Column buckling of multiwalled carbon nanotubes with interlayer radial displacements

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An elastic model is presented for column buckling of a multiwalled carbon nanotube embedded within an elastic medium. The emphasis is placed on the role of interlayer radial displacements between adjacent nanotubes. In contrast to an existing model which treats the entire multiwalled nanotube as a single column, the present model treats each of the nested tubes as an individual column interacting with adjacent nanotubes through the intertube van der Waals forces. Based on this model, a condition is derived in terms of the parameters describing the van der Waals interaction, under which the effect of the noncoincidence of all deflected column axes is so small that it does not virtually affect the critical axial strain. In particular, this condition is met for carbon multiwalled nanotubes provided that the half-wavelength of the buckling mode is much larger than the outermost diameter. In this case, the critical axial strain can be predicted correctly by the existing single-column model. On the other hand, the existing model could overestimate the critical axial strain when the half-wavelength of the buckling mode is close to or smaller than the outermost radius.

I. INTRODUCTION

The discovery of carbon nanotubes[1] attracted wide attention and stimulated extensive experimental and theoretical studies.[2] Numerous studies showed that carbon nanotubes exhibit superior mechanical properties over other known materials, and hold substantial promise as superfibers for composite materials.[3] Hence one of the most promising applications of carbon nanotubes is likely to take advantage of their exceptionally high stiffness combined with their excellent resilience. Owing to their large aspect ratio and hollow geometry, various buckling behaviors of carbon nanotubes, as a shell or a column, and under bending[4] or axial compression,[5–9] have been the subjects of numerous recent experimental and molecular-dynamics simulations. More recently, considerable attention was directed to mechanical behavior of carbon nanotubes embedded in a polymer or metal matrix.[7,10–13] These prior studies indicated that ''the laws of continuum mechanics are amazingly robust and allow one to treat even intrinsically discrete objects only a few atoms in diameter'' (Yakobson and Smalley[2]). Thus, because atomistic modeling remains expensive for MWNT's (multiwalled nanotubes), continuum mechanics models are particularly useful for the study of carbon nanotubes. However, there is strong evidence[14] that the interlayer displacements and the associated van der Waals forces could have a crucial effect on the mechanical behavior of carbon MWNT's. For this reason, existing continuum models (for instance, elastic column, and shell models) cannot be directly applied to MWNT's. This raises a major challenge to conventional continuum models.

It is known[5] that actual bending stiffness of carbon SWNT's (single-walled nanotubes) is low (about 25 times lower than that predicted by the elastic shell model if a representative thickness of 0.34 nm is used). Hence one could consider MWNT's as candidates to improve bending stiffness. Carbon MWNT's are distinguished from traditional single-layer elastic tubes due to their hollow multilayer structure and the associated intertube van der Waals forces. On the one hand, it is known that the friction energy barrier between adjacent tubes in carbon MWNT's is so low that the latter could freely slide and rotate toward each other[15] (see Ref. 16 for the curvature effect). On the other hand, although the van der Waals forces[17,18] resist interlayer radial displacements, it is not clear whether the magnitude of the van der Waals interaction in carbon nanotubes is strong enough to prevent any significant interlayer radial displacement. Hence it is anticipated[5] that interlayer displacements could affect the overall buckling behavior of carbon MWNT's. In spite of this, almost all previous works (see, e.g., Ref. 7 and Falvo *et al.* and Govindjee and Sackman in Ref. 4) ignored interlayer displacements, and treated a MWNT as a single shell or column described by a single deflection function. The validity and limitations of such a simplified model call for a thorough study. To our knowledge, this issue has yet to be investigated.

To study this issue, a double-shell model was recently suggested by the present author[19] for axially compressed shell buckling of carbon DWNT's (double-walled nanotubes). It was shown[19] that inserting an inner tube into a SWNT does not change the critical axial strain of the shell buckling, provided the inner tube is allowed to freely slide with respect to the outer one.[15] This conclusion is in sharp contrast to the single-layer shell model (see Ref. 20), which predicted that the critical axial strain of a cylindrical shell doubles when its thickness doubles. This remarkable discrepancy clearly indicates the significant effect of interlayer displacements. It is this idea that encourages us to study the effect of interlayer displacements on column buckling of carbon MWNT's.

In this paper, a multiple-column model is presented for linearized column buckling of carbon MWNT's. In contrast to the existing single-column model, which ignores interlayer displacements and treats a MWNT as a single column, the present model assumes that each of the nested concentric nanotubes is an individual hollow column, and that the deflections of all columns are coupled to each other through the van der Waals interaction between adjacent nanotubes.

