Nonlinear Analysis of Oscillatory Indentation in Elastic and Viscoelastic Solids

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Determining the mechanical properties at micro- and nanometer length scales using nanoindentation or atomic force microscopy is important to many areas of science and engineering. Here we establish equations for obtaining storage and loss modulus from oscillatory indentations by performing a nonlinear analysis of conical and spherical indentation in elastic and viscoelastic solids. We show that, when the conical indenter is driven by a sinusoidal force, the square of displacement is a sinusoidal function of time, not the displacement itself, which is commonly assumed. Similar conclusions hold for spherical indentations. Well-known difficulties associated with measuring contact area and correcting thermal drift may be circumvented using the newly derived equations. These results may help improve methods of using oscillatory indentation for determining elastic and viscoelastic properties of solids.

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Atomic force microscopy and nanoindentation techniques developed in the past two decades have become powerful tools for measuring mechanical properties at micro- and nanometer length scales for ceramics, metals, polymers, and biomaterials [1–23]. These force microscopy techniques can be operated in either quasistatic [1–12] or oscillatory modes [11–23]. In the oscillatory mode, a sinusoidal force is typically superimposed on a quasistatic load on the indenter [11–23]. The indentation displacement response and the out-of-phase angle between the applied harmonic force and the corresponding harmonic displacement may be recorded at a given excitation frequency or multiple frequencies. A number of authors [11–17] have proposed analysis procedures for determining the complex Young's modulus $E^*(\omega) = E'(\omega) + iE''(\omega)$, where $E'(\omega)$ is the storage modulus and $E''(\omega)$ is the loss modulus, from oscillatory indentations using the following equations:

$$\frac{E'}{1-\nu^2} = \frac{\sqrt{\pi}}{2}\frac{S}{\sqrt{A}} \quad \text{and} \quad \frac{E''}{1-\nu^2} = \frac{\sqrt{\pi}}{2}\frac{C\omega}{\sqrt{A}}, \tag{1}$$

where ν is Poisson's ratio, S is contact stiffness, C is damping coefficient, and A is contact area between the indenter and the sample. Several authors [11,12,14–17] have developed models for contact stiffness and damping coefficient. Applying these equations to an ideal indenter with infinite system stiffness and zero mass, the contact stiffness and damping coefficient are given by $S = |\Delta F/\Delta h|\cos\phi$ and $C\omega = |\Delta F/\Delta h|\sin\phi$, where ΔF is the amplitude of sinusoidal force with angular frequency ω, Δh is the amplitude of oscillatory displacement, and ϕ is the phase angle of the displacement response. Thus, by measuring displacement amplitude and phase angle under harmonic oscillation, the reduced storage and loss modulus $E'/(1-\nu^2)$ and $E''/(1-\nu^2)$ can be obtained from

$$\begin{aligned} \frac{E'}{1-\nu^2} &= \frac{\sqrt{\pi}}{2\sqrt{A}}\left|\frac{\Delta F}{\Delta h}\right|\cos\phi, \\ \frac{E''}{1-\nu^2} &= \frac{\sqrt{\pi}}{2\sqrt{A}}\left|\frac{\Delta F}{\Delta h}\right|\sin\phi. \end{aligned} \tag{2}$$

In this Letter, we show that Eq. (2) is the result of a linear approximation of the oscillatory indentation. By performing a nonlinear analysis, we derive the corresponding set of equations without evoking the small amplitude oscillation assumption. We begin with oscillatory conical indentation in purely elastic solids, then in viscoelastic solids, and end with oscillatory spherical indentation in viscoelastic solids.

We consider a rigid, smooth, and frictionless conical indenter with half-angle θ indenting a purely elastic solid [Fig. 1(a)]. The well-known load-displacement relation is given by [24]

$$h^2 = \frac{\pi(1-\nu^2)}{2E\tan\theta}F. \tag{3}$$

Equation (3) is valid for both increasing and decreasing loads since purely elastic solids respond to time-varying loads instantaneously. Thus, Eq. (3) is also valid for oscillatory loading and unloading. We consider a sinusoidal load superimposed on a step load F_m:

$$F(t) = F_m + \Delta F\sin(\omega t), \tag{4}$$

where ΔF and ω are the amplitude and angular frequency of the sinusoidal load, respectively. The displacement is then given by

$$h^2(t) = \frac{\pi(1-\nu^2)}{2E\tan\theta}F_m\left(1 + \frac{\Delta F}{F_m}\sin\omega t\right). \tag{5}$$

