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Pulsed contact resonance for atomic force microscopy nanomechanical measurements

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We demonstrate an improved technique for nanomechanical imaging in atomic force microscopy. By merging the sensitivity to contact stiffness inherent to contact resonance (CR) spectroscopy with the delicate nature and potential for adhesion data of pulsed-force mode, we address major shortcomings of both techniques. Fast CR data are recorded during each pulsed cycle by driving the sample at two frequencies near the CR frequency and modeling the contact as a harmonic oscillator. The technique provides nanomechanical parameters including frequency, quality factor, and adhesion force. Compared to continuous contact, the technique should reduce damage and support more complex analysis models. [doi:10.1063/1.3680212]

The resonance response of an atomic force microscopy (AFM) cantilever in repulsive contact with a substrate can provide nanomechanical information about the material. In contact resonance (CR) spectroscopy,1,2 the cantilever is brought in contact with a sample of interest, and then either the sample or the cantilever is vibrated to determine the amplitude versus frequency response of the cantilever. By comparing the resonance frequency \( f_{\text{CR}} \) of a cantilever in contact to the corresponding frequency \( f_0 \) in free space and applying a suitable beam mechanics model, the tip-sample contact stiffness can be determined.3 From contact stiffness, a contact mechanics model can be used to determine material properties such as elastic modulus. CR techniques afford excellent sensitivity to contact stiffness in comparison to force-distance spectroscopy approaches, which may be hindered by the limited deflection sensitivity of a given cantilever.3,4 Furthermore, by measuring the quality factor \( Q \) of the contact resonance, viscoelastic properties (e.g., loss modulus, storage modulus, loss tangent) can be determined.5,6 Although originally a tool for point spectroscopy, CR techniques have also been adapted for continuous scanning in contact mode.7–11

However, scanning with contact-mode feedback is difficult, and in some cases, impossible. The lateral forces in contact-mode scanning can be sufficiently large to damage the tip and sample, thus limiting resolution and preventing application to delicate samples. Continuous contact also precludes the measurement of local adhesion forces, prohibiting analysis with more complex contact models such as Derjaguin-Müller-Toporov and Johnson-Kendall-Roberts.12

For some other AFM modes including force modulation microscopy,13 fixed-frequency ultrasonic AFM,14 and conductive AFM,15 shortcomings of contact-mode scanning have been addressed by integrating the techniques into pulsed-force16 and peak-force17 AFM modalities, where a sufficiently large amplitude oscillation breaks the tip-sample contact during each cycle. In pulsed-force and peak-force modes, the drive frequency \( f_{\text{pulse}} \) is sufficiently fast (~100 Hz to ~2 kHz) that durations of contact, and thus destructive lateral forces, are minimized. By monitoring the response of the cantilever deflection at \( f_{\text{pulse}} \), it is possible to measure adhesion force, contact stiffness, electrostatic interactions, and dissipation in the contact.16 A drawback of most pulsed-force and peak-force measurements is that contact stiffness analysis relies on the deflection sensitivity of the cantilever. To characterize materials with high elastic modulus, tip-sample deformations large enough to resolve in the cantilever deflection signal often require high enough forces to cause reduced spatial resolution and permanent damage to the tip and sample.

Marrying the delicate nature and potential for adhesion data of pulsed-force mode with the sensitivity to contact stiffness and damping of CR spectroscopy could provide an improved tool for nanomechanical characterization. However, existing CR spectrum acquisition techniques are too slow to acquire sufficient data over force cycles of such short duration.8,10,18 Multifrequency techniques provide a possible solution. Instead of acquiring a complete resonance spectrum, the cantilever is driven at a limited number of fixed19 or variable7 frequencies near the resonance peak. The amplitude and phase of the cantilever response at each frequency are detected with lock-in amplifiers and then used together to reconstruct the resonance response with use of a damped simple harmonic oscillator (DSHO) model. Such approaches have been demonstrated for tapping mode operation19 and scanning CR operation in contact mode.9,13 but not for CR measurements of \( f_{\text{CR}} \) and \( Q \) during a short pulsed-force cycle where the speed requirements are especially stringent.

