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Review of “The average conformation tensor of inter-atomic bonds as an alternative state variable to the strain tensor: definition and first application—the case of nanoelasticity”

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Review of version 1

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Reviewer 1 (Ranganathan PARTHASARATHY)

Reviewer This paper proposes an alternative kinematic state variable, denoted as a conformation tensor, to the strain tensor based on an atomic description of the material. One of the motivations the author touches upon is that the strain tensor needs a reference configuration in order to be defined, while no such reference is needed in an atomistic model.

Author These few lines from the introduction of the report perfectly summarize the point of view that I develop in my study. The notion of deformations requires indeed to specify the configuration of the considered solid with respect to which this deformation is expressed. Mechanically speaking, it amounts to linking only the current configuration (at time t) to a reference configuration (at time t_0 prior to t), whatever the evolution of the configuration between t_0 and t , via the Lagrangian gradient of the transformation (or deformation gradient), i.e. the Lagrangian description of the solid motion.

From a purely kinematic point of view, such a way of doing things does not really pose a problem, as it allows us to express the local variations of lengths, angles and volumes between t_0 and t . On the other hand, from a sthenic point of view, i.e. when it comes to the state of stress at any time, it does not seem so obvious to me that it must necessarily depend on the deformations. This is the key point of my paper, where I try to show that the notion of conformation—which makes sense a any time, including t_0 where conformation, unlike deformations, can be non-zero—can be substituted for that of deformations in the expression of Cauchy stresses, at least in the case of elasticity.

The corollary of this choice is that, contrary to a deformation tensor, the conformation tensor at time t is only accessible through its evolution (its material derivative) from an initial time, for example t_0 . However, this evolution, like that of any state variable, is constrained by Thermodynamics, as I show in Section 5 of the paper (“Thermodynamics and material derivative of the average conformation tensor of interatomic bonds”).

The other important point of the study is the physical interpretation of conformation. I have tried to show that it is essentially geometric, but I may not have succeeded completely.

Some of the reviewer's comments seem to indicate that I did not detail enough what I meant by reference length (of an interatomic bond). I have tried to clarify my point in some of my answers below, and I also came back to the various ways of expressing the Cauchy stress tensor and the interaction potentials between atoms. In one way or another, these answers have also been included in the new version of the paper.

Reviewer In the introduction, questions have been raised on the physical relevancy of strain decomposition used in continuum models of elastoplasticity. However, these questions have not been discussed anywhere in the manuscript, although some conclusions regarding this have been presented.

Author It is quite true that I only deal with elastic behavior in this study. The last part of the title of the paper clearly states this ("...—the case of nanoelasticity."). It is also true that, in the introduction, I devoted a whole paragraph to the inelastic models usually used in mechanics of materials, to which I do not return at all in the following sections.

I thought it would be useful to mention these inelastic models (elastoplastic, to be precise) because they confirm the interest of looking for an alternative kinematic variable to a strain tensor. But in the current version of the paper, I also wrote that: "... Moreover, the present study is limited to the elastic case. Although the issues linked to the usual way of modeling the elastoplastic strains are one of the reasons to look for an alternative to a strain tensor as a state variable, it is indeed necessary to demonstrate that an alternative variable to S can be found in elasticity since, in most of the materials, the mechanical behavior is first elastic before becoming, possibly, elastoplastic..."

However, this important clarification only appears in the paragraph following Eq. (2), i.e. at the end of the introduction. The reviewer's remark seems to indicate that it could have been made earlier. I have nevertheless left it where it is, but I have also completed, still in the introduction, the paragraph beginning with "The same question is both relevant and interesting..." so as to leave no doubt that I am dealing only with the elastic case in my study. More precisely, I added these sentences in the new version on the paper: "Such models are clearly outside the scope of this study, which is only devoted to elasticity. It is however interesting to mention them, but only in this introduction, because they confirm the interest of looking for an alternative kinematic variable to a strain tensor."