Based on this model, a general condition is derived which ensures that the effect of interlayer radial displacements on column buckling is negligible. Under this condition, the critical axial strain predicted by the present model is identical to that given by the existing single-column model.

II. MULTIPLE-COLUMN MODEL

The existing single-column model[4,7] is based on the assumption that all individual tubes of a MWNT remain coaxial during deformation, and thus can be described by a single deflection curve. In reality, due to different geometric radii and external conditions (such as different end conditions and surrounding conditions), individual tubes of a MWNT could deform independently with nonzero interlayer radial displacements, while their individual deformations are couped through the intertube van der Waals forces. In particular, the interlayer radial displacements and the generated noncoincidence of the deflected column axes could affect overall mechanical behavior of the MWNT. It is these ideas that motivate the present multiple-column model for carbon MWNT's.

It is known[20] that the deflection curve of an elastic column under constant axial load and distributed lateral pressure is governed by

$$p(x)+F\frac{d^2w}{dx^2}=EI\frac{d^4w}{dx^4}, \tag{1}$$

where x is the axial coordinate, $w(x)$ is the deflection of the column, F is the constant axial force, E is the Young's modulus, I is the moment of inertia of the cross-section, and $p(x)$ is the distributed lateral pressure per unit axial length (measured positive in the direction of the deflection). Because all inner tubes of carbon MWNT's are found to be well stressed when a MWNT's is compressed,[12] throughout the paper, the axial compressive stress σ_x^0 prior to buckling is assumed to be uniform over the entire cross-section.

Equation (1) can be used to each of all nested tubes of a carbon MWNT embedded within an infinite elastic medium. Here, the subscripts 1,2, . . . ,N are used to denote the quantities associated with the innermost tube, its neighboring tube, etc., and the outermost tube, respectively. Thus, it follows from Eq. (1) that

$$\begin{aligned}
&p_{12}(x)+F_1\frac{d^2w_1}{dx^2}=EI_1\frac{d^4w_1}{dx^4},\\
&p_{23}(x)-p_{12}(x)+F_2\frac{d^2w_2}{dx^2}=EI_2\frac{d^4w_2}{dx^4},\\
&p_{(N-1)N}(x)-p_{(N-2)(N-1)}(x)+F_{N-1}\frac{d^2w_{N-1}}{dx^2}\\
&\quad=EI_{N-1}\frac{d^4w_{N-1}}{dx^4},\\
&p_N(x)-p_{(N-1)N}(x)+F_N\frac{d^2w_N}{dx^2}=EI_N\frac{d^4w_N}{dx^4},
\end{aligned} \tag{2}$$

where all nanotubes are assumed to have the same Young's modulus, p_{12} denotes the resultant van der Waals pressure per unit axial length exerted on tube 1 by tube 2 (the interaction forces are equal and opposite to any two adjacent tubes), and p_N is the interaction pressure per unit axial length between the outermost tube and the surrounding elastic medium.

The van der Waals interaction energy potential, as a function of the interlayer spacing between two adjacent tubes, can be estimated by the Lennard-Jones model.[17,18] In view of the linearized characteristic of the present buckling analysis, the van der Waals interaction pressure at any given point x between two adjacent tubes should be a linear function of the deflection jump at that point. On the other hand, because the resultant interaction pressure, or its energy potential, is defined per unit axial length, it should be proportional to the circumferential dimension (for instance, the inner radius) of the adjacent tubes. Thus one can assume that the interaction energy potential per unit axial length between any two adjacent tubes is $2R_ig(\delta)$, where $g(\delta)$ is a universal function of the interlayer spacing δ, and R_i is the inner radius. Note that the radii of nanotubes are usually not smaller than 0.5 nm,[16] $g(\delta)$ can be well estimated by using the energy potential per unit area between two flat graphite sheets, as given by Girifalco and Lad.[18] In doing so, the resultant interaction pressure (from both sides) between any two adjacent tubes of the inner radius R_i is given by

$$R_i\left.\frac{dg}{d\delta}\right|_{\delta=t}+c(\Delta w),\quad c=2R_i\left.\frac{d^2g}{d\delta^2}\right|_{\delta=t}, \tag{3}$$

