Prismatic dislocation loops in strained core-shell nanowire heterostructures

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The formation of prismatic dislocation loops in a core nanowire coated with a thin shell has been investigated in the hypothesis where the core-shell interface is coherent leading to misfit stress. In the framework of a thermodynamic equilibrium approach, the energy variation from the dislocation-free heterostructure has been calculated and the equilibrium positions of the loops have been determined as a function of the misfit stress, the nanowire radii, and the ratio of the shear modulus between the two phases. Depending on misfit strain and radii, it is found that for a sufficiently soft core with respect to the shell, prismatic dislocation loops may form into the inner nanowire with equilibrium positions located at a few interatomic distances from the interface.

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Nanowires and nanowhiskers have recently attracted much attention because of their increasing importance in fundamental research on electrical transport[1] and growth processes[2] at the nanoscale or on optical properties of low-dimensional structures.[3,4] They are also used for a wide range of applications in nanoelectronics, including field-effect transistors' logical gates and single-electron transistors[5,6] or optoelectronic devices.[7,8] Composite nanowires can be produced either by one-dimensional modulation of nanowire composition and doping[9–11] or by modulation[12] of the radial composition and doping in the nanowires. When coherent interfaces are present in such heterostructures, the resulting misfit stress may affect their plasticity. In the case of a misfit layer coherently grown at the top of a free-standing nanowhisker, a theoretical study of the nucleation of interfacial dislocations has been performed in the framework of continuum mechanics and a critical layer thickness depending on the radius of the structure has been determined.[13] The nucleation of dislocations in strained core nanowires embedded in matrices has been also considered. In the case of a stressed core nanowire embedded in a finite-size shell of identical elastic constants, Ovid'ko *et al.*[14,15] have determined the critical thickness of the inner wire beyond which edge dislocations and prismatic dislocation loops can develop at the coherent interface to release the misfit stress. In a strained annular film grown on a nanopore incorporated into an infinite-size matrix, it has been demonstrated that the ratio of the shear modulus between the two phases influences the critical thickness of the film beyond which the formation of screw misfit dislocations at the film/matrix interface is energetically favorable.[16] Likewise, the formation of screw dislocation dipoles to release the misfit stress has been investigated for a strained cylindrical precipitate embedded in a matrix of infinite size with a coherent precipitate/matrix interface.[17] It has been found that for a softer matrix than the precipitate, the formation of such dipoles may be energetically favorable in the matrix at a finite distance from the interface, this distance increasing with the shear modulus ratio between the matrix and the precipitate. When a core-shell nanowire is embedded in an infinite matrix, the equilibrium positions of edge dislocations have been also found to depend on the interfacial stress and the elastic constants of the two phases.[18] The shear modulus effect has already been analyzed in a planar structure composed of a thin film coherently strained on a semi-infinite substrate.[19] It has been demonstrated that rows of misfit edge dislocations are repelled from the interface into the softer material. High-resolution TEM observations of the interface between an alumina precipitate and a niobium matrix, with a relative shear modulus between the precipitate and the matrix on the order of 3.7, have also evidenced the presence of misfit dislocations in the matrix at four interatomic distances from the interface.[20]

In this paper, the problem of the formation of prismatic dislocation loops is investigated in a core-shell heterostructure composed of a core nanowire coherently strained in a thin shell matrix of different shear modulus. In the framework of a thermodynamic equilibrium approach, the energy variation from the configuration free of dislocation is determined and the loop equilibrium positions with respect to the interface have been studied as a function of the ratio of the shear modulus, the inner and outer radii and the misfit stress.

