Nonlinear mechanical resonators for ultra-sensitive mass detection

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ABSTRACT

The fundamental sensitivity limit of an appropriately scaled down mechanical resonator can approach one atomic mass unit when only thermal noise is present in the system. However, operation of such nanoscale mechanical resonators is very challenging due to minuteness of their oscillation amplitudes and presence of multiple noise sources in real experimental environments. In order to surmount these challenges, we use microscale cantilever resonators driven to large amplitudes, far beyond their nonlinear instability onset. Our experiments show that such a nonlinear cantilever resonator, described analytically as a Duffing oscillator, has mass sensing performance comparable to that of much smaller resonators operating in a linear regime. We demonstrate femtogram level mass sensing that relies on a bifurcation point tracking that does not require any complex readout means. Our approaches enable straightforward detection of mass changes that are near the fundamental limit imposed by thermo-mechanical fluctuations.

Keywords: Mechanical nanoresonators, nonlinear oscillators, bifurcation point, mass sensing

1. INTRODUCTION

Over the last decade nanoscale and microscale resonators similar to microcantilevers used in atomic force microscopy have been successfully explored as mass and force sensing devices with unprecedented sensitivity. [1-5] The fundamental limit of performance for an appropriately designed mechanical oscillator can approach one atomic mass unit provided that only thermal noise is present in the system. [4, 6-9] Such predictions are based on evaluations of a classical model of a weakly damped mechanical oscillator in a thermal bath, which predicts a decrease in the resonance frequency as a result of mass loading while thermomechanical noise [6, 8-10] imposes an ultimate limit on the accuracy of resonance frequency measurements. However, experimentally achievable performance of nanomechanical oscillators is often compromised by multiple sources of noise in addition to the thermal noise. [8, 9, 11] In particular, a non-ideal readout may decrease experimentally observed signal-to-noise ratios by orders of magnitude.

A previously established trend in improving mass sensing performance of mechanical resonators is to decrease their mass, increase their frequency and maximize their mechanical quality factor (Q-factor). For instance, in order to detect femtogram level mass changes, devices with characteristic lengths of just a few micrometers and resonance frequencies in the MHz range are typically required. [7-9, 12] Operation of high frequency nanoscale mechanical oscillators and their mass sensing performance in the attogram [13] and even zeptogram range has been demonstrated more recently [4, 14] Although remarkable improvements in nanomechanical mass sensing spanning five orders of magnitude have been demonstrated over the last decade, these measurements involve significant experimental challenges. [3, 4, 14] Indeed, majority of the previous studies in this area have used cantilevers operating in the linear regime, which typically corresponds to oscillations amplitudes less than 10% of the cantilever length, *l*. Therefore, measurements and analysis of oscillation amplitudes as small as 10^{-9} m are typically required. Such measurements rely on the use of complex low-noise electronic and sophisticated optical components.

In our present efforts we seek to overcome these experimental challenges of mass sensitive nano- and micro-resonators by driving them to large amplitudes and operating them in a strongly nonlinear regime. The mass sensing performance of a linear mechanical oscillator limited by its thermal fluctuations can be expressed as a function of the oscillator parameters [6, 15]

$$\mathrm{D} m_{th} = \left(k_B T B\right)^{1/2} \frac{8G}{A} \frac{m_0^{5/4}}{k^{3/4} Q^{1/2}} \qquad (1)$$

where k_B is the Boltzmann constant, T is the temperature of the device, B is the bandwidth of the measurement, G is a geometric factor (close to unity for rectangular cantilevers), k is the force constant, Q is the mechanical quality factor, m_0 is the suspended oscillating mass, and A is the rms amplitude of the oscillator motion. One unambiguous conclusion that follows from Equation (1) is that large oscillation amplitudes and a high mechanical quality factor, Q, i.e. low dissipation are important. Multiple damping mechanisms present in mechanical resonators determine typical Q-factors to be $< 10^3$ for structures operated under the ambient conditions while Q-factors close to 10^6 can only be achieved using certain types of mechanical resonating structures in vacuum. On the other hand, there are no obvious fundamental limitations imposed on the oscillation amplitude until it becomes comparable to the total resonator size.