Clearly, the *square* of the indenter displacement $h^2(t)$, not the displacement itself, is a sinusoidal function. Let $h^2(t) \equiv h_m^2 + \Delta_2 h\sin(\omega t)$, where h_m^2 and $\Delta_2 h$ are the re-

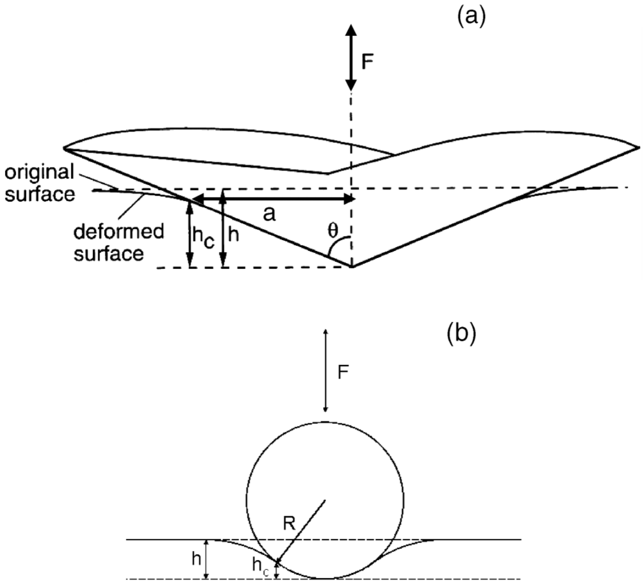

FIG. 1. A schematic illustration of oscillatory (a) conical and (b) spherical indentations.

spective square displacement and the amplitude of the oscillatory square displacement; we obtain a pair of equations:

$$h_m^2 = \frac{\pi(1-\nu^2)}{2E\tan\theta} F_m, \tag{6}$$

$$\frac{E}{1-\nu^2} = \frac{\pi}{2\tan\theta}\frac{\Delta F}{\Delta_2 h}. \tag{7}$$

Equation (6) is a restatement of Eq. (3), which represents the static response due to F_m. Equation (7) describes the dynamic response due to the oscillatory component of load ΔF. Since ΔF is known, the reduced modulus $E/(1-\nu^2)$ can be obtained by measuring the amplitude of the square displacement $\Delta_2 h$.

We now show that Eq. (2) is a linear approximation of Eq. (7) for oscillatory conical indentation in purely elastic solids. Making the commonly used assumption that the displacement is a sinusoidal function of time, i.e., $h(t) = h_m + \Delta h \sin(\omega t)$, where h_m and Δh are the respective displacement and displacement amplitude, we obtain, in the linear approximation, $\Delta_2 h \approx 2h_m \Delta h$. Using the fact that, for conical indentation in elastic solids [24], $h_m(t) = (\pi/2)h_c(t)$, $a = h_c \tan\theta$, and $A = \pi a^2$, where h_c, a, and A are the respective contact depth, contact radius, and contact area [Fig. 1(a)], we see that Eq. (7) becomes

$$\frac{E}{1-\nu^2} = \frac{\sqrt{\pi}}{2\sqrt{A(t)}}\frac{\Delta F}{\Delta h}. \tag{8}$$

When the amplitude of displacement oscillations is small, i.e., $\Delta h/h_m \ll 1$, the contact area is approximately independent of displacement oscillation amplitude. Equation (8) is then identical to Eq. (2) when it is applied to purely elastic solids, since $\phi = 0$ and $E = E'$.

The difference between nonlinear analysis and linear approximation can be demonstrated by comparing the $h^2(t)$ and $h(t)$ for various oscillation amplitudes $\Delta F/F_m$. According to the nonlinear analysis [Eq. (5)], $h(t)$ can be written, in linear approximation, as

$$h_{\mathrm{approx}}(t) = \sqrt{\frac{\pi(1-\nu^2)}{2E\tan\theta} F_m}\left(1 + \frac{1}{2}\frac{\Delta F}{F_m}\sin\omega t\right). \tag{9}$$

Figures 2(a) and 2(b) plot $h^2(t)/\{[\pi(1-\nu^2)/2E\tan\theta]F_m\}$ vs time for $\Delta F/F_m = 0.5$ and 0.8 for both linear and nonlinear cases. As expected, the results from nonlinear analysis and linear approximation are very similar when $\Delta F/F_m < 0.1$. However, the discrepancy increases with $\Delta F/F_m$. When the oscillation amplitude is large, the displacement is no longer sinusoidal, but the square of displacement is. Furthermore, we note $\Delta_2 h = (1/2)[h_{\mathrm{approx}}{}^2(\max) - h_{\mathrm{approx}}{}^2(\min)]$. Thus, the square amplitude can be obtained by measuring the peak $h_{\mathrm{approx}}(\max)$ and valley $h_{\mathrm{approx}}(\min)$ of oscillations in the linear plot of $h(t)$ vs t.