A cylindrical inner nanowire of radius ρ_i, shear modulus μ_i, and Poisson's ratio ν_i is considered in a thin shell of outer radius ρ_o, shear modulus μ_o, and Poisson's ratio ν_o. Classical cylindrical coordinate system (ρ,θ,z) is used and both nanowires are supposed to be infinitely long along their common (Oz) axis. The misfit stresses in both materials resulting from the coherent interface between the two phases has already been determined in a number of papers.[14,15,21–24] Raychaudhuri and Yu[21] have studied the case where both core and shell nanowires have the wurtzite crystal structure with the [0001] direction along the axis of the wire. Considering the full elastic stiffness matrices of the materials, they have analyzed the stress field when the longitudinal misfit strain is different from the radial one. Likewise, in the case of two cubic crystals, Trammel *et al.*[22] have calculated the stress as a function of the stiffness matrix coefficients when the lattice parameters of the core (i) and shell (o) are such that $a_\theta^i = a_\theta^o$, $a_z^i = a_z^o$, and $a_\rho^i \neq a_\rho^o$. Using isotropic and linear elasticity, Ovid'ko *et al.*[15] have also investigated the stress field in nanowires when a dilatational misfit strain $\epsilon_{ij}^* = \epsilon^* \delta_{ij}$ is present in the inner nanowire and the elastic constants of both materials are equal, with ϵ^* a constant and $\delta_{ij}=1$ when $i=j$ and $=0$ when $i \neq j$. In this paper, the formalism used by the previously cited authors has been considered to determine, within the framework of linear and isotropic elasticity, the stress in a core-shell heterostructure when the lattice parameters of both core and shell materials of different elastic constants lead to isotropic eigenstrain which can be located in the inner nanowire: $\epsilon^*_{\rho\rho}=\epsilon^*_{\theta\theta}=\epsilon^*_{zz}=\epsilon^*$, with ϵ^* a constant. The effect of interface stress and anisotropy of the eigenstrain as the size of the nanowires reduces are beyond the scope of the present analysis. The different steps of the calculation can be then summarized as follows. The resulting elastic state of the core and shell has been determined using the following form[24,25] of the misft elastic displacement field $u_\rho^{m,p}(\rho,z)=A_p^m+B_p^m/\rho$, $u_\theta^{m,p}(\rho,z)=0$, $u_z^{m,p}(\rho,z)=C_p^m z$, with A_p^m, B_p^m and C_p^m three coefficients and $p=i,o$ in the inner and outer nanowire, respectively. The misft strain and stress tensors, respectively, labeled $\bar{\bar{\epsilon}}^{m,p}$ and $\bar{\bar{\sigma}}^{m,p}$ are then determined from $\mathbf{u}^{m,p}$ using the classical laws of linear and isotropic elasticity expressed in cylindrical coordinates.[25] Assuming a finite value of the elastic displacement at $\rho=0$, one takes $B_i=0$ and the other coefficients are determined with the help of the following conditions that guarantee the mechanical equilibrium of the heterostructure. At the interface ρ_i, one writes $(\bar{\bar{\sigma}}^{m,i}-\bar{\bar{\sigma}}^{m,o})\cdot\mathbf{n}^i=0$ and $\mathbf{u}^{m,i}+\epsilon^*\cdot\mathbf{x}=\mathbf{u}^{m,o}$, with $\mathbf{n}^i$ the unit normal vector to the interface pointing into the shell and $\mathbf{x}$ the position vector. At the outer radius ρ_o, one takes $\sigma^{m,o}\cdot\mathbf{n}^o=0$ with $\mathbf{n}^o$ the unit normal vector to the free surface pointing into the vacuum, and $(\rho_o^2-\rho_i^2)\sigma_{zz}^{m,o}+\rho_i^2\sigma_{zz}^{m,i}=0$ in the cross section of the heterostructure. The final cumbersome expressions of the misfit strain and stress tensor components determined with the help of a CALCULUS software[30] are not displayed in this paper. The elasticity problem of prismatic dislocation loops lying into the interface between the core and shell nanowires of equal elastic coefficients has already been considered.[14,15,21,22] In the following, the case of an isolated dislocation loop is addressed when the loop is not constraint into the interface, the core and shell nanowires having different elastic constants. At this point, it can be underlined that the study of the full relaxation mechanism of misft stresses would require to consider a periodic array of dislocation loops lying along the nanostructure axis. However, it is assumed that when the misft is sufficiently small such that the distance between two consecutive loops is greater than the diameter of the core nanowire (and much greater than the dislocation-interface distance), the effect of the other dislocations onto the equilibrium position of each loop can be neglected. The following calculation has been performed in this hypothesis. A prismatic dislocation loop whose center is located at the center line $\rho=0$ is first considered in an infinite media of the same elastic constants as the inner nanowire, with (Oz) its symmetry axis. The radius of the loop is labeled ρ_d, its Burgers vector $\mathbf{b}=b_z\mathbf{e}_z$, with $\mathbf{e}_z$ a unit vector along (Oz) axis. It is well known that the elastic stress tensor and displacement field of such a loop are fully determined from a biharmonic stress function ϕ_∞^i satisfying $\Delta^2\phi_\infty^i(\rho,z)=0$ and given by[26,27]

