Comment on "Effects of shear methods on shear strengths and deformation modes of two typical transition metal carbides and their unification"

Marcin Maździarz*
Institute of Fundamental Technological Research, Polish Academy of Sciences, Pawińskiego 5B, 02-106 Warsaw, Poland

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Recently, Li *et al.* [C. Li *et al.*, Phys. Rev. B **107**, 224106 (2023)] investigated the mechanical behavior of cubic HfC and TaC under simple shear and pure shear using first-principles calculations. Unfortunately, the paper contains some serious and fundamental flaws in the field of continuum mechanics and nanomechanics. The results presented appear to be qualitatively and quantitatively incorrect; they would be correct if we were in the small/linear deformation/strain regime, which we are not. A correct description therefore requires a finite/nonlinear deformation/strain apparatus. The solution for simple shear, even from density functional theory calculations, must follow Frenkel's sinusoidal solution.

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In the paper by Li *et al.* [1], cubic HfC and TaC crystals were subjected to simple shear (SS) and pure shear (PS) by density functional theory (DFT) simulations. The paper mistakes concepts from the continuum mechanics, incorrectly enforces deformations, and it appears that the computational results are qualitatively and quantitatively incorrect. Even when we perform DFT calculations, they must not be inconsistent with classical solutions, and so for simple shear they must be consistent with Frenkel's classical sinusoidal solution [2].

Li *et al.* write the following: "During deformation, the lattice vectors need to be changed,. . .. Hence, the deformation can be imposed by transforming the $i-1$th step lattice vector matrix $\mathbf{R}^{i-1}$ to the deformed ith step lattice vector matrix $\mathbf{R}^{i}$ as follows [36]:

$$\mathbf{R}^{i}=\mathbf{R}^{i-1}\left[\mathbf{I}+\begin{pmatrix}0 & 0 & 0\\ 0 & 0 & 0\\ 0 & \Delta\varepsilon_{zy} & 0\end{pmatrix}\right].\text{"} \tag{1}$$

This formula is incorrect.

In the nonlinear mechanics of crystals there is a well-known hypothesis, called the Cauchy-Born rule [3], which assumes that under a homogeneous macroscopic deformation, the primitive Bravais lattice vectors of a three-dimensional crystal deform in an affine manner via a 3×3 matrix $\mathbf{F}$,

$$\mathbf{a}_i=\mathbf{F}\mathbf{A}_i, \tag{2}$$

where $\mathbf{a}_i$ stands for spatial lattice vectors, $\mathbf{A}_i$ denotes reference vectors, and $\mathbf{F}$ is the deformation gradient [4].

Continuum mechanics has a well-established methodology for describing arbitrary finite (not small, linearized) deformations. The Cauchy-Born rule does nothing more than replace infinitesimal elements dx and dX by lattice vectors. The definition and interpretation of the deformation gradient $\mathbf{F}$ is also known; it is a multiplicative assembly of rotation and stretch (see polar decomposition of the deformation gradient [5]).

If we even wanted to do it in steps, it is clear from the tensor calculus point of view that Eq. (1) should be

$$\mathbf{R}^{\mathrm{def}}=\mathbf{F}\cdot\mathbf{R}^{\mathrm{ini}}. \tag{3}$$

The vector, or matrix of vectors, is multiplied by the matrix on the left; in continuum mechanics, using $\mathbf{F}$ on the right side of a vector is unimaginable. However, it may be worthwhile to use well-established approaches, especially as they come from such giants of intellect like Cauchy and Born. Moreover, the components of the deformation gradient $\mathbf{F}$ are not, in general, components of the strain tensor.

Li *et al.* write further: "For PS, after each ε_{zy}, the atomic coordinates and the other five independent strain components (except ε_{zy}) are optimized simultaneously to reach a stress state with $\sigma_{xx}=\sigma_{yy}=\sigma_{zz}=\sigma_{xy}=\sigma_{zx}=0$ [35,37,38]. For SS, after applying ε_{zy}, the atomic coordinates are optimized but remain $\varepsilon_{xx}=\varepsilon_{yy}=\varepsilon_{zz}=\varepsilon_{xy}=\varepsilon_{zx}=0$ [14,39]. The schematics of PS and SS are illustrated in Figs. 1(a) and 1(b), respectively."