We now perform nonlinear analysis of oscillatory conical indentation in viscoelastic solids. The solid may be described by the constitutive relationships [25] between deviatoric stress and strain s_{ij} and d_{ij} and between dilatational stress and strain σ_{ii} and ε_{ii}:

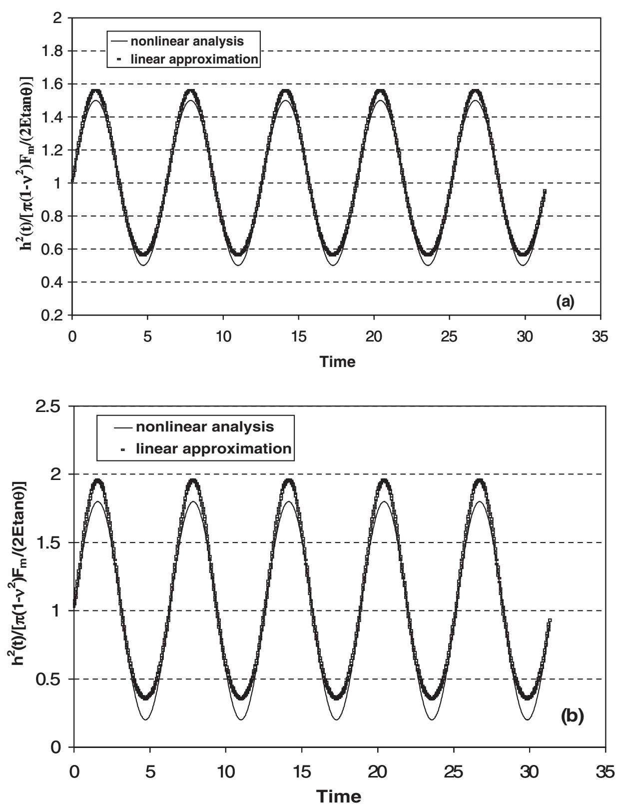

FIG. 2. Comparison of nonlinear analysis and linear approximation for oscillatory indentation with $\omega = 1$ in elastic solids: $h^2(t)/\{[\pi(1-\nu^2)]/2E\tan\theta\}F_m$ vs time for $\Delta F/F_m = 0.5$ (a) and 0.8 (b).

$$2d_{ij}(t) = \int_{-\infty}^{t} J_1(t-\tau)\frac{\partial s_{ij}(\tau)}{\partial\tau}d\tau,$$
$$3\varepsilon_{ii}(t) = \int_{-\infty}^{t} J_2(t-\tau)\frac{\partial \sigma_{ii}(\tau)}{\partial\tau}d\tau, \tag{10}$$

where $J_1(t)$ is the shear compliance and $J_2(t)$ is the volumetric compliance. The shear and volumetric compliances are related to the relaxation modulus in shear $G(t)$ and relaxation modulus in dilatation $K(t)$. The time dependent Young's modulus and Poisson's ratio are given by $E(t) = [9K(t)G(t)]/[3K(t) + G(t)]$ and $\nu(t) = [E(t)/2G(t)] - 1$, respectively. In the following, we assume that Poisson's ratio is time independent. Consequently, $J_1(t)$ and ν are sufficient to describe the viscoelastic behavior.

With force as the independent variable, the relationship between displacement $h(t)$ and force $F(t)$ is given by [26,27]:

$$h^2(t) = \frac{\pi(1-\nu)}{4\tan\theta}\int_{-\infty}^{t} J_1(t-\tau)\frac{dF(\tau)}{d\tau}d\tau. \tag{11}$$

We consider a harmonic force superimposed on a quasistatic force, i.e.,

$$F(t) = F_m f(t) + \Delta F\sin(\omega t), \tag{12}$$

where $f(t)$ is a monotonically nondecreasing function of time $|f(t)| \leq 1$. Inserting Eq. (12) into (11) and using the definition of the storage and loss shear compliances, $J'(\omega) = \omega\int_0^\infty J_1(s)\sin(\omega s)ds$ and $J''(\omega) = -\omega\int_0^\infty J_1(s)\cos(\omega s)ds$, we obtain

$$h^2(t) = \frac{\pi(1-\nu)}{4\tan\theta}\left\{F_m\int_{-\infty}^{t} J_1(t-\tau)\frac{df(\tau)}{d\tau}d\tau + J'(\omega)\Delta F\sin(\omega t) - J''(\omega)\Delta F\cos(\omega t)\right\}. \tag{13}$$