Therefore, our key hypothesis is that very large oscillation amplitudes of cantilever resonators can significantly improve their frequency stability by minimizing dephasing effects of the thermal noise while additional advantages can be gained due to the nonlinear dynamics. When mechanical oscillators are driven to progressively larger amplitudes they inevitably exhibit a nonlinearity onset due to the intrinsic nonlinear elasticity of materials, geometrical nonlinearity, nonlinearity of external non-mechanical components or a combination of these factors. [12, 16-19] Initially, the nonlinearity manifests itself as an asymmetrically shaped resonance curve indicative of effective softening or stiffening of a spring. At larger amplitudes, an onset of the nonlinear instability is typically observed. In particular, a bifurcation point as well as a hysteretic behavior characteristic of a Duffing oscillator appears above certain oscillation amplitudes as a result of the strongly nonlinear regime. [18, 20] Aldridge *et al.* evaluated the nonlinear response of a radio frequency mechanical resonator and concluded that their nonlinear operation can lead to a 100-fold improvement in precision of frequency measurements with respective implications in mass sensing. [21] Nonetheless, no mass sensing device that realizes these advantages has been reported so far. It is important to emphasize that, above the nonlinear instability onset, mass and force loading of nonlinear resonators cannot be analyzed using methodology developed for linear resonators. In particular, mass loading of a nonlinear resonator cannot be determined by fitting the resonance curve to Lorentzian or measuring the output frequency of a self-oscillating circuitry based on a phase-locked loop. However, in this work we demonstrate that nonlinear dynamics combined with large oscillation amplitudes facilitates highly accurate mass loading analysis of a cantilever resonator.

2. BIFURCATION POINT TRACKING

Center the paper title at the top of the page in 16-pt. bold. Only the first word, proper nouns, and acronyms are capitalized. Keep titles brief and descriptive. Spell out acronyms unless they are widely known. Avoid starting with articles or prepositions, e.g., “The study of … ,” or, “On the ….”

Our experimental approach to analysis of mass loading of a cantilever resonator operating in a strongly nonlinear regime is based on a bifurcation point tracking. The dynamics of a nonlinear resonator can be described by the Duffing equation [12]

$$\frac{d^2x}{dt^2} + \delta\frac{dx}{dt} + \omega_0^2 x + \beta x^3 = \gamma F\cos(\omega t + \varphi) \tag{2}$$

where x is the displacement (amplitude), ω is the angular frequency, φ is the phase shift, F is the driving force, and β, γ and δ, and are parameters related to, respectively, the resonator mass, elasticity and losses. Solutions to Equation (2) plotted in **Figure 1** are analogous to the experimentally measured resonance curves for nanomechanical and micromechanical resonators driven to large amplitudes. [12, 16-19]

Equation 2 predicts a bifurcation point when oscillation amplitudes are above a certain value. Notably, the oscillation amplitude abruptly collapses at the bifurcation point upon forward frequency sweeping (dotted arrow in **Figure 1**). As we show below, the frequency at which this extremely abrupt amplitude decrease occurs can be tracked by facile means with high accuracy in order to provide a measure of mass or force loading of the resonating structure.

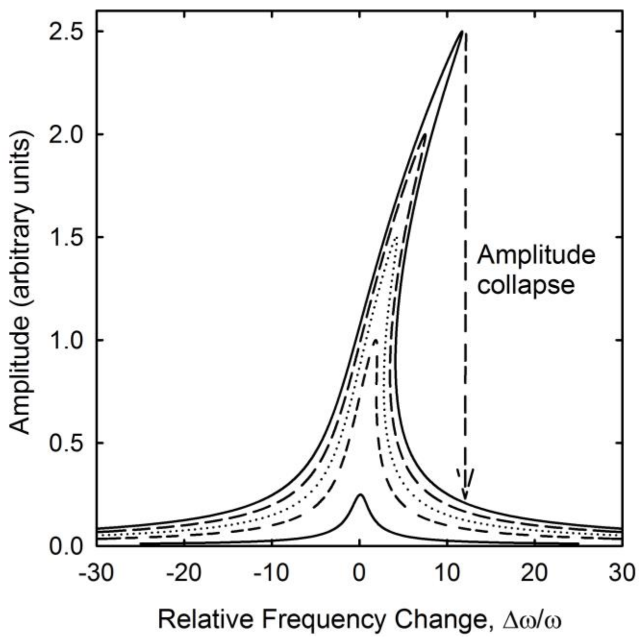

Figure 1. Frequency responses of a mechanical resonator driven with progressively large amplitudes. A nearly symmetric bell shaped response (black line) observed at small driving amplitudes is characteristic of a linear regime. Asymmetrically shaped frequency response curves are observed at larger driving amplitudes. At the highest driving force, and amplitude collapse (indicated dashed red arrow) is observed upon forward frequency sweeping.