$$\phi_\infty^i(\tilde{\rho},\tilde{z})=\frac{\mu_i b_z\rho_d^2}{2(1-\nu_i)}\int_0^{+\infty}\left(2\nu_i\frac{|\tilde{z}|}{z}+k|\tilde{z}|\right)J_1(\tilde{k})J_0(k\tilde{\rho})\frac{e^{-k|\tilde{z}|}}{k^2}dk \tag{1}$$

with Δ the Laplacian, J_0 and J_1 the Bessel functions of the first kind of zero and first order, respectively, $\tilde{z}=z/\rho_d$ and $\tilde{\rho}=\rho/\rho_d$. When the loop is now introduced into the inner nanowire coated with its thin shell, the stress function is modified as follows: $\phi^p(\tilde{r},\tilde{z})=\phi_\infty^p(\tilde{\rho},\tilde{z})+\phi_{rel}^p(\tilde{\rho},\tilde{z})$ with $p=i,o$ and $\phi_\infty^o=0$. The general expression of the relaxation part of the stress function ϕ_{rel}^p due to the core/shell interface and shell free surface is written as[25]

$$\phi_{rel}^p(\tilde{\rho},\tilde{z})=\frac{\mu_i b_z\rho_d^2}{2(1-\nu_i)}\int_0^{+\infty}[I_0(k\tilde{\rho})g_1^p(k)-k\tilde{\rho}I_1(k\tilde{\rho})g_2^p(k)+K_0(k\tilde{\rho})g_3^p(k)-k\tilde{\rho}I_1(k\tilde{\rho})g_4^p(k)]\sin k\tilde{z}\,dk, \tag{2}$$

where I_n and K_n are the modified Bessel functions of the first and second kind, respectively, of order n, with $n=0,1$. The g_j^p coefficients, with $j=1,2,3,4$ and $p=i,o$, are determined with the help of the mechanical equilibrium equations which have already been used in the first step of this work to determine the misft stress, i.e., continuity of the normal traction and total displacement at the core/shell interface, the shell free surface being traction free. Following Refs. 14, 15, and 25, the sine and cosine Fourier transforms of the initial stress components $\sigma_{ij}^{\infty,p}$ have been first expressed using integrals of Lipschitz-Hankel type.[28] The above-mentioned equations resulting from the equilibrium conditions at the interface and free surface have been then solved in the Fourier space.[30] The heavy but straightforward calculations are not detailed in this paper, the cumbersome expressions of the g_j^p coefficients are neither given. Once the total stress tensor $\bar{\bar{\sigma}}^p=\bar{\bar{\sigma}}^{\infty,p}+\bar{\bar{\sigma}}^{rel,p}$ has been determined in the heterostructure, with $\bar{\bar{\sigma}}^{\infty,p}$ and $\bar{\bar{\sigma}}^{rel,p}$ the stress tensors derived within elasticity theory[25] from ϕ_∞^p and ϕ_{rel}^p, respectively, the formation of the prismatic dislocation loop has been investigated from an energetic point of view, the kinetics of the loop being beyond the scope of the present analysis. Likewise, the nucleation process of the loop has not been addressed in this work based on a continuum mechanics approach and the "initial state" for the dislocation has been taken to be a small loop of core radius $\rho_c=b_z$ located at the center of the nanowire. The energy variation $\Delta\mathcal{F}$ due to the spreading of the loop from the center of the core nanowire until a given radius $\rho_d<\rho_i$ is determined with respect to the dislocation free heterostructure.[14,15,29] It is the sum of three terms, the proper energy $\Delta\mathcal{F}_{pr}$ defined by