Reviewer "The important point that must be emphasized here is that all these models are actually based on an implicit assumption, namely that the only kinematic variable which can be associated with the Cauchy stress tensor is a strain tensor". The author must note that there has been other research that has explored other kinematic variables. For example, please see:

- Kuzkin, V. A. and A. M. Krivtsov: An analytical description of transient thermal processes in harmonic crystals, *Physics of the Solid State*, vol. 59, no. 5, pp. 1051-1062, (2017)
- Kuzkin, V. A., A. M. Krivtsov, R. E. Jones, and J. A. Zimmerman: Material frame representation of equivalent stress tensor for discrete solids, *Physical Mesomechanics*, vol. 18, no. 1, pp. 13-23, (2015)
- Kuzkin, V. A. and A. M. Krivtsov: Nonlinear positive/negative thermal expansion and equations of state of a chain with longitudinal and transverse vibrations, *physica status solidi (b)*, vol. 252, no. 7, pp. 1664-1670, (2015)
- Krivtsov, A. M. and V. A. Kuzkin: Derivation of equations of state for ideal crystals of simple structure, *Mechanics of solids*, vol. 46, no. 3, p. 387, (2011)
- Krivtsov, A. M. and V. A. Kuzkin: Discrete and Continuum Thermomechanics. (2017)
- Parthasarathy, R., A. Misra, and L. Ouyang: Finite-temperature stress calculations in atomic models using moments of position, *Journal of Physics: Condensed Matter*, vol. 30, no. 26. p. 265901, (2018)
- Parthasarathy, R.: Thermal vibration contribution to continuum stress in the elastic regime, *Mechanics Research Communications*, vol. 92, pp. 101-106, (2018)

Author I admit without any hesitation that the sentence of my paper quoted by the reviewer is too peremptory. I must also admit that I was not aware of any of the works he cites. Among these, the paper entitled "Material frame representation of equivalent stress tensor for discrete solids" by V. Kuzkin *et al.* is the one that caught my attention the most, where, for discrete solids, the

Cauchy stress tensor, among others, is expressed in terms of average distances and average forces between particles. There are undeniably similarities with the approach I propose to define the Cauchy stress tensor, σ , starting with the consideration of the discrete nature of matter (at the nanoscopic scale in my case, but the approach seems to be potentially applicable to larger scales).

Unlike the above-mentioned study, however, I did not try in mine to use or interpret the Cauchy tensor in the discrete case. I have only defined it in the continuous case, *i. e.* after having associated to the real, discrete medium, an “equivalent continuous” one, fictitious but verifying the conditions stated at the beginning of Section 4 of my paper (“Average conformation tensor of interatomic bonds, average internal forces tensor and Cauchy stress tensor: continuum approach”). Since all the rest of my study is in a continuum framework, I have not tried to give an interpretation of the interatomic forces existing in the real (discrete) medium from the Cauchy stress tensor. Of course, it is possible to define, from σ , the stress vector for any direction in space and thus, in particular, for the direction of an interatomic bond— $\pm \mathbf{n}^{1,j+1}$ according to the notations I used in the paper –, that is to say:

$$\pm T^{1,j+1} = \sigma \cdot (\pm \mathbf{n}^{1,j+1}) \quad (1)$$

The normal component of this stress vector, *i. e.* $T_n^{1,j+1} = (\pm T^{1,j+1}) \cdot (\pm \mathbf{n}^{1,j+1})$, can then be interpreted, in the real (discrete) medium, as the value of the force per unit area—the average force per unit area, in fact, since the Cauchy stress tensor is an average one, directly deduced from the average conformation tensor—acting between atoms 1 and $j + 1$. From $T_n^{1,j+1}$, it is however not at all trivial, I readily admit, to define the force (in Newton, therefore, and not in Pascal) acting between atoms i and $j + 1$. This difficulty is all the greater since, as I indicate at the very beginning of Section 2 (“Conformation tensor of an interatomic bond and internal force tensor: definitions”), the atomic nuclei are assimilated to points in my study—so as to precisely define the notion of interatomic distance—, *i. e.* they have zero volume.

On the other hand, it is quite possible to calculate the average interatomic force acting on a bond of orientation $\pm \mathbf{n}^{1,j+1}$ from the average internal forces tensor \widehat{F}^1 defined by Eq. (19) in Section 3 (“Average conformation tensor of interatomic bonds and average internal forces tensor: discrete case”). The algebraic value of the latter is indeed simply given by:

$$\widehat{F}^{1,j+1} = \widehat{F}^1 : ((\pm, \mathbf{n}^{1,j+1}) \otimes (\pm, \mathbf{n}^{1,j+1})) = (\pm, \mathbf{n}^{1,j+1}) \cdot \widehat{F}^1 \cdot (\pm, \mathbf{n}^{1,j+1}) \quad (2)$$

which can also be written, using the characteristic tensor of the direction of an interatomic bond $\mathbf{N}^{1,j+1} = (\pm, \mathbf{n}^{1,j+1}) \otimes (\pm, \mathbf{n}^{1,j+1})$, see also Eq. (4):

$$\widehat{F}^{1,j+1} = \widehat{F}^1 : \mathbf{N}^{1,j+1} \quad (3)$$

In the new version of the paper, I added some sentences and two equations in the very last paragraph of Section 3 to specify this relation between the average internal forces tensor and the average force acting on an interatomic bond. I will also come back to this notion of forces acting on an atom in the rest of this text (see my answer to comment 4).