3. RESULTS AND DISCUSSION

In this work we primarily used cantilevers with 130 μm in length, and 30 μm in width. **Figure 2** shows images [optical 3(a) and SEM 3(b)] of the chip with the cantilever resonators, which was mounted on a piezoelectric transducer. The experimental setup used in the present studies is shown in **Figure 3**.

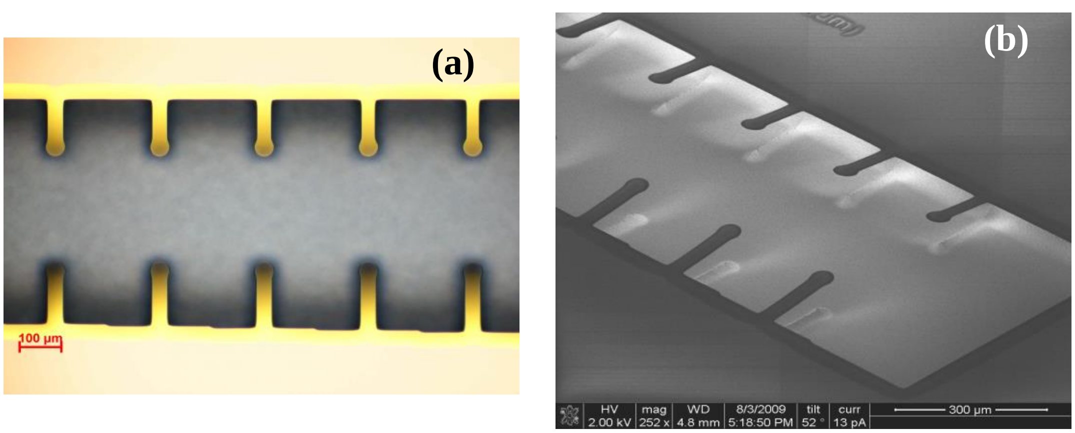

Figure 2. (a) Optical microscopy and (b) SEM image of cantilever array structures used in our present studies.

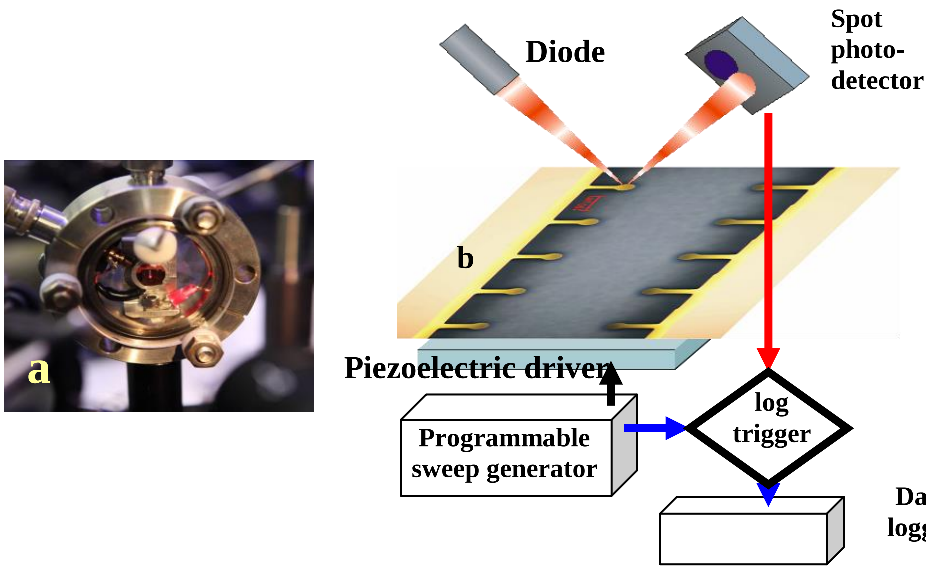

Figure 3. (a) The flow cell housing the nonlinear resonator and (b) a schematic diagram of tracking frequency shifts in bifurcation point.