In this Letter, we introduce a hybrid method dubbed pulsed contact resonance (pCR) for improved quantitative AFM nanomechanical measurements. The method uses a multifrequency technique to calculate \( f_{\text{CR}} \) and \( Q \) during the repulsive contact segment of the pulsed force cycle. After describing the experimental setup and data analysis, we present experimental results with pCR for a polymer film and an adjacent exposed silicon substrate. The data clearly reveal the nanomechanical contrast between the two materials, demonstrating the potential of this approach.

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As shown in Fig. 1(a), the experimental setup represents a hybrid of existing pulsed-force and multifrequency techniques. A function generator drives the z-piezo of the AFM instrument at a frequency $f_{\text{pulse}}$ and amplitude $Z$. For these proof-of-concept experiments, we used $f_{\text{pulse}} = 100$ Hz and $Z \approx 600$ nm; however, a range of frequencies and amplitudes are possible depending on cantilever bandwidth and resonances in the AFM instrument head. Two additional function generators drive a piezoelectric actuator beneath the sample at fixed frequencies $f_{\text{LI1}}$ and $f_{\text{LI2}}$, which are chosen to coincide with the range of contact resonance frequencies $f_{\text{CR}}$ expected for the specimen. The cantilever vibration amplitude and phase at $f_{\text{LI1}}$ and $f_{\text{LI2}}$ are detected by two lock-in amplifiers. Here, the lock-in acquisition rate was 25.0 kHz, but it could be increased or decreased depending on capabilities of the electronics. For these experiments, we used a rectangular silicon contact-mode cantilever with spring constant $k_1 = (0.14 \pm 0.02)$ N/m determined from the thermal method\cite{20} and free resonance frequencies of $f_1^{0} = (11.2 \pm 0.01)$ kHz and $f_2^{0} = (70.3 \pm 0.01)$ kHz for the first and second flexural eigenmodes, respectively. Compared to the first (lowest) eigenmode, the second mode is expected to provide increased frequency shifts for detecting contact stiffness and damping contrast between the materials for these experimental conditions.\cite{21} Still, the frequency shift must remain small enough to ensure sufficient amplitudes at both lock-in frequencies throughout most of the repulsive force cycle (i.e., the peaks should not shift so far that the cantilever response is indistinguishable from the noise floor). For these reasons, the second flexural eigenmode was excited during our proof-of-concept experiments. All phase, frequency, and quality factor data for $A_1 < 0.5$ mV were discarded due to spurious phase signals that occurred when the cantilever was out of contact.

The test sample was a (001) silicon (Si) wafer partially covered with a film of epoxy photoresist material $\sim 2 \mu$m thick. Figure 1(b) shows conventional CR amplitude versus frequency spectra for single-point measurements acquired on the epoxy and Si portions of the sample. It can be seen that the CR frequencies are in the range of 165 kHz to 175 kHz. As expected from modulus values for the bulk materials, the Si region exhibits higher CR frequencies than epoxy. From the point spectra in Fig. 1(b), $f_{\text{LI1}}$ and $f_{\text{LI2}}$ for pCR were set to 159 kHz and 172 kHz to sufficiently optimize amplitude, signal-to-noise ratio, and stability at the same time.

pCR measurements were initially made separately on epoxy and Si regions with both fast and slow scanning dis-
indicating higher elastic stiffness and lower damping in the Si region compared to epoxy. The CR frequency increases with deflection force $F$ on both regions, as expected for a conical or hemispherical indenter whose contact area increases as a function of load.\textsuperscript{12} The ability to resolve the frequency-versus-load dependence during the short pulsed cycle highlights the temporal resolution of the technique.