where t is the initial interlayer spacing prior to buckling, (Δw) is the deflection jump due to buckling, and all higher-order terms have been neglected because the present analysis is linearized in nature. For the present model, the initial interlayer spacing (about 0.34 nm) is equal or very close to the equilibrium interlayer spacing at which $dg/d\delta=0$,[18] then the first term of Eq. (3) vanishes or is negligible everywhere, implying that all initial interlayer pressures vanish prior to buckling. It then follows from Eq. (3) that the interaction pressure per unit axial length is $c(\Delta w)$, and we have

$$\begin{aligned}
&p_{12}=c_{12}[w_2-w_1],\quad p_{23}=c_{23}[w_3-w_2],\ \ldots,\\
&p_{(N-1)N}=c_{(N-1)N}[w_N-w_{N-1}],
\end{aligned} \tag{4}$$

where

$$\begin{aligned}
&c_{12}=2R_1\left.\frac{d^2g}{d\delta^2}\right|_{\delta=t},\quad c_{23}=2R_2\left.\frac{d^2g}{d\delta^2}\right|_{\delta=t},\ \ldots,\\
&c_{(N-1)N}=2R_{N-1}\left.\frac{d^2g}{d\delta^2}\right|_{\delta=t}.
\end{aligned} \tag{5}$$

In addition, assuming that the bond strength between the outermost nanotube and the surrounding elastic medium is strong,[10–12] the interaction pressure per unit axial length between the outermost tube and the elastic medium can be described by a Winkler-like model[7,21]

$$p_N(x)=-kw_N(x), \tag{6}$$

where k is the spring constant of the surrounding elastic medium (it is twice that used in Ref. 7, because the resultant

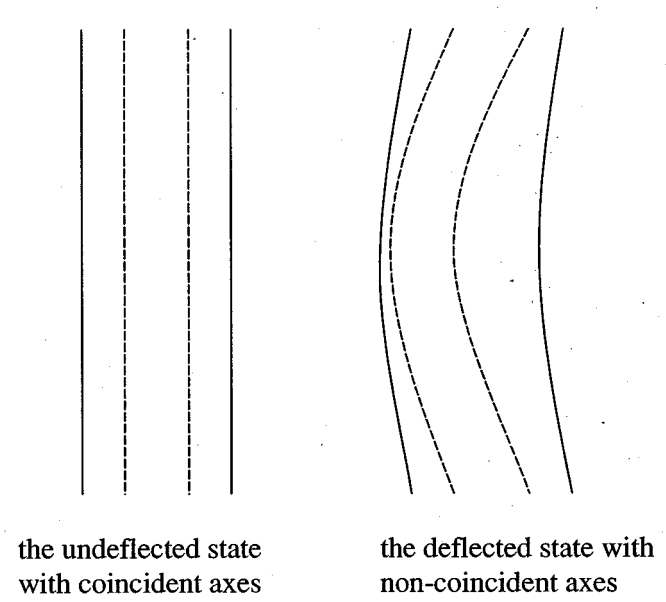

FIG. 1. Column buckling of a multiwalled nanotube with non-coincident deflected axes.

forces on two sides of each column are equal). Now, substitution of Eqs. (4) and (6) into Eq. (2) gives

$$c_{12}[w_2-w_1]+F_1\frac{d^2w_1}{dx^2}=EI_1\frac{d^4w_1}{dx^4},$$

$$c_{23}[w_3-w_2]-c_{12}[w_2-w_1]+F_2\frac{d^2w_2}{dx^2}=EI_2\frac{d^4w_2}{dx^4},$$

$$\ldots, \tag{7}$$

$$c_{(N-1)N}[w_N-w_{N-1}]-c_{(N-1)(N-2)}[w_{N-1}-w_{N-2}]+F_{N-1}\frac{d^2w_{N-1}}{dx^2}=EI_{N-1}\frac{d^4w_{N-1}}{dx^4},$$

$$-kw_N(x)-c_{(N-1)N}[w_N-w_{N-1}]+F_N\frac{d^2w_N}{dx^2}=EI_N\frac{d^4w_N}{dx^4}.$$

These equations are coupled to each other due to the van der Waals interaction terms, which vanish only when all deflected column axes are coincident; see Fig. 1. Here, all parameters $c_{i(i+1)}$ $[i=1,2,\ldots,(N-1)]$ defined by Eq. (5) at the initial interlayer spacing are bounded. It is seen from Eqs. (7) that the exact solution with coincident deflected column axes [that is, $w_i(x)=w(x)$ for $i=1,2,\ldots,N$] does not exist because any single function cannot satisfy all N different equations in Eqs. (7). This means that, although the van der Waals repulsive forces could largely restrict the noncoincidence of the deflected column axes, the actual column axes cannot be coincident. In particular, due to linearized characteristic of column buckling, it is expected that even a small noncoincidence of the deflected column axes could affect the critical axial strain of column buckling. Hence it is significant to quantify the effect of the non-coincidence of the deflected column axes on the column buckling.