$$\Delta\mathcal{F}_{pr}=\frac{b_z}{2}\int_0^{2\pi}\int_0^{\rho_d-b_z}[\sigma_{zz}^{\infty,i}(\tilde{\rho},\tilde{z}=0)+\sigma_{zz}^{rel,i}(\tilde{\rho},\tilde{z}=0)]\rho d\rho d\theta, \tag{3}$$

the energy term $\Delta\mathcal{F}_{mi}$ characterizing the interaction between the misft stress and the loop,

$$\Delta\mathcal{F}_{mi}=b_z\int_0^{2\pi}\int_0^{\rho_d}\sigma_{zz}^{m,i}(\tilde{\rho},\tilde{z}=0)\rho d\rho d\theta, \tag{4}$$

and the core energy $\Delta\mathcal{F}_{co}$,

$$\Delta\mathcal{F}_{co}=\frac{\mu_i b_z^2\rho_d}{2(1-\nu_i)}. \tag{5}$$

One finally gets

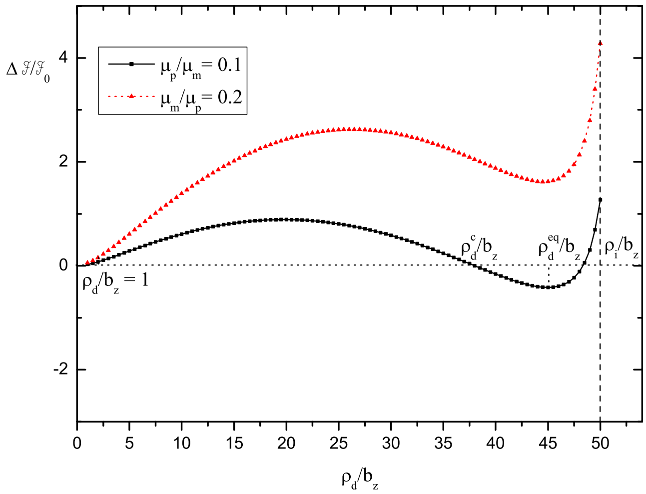

FIG. 1. (Color online) $\Delta\mathcal{F}/\mathcal{F}_0$ versus ρ_d/b_z for μ_i=10 GPa and μ_i=20 GPa, with μ_o=100 GPa, $\nu_i=\nu_o$=0.3, $r_i=50b_z$, and $r_o=65b_z$.

$$\Delta\mathcal{F}=\mathcal{F}_0\frac{\mu_i}{\mu_o}\left\{\frac{1}{\pi}\frac{\rho_d}{b_z}+4(1-\nu_p)\left(\frac{\rho_d}{b_z}\right)^2\epsilon^*\frac{\chi_1}{\chi_2}+\frac{2}{\pi}\frac{\rho_d}{b_z}[K(\tilde{x})-E(\tilde{x})]+\frac{\rho_d}{b_z}\int_0^{+\infty}\psi(k,\tilde{x})dk\right\} \tag{6}$$

with K and E the complete elliptic integrals of the first and second kind, respectively, $\mathcal{F}_0=\mu_o b_z^3\pi/[2(1-\nu_i)]$ and $\tilde{x}=1-b_z/\rho_d$. The functions χ_1, χ_2, and ψ are defined by

