Parametric resonance based scanning probe microscopy

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We propose a mode of dynamic scanning probe microscopy based on parametric resonance for highly sensitive nanoscale imaging and force spectroscopy. In this mode the microcantilever probe is excited by means of a closed-loop electronic circuit that modulates the microcantilever stiffness at a frequency close to twice its natural resonance frequency. Under ambient conditions this parametric pumping leads to self-sustained oscillations in a narrow frequency bandwidth thereby resulting in exquisitely sharp, controllable, and non-Lorentzian resonance peaks. We discuss and demonstrate the potential of imaging and force spectroscopy using this mode. © *2006 American Institute of Physics.* [DOI: 10.1063/1.2202132]

Dynamic scanning probe microscopy (SPM) has become a cornerstone technique for nanoscale imaging and force spectroscopy.[1,2] The method relies on controlling the amplitude, resonance frequency, or phase of a microcantilever probe as it is scanned over a sample. However, under ambient conditions the sensitivity and the minimum force exerted on the sample are fundamentally limited by the low quality factor (Q factor) of the SPM microcantilever.

In an effort to overcome these fundamental challenges, researchers have investigated the so-called "Q-control" methods[3–6] to enhance the Q factor of microcantilevers by means of electronic feedback circuits. The benefits of this approach have been debated extensively in the literature, however, it is generally agreed upon that enhancing the resonance sharpness by Q control reduces greatly the scanning speed[3] because of the inherent transients or "ringing" effects. There is also evidence[6] that the use of Q control increases the background noise in the system.

In this letter we propose the use of parametric resonance via a feedback circuit as a means to enhance the sensitivity in dynamic SPM without incurring some of the disadvantages of Q control. Parametric resonance is a phenomenon that underlies the physics of swings[7] and of the Botafumeiro censer in the cathedral of Santiago de Compostela in Spain,[8] and the creation of surface wave patterns in liquids.[9] More recently, parametric resonances have been realized in micro- and nanoelectromechanical systems.[10–12]

In the context of SPM, the use of parametric excitation has been proposed as a mechanical preamplifier to sharpen the resonance peaks of microcantilevers.[13–15] However, in contrast to the present work, the strength of parametric excitation was kept below that required for self-sustained oscillations. More recently self-sustained parametric resonances

in electrostatic force microscopy (EFM) have been investigated theoretically.[16] The use of parametric resonance in noncontact SPM has also been proposed via periodic excitation of the sample.[17]

Our implementation of parametric resonance in SPM is a simple and yet general protocol that can be used in ambient, vacuum, or liquid environments. The cantilever deflection signal from the photodiode[18] is passed through a high pass filter (with cutoff frequency $\sim$100 Hz) to isolate the ac component. As seen in Fig. 1, this is then amplified via a variable gain amplifier and multiplied with a periodic driving signal whose variable frequency ω is close to twice the natural resonance frequency ($2\omega_0$) of the microcantilever. The resulting signal is simply fed back to the dither piezo. Because the photodiode signal is not differentiated or integrated, this feedback circuit is extremely fast with minimal delay between measurement and actuation.

To understand why this circuit leads to a parametric resonance of the cantilever, consider a simple single degree-of-freedom model of the cantilever dynamics. Let m be the modal mass of the cantilever and tip, k be the modal stiffness or cantilever spring constant, and c be the velocity proportional modal damping coefficient representing linear hydrodynamic losses. Let $z(t)$ be the base motion of the dither piezo, $x(t)$ the instantaneous displacement of the tip, both quantities being measured in an inertial reference frame, and d_0 the equilibrium gap between the tip and the fixed sample. Because the photodiode detects the bending of the cantilever $x(t)-z(t)$ (plus an arbitrary dc component that is removed by the high pass filter of our circuit), the dither piezo motion must obey $z(t)=G[x(t)-z(t)]\cos(\omega t)$, where G is the overall gain of the optical measurement system, the feedback loop shown in Fig. 1, and the dither piezo actuator. In experiments G is usually changed conveniently by changing the feedback
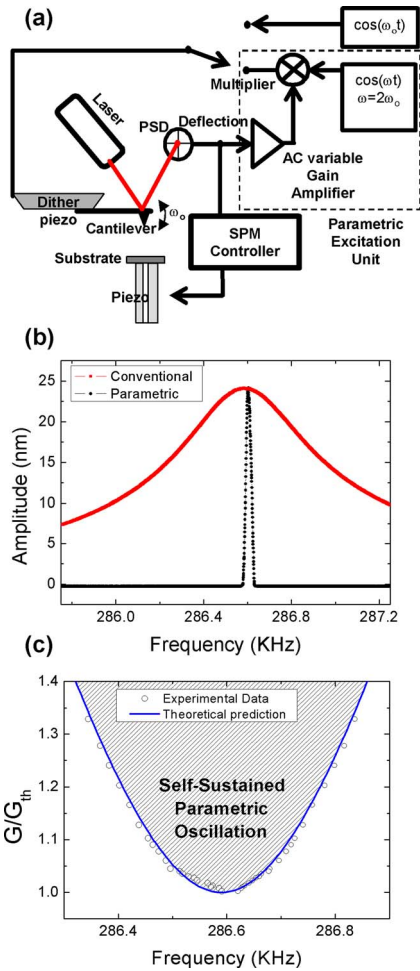
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FIG. 1. (Color online) (a) A schematic showing the conventional SPM and the parametric resonance unit required to implement parametric resonance in SPM. (b) Comparison of the frequency response of conventional (solid squares) vs parametric resonance (solid dots). The magnitude of conventional excitation has been adjusted so that the cantilever amplitude at resonance is comparable to that obtained with parametric resonance. For the case of parametric resonance (solid dots), the actual excitation frequency (ω) is twice the frequency at which each data point is plotted, (c) The instability tongue in the feedback gain-excitation frequency domain. The experimental data are in circles, while the solid line is the theoretical prediction using Eq. (3) and using a measured Q factor of 522. The instability region corresponds to gain and frequency combinations that lead to self-sustained parametric oscillations.