III. CRITICAL AXIAL STRAIN

To identify the magnitude of the parameters $c_{i(i+1)}$ $[i=1,2,\ldots,(N-1)]$ which assures that the noncoincidence of the deflected column axes is negligible, let us consider a free-standing DWNT. When $N=2$ and $k=0$, Eqs. (7) become

$$c_{12}[w_2-w_1]+\sigma_x^0A_1\frac{d^2w_1}{dx^2}=EI_1\frac{d^4w_1}{dx^4},$$
$$-c_{12}[w_2-w_1]+\sigma_x^0A_2\frac{d^2w_2}{dx^2}=EI_2\frac{d^4w_2}{dx^4}, \tag{8}$$

where the axial stress is uniform over the entire cross section. Let us consider the hinged boundary conditions.[20,22] Thus, substituting the expressions

$$w_1=f_1\sin\frac{n\pi}{L}x, \quad w_2=f_2\sin\frac{n\pi}{L}x \tag{9}$$

into Eqs. (8), the existence condition for a nonzero solution of the coefficient pair (f_1,f_2) is

$$a(\sigma_x^0)^2+b\sigma_x^0+c=0, \tag{10}$$

where

$$a=\left(\frac{n\pi}{L}\right)^4A_1A_2,$$

$$b=\left(\frac{n\pi}{L}\right)^2A_1\left[EI_2\left(\frac{n\pi}{L}\right)^4+c_{12}\right]+A_2\left(\frac{n\pi}{L}\right)^2\times\left[c_{12}+EI_1\left(\frac{n\pi}{L}\right)^4\right], \tag{11}$$

$$c=EI_1\left(\frac{n\pi}{L}\right)^4\left[EI_2\left(\frac{n\pi}{L}\right)^4+c_{12}\right]+c_{12}EI_2\left(\frac{n\pi}{L}\right)^4.$$

Hence the critical axial strain is

$$-\frac{\sigma_x^0}{E}=\frac{1}{2A_2}\left[I_2\left(\frac{n\pi}{L}\right)^2+\frac{c_{12}}{E(n\pi/L)^2}\right]+\frac{1}{2A_1}\left[\frac{c_{12}}{E(n\pi/L)^2}+I_1\left(\frac{n\pi}{L}\right)^2\right]-\left(\left\{\frac{1}{2A_2}\left[I_2\left(\frac{n\pi}{L}\right)^2+\frac{c_{12}}{E(n\pi/L)^2}\right]-\frac{1}{2A_1}\left[\frac{c_{12}}{E(n\pi/L)^2}+I_1\left(\frac{n\pi}{L}\right)^2\right]\right\}^2+\frac{c_{12}^2}{A_1A_2E^2(n\pi/L)^4}\right)^{1/2}. \tag{12}$$

When the interlayer radial displacements are prohibited, the DWNT is equivalent to a single hollow column of the thickness $(2t)$. In this case, the critical axial strain is given by the classic formula[20]

$$-\frac{\sigma_x^0}{E}=\frac{I}{A}\left(\frac{n\pi}{L}\right)^2, \quad I=I_1+I_2, \quad A=A_1+A_2, \tag{13}$$

where I and A are the total area and total moment of inertia of the entire cross section. Hence the difference between the critical strains given by Eqs. (12) and (13) represents the effect of the interlayer radial displacement on column buckling. Now let us find out the condition under which the expression (12) reduces to Eq. (13).

It is expected that the magnitude of the parameter c_{12} is essential. To see this, first, if the parameter c_{12} is so small that

$$\frac{c_{12}}{EAR^2}\left(\frac{L}{n\pi}\right)^4 \ll 1, \tag{14}$$

then it can be verified that expression (12) reduces to

$$-\frac{\sigma_x^0}{E}=\frac{I_1}{A_1}\left(\frac{n\pi}{L}\right)^2. \tag{15}$$

The interpretation of Eq. (15) is simple: when the van der Waals interaction is absent, the innermost tube behaves like an isolated tube and then buckles first. In this case, the critical strain [Eq. (15)] is lower than Eq. (13), indicating that the critical strain of the DWNT is determined by the innermost tube of lower buckling strength. It is easy to understand that this reduction of the critical strain could be substantial when the number of the inner layers is so large that the innermost tube has a very small radius and buckling strength. Hence the existing single-column model could substantially overestimate the critical axial strain for linearized column buckling of a MWNT when the van der Waals interaction is extremely weak.