$$\chi_1=-\mu_i(1+\nu_o)(1+\nu_i)(\rho_o^2-\rho_i^2)[(\mu_o+\mu_i)\rho_o^2+(\mu_i-\mu_o)\rho_i^2], \tag{7}$$

$$\chi_2=(1+\nu_i)\mu_i^2\rho_i^2[\rho_o^2+(1-2\nu_o)\rho_i^2]+(1+\nu_o)(1-2\nu_i)\mu_o^2(\rho_o^2-\rho_i^2)^2+\mu_i\mu_o(\rho_o^2-\rho_i^2)[(1+\nu_o)\rho_o^2+(2-\nu_o-\nu_i-4\nu_i\nu_o)\rho_i^2], \tag{8}$$

$$\psi(k,\tilde{x})=k^2\tilde{x}I_1(k\tilde{x})[g_1^i(k)-2(2-\nu_i)g_2^i(k)]-kg_2^i(k)[k^2\tilde{x}^2I_0(k\tilde{x})-2k\tilde{x}I_1(k)]. \tag{9}$$

Following Ovid'ko *et al.*,[14,15] the formation of prismatic dislocation loops of radius ρ_d is assumed to be energetically favorable when $\Delta\mathcal{F}<0$. To investigate the effect of misft stress and shear moduli of both phases on the formation into the core nanowire of the dislocation loop, the last integral in the expression of the total-energy variation given in Eq. (6) has been numerically estimated[30] and $\Delta\mathcal{F}/\mathcal{F}_0$ has been plotted in Fig. 1 as a function of ρ_d with $b_z\le\rho_d<\rho_i$, for two different values of μ_i and for $\rho_i=50b_z$, $\rho_o=65b_z$, $\epsilon^*=0.66\%$, μ_o=100 GPa, and $\nu_i=\nu_o$=0.3. It is found that whereas $\Delta\mathcal{F}/\mathcal{F}_0$ is always positive for μ_i=20 GPa, such that the formation of the loop inside the core nanowire is not energetically favorable, $\Delta\mathcal{F}/\mathcal{F}_0$ becomes negative beyond $\rho_d^c\sim 38b_z$ for μ_i=10 GPa and is minimum (and negative) for $\rho_d^{eq}\sim 45b_z$. It is thus suspected that when the core is sufficiently soft with respect to the shell, one possible relaxation mechanism of the strained heterostructure may result in the formation of prismatic dislocation loops whose equilibrium positions, depending on misft strain, are not into the core/shell interface but in the nanowire at a few interatomic distances from it. This result can be qualitatively understood by analyzing the sign of the different forces acting onto the loop. Indeed, whereas the misft stress attracts the loop into the interface, the force resulting from the difference between both shear moduli, in addition with the shrinking effect of the line tension, is supposed to repel the dislocation from it. Depending on the relative values of the different forces, the equilibrium positions of the loop can thus be located in the nanowire. These coupled effects between the shear moduli and misft strain onto the loop equilibrium positions has been illustrated in Fig. 2 where ρ_d^{eq} has been plotted as a function of ϵ^* for different values of the ratio of the shear modulus between the two phases (μ_o being constant). Since the spreading of the core of the dislocation into the interface is assumed to be beyond the scope of this paper, the determination of ρ_d^{eq} has been cut off in the following when ρ_d^{eq} reaches $\rho_i-0.49b_z$. It is observed that for fixed values of the