gain. G is the strength of the parametric excitation. If $G \ll 1$, the above condition can be rewritten as

$$z(t) = \frac{Gx(t)\cos(\omega t)}{1 + G\cos(\omega t)} \sim Gx(t)\cos(\omega t) + O(G^2). \quad (1)$$

Using (1) the equations of motion for the tip can be written in the following form:

$$\ddot{x}(t) + \omega_0^2 x(t)[1 - G\cos(\omega t)] + \frac{\omega_0}{Q}\dot{x}(t)$$

$$= \frac{1}{m}[F_{\text{cantilever}}(x,\dot{x}) + F_{\text{fluidic}}(x,\dot{x}) + F_{\text{tip-sample}}(x,\dot{x},d_0)], \quad (2)$$

where $\omega \sim 2\omega_0$ and $F_{\text{cantilever}}(x,\dot{x})$, $F_{\text{fluidic}}(x,\dot{x})$, and $F_{\text{tip-sample}}(x,\dot{x},d_0)$ represent, respectively, the intrinsic geometric and inertial nonlinearities of the microcantilever, the dissipative nonlinear fluid forces, and the tip-sample nonlin-

ear forces. Further $\omega_0 = \sqrt{k/m}$ is the natural frequency of the microcantilever, and $Q = m\omega_0/c$ is the natural quality factor of the resonance in the absence of the feedback circuit. Setting the right-hand side of (3) to zero yields the damped Mathieu's equation.[19–22]

Accordingly if the feedback gain is greater than a threshold value and the conditions for principal parametric resonance exist ($\omega/2 \sim \omega_0$), then sustained periodic oscillations at frequency $\omega/2$ occur over a small bandwidth of excitation frequencies. Within the bandwidth of parametric resonance a balance between the gain and the nonlinear terms in Eq. (1) determines the amplitude and phase of self-sustained oscillations. The boundaries in $G-\omega$ parameter space that demarcate the transition between zero amplitude and finite amplitude self-sustained oscillations are known as the instability tongues and can be approximated using perturbation methods. Assuming $\varepsilon = G(\omega_0^2/\omega^2) \ll 1$ and small damping $1/Q = O(\varepsilon)$ it can be shown[23] that the width of the instability tongue for principal parametric resonance is given by

$$\left(\frac{\omega}{\omega_0}\right)^2 = 4 \pm 2\sqrt{G^2 - \frac{4}{Q^2}} + O(\varepsilon^2). \quad (3)$$

Finally it can be shown[24] through a nonlinear analysis of the equations of motion (2) with specific forms of nonlinearities, that if the steady state oscillations at parametric resonance are perturbed slightly, the transients decay away with the time constant $2Q/\omega_0$ of the original system. In other words the transient response about the steady oscillation state is not compromised as long as the perturbation is small.

Parametric resonance in SPM was implanted using a Nanotec Electrónica SPM system modified as shown in Fig. 1(a). In these initial studies, two different cantilevers were investigated: cantilever 1 (Olympus Inc. $k \sim 0.75$ N/m, $\omega_0 \sim 80$ kHz) and cantilever 2 (Nanosensors Gmbh., $k \sim 40$ N/m, $\omega_0 \sim 286$ kHz). In what follows, we report the results obtained from the cantilever 2 under ambient conditions although similar results were also obtained for the cantilever 1.