Here it should be stated that mechanical and electronic properties of carbon MWNT's could be very sensitive to even small radial distortion (see, e.g., Ref. 14) or buckling deflection of individual inner tubes in some applications. For instance, carbon MWNT's have been used as tips for atomic force microscopy.[23] In that case, interlayer radial displacements caused by column buckling of the inner tubes could significantly affect the mechanical and electronic performance of the MWNT. This also justifies the need to quantify the interlayer radial displacements and their effect on linearized column buckling of carbon MWNT's.

On the other hand, if the parameter c_{12} is extremely large, so that

$$\frac{c_{12}}{E\pi R^4}\left(\frac{L}{n\pi}\right)^4 \gg 1, \tag{16}$$

it can be verified that the square root in Eq. (12) becomes

$$\frac{1}{2}\left(\frac{1}{A_2}+\frac{1}{A_1}\right)\frac{c_{12}}{E\left(\frac{n\pi}{L}\right)^2}+\frac{\left(\frac{1}{A_2}-\frac{1}{A_1}\right)\left(\frac{I_2}{A^2}-\frac{I_1}{A_1}\right)}{2\left(\frac{1}{A_2}+\frac{1}{A_1}\right)}\left(\frac{n\pi}{L}\right)^2,$$

and then the critical axial strain [Eq. (12)] is given by

$$-\frac{\sigma_x^0}{E}=\frac{1}{2}\left(\frac{n\pi}{L}\right)^2\left[\frac{I_2}{A_2}+\frac{I_1}{A_1}-\frac{(A_1-A_2)(A_1I_2-I_1A_2)}{A_1A_2(A_1+A_2)}\right]$$
$$=\left(\frac{n\pi}{L}\right)^2\frac{I_1+I_2}{(A_1+A_2)}, \tag{17}$$

which is identical to Eq. (13), given by the existing single-column model. Hence the existing single-column model gives the correct critical axial strain when the van der Waals interaction between adjacent nanotubes is strong enough that condition (16) is met. Because the coefficient c_{12} is proportional to the radius, it is noted that condition (16) is met for a MWNT if it is met for its outermost radius.

To examine the implications of condition (16), let us discuss carbon nanotubes. Let the interaction energy potential (per cm^2) between two monolayer graphite sheets be $g(\delta)$, according to the data given in Girifalco and Lad,[18] it is found that

$$\left.\frac{d^2g}{d\delta^2}\right|_{\delta=2.4d}\sim\frac{200\ \text{erg/cm}^2}{0.16d^2},\quad d=1.42\times10^{-8}\ \text{cm},$$

then

$$c_{12}=\frac{200(2R)\ \text{erg/cm}^2}{0.16d^2},$$

where R (measured in cm) denotes the inner radius of each pair of nanotubes. For instance, if $R=5$ nm, then it is found that

$$c_{12}=10^{13}\ \text{erg/cm}^3=1\ \text{TPa}. \tag{18}$$

Since the order of magnitude of E for carbon nanotubes is about 1 TPa (for a review of recent results, see Popov, Doren, and Balkanski in Ref. 3), condition (16) for carbon nanotubes of the outermost radius 5 nm reads, approximately,

$$\frac{1}{\pi}\left(\frac{L}{n\pi R}\right)^4 \gg 1. \tag{19}$$

For carbon MWNT's of smaller outermost radius, condition (16) has a form close to Eq. (19). For instance, when the outermost radius is 1 nm and $E=1$ TPa, condition (16) becomes

$$\frac{1}{5\pi}\left(\frac{L}{n\pi R}\right)^4 \gg 1.$$

Because the above condition and condition (19) are proportional to the fourth power of (L/R), the restriction imposed on (L/R) by the above condition is only slightly different from that imposed by condition (19). Hence, without loss of the generality, all subsequent discussions are given for condition (19).

Roughly, condition (19) requires that the half-wavelength of the buckling mode is much (say, at least a few times) larger than the outermost diameter of the MWNT. In other words, the van der Waals forces between adjacent nanotubes can be regarded as ''infinitely'' strong if condition (19) is met.

