

# Reply to the comment by M. Mazdziarz on the article "Ab initio calculations of pressure-dependence of high-order elastic constants using finite deformations approach" [Computer Physics Communications 220 (2017) 20-30]

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Reply to the Comment by M. Mazdziarz on the article "Ab initio calculations of pressure-dependence of high-order elastic constants using finite deformations approach" [Computer Physics Communications 220 (2017) 20-30]

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The central point of the Comment [1] is the criticism of Eq. (13) in our original paper [2]

$$\alpha_{ij} = \delta_{ij} + \eta_{ij} - \frac{1}{2}\eta_{ki}\eta_{kj} + \frac{1}{2}\eta_{rk}\eta_{ri}\eta_{kj} + \dots \quad (7)$$

where  $\alpha_{ij} = \partial r_i / \partial R_j$  stands for components of the deformation gradient tensor ( $r_i$  and  $R_j$  are the Cartesian coordinates of a selected point of solid in strained and initial states;  $i$  and  $j$  are Cartesian indexes), and  $\eta_{ij}$  are the components of the Lagrange finite strain tensor.

The author of the comment [1] states:

«In general, tensor  $\alpha$  is nonsymmetric and has nine components. Deformation gradient  $\alpha$  cannot be expressed by a function of  $\eta$ :

- Tensor  $\alpha$  is in general non-symmetric while  $\eta$  is symmetric, thus the right sides of this equations in Ref. [2, Eq.(13) and Eq.(18)] are always symmetric.»

The author of the Comment [1] would be right if one discuss a general case of deformations including rotations. However, the author overlooked the fact that in our paper [2] we discuss the crystal energy that changes only due to deformations and is invariant with respect to rotations of the crystal as a whole. This is a standard case in solid state physics, in which only symmetric deformations are considered. Therefore for our problem  $\alpha_{ij} = \alpha_{ji}$ , and in Eqs. (13) and (18) of Ref. [2] the left- and right-hand sides are symmetric.

Let us demonstrate it in more detail. To derive the expressions for higher order elastic constants at high pressures, the decomposition of the Gibbs free energy over components of the Lagrange finite strain tensor  $\eta_{ij}$  is used. According to Eq. (1.5) of Ref. [3]

$$\eta_{ij} = \frac{1}{2}(\alpha_{ki}\alpha_{kj} - \delta_{ij}), \quad (1)$$

where  $\delta_{ij}$  is the Kronecker delta.

Then, for each vector of a crystalline lattice in the deformed and in the initial state  $r_i = \alpha_{ij}R_j$ . The tensor  $\alpha_{ij}$  describes both pure deformations and rotations of the crystal. Now we express  $\alpha_{ij}$  via  $\eta_{kl}$ , taking into account that the energy is not changed with any crystal rotation. That is we consider only pure deformations and no rotations. We express  $\alpha_{ij}$  via the displacement gradient  $u_{ij} = \partial u_i / \partial R_j$  ( $u_i = r_i - R_i$ ) (see Eq.(1.4) of Ref. [3]) as

$$\alpha_{ij} = \delta_{ij} + u_{ij} \quad (2)$$

Then according to Eq. (1.5) of Ref. [3]

$$\eta_{ij} = \frac{1}{2}(u_{ij} + u_{ji} + u_{ki}u_{kj}) \quad (3)$$

In the case of pure deformations without rotations  $u_{ij} = u_{ji}$ ,  $\alpha_{ij} = \alpha_{ji}$  (see Eq. (8.13) of Ref.[3]).

Therefore

$$\eta_{ij} = u_{ij} + \frac{1}{2}u_{ki}u_{kj}. \quad (4)$$

For this case (the symmetric deformations) we express  $\alpha_{ij}$  via  $\eta_{kl}$ . From Eq. (4) we get

$$u_{ij} = \eta_{ij} - \frac{1}{2}u_{ki}u_{kj} \quad (5)$$

and

$$\alpha_{ij} = \delta_{ij} + \eta_{ij} - \frac{1}{2}u_{ki}u_{kj}, \quad (6)$$

$$u_{ki} = \eta_{ki} - \frac{1}{2}u_{lk}u_{li}; \quad u_{kj} = \eta_{kj} - \frac{1}{2}u_{mk}u_{mj}$$

Substituting the expressions for  $u_{ki}$  and  $u_{kj}$  in Eq. (6) and truncating the expansion at the third-order term in  $\eta$ , we get

$$\alpha_{ij} = \delta_{ij} + \eta_{ij} - \frac{1}{2}\eta_{ki}\eta_{kj} + \frac{1}{2}\eta_{rk}\eta_{ri}\eta_{kj} + \dots \quad (7)$$

It corresponds to Eq. (13) in our Ref. [2]. Taking into account the fourth order term, we can derive an expression for  $\alpha_{ij}$  corresponding to Eq. (18) in Ref. [2].

Further the Comment [1] states that on p.21 of Ref. [2] we use the term “tensor of transformation coefficients” for  $\alpha_{ij}$  instead of conventional “deformation gradients”. We notice that the term “transformation coefficients  $\alpha_{ij}$ ” is taken from p.306 of Ref. [3], where its meaning is clearly explained. We use this term for the same purpose.

The Comment [1] also points out that in the first term of Eq. (1) of Ref. [2] the order of indexes in brackets is violated ( $\alpha_{ik}$  is written instead of  $\alpha_{ki}$ ). Certainly, that is a misprint, which we unfortunately overlooked in the proofs of our paper.

We notice that the relation in Eq. (7) (without the third order contribution), corresponding to the deformation (i.e. when rotations are absent), is given by Eq.(8.15) in Ref. [3] and also by Eq. (19) in Ref. [4]. It is used in our Ref. [2] as well as in Refs. [4], [5], and [6] for setting up deformations when calculating the third and fourth order elastic constants at ambient pressure. The references to these papers are given in our Ref. [2] (as Refs. [10] and [11]). In the case addressed in Ref. [2] the problem is far more complicated because of non-zero pressure. One needs to take into account the change of the Gibbs free energy due to  $P\Delta V/V_0$  term (see Eq.(12) of Ref. [2]), where  $P$  stands for pressure,  $V_0$  for the equilibrium volume at pressure  $P$ , and  $\Delta V$  for the volume change at pressures  $P$  when a small deformation  $\eta$  is applied. Herewith the volume change does not depend on the rotation of the whole crystal either. It is determined solely by the symmetric deformations.

To conclude, the statement made by M. Mazdziarz is irrelevant for the problem discussed in our paper [2]. All the results presented in Ref. [2] are therefore correct.

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