponent H that characterizes short wavelength roughness at length scales. The chosen roughness amplitudes are close to values found in real resonators as, for example, in SiC/Si NEMS with $w=7.1$ nm operating in the VHF range.²⁰ Although for H and ξ there are not yet systematic studies still available in NEMS, we used values that were observed in a variety of Si based experimental systems.^{25,27} Nonetheless, further studies will be required to account effects arising due to morphology dependence of the sticking probability and the residence time (through the binding energy) of the adsorbed molecules on the resonator surface.

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