The above excerpt from Ref. [1] contains several misrepresentations. In general, the Lagrangian finite strain tensor $\mathbf{E}$, also called the Green-Lagrangian strain tensor or Green–St-Venant strain tensor, is defined as

$$\mathbf{E}=\tfrac{1}{2}(\mathbf{F}^{T}\mathbf{F}-\mathbf{I}) \tag{4}$$

and its linear approximation, the infinitesimal strain tensor ε, also called the Cauchy strain tensor, linear strain tensor, or small strain tensor, takes the form

$$\varepsilon=\tfrac{1}{2}(\mathbf{F}^{T}+\mathbf{F})-\mathbf{I}. \tag{5}$$

The classical finite simple shear deformation is an isochoric plane deformation defined by the deformation gradient tensor $\mathbf{F}$ in the following form [6] (and this implies the form

*mmazdz@ippt.pan.pl

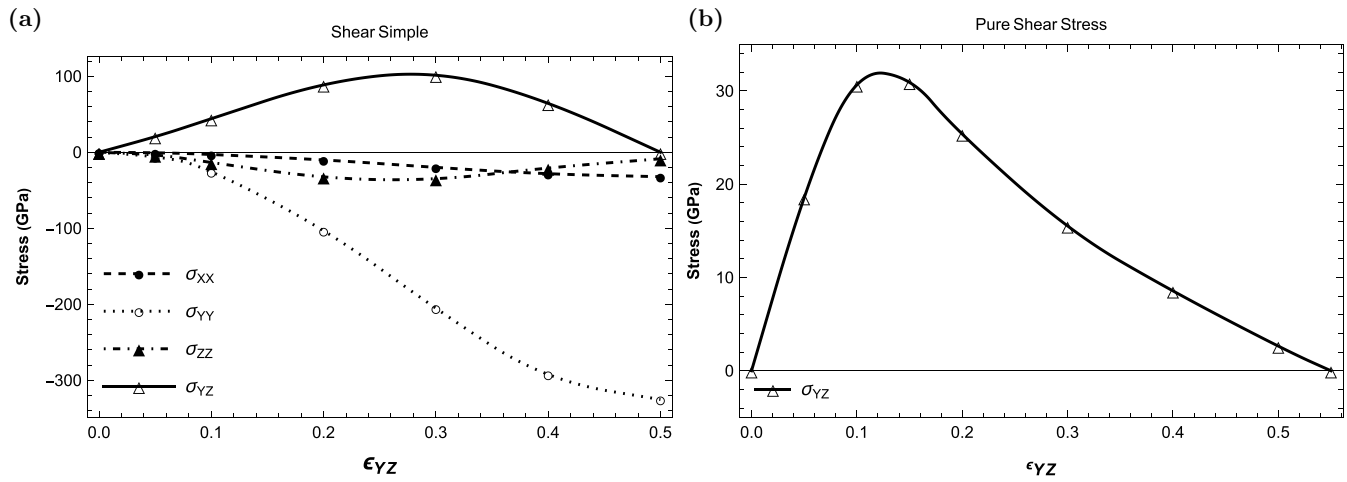

FIG. 1. Results for HfC $[1\bar{1}0](110)$ for (a) simple shear and (b) pure shear stress.

of the tensor $\mathbf{E}$ and ε):

$$\mathbf{F}^{\mathrm{SS}} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & \gamma \\ 0 & 0 & 1 \end{bmatrix} \Rightarrow \mathbf{E}^{\mathrm{SS}} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \frac{\gamma}{2} \\ 0 & \frac{\gamma}{2} & \frac{\gamma^2}{2} \end{bmatrix}$$
$$\Rightarrow \varepsilon^{\mathrm{SS}} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \frac{\gamma}{2} \\ 0 & \frac{\gamma}{2} & 0 \end{bmatrix}. \tag{6}$$

It can be seen here that $\mathbf{E}^{\mathrm{SS}}_{zz} \neq 0$. This would be the case if we were using the small strain tensor $\varepsilon^{\mathrm{SS}}_{zz}$. The deformations in this work are not small. It is important to remember what the consequences of overusing linear theories can be. Because $\mathbf{F}$ is not objective and hence small, a strain tensor $\boldsymbol{\varepsilon}$ and a rigid rotation can induce any stresses in a deformable body and they should not (see [7]).