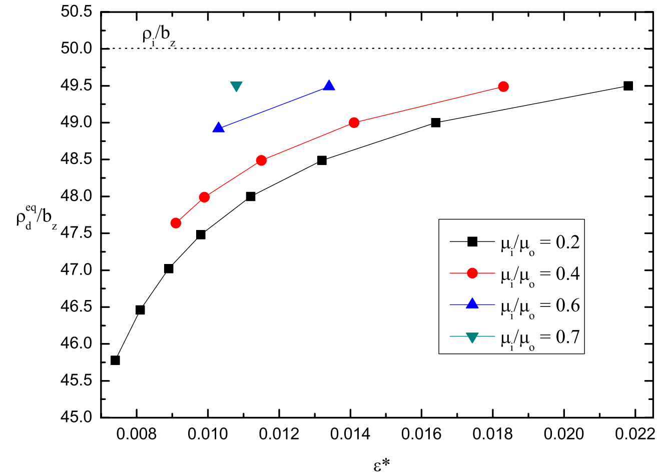

FIG. 2. (Color online) ρ_d^{eq}/b_z versus ϵ^* for different values of μ_i, with μ_o=100 GPa, $\nu_i=\nu_o$=0.3, $r_i=50b_z$, and $r_o=65b_z$.

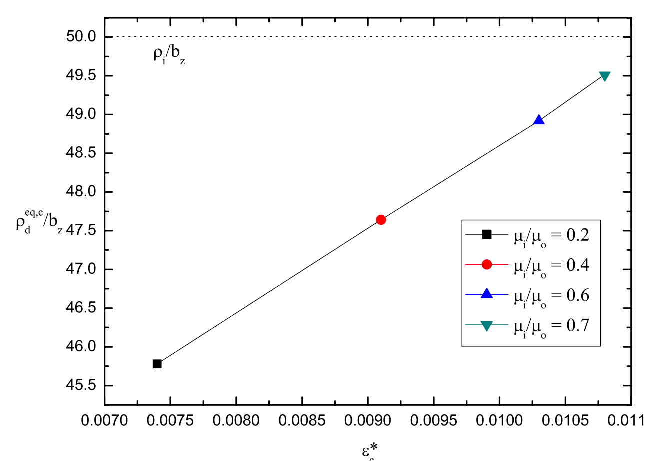

FIG. 3. (Color online) $\rho_d^{eq,c}/b_z$ versus ϵ_c^* for different values of μ_i, with μ_o=100 GPa, $\nu_i=\nu_o$=0.3, $r_i=50b_z$, and $r_o=65b_z$.

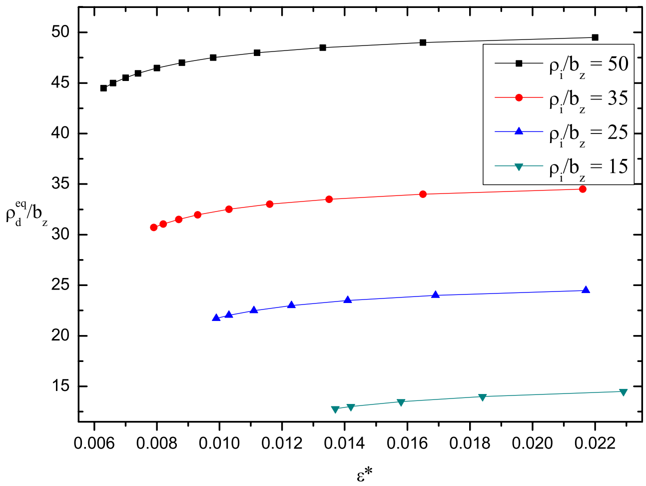

FIG. 4. (Color online) ρ_d^{eq}/b_z versus ϵ^* for different values of ρ_i and ρ_o, with $\rho_o-\rho_i=15b_z$, $\nu_i=\nu_o=0.3$, $\mu_i=10$ GPa, and $\mu_o=100$ GPa.