Additional experiments were performed to demonstrate the utility of pCR techniques during scanning. Because we had not yet implemented topographic pulsed-force feedback, we improvised pCR imaging using the AFM lift or interleaf mode.\textsuperscript{22} In this manner, we acquired the topography of the sample in the trace direction with the pulsed force drive disabled and either AC or contact feedback. The z-piezo’s average position was then raised 100 nm from the surface, and the pulsed-force drive was enabled for the retrace direction. With the interleaf method, the maximum normal applied force during pCR operation ranged between $(34.2 \pm 4.9) \text{nN}$ and $(38.2 \pm 5.5) \text{nN}$. Figure 3(a) shows a topographic scan of the sample and the corresponding line traces of $f_{\text{CR}}$, Q, adhesion force $F_{\text{adh}}$, and force-distance slope $dF/dZ$. Because we capture data for the entire force cycle, $f_{\text{CR}}$ and $Q$ can be calculated for any instantaneous force. The $f_{\text{CR}}$ and $Q$ data shown in Fig. 3 were extracted from each pCR cycle when the applied force was $(28 \pm 0.1) \text{nN}$ and only during the approach portion of the cycle. Figures 3(b) and 3(c) show the strong contrast in CR frequency and quality factor between the two materials, with $f_{\text{CR}}$(epoxy) = $(167.9 \pm 0.5) \text{kHz}$ and $Q$(epoxy) = $48.2 \pm 9.8$, and $f_{\text{CR}}$(Si) = $(170.3 \pm 0.2) \text{kHz}$ and $Q$(Si) = $88.9 \pm 9.0$. Similar to the stationary measurements in Fig. 2, $f_{\text{CR}}$ and $Q$ are higher on Si than on epoxy. From Fig. 3(d), it can be seen that compared to $f_{\text{CR}}$ and $Q$ there is less contrast between materials in the average adhesion force, with $F_{\text{adh}}$(epoxy) = $(8.5 \pm 0.5) \text{nN}$ and $F_{\text{adh}}$(Si) = $(8.9 \pm 0.9) \text{nN}$.
Finally, Fig. 3(e) shows the slope $dF/dZ$ of the force-distance curve extracted in the conventional pulsed-force manner. Like the CR frequency in Fig. 3(b), $dF/dZ$ should be monotonically correlated to the contact stiffness of the sample. However, the average slopes obtained by analysis of the retraction portion of the pulsed force cycle are

$$
\frac{dF}{dZ} \text{(epoxy)} = (49.6 \pm 0.8) \text{nN/\mu m}
$$

and

$$
\frac{dF}{dZ} \text{(Si)} = (48.3 \pm 0.6) \text{nN/\mu m},
$$

incorrectly indicating that epoxy is elastically stiffer than Si. Furthermore, compared to the CR frequency the pulsed-force slope shows much less contrast between the two materials (i.e., the standard deviations overlap). The failure of the $dF/dZ$ signal to provide meaningful contrast is attributed to the limited sensitivity of such a compliant cantilever over the contact stiffness range of the sample. Thus, the pCR frequency signals are seen to provide improved measurement sensitivity compared to quasistatic deflection for the same experimental conditions.

In summary, we have demonstrated contact resonance imaging of frequency and quality factor during pulsed-force AFM mode. Capturing pCR data in addition to conventional pulsed-force signals achieves improved stiffness sensitivity and enables use of more sophisticated contact mechanics models for CR data analysis. With pCR, we could clearly distinguish elastically dissimilar materials, even though the conventional pulsed-force results were inconclusive. These results demonstrate an improved technique for quantitative nanomechanical characterization of fragile materials and materials with substantial adhesion. Although the range of detectable frequencies may initially appear somewhat limited, it could be significantly expanded by use of additional drive frequencies and lock-in detectors. The technique is also expected to be readily applicable to peak-force tapping. The benefits of pCR show additional promise for piezoresponse force microscopy, where contact resonance methods are used to increase sensitivity to piezoelectric displacement and provide dissipation information.

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