Effect of Surface Stress on the Stiffness of Cantilever Plates

Michael J. Lachut and John E. Sader*

Department of Mathematics and Statistics, The University of Melbourne, Victoria 3010, Australia (Received 2 May 2007; published 15 November 2007)

Measurements over the past 30 years have indicated that surface stress can significantly affect the stiffness of microcantilever plates. Several one-dimensional models based on beam theory have been proposed to explain this phenomenon, but are found to be in violation of Newton's third law, in spite of their good agreement with measurements. In this Letter, we review this work and rigorously examine the effect of surface stress on the stiffness of cantilever plates using a full three-dimensional model. This study establishes the relationship between surface stress and cantilever stiffness, and in so doing elucidates its scaling behavior with cantilever dimensions. The use of short nanoscale cantilevers thus presents the most promising avenue for future investigations.

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Surface stress is an essential property of a solid surface that has been widely studied using a range of experimental techniques [1,2] and first principles calculations [3,4]. An ability to control its effects lies at the core of many applications including the deposition of metal coatings [1], epitaxial growth of semiconductor films [4], formation of self-assembled monolayers [2], and design and construction of ultrasensitive mechanical sensors [5]. One such sensing platform that has received considerable attention is the microcantilever, due to its ease of construction, implementation, and versatility [5]. In spite of this activity, knowledge of the effect of surface stress on the stiffness of cantilever plates remains an outstanding problem in the physical sciences.

Over the past 30 years, measurements have suggested that the stiffness of microcantilever plates can be tuned by varying their surface stress. This phenomenon was first reported by Lagowski et al. [6], who studied GaAs cantilevers as a function of surface preparation. These authors proposed a one-dimensional model based on classical beam theory for the effect that changes in strainindependent surface stress [6-8] have on the resonant frequency; see Eq. (5) of Ref. [6]. However, this model was later shown to be incorrect by Gurtin et al. [8] who wrote, "Within the framework of classical beam theory it is shown that strain-independent surface stress has no effect on the natural frequency of a thin cantilever beam. Therefore, the experimental results of Lagowski, Gatos, and Sproles must have a different explanation." Since that time, numerous other models based essentially on the Lagowski hypothesis have been proposed [9-12]. Indeed, such models have recently been used to argue that strainindependent surface stress dominates the dynamic response of microcantilevers in biomolecular recognition measurements [11]. It is thus of critical importance to assess the validity of such models.

Gurtin *et al.* [8] also examined the effects of surface elasticity, i.e., "strain-dependent" surface stress, by considering a general constitutive model for surface stress [7], and showed theoretically that this can affect cantilever

stiffness. However, they concluded that this effect is negligible, leaving the experimental results of Ref. [6] unexplained. Other authors have subsequently adopted this idea of surface elasticity [12-14] in an attempt to account for the experimental measurements.

In this Letter, we return to the original question: What is the relationship between strain-independent surface stress change and the stiffness of cantilever plates?

As we shall discuss, the origin of this relationship lies in an alternative mechanical process to that proposed previously in Refs. [6,8-12].

To determine this relationship, we abandon the conventional approach that uses (approximate) classical beam theory and replace it with a rigorous three-dimensional treatment of the deformation problem, within the framework of the theory of linear elasticity. There are two equivalent approaches to examining the effect of surface stress change on cantilever stiffness: (i) monitor the change in deflection for a given applied normal load, or (ii) monitor the resonant frequency change. Since the latter is most commonly reported and easily measured, we focus our investigation on the change in the fundamental resonant frequency, while noting that our conclusions also apply to the first approach.

We study a rectangular cantilever plate under a uniform and isotropic strain-independent surface stress loading on both faces, i.e., σ_s^+ and σ_s^- on upper and lower faces, respectively; see Fig. 1. Note that σ_s^+ and σ_s^- are taken to be the changes in surface stress from their base (intrinsic) values. We define a differential surface stress, $\Delta \sigma_s =$ $\sigma_s^+ - \sigma_s^-$, and a total surface stress, $\sigma_s^T = \sigma_s^+ + \sigma_s^-$. Importantly, differential surface stress $\Delta \sigma_s$ will not affect cantilever stiffness within the framework of linear elasticity theory, since its only effect is to induce an effective bending moment at the edges [15]. As such, differential surface stress and the subsequent bending are ignored in this study, and we focus our attention on the total surface stress σ_s^T and its effect on cantilever stiffness. The application of a uniform and isotropic σ_s^T will give rise to inplane deformation of the plate, which we now examine.