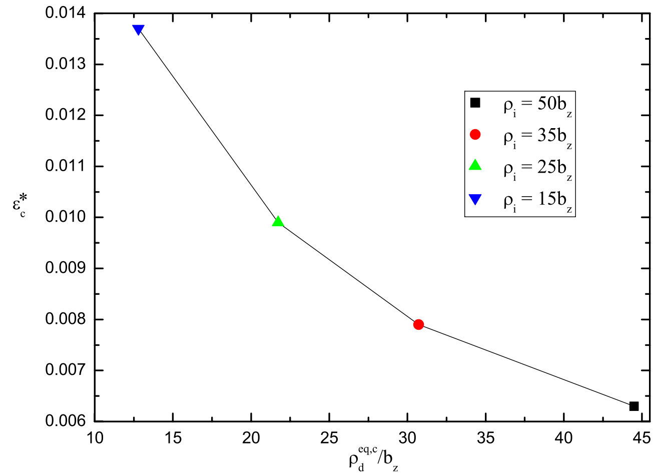

FIG. 5. (Color online) ϵ_c^* versus $\rho_d^{eq,c}/b_z$ for different values of ρ_i and ρ_o, with $\rho_o-\rho_i=15b_z$, $\nu_i=\nu_o=0.3$, $\mu_i=10$ GPa, and $\mu_o=100$ GPa.

ρ_i and ρ_o radii, the range of ρ_d^{eq} values increases as the ratio μ_i/μ_o decreases (with $\mu_o=100$ GPa). A critical misft strain ϵ_c^* can also be defined from Fig. 2 below which the total-energy variation $\Delta\mathcal{F}$ becomes positive such that no equilibrium position in the inner nanowire can be found for the loop. It corresponds to the strain threshold for which the minimum of $\Delta\mathcal{F}$ goes to zero (with negative values) as the increasing loop/interface distance reaches its maximum value. It allows thus to define $\rho_d^{eq,c}$ the critical equilibrium radius of the loop in the inner nanowire corresponding to the minimum value of ρ_d^{eq}. It can be underlined that $\rho_d^{eq,c}$ obtained for ϵ_c^*, also decreases with μ_i/μ_o. On the other hand, for $\mu_i/\mu_o \geq 0.7$, the loop is suspected to develop with an equilibrium radius $\rho_d^{eq}>49.51b_z$ such that it has been considered as laying into the interface. In Fig. 3, $\rho_d^{eq,c}$ has been plotted versus ϵ_c^* for the set of shear modulus ratios already used in Fig. 2 (with $\mu_o=100$ GPa). The almost linear growth of this critical equilibrium radius with the critical misft strain is then clearly illustrated in the range $\epsilon_c^* \in [0.0074, 0.0108]$. The geometric effect of the radii onto the loop equilibrium positions has been finally characterized in Figs. 4 and 5 in the hypothesis where the thickness of the shell $\rho_o-\rho_i$ remains constant and egal to $15b_z$, with $\mu_i=10$ GPa and $\mu_o=100$ GPa. In Fig. 4, it can be observed that as the radius of the inner nanowire decreases from $50b_z$ to $15b_z$, the equilibrium position of the loop can still be inside the nanowire rather than into the interface, depending on the misft strain whose range of values decreases with ρ_i. The critical misft strain ϵ_c^* has been plotted in Fig. 5 versus the critical equilibrium radius $\rho_d^{eq,c}$. It is found that in the radius range $[15b_z, 50b_z]$, the misft strain required to reach this loop equilibrium position is increased at constant shell thickness as the inner radius ρ_i decreases. Finally, it can be emphasized that the results obtained in this work may apply to a wide range of oxide/metal interfaces in nanostructures of technological interest (in microelectronics or aeronautics for example) for which the shear modulus ratio can take high values, for example on the order of 5.5 for Al_2O_3/Al nanocomposites. Indeed, since the strength and plasticity of such nanowires are structure dependent, it appears that the control of their nanoscale interface properties through the control of the dislocation positions is of paramount importance.

In this paper, the possibility of formation of prismatic dislocation loops in a nanowire, at a few interatomic distances from its interface with a harder thin outer shell has been investigated and the coupling effects between the shear moduli, the inner and outer radii and the misft strain has been identified. It is believed that this study raises further problems concerning the plasticity of confined nanostructures among which are the localization of the preferential nucleation areas of the loop (the inner nanowire, the free surface of the shell?) and the possible interface crossover of the loops. A coupled approach based on continuum mechanics and atomistic-scale description of the dislocation formation through molecular-dynamics simulations would help in answering to these fundamental questions.

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