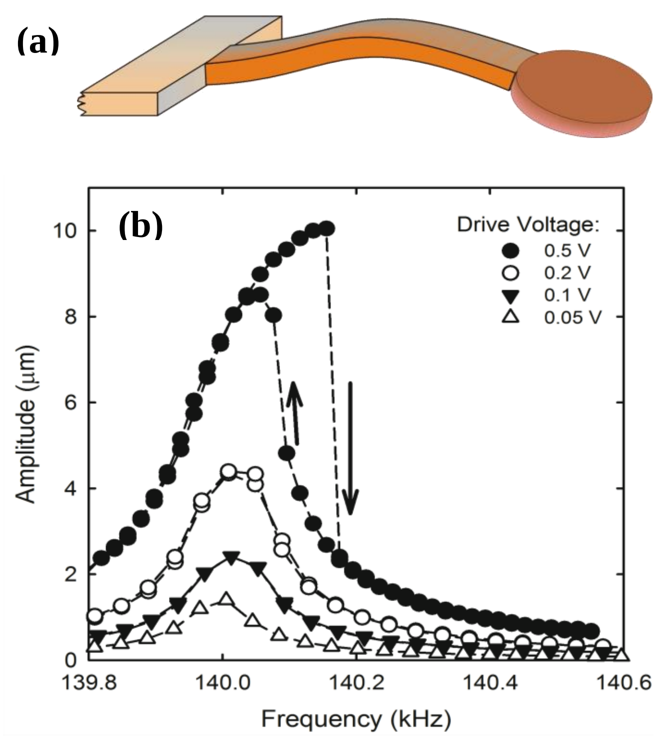

Figure 4. (a) Schematic representation of a cantilever resonating structure excited at its second mode and (b) nonlinear resonances acquired by driving a piezoelectric element at incrementally larger voltages in the range of 0.05 to 0.5 V. A nearly symmetric bell shaped response (curve corresponding to 0.05V) is observed at small driving amplitudes, which is characteristic of a linear regime. Asymmetrically distorted frequency response curves are observed at larger driving amplitudes. At a driving voltage of 0.5 V, hysteresis (shown by the arrows) and amplitude collapse is observed upon forward frequency sweeping.