My last remark on the average interatomic forces, however, does not answer the question of the relation between \widehat{F}^1 and σ . This problem would certainly deserve to be explored further. I have not done it at all in the present study which, if it does start from discrete notions, then focuses on the continuous quantities which can be associated with them. It is nevertheless true that papers do exist where a deformation tensor is not the variable which is associated with the Cauchy stress tensor. In the paragraph following Eq. (2) of my paper, I have therefore added, in its new version, a few lines to refer to the paper by V. Kuzkin *et al.* that I mentioned earlier. Insofar as, by choice, I did not take into account the temperature at all in my study (which I clearly wrote at the very end of the introduction, where we read: “Note finally that all the arguments, hypotheses and equations detailed in this study concern a “frozen” state of a pure substance in the solid state, observed at the generic time t . In other words, the thermal and viscid effects are not taken into account.”), I do not quote five of the six other papers mentioned by the reviewer in comment 2 (however, I do quote the one by Parthasarathy *et al.*, for the reason I explain below, in my answer to comment 4). I am nevertheless convinced of the interest of taking thermal effects

into consideration, if only because they are closely coupled to deformations—and therefore, according to the point of view I defend in my study, to conformation. I obviously do not exclude the idea of integrating them in future studies on thermomechanics of solid media based on the notion of conformation.

Reviewer When the author refers to “pure substance”, does it refer to a perfect defect-free crystal with only one atom in the primitive cell (such as aluminum or copper as mentioned in the abstract)? From the uniaxial tension case examined later in the paper, it seems that defects are permitted.

Author The average conformation tensor Γ can be defined if defects—punctual (*e. g.* vacancy defects) or linear (dislocations)—exist in the pure substance. This is clearly indicated in the current version of the paper. At the end of the first paragraph of Section 5, it is indeed written: “From a nanoscopic point of view, this means that, at any time of the evolution of the pure substance considered in the solid state:

- each atom has the same first neighbors. Defects such as dislocations can exist in the lattice, but in constant number and immobile (in other words: no plasticity);
- each atom is always bonded to its first neighbors by active interatomic bonds. These bonds can vary in length and direction but they cannot disappear or break (in other words: no damage).”

However, the reviewer’s comment seems to indicate, again, that this important clarification could have been made earlier in the paper. I fully agree. In the new version of the paper, I have therefore added a few sentences to this effect, on the one hand in the introduction (paragraph beginning with “For the sake of enhancement of the main, new ideas,...”), on the other hand in the very last paragraph of Section 3.

Reviewer For Eq. (3), “the length of the bond when no force is applied can be considered as a characteristic length, which will be denoted by r_r ”. Is the force mentioned here the force acting on each of the atoms, applied by all other atoms in the crystal? Or the force exerted by the two atoms on each other? Please clarify.

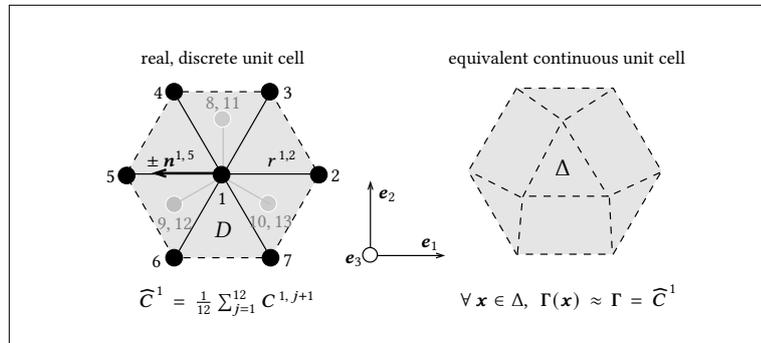
Author The characteristic length r_r involved in Eq. (3) is the one that appears again in Eq. (7), which gives the expression of the force f existing between two atoms. And it is indeed this force f that cancels when the distance between the two atoms, r , is equal to r_r (and thus $\bar{r} = r/r_r = 1$). This force is itself supposed to derive from an interaction potential which, in this case, can only be a pair potential (Lennard-Jones for example, but many other potentials exist) since the system is reduced to two atoms. I will come back to this last, important notion in some of my other answers to the reviewer.