Equation (13) implies that

$$h^2(t) = B(t) + \Delta_2 h\sin(\omega t - \phi), \tag{14}$$

where ϕ is the phase shift. Comparing Eq. (14) with Eq. (13) and using the relationship [25] between the complex modulus E^* and the complex shear compliance J^*, i.e., $E' = 2(1+\nu)[J'/(J'^2 + J''^2)]$ and $E'' = 2(1+\nu)\times[J''/(J'^2 + J''^2)]$, we obtain

$$B(t) = \frac{\pi(1-\nu)}{4\tan\theta}F_m\int_{-\infty}^{t} J_1(t-\tau)\frac{df(\tau)}{d\tau}d\tau, \tag{15}$$

$$E' = \frac{\pi(1-\nu^2)}{2\tan\theta}\frac{\Delta F}{\Delta_2 h}\cos\phi,$$
$$E'' = \frac{\pi(1-\nu^2)}{2\tan\theta}\frac{\Delta F}{\Delta_2 h}\sin\phi. \tag{16}$$

On the other hand, if we use the common assumption that the displacement is a sinusoidal function of time, i.e., $h(t) = h_m(t) + \Delta h\sin(\omega t - \psi)$, where h_m and Δh are the respective displacement and displacement amplitude, then in the linear approximation, we have $\Delta_2 h \approx 2h_m(t)\Delta h$, $\psi = \phi$, and

$$E' = \frac{\pi(1-\nu^2)}{4h_m(t)\tan\theta}\frac{\Delta F}{\Delta h}\cos\phi,$$
$$E'' = \frac{\pi(1-\nu^2)}{4h_m(t)\tan\theta}\frac{\Delta F}{\Delta h}\sin\phi. \tag{17}$$

Equation (17) is analogous to that derived recently by Lu and co-workers for spherical indentation in viscoelastic solids [23]. By further assuming that $h_m(t) = (\pi/2)h_c(t)$ when $\Delta h \ll h_m(t)$, $a = h_c\tan\theta$, and $A = \pi a^2$, we transform Eq. (17) to Eq. (2). From this derivation, it is evident that Eq. (2) is a linear approximation of Eq. (16). Furthermore, Eq. (2) assumes that $h_m(t) = (\pi/2)h_c(t)$ holds during oscillatory indentation, which is not true for large magnitude unloading [10] or oscillations [23].

To further illustrate the differences between linear and nonlinear analysis, we consider a sinusoidal oscillation superimposed on a step loading. The nonlinear and linear relationships obtained from Eq. (13) are then given by

$$h^2(t) = \frac{\pi(1-\nu)}{4\tan\theta}F_m J_1(t)\left\{1 + \frac{\Delta F|J^*(\omega)|}{F_m J_1(t)}\sin(\omega t - \phi)\right\}, \tag{18}$$

$$h_{\text{approx}}(t) = \sqrt{\frac{\pi(1-\nu)}{4\tan\theta}F_m J_1(t)} \times\left\{1 + \frac{1}{2}\frac{\Delta F|J^*(\omega)|}{F_m J_1(t)}\sin(\omega t - \phi)\right\}, \tag{19}$$

where $|J^*(\omega)| = \sqrt{J'(\omega)^2 + J''(\omega)^2}$ is the magnitude of the complex compliance and the phase angle is given by $\cos\phi = J'/|J^*|$ and $\sin\phi = J''/|J^*|$. Unlike the purely elastic case where $\Delta F/F_m$ determines the amplitude of displacement oscillation, the parameter $\Delta F|J^*(\omega)|/F_m J_1(t)$ affects the amplitude of oscillation and nonlinearity for the viscoelastic case. Since $J^*(\omega)$ and $J_1(t)$ depend on the frequency and time, the amplitude of displacement oscillation and nonlinearity are also functions of frequency and time. Similar to the purely elastic case, $\Delta_2 h = (1/2)|h^2_{\text{approx}}(\max) - h^2_{\text{approx}}(\min)|$.