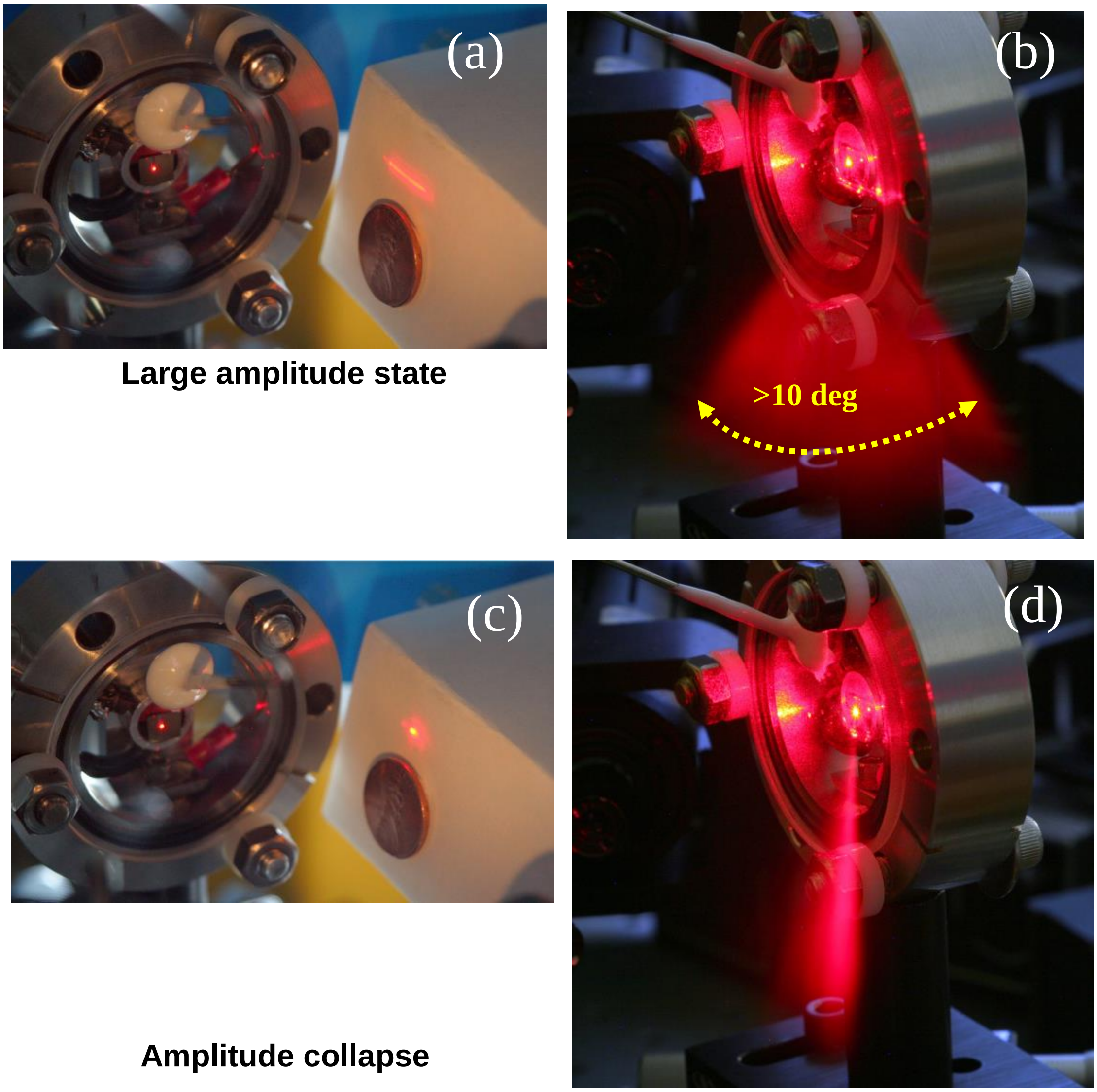

Figure 5. Amplitude collapse at the bifurcation points can readily be visualized and monitored using a simple spot photodetector. In the large amplitude state [(a) and (b)] the laser reflected off the transducer forms a line that is nearly 2 cm long on a screen at a distance ~5 cm. When cantilever amplitude collapses, the reflection of the laser beam changes from a centimeter-long scan to nearly a point [(c) an (d)]. The frequency at which the amplitude collapse occurs can be used as a measure of the transducer mass loading.

By driving a piezoelectric element at incrementally increasing voltages in the range of 0.05 to 0.5 V, we observed excitation of the cantilever resonators in their second bending mode shown schematically in **Figure 4(a).** **Figure 4(b)** shows a typical series of resonance curves obtained when driving one of the cantilevers with progressively larger amplitudes. A nearly symmetric bell shaped response (curve corresponding to a drive voltage of 0.05V) characteristic of a linear regime is observed at small oscillation amplitudes. Asymmetric frequency responses are observed at larger driving amplitudes. Finally, at the drive voltage of 0.5 V, a hysteresis (depicted by the arrows) is observed along with the amplitude collapse during the forward frequency sweeping.

In order to take advantage of the observed nonlinear behavior for mass detection, we relied on tracking of the frequency values that corresponded to the amplitude collapse. Please note that the maximum deflections of the cantilever operating in a strongly nonlinear regime observed during the forward frequency sweeping immediately before the amplitude collapse exceeded several micrometers. Such amplitudes were sufficient to deflect the read laser beam focused on the cantilever by 10 angular degrees (see top right panel in **5**). As can be seen in **Figure 5(a)**, this level of laser beam deflection corresponded to approximately 1.5 cm span of the laser spot projected on the screen positioned 5 cm from the resonator [see Figure 5(a), 5(c)].

During every frequency sweep cycle, a gradual increase in the cantilever oscillation amplitude was followed by its abrupt decrease. This sudden change in the amplitude is clearly attributable to the bifurcation point predicted for a Duffing oscillator (**Figure 1**). Importantly, the amplitude collapse could readily be observed by the naked eye by watching the laser beam projection reflected off the oscillating cantilever. Therefore, the change in the reflected spot shape [see **Figure 5(a)** and **Figure 5(c)**], associated with the amplitude collapse could also be detected with a simple spot photodetector as shown in **Figure 3(b)**.