The pure shear deformation [5] is a deformation in which the body is elongated in one direction while being shortened perpendicularly and is defined by

$$\mathbf{F}^{\mathrm{PS}} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \beta & 0 \\ 0 & 0 & \frac{1}{\beta} \end{bmatrix} \Rightarrow \mathbf{E}^{\mathrm{PS}} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & \frac{\beta^2-1}{2} & 0 \\ 0 & 0 & \frac{\beta^{-2}-1}{2} \end{bmatrix}$$
$$\Rightarrow \varepsilon^{\mathrm{PS}} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & \beta - 1 & 0 \\ 0 & 0 & \beta^{-1} - 1 \end{bmatrix}. \tag{7}$$

The details of the PS in the rotated coordinate frame along the shear plane can be found in [6]. The PS of Li *et al.* is not in fact a PS, but another deformation.

The pure shear stress (PSS) is such a deformation for which the Cauchy stress tensor is a pure shear stress tensor of the form $\sigma = \tau(e_2 \otimes e_3 + e_3 \otimes e_2)$, with $\tau \in \mathbb{R}$ [6]. To enforce such a deformation $\mathbf{F}$ must take the form of a simple shear composed with a triaxial stretch:

$$\sigma^{\mathrm{PSS}} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \tau \\ 0 & \tau & 0 \end{bmatrix} \Rightarrow \mathbf{F}^{\mathrm{PSS}} = \begin{bmatrix} a & 0 & 0 \\ 0 & b & c\eta \\ 0 & 0 & c \end{bmatrix}$$
$$\Rightarrow \mathbf{E}^{\mathrm{PSS}} = \begin{bmatrix} \frac{a^2-1}{2} & 0 & 0 \\ 0 & \frac{b^2-1}{2} & \frac{bc\eta}{2} \\ 0 & \frac{bc\eta}{2} & \frac{\eta^2c^2+c^2-1}{2} \end{bmatrix}. \tag{8}$$

In Fig. 2 in [1], in which the "[m]echanical response of samples sheared along $[1\bar{1}0](110)$" is shown, including "[v]ariations of stress components of HfC and TaC with ε_{zy} under SS and PS, respectively . . .," Li *et al.* presented the results of their DFT shear computations. The HfC and TaC crystals analyzed here have cubic symmetry and there is no reason for them to behave differently in shear than other cubic crystals (see [8]). It is known that for centrosymmetric lattices the Cauchy-Born rule applies directly and for the noncentrosymmetric lattices the relaxation of atoms in the periodic cell must be taken into account as well. The analyzed HfC and TaC structures crystallizes in the cubic $Fm\bar{3}m$, No. 225 space group, which is centrosymmetric. The shear component of the stress during SS deformation, as well as for PSS, should have a sinelike character (see Frenkel's classical sinusoidal solution [2]). In order to verify this, the HfC $[1\bar{1}0](110)$ crystal was subjected to SS and PSS by *ab initio* calculations based on DFT [9,10] within the pseudopotential plane-wave approximation performed by the ABINIT [11] code. The projector augmented wave method (PAW) pseudopotentials used for the Perdew-Burke-Ernzerhof [12] generalized gradient approximation exchange-correlation functionals were obtained from the PseudoDojo project [13]. The valence electron configurations and the cutoff energy used for Hf and C atoms were consistent with those utilized in the PAW pseudopotentials. The calculation accuracy settings correspond to those used in [14].

It can be seen from Fig. 1 that the results presented here are substantially different from those presented by Li *et al.* in their

Fig. 2 for HfC. For SS, the σ_{yz} shear component of the stress has a proper sinelike character and the σ_{yy} component of the stress is about twice as large. Zeroing the normal stresses in the PSS reduces the shear stress and introduces its asymmetry. These results also differ substantially from those presented by Li *et al.* A look at other results in the article also raises further questions.

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