The same approach can be applied to spherical indentation in viscoelastic solids. With force as the independent variable, the relationship between $h(t)$ and $F(t)$ is given by [26,27]:

$$h^{3/2}(t) = \frac{3(1-\nu)}{8\sqrt{R}}\int_{-\infty}^{t} J_1(t-\tau)\frac{dF(\tau)}{d\tau}d\tau, \tag{11'}$$

where R is the radius of the spherical indenter [Fig. 1(b)]. Substituting Eq. (12) into Eq. (11′) for a harmonic force superimposed on a quasistatic force, we observe that $h^{3/2}(t)$, not $h(t)$, is a sinusoidal function of time, i.e.,

$$h^{3/2}(t) = B(t) + \Delta_{3/2}h\sin(\omega t - \phi), \tag{14'}$$

where ϕ is the phase shift and $\Delta_{3/2}h$ is the amplitude of the oscillatory component of $h^{3/2}(t)$. In particular, we find

$$B(t) = \frac{3(1-\nu)}{8\sqrt{R}} F_m \int_{-\infty}^{t} J_1(t-\tau)\frac{df(\tau)}{d\tau} d\tau, \qquad (15')$$

$$E' = \frac{3}{4}\frac{(1-\nu^2)}{\sqrt{R}}\frac{\Delta F}{\Delta_{3/2}h}\cos\phi,$$
$$E'' = \frac{3}{4}\frac{(1-\nu^2)}{\sqrt{R}}\frac{\Delta F}{\Delta_{3/2}h}\sin\phi. \qquad (16')$$

Thus, by measuring ϕ and $\Delta_{3/2}h$, the reduced storage and loss modulus can be obtained from oscillatory spherical indentation using Eq. (16′).

Similar to the conical indentation case, if we use the common assumption that the displacement is a sinusoidal function of time, i.e., $h(t) = h_m(t) + \Delta h \sin(\omega t - \psi)$, then in the linear approximation, we have $\Delta_{3/2}h = (3/2)\Delta h h_m^{1/2}$ and $\psi = \phi$. Equation (16′) becomes

$$E' = \frac{1}{2}\frac{(1-\nu^2)}{\sqrt{Rh_m}}\frac{\Delta F}{\Delta h}\cos\phi,$$
$$E'' = \frac{1}{2}\frac{(1-\nu^2)}{\sqrt{Rh_m}}\frac{\Delta F}{\Delta h}\sin\phi. \qquad (17')$$

Equation (17′) is equivalent to that derived recently by Lu and co-workers [23]. Equation (17′) becomes Eq. (2) since the contact area $A = \pi R h$ for small amplitude spherical indentation in elastic and viscoelastic solids.

The above results are derived by assuming that Eqs. (11) and (11′) are applicable to oscillatory indentation in viscoelastic solids. Although Eqs. (11) and (11′) were derived under the condition of monotonically increasing contact area with time [26,27], our recent work [7–10] has shown that Eqs. (11) and (11′) can be applied to initial unloading for conical and spherical indentation in viscoelastic solids, respectively. A recent paper by Lu and co-workers [23] suggested that Eq. (17′) derived from Eq. (11′) under the assumption of small amplitude oscillations is indeed a very good approximation for spherical indentation in viscoelastic solids.

The results from nonlinear analysis may have significant ramifications on how to use oscillatory indentation to determine the storage and loss modulus of viscoelastic solids. The results from the linear approximation [Eqs. (2), (17), and (17′)] show that either the contact area A or indenter position h_m must be measured in order to obtain the storage and loss modulus. There are, however, well-known difficulties in measuring the contact area [1,2,9–12]. Furthermore, both the contact area and the absolute position of the indenter are affected by thermal drift during measurements. In contrast, Eqs. (7), (16), and (16′) derived from nonlinear analysis do not require the measurement of the contact area or the absolute position of the indenter, thus removing significant difficulties associated with contact area measurement and thermal drift. Instead, the amplitude of the square (or 3/2 power) displacement $\Delta_2 h$ (or $\Delta_{3/2}h$) and phase angle ϕ should be measured to obtain the storage and loss modulus from oscillatory conical (or spherical) indentations using Eq. (16) [or Eq. (16′)] for elastic and viscoelastic solids. While these equations are not limited to very small amplitude oscillations, their advantages over that of linear approximation [Eqs. (17) and (17′)] are apparent even for small amplitude oscillatory indentations. The results from the nonlinear approach to oscillatory indentation should, therefore, help improve methods of using oscillatory indentation for determining mechanical properties of a variety of materials, ranging from polymers, composites, and biomaterials.

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