FIG. 1. Schematic of rectangular cantilever plate showing coordinate system and applied surface stresses. Origin of coordinate system is at center of mass of the clamped end.

To begin, we consider the related problem of an unrestrained plate. Here, the no-traction boundary condition along all edges ensures that the plate is in a state of uniform plane stress, where the integral of the stress over the thickness h is zero everywhere; i.e., the net in-plane stress is zero. The displacement field for this problem is

$$(u, v) = -(1 - v)\sigma_s^T(x, y)/(Eh),$$
(1)

where u and v are displacements in the x and y directions, respectively, E and v are the Young's modulus and Poisson's ratio, respectively. Since the net in-plane stress is zero, the resonant frequency of an unrestrained (free) plate is independent of the applied surface stress.

In contrast, application of a uniform isotropic surface stress σ_s^T to a cantilever plate will not yield uniform inplane deformation. To account for this behavior, the effect of the clamp must be included. This is achieved by decomposing the original cantilever problem into two subproblems.

Subproblem (1).—Deformation of an unrestrained plate under application of a total surface stress σ_s^T .

Subproblem (2).—Cantilever plate with no surface stress load and a specified in-plane displacement at its clamped end: u = 0, $v = (1 - \nu)\sigma_s^T y/(Eh)$, w = 0.

Superposition of these subproblems gives the required in-plane deformation of the original cantilever problem, with exact satisfaction of free edge and clamped boundary conditions. The in-plane stress distribution in Subproblem (2) is identical to the original cantilever problem, since Subproblem (1) has zero net in-plane stress. As such, resonant frequencies of the original cantilever problem are given by those of Subproblem (2).

An exact analytical solution to Subproblem (2) poses a formidable challenge. To gain insight into the dependence of surface stress on cantilever stiffness, we initially examine its scaling behavior. The load at the clamped end in Subproblem (2) will induce nonzero in-plane stresses that decay in the *x* direction with a characteristic length scale given by the cantilever width *b*. As such, there exists a region near the clamp, x < O(b), where nonzero in-plane stresses exist, outside of which in-plane stress is zero. Consequently, increasing the aspect ratio, L/b, of the cantilever will reduce the overall effect of the applied surface stress. To derive the scaling law, we use the (twodimensional) governing equation for small deflection of a thin cantilever plate, $D\partial_{ii}\partial_{jj}w - N_{ij}\partial_{ij}w = q$, where *w* is the deflection in the *z* direction, *D* is the flexural rigidity, *N* is the in-plane stress tensor, and *q* is the applied load per unit area. Making use of this plate equation and Eq. (1) then leads to the following leading order scaling dependence for the effective flexural rigidity D_{eff} ,

$$\frac{D_{\text{eff}}}{D} - 1 \sim O\left(\frac{(1-\nu)\sigma_s^T}{Eh}\left(\frac{b}{L}\right)\left(\frac{b}{h}\right)^2\right).$$
(2)

This result contrasts directly with the result obtained from classical beam theory, which predicts a zero surface stress effect. Since cantilever stiffness is proportional to the flexural rigidity, the leading order dependence of the relative change in resonant frequency due to an applied surface stress is

$$\frac{\Delta\omega}{\omega_0} = \phi(\nu)\bar{\sigma}\left(\frac{b}{L}\right)\left(\frac{b}{h}\right)^2,\tag{3}$$

where ω_0 is the resonant frequency in the absence of an applied surface stress, $\Delta \omega = \omega - \omega_0$, $\bar{\sigma} = (1 - \nu)\sigma_s^T/(Eh)$ is the dimensionless surface stress change, and $\phi(\nu)$ is a function purely dependent on Poisson's ratio ν . This expression is expected to be valid in the limit $L \gg b \gg h$, and when surface stress has a small effect on stiffness, which is frequently encountered in practice.