When the van der Waals forces are infinitely strong, all jumps of the deflection between any two adjacent nanotubes should be infinitely small (compared to the deflection itself) because all van der Waals interaction terms in Eq. (7) must be bounded. This implies that all deflection curves are identical and then $w_i(x)=w(x)$ for $i=1,2,\ldots,N$. In this extreme case, summing all of Eqs. (7), one can obtain

$$-kw(x)+\sigma_x^0(A_1+A_2+\cdots+A_N)\frac{d^2w}{dx^2}$$

$$=E(I_1+I_2+\cdots+I_N)\frac{d^4w}{dx^4}, \tag{20}$$

where all subscripts for $w(x)$ have been dropped. Obviously, Eq. (20) is identical to the governing equation of a MWNT when it is treated as a single column having the total area A and the moment of inertia I. In this case, if the hinged end conditions are assumed, Eq. (20) leads to the critical axial stress

$$-\sigma_x^0=\frac{k}{A(n\pi/L)^2}+\frac{EI}{A}\left(\frac{n\pi}{L}\right)^2, \tag{21}$$

which is equivalent to the formula used in Ref. 7 (where a factor of 4 was multiplied to the first term because they assumed clamped end conditions). Hence, when the van der Waals interaction is extremely strong, as defined by Eq. (19), the interlayer radial displacements can be neglected as compared to the deflection itself, and the existing single-column model [Eq. (20)] can be applied to carbon MWNT's.

For example, when a very compliant surrounding elastic medium (such as a polymer) is used, the half wave number n is very small (for instance, it is 1 or 2 in Ref. 7) for carbon MWNT's of moderate aspect ratio. In this case, condition (19) holds well, and the existing single-column model can be used to study column buckling. On the other hand, if the surrounding elastic medium is reasonably stiff and the aspect ratio of the MWNT is sufficiently large so that

$$\frac{kL^4}{EI}\gg 1, \tag{22}$$

then the integer n is large compared to unity, and the half-wavelength is given by

$$\frac{L}{n\pi}=\left(\frac{EI}{k}\right)^{1/4}. \tag{23}$$

In this case, condition (19) reads

$$\frac{EI}{kR^4}\gg 1. \tag{24}$$

It is noted that condition (24) could be consistent with Eq. (22) only when the spring constant k [defined for per unit axial length; see Eq. (6)] is much smaller than the Young's modulus E, but the length of the column is much larger than the outermost radius. For instance, when a MWNT surrounded by many other nanotubes (such as nanotube ropes) is considered, it seems that the spring constant k could have an order of magnitude close to c_{12} estimated by Eq. (18). If it is the case, condition (22) holds for MWNT's of large aspect ratio, but condition (24) could fail. This suggests that the existing single-column model cannot be applied to a MWNT embedded in a very stiff elastic medium whose spring constant is of an order of magnitude close to the Young modulus of carbon nantotubes.

IV. CONCLUSIONS

An elastic model is presented for column buckling of carbon MWNT's, which allows for interlayer radial displacements between adjacent nanotubes. It is found that the noncoincidence of the deflected column axes is negligible provided the van der Waals interaction is so strong that condition (16) is met. For carbon nanotubes, condition (16) holds if the half-wavelength of buckling mode is much (say, at least a few times) larger than the outermost diameter. Under this condition, the existing single-column model is applicable for column buckling of carbon MWNT's. However, the existing model is questionable when the half-wavelength is close to or smaller than the outermost radius. For example, this suggests that the existing single-column model cannot be applied to a MWNT surrounded by a very stiff elastic medium.

These results on column buckling are in sharp contrast to axially compressed shell buckling of carbon MWNT's, for which study[19] has indicated that interlayer displacements drastically reduce the critical axial strain of DWNT's, regardless of the magnitude of the van der Waals forces. This distinction is due to the fact that the bending stiffness of all tubes of a MWNT, as a column, is measured from the common (undeformed) central axis, and then will not change when some inner tubes are inserted or removed. On the other hand, the bending stiffness of a MWNT as a shell is measured from its middle face, and then changes when some inner tubes are inserted or removed. This explains why interlayer displacements have substantial influence on shell buckling of MWNT's, as shown in Ref. 19 for DWNT's. Finally, it should be stated that the multiple-column model proposed here could be used to study other phenomena caused by interlayer displacements in carbon MWNT's, such as intrinsic dynamic damping due to interlayer friction, and the end deformation of clamped MWNT's in which all inner tubes are free or hinged at the end whereas the outermost tube is clamped.

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