In the next series of experiments, we were continuously recording the drive frequency and the cantilever oscillation amplitude (measured using the PSD) as a function of time. In **Figure 6** we plotted the data obtained from the experiment when the frequency was repeatedly swept from 135,930 Hz to 136,020 Hz. The step-like change in the PSD signal associated with every amplitude collapse event was used to trigger the frequency recording.

According to the theory of a Duffing oscillator, the frequency at which the amplitude collapse occurs is sensitive to mass changes and we used it as a measure of the transducer mass loading. By applying this approach to the detection of a model analyte (4-tert-butyl-calix[6]arene) we sought to achieve high sensitivity and fast response times. Indeed, **Figure 7** shows the effect of mass loading on the frequency that corresponds to the amplitude collapse when while approximately 13 fg of 4-tert-butyl-calix[6]arene was deposited on the cantilever. As can be seen in **Figure 7**, mass loading caused an approximately 0.05 Hz decrease in the frequency values recorded at the point of amplitude collapse. This was a clearly resolvable change consistent with the amount of analyte deposited on the cantilever. This level of mass sensing was previously achievable only by using much smaller resonators and sophisticated laser beam focusing and analysis of the nanoscale beam displacements using position sensitive detectors. [7, 25]

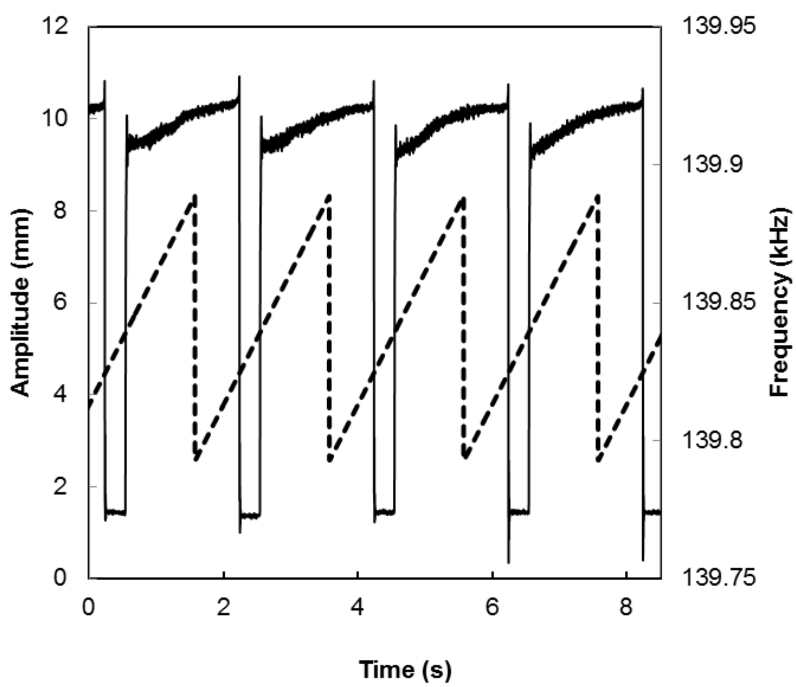

Figure 5. The frequency sweep and amplitude collapse near the bifurcation points as a function of sweep time. In the large amplitude state the frequency at which the amplitude collapse occurs can be used as a measure of the transducer mass loading

4. CONCLUSIONS

We present a new approach to sensitive detection of small mass changes using externally driven cantilever resonators in a strongly nonlinear regime. This approach takes advantage of intrinsic mechanical nonlinearity of cantilever structures and provides a facile means to detect mass loading without the need for any low noise electronics analog signal processing, and accurate measurements of oscillation amplitudes. We demonstrate that cantilevers with lengths of approximately 100 micrometers can be driven to amplitude sufficiently large to provide macroscopic rastering of a laser beam. Such large amplitudes greatly facilitate tracking of the amplitude collapse associated with the bifurcation point. We demonstrate that the amplitude collapse can clearly be observed as a drastic decrease in the length of the raster line projected by the oscillating cantilever by the naked eye. Alternatively, a stepwise signal generated by a spot photodiode provides a triggering signal for an extremely accurate detection of the bifurcation point.

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