Next we solve Subproblem (2) using a full threedimensional finite element analysis [16]. To begin, we examine the prediction of Eq. (3) that the change in resonant frequency varies in proportion to the square of width to thickness ratio b/h, for a fixed aspect ratio L/b. Figure 2 presents results for the relative change in resonant frequency, $\Delta \omega / \omega_0$, which have been scaled by $(b/h)^2$, in accordance with Eq. (3). From Fig. 2, it is strikingly evident that Eq. (3) accurately captures the dominant width ratio b/h dependence, with all curves collapsing onto each other for a given Poisson's ratio ν . Also note that the frequency shift depends strongly on Poisson's ratio, with increasing ν enhancing the effect. This is to be expected, since the applied load is a deformation in the y direction, and the stiffness probed is predominantly in the x direction. Interestingly, we find that for $\nu = 0$, varying the surface stress has a negligible effect on stiffness in comparison to nonzero ν .

The numerical results in Fig. 2 include all nonlinear surface stress effects, which have been ignored in the formulation of Eq. (3). Consequently, to make a quantitative and rigorous comparison to Eq. (3), we henceforth



FIG. 2. Results for relative frequency shift $\Delta \omega / \omega_0$ vs $\bar{\sigma} (b/h)^2$ for L/b = 25/3. Three groups of Poisson's ratio shown: $\nu = 0$, 0.25, 0.49. Each group contains b/h = 16, 19.2, 24, 32, 48.

extract the linear portion of these numerical results using linear regression and report these results only.

In Fig. 3, we assess the aspect ratio (L/b) dependence predicted in Eq. (3). Results are presented for a range of aspect ratios L/b, Poisson's ratio $\nu = 0.25$, with width ratios b/h corresponding to those used in Fig. 2. Results for other nonzero Poisson's ratios are similar to those in Fig. 3, apart from a change in magnitude. The vertical axis is scaled in accordance with Eq. (3) to examine its validity. From Fig. 3, it is clear that Eq. (3) also captures the dominant aspect ratio dependence for large L/b, which is the regime in which it was derived, while a higher order dependence on aspect ratio is also visible for smaller L/b; this higher order dependence can be calculated from results in Fig. 3 if required. These results confirm the validity of the scaling argument behind Eq. (3).

To determine $\phi(\nu)$, the data in Fig. 3 are extrapolated in the binary limit $L/b \rightarrow \infty$ and $b/h \rightarrow \infty$. Given the linearity of the data in this limit, extrapolation is robust and accurate. Results of extrapolation for various Poisson's ratio are then used to evaluate $\phi(\nu)$. We find that $\phi(\nu)$ varies approximately linearly with Poisson's ratio and is well described by $\phi(\nu) \approx -0.042\nu$ [17]. Substituting this



FIG. 3. Results for normalized frequency shift $\Omega = |\Delta \omega_{\text{lin}}/\omega_0|/|\bar{\sigma}(b/L)(b/h)^2|$; b/h = 16, 19.2, 24, 32, 48; $\nu = 0.25$. Subscript "lin" indicates result from linear regression.

result into Eq. (3) gives the required dependence of the relative frequency shift on surface stress change,

$$\frac{\Delta\omega}{\omega_0} \approx -0.042 \frac{\nu(1-\nu)\sigma_s^T}{Eh} \left(\frac{b}{L}\right) \left(\frac{b}{h}\right)^2. \tag{4}$$

We emphasize that this result is not inconsistent with the null result of Gurtin *et al.* [8], since they implicitly considered the formal limit $L/b \rightarrow \infty$ using classical beam theory. In this limit, Eq. (4) also predicts that surface stress change has no effect. The mechanism giving rise to surface stress induced stiffness changes lies in development of inplane stresses near the clamp, which are inherently ignored in beam theory.

The resonant frequency change is dictated by the ratio of the modified total surface stress $\nu(1-\nu)\sigma_s^T$ to the stiffness $k_{\rm ref} = Eh^3L/b^3$. Increasing the length L therefore has a relatively weak effect in comparison to changing the thickness or width. Importantly, this scaling dependence differs considerably from that for surface elasticity effects (straindependent surface stress) [13] and can be used as a signature to investigate the presence of strain-independent surface stress effects. Indeed, probing the scaling dependence of strain-independent and strain-dependent surface stress contributions with cantilever geometry allows for determination of the underlying mechanism driving stress induced changes in cantilever stiffness. In principle, Eq. (4) could be combined with static bending measurements, which probe differential surface stress, to determine surface stress changes on each face of the cantilever, i.e., individual measurement of σ_s^+ and σ_s^- . Such application would, of course, be contingent on the frequency resolution achievable in practice. Equation (4) establishes that use of low aspect ratio cantilevers and reduction in thickness will yield the greatest sensitivity. We emphasize that our study is applicable to nanoscale structures where the classical theory of elasticity is valid, the subject of which was investigated recently [18]. Miniaturization to the nanoscale thus presents a promising avenue for future developments.