In Fig. 1(b) we compare the parametric resonance curve with that using conventional excitation. The conventional resonance is at $\omega_0 \sim 285.75$ kHz with $Q \sim 520$. To achieve parametric resonance, the excitation frequency is tuned so that $\omega = 2\omega_0$ and the feedback gain increased gradually until a significant cantilever response is observed at half the driving frequency. The threshold value for G can be determined in this way.[25] Then G is fixed and the excitation frequency is gradually increased across the principal parametric resonance and the cantilever response is recorded. The cantilever response curve is extremely sharp and is nearly zero outside a 50 Hz bandwidth. Moreover, closer observation shows that the resonance peak is asymmetric and slightly bent towards lower frequencies.[26,27]

The bandwidth of parametric resonance and the maximum amplitude in the resonance bandwidth can be controlled precisely by varying the feedback gain. This is shown in Fig. 1(c) where beyond a threshold feedback gain, the bandwidth becomes progressively broader with increasing feedback gain. For comparison, the theoretical frequency range over which parametric oscillation is sustained [Eq. (3)] is plotted against the experimental linewidths and found to be in excellent agreement.
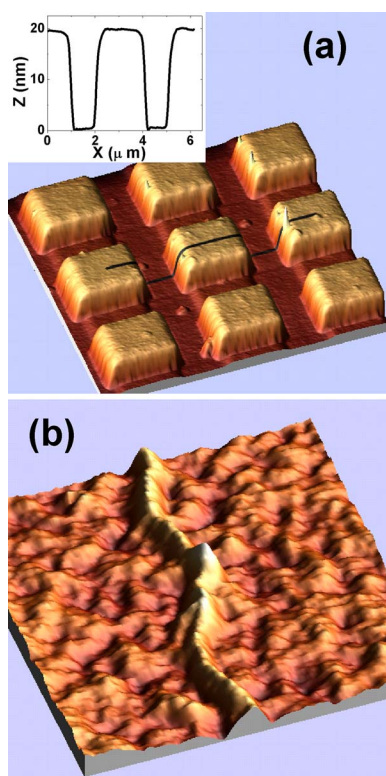
FIG. 2. (Color online) (a) Image of a silicon grating (see inset for details), and (b) image of a DNA strand on mica (image size 700×700 nm^2) taken with the parametrically excited microcantilever.

Several amplitude versus distance experiments were performed to investigate the behavior of the parametrically resonant probe as it approaches a sample surface. Based on these experiments we have observed that (a) when the feedback gain is kept just above the threshold value and the oscillation amplitude is very small, the probe oscillations are extremely sensitive to long-range tip-sample interactions such as those due to surface potentials. (b) At larger feedback gain and larger oscillation amplitudes, the decrease in oscillation amplitude upon approach to the sample is similar to that of the conventional tapping mode, and (c) by meticulous adjustment of operating parameters the slope of the amplitude-distance curve for the parametrically excited case can be made steeper than for the conventional case.

The exquisite sensitivity of the parametric resonance peak raises the following question: if it is possible to approach the sample in a controlled manner and image stably at the nanoscale? To demonstrate this we provide two images acquired under ambient conditions using the cantilever above. Specifically in Fig. 2(a) an image of a silicon grid is shown (each feature is of size ~ 1.5 μm $\times 1.5$ μm $\times 19.5$ nm), while in Fig. 2(b) an image of a DNA strand on a mica substrate is shown. For imaging the silicon grating, the unconstrained vibration amplitude was set to ~ 60 nm while the setpoint amplitude was ~ 20 nm. For the DNA on mica image, the unconstrained vibration amplitude was ~ 200 nm while the setpoint amplitude was ~ 60 nm. In contrast with the ringing artifacts typical in high Q systems, these images are clean even though the images are taken at normal scanning speeds (~ 1.5 Hz).

In conclusion, we have demonstrated a mode of dynamic SPM based on parametric resonance that has the potential to lead to highly sensitive nanoscale imaging and force spectroscopies especially for applications including electric force microscopy, magnetic force microscopy, and for SPM in liquid environments. Given the tunable sharpness of the parametric resonance peak at gains just above threshold, any shift in resonance frequency due to tip-sample interaction will produce a corresponding change in the cantilever's amplitude with sensitivity comparable to that found in UHV conditions under conventional excitation.

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[25] The threshold feedback gain typically changes when the same cantilever is remounted. This is because the optical feedback gain changes each time the laser is adjusted.
[26] This asymmetric resonance peak is typical in parametric resonance and is a consequence of the cantilever and fluidic nonlinearities. For cantilevers oscillating in their first bending mode it is known that the curvature and inertial nonlinearities possess opposite signs,[27] thereby reducing the overall effect of cantilever nonlinearities. The presence of fluidic nonlinearities also serves to minimize the asymmetry of the parametric resonance peak.
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