We now examine the validity of recent models [9-12]derived using beam theory. Similar to Lagowski's model, these models attempt to simplify the problem by replacing surface stress with an external axial force equal to the total surface stress σ_s^T integrated over the cantilever dimensions. However, such approaches are not physically justified since the cantilever free end is unrestrained with zero net force. Application of a (uniform) axial force along the beam is therefore in direct violation of Newton's third law. As explained by Gurtin et al. [8], application of surface stress will result in a commensurate stress of opposite sign within the beam material, ensuring zero net in-plane stress across the beam [13]. Therefore, the physical basis of all such one-dimensional models based on beam theory is flawed. The true effect of surface stress change can only be captured using higher dimensional models that account for nonuniform distribution of inplane stresses in the cantilever, as given above.

Next, we compare predictions of Eq. (4) with published experimental results for surface stress induced changes in cantilever stiffness. While the primary focus of Ref. [6] was (intrinsic) residual stress, modification of surface properties by etching was also examined; see Fig. 4 of Ref. [6]. Variations in resonant frequency from a few percent to nearly 100% are reported. Using their unphysical model (see above), surface stress changes in the vicinity of 0.2 N/m are calculated, which are in agreement with expected values. While a direct comparison with their measurements is not possible, due to omission of specific cantilever dimensions with resonant frequency shifts in their study, we can provide an estimate of the relative frequency shift. Using typical values for the material properties of GaAs and the limiting cantilever geometry L/b =6, b/h = 500 [6], yields a relative frequency shift $\Delta\omega/\omega_0 \approx 10^{-4}$, which is significantly smaller than the shift observed in Ref. [6]. The observations in Ref. [6] are therefore not due to strain-independent surface stress; Gurtin et al. [8] obtained the same conclusion based on the null result from beam theory.

Recent measurements use silicon and silicon nitride microcantilevers and monitor resonant frequency change as the surface is modified. Since absolute surface stress change is not measured, direct comparison with Eq. (4) is again not possible. Instead, we determine the required surface stress change to recover the experimentally observed frequency shifts. While significant variation exists between measurements in different studies, the range of frequency shifts is $\Delta \omega / \omega_0 \approx 0.002 - 0.06$. Reference [10] used gold coated silicon rectangular cantilevers with dimensions $499 \times 97 \times 0.8 \ \mu m^3$ and reported frequency shifts up to $\Delta \omega / \omega_0 \approx 0.01$ following an amino-ethanethiol-gold adsorption binding event. These authors also measured surface stress change from the static deflection of the cantilever and compared it to dynamic measurements based on an (unphysical) axial stress model; see above. Good agreement was found between these independent measurements and the axial stress model for the first six modes. However, from Eq. (4), we find that the total surface stress change required to achieve this maximum frequency shift is $\bar{\sigma_s^T} \approx 60$ N/m. This value is orders of magnitude higher than typical surface stress changes, and $\sim 10^5$ times larger than surface stresses reported in Ref. [10]. A similar conclusion is drawn on measurements in Refs. [9,19]. This analysis indicates that these experimental results are not due to the effects of strainindependent surface stress, as has been implicitly assumed.

We have investigated the effects of strain-independent surface stress change on the stiffness of cantilever plates using a three-dimensional analysis and scaling argument. This overcomes the limitations of beam theory, which predicts a null effect of strain-independent surface stress. We also discussed the invalidity of so-called axial stress models that proliferate the current literature. Our analysis indicates that current measurements of surface induced stiffness changes are not due to strain-independent surface stress. While changes in surface elasticity have been proposed to explain these experiments, a quantitative comparison with measurements remains elusive. The scaling laws of these complementary approaches can thus be used to investigate the true nature of surface induced changes in cantilever stiffness and presents a rigorous avenue for elucidating the underlying mechanism. The use of low aspect ratio nanoscale cantilevers presents the most promising approach for investigating the effects of strain-independent surface stress change on cantilever stiffness.

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*jsader@unimelb.edu.au

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