More generally, the whole Section 2 of the paper is devoted to the case, elementary but indispensable for the continuation of the study, of a system reduced to 2 atoms. In a real solid, these two atoms are of course surrounded by many others, with which they interact. In Section 2 however, they are considered as an isolated system, in the sense that Thermodynamics gives to this word. Consequently, the only force that exists in the system is the one between the two atoms. To remove any ambiguity on this important point, I have added, in the new version of the paper, a few lines at the very beginning of Section 2, namely: “... In this paragraph, these two atoms are assimilated to an isolated system, in the thermodynamic sense of the word. They are therefore assumed to have no interaction of any kind with the other atoms of the pure substance that surround them. On the other hand, they do interact with each other, a force resulting from this interaction.” I also insisted, in the paragraph devoted to the interaction force, on the nature of the interaction potential, adding to it: “...—a pair potential, in this case, since the system under consideration reduces to two atoms.”

Reviewer If it is just two atoms exerting force on each other, how can this scenario represent a pure crystal such as aluminum or copper? From the description in the last paragraph of page 17, “However, and in order to facilitate the presentation of the main results... initially belong to a sphere with a radius r_r ”, it seems that the author expects $\Gamma = 0$ in the case of a perfect crystal in the frozen state (0 K). But, in order to associate this condition with a zero value for the conformation tensor of a particular atom, this atom needs to experience zero stress. For the concept of stress-per-atom, the author can refer:

- Egami, T.: Atomic level stresses, Progress in Materials Science, vol. 56, no. 6. Elsevier, pp. 637 – 653, (2011)
- Parthasarathy, R., A. Misra and L. Ouyang: Finite-temperature stress calculations in atomic models using moments of position, Journal of Physics: Condensed Matter, vol. 30, no. 26, p. 265901, (2018)

Author To answer this new question of comment 4 as clearly as possible, let me first reproduce Figure 4 of my paper (without its legend). Let’s start by considering the discrete case, *i. e.* the left part of



the figure. The example presented concerns mainly atom 1, for which the average conformation tensor \widehat{C}^1 is defined (see the lower part of the figure). It is surrounded by 12 atoms identical to itself (only 6 in the plane of the figure), which form the vertices of a cuboctahedron—the one whose edges are dotted on the left side of the figure. Since all these atoms are identical, each of them has the same interaction potential—pair potential, see previously—with each of its first neighbors: the reference length r_r previously introduced is indeed the same for all these atoms, in particular for atom 1 but also for atoms 2, 3... whose first neighbors are not all represented on the figure but for each of which, from the same reference length r_r , an average conformation tensor can be defined which, like \widehat{C}^1 , is generally non-zero. It is therefore possible, with the conformation tensor presented in my study, to “... *represent* (but only on average, this is an intrinsic limitation of my approach) *a pure crystal such as aluminum or copper.*”

Moreover, atom 1 is subjected, by its 12 first neighbors, to 12 forces (see my previous answer to comment 2), each of them deriving from the same pair potential. These forces are generally non-zero (for this, it is sufficient that the distance between atom 1 and one of its first neighbors is not equal to r_r) but they must be balanced, that is to say that the net force acting on atom 1 must be equal to zero (see again my response to comment 2). But in no way does the mechanical equilibrium of the discrete forces correspond, in the continuous case (see the right-hand side of the figure), to a state of zero stress. On the contrary, at least if the behavior of the material is assumed to be elastic, as in this study, it is sufficient that the mean conformation tensor Γ —which is assumed to be equal to that obtained in the discrete case, \widehat{C}^1 , in my study, see the lower part of the figure—is non-zero for the stress tensor σ to be as well. This is particularly the case when the trace of \widehat{C}^1 is non-zero, *i. e.*, following Eq. (13) of the current version of the paper, when at least one of the interatomic bond (at least two, actually, for the static equilibrium of atom 1 to be verified) is such that its length is different from the reference length r_r . Note also that, according to Eq. (14) in the current version of the paper, the trace of \widehat{C}^1 is zero if (and only if) the geometric mean of the distances between atom 1 and its first neighbors is zero: this certainly does not mean that all these distances are equal to r_r (in which case not only the trace of \widehat{C}^1 but its three eigenvalues and thus its deviatoric part would also be zero), and this can happen whatever the temperature.

The argument I have just developed is probably a bit long. I hope, however, that it will have convinced the reviewer that with a single characteristic length, it is indeed possible to describe on average, at the nanoscopic scale, the kinematics and sthenics of pure substances. However, I would like to add two things:

- there is no doubt that the characteristic length r_r that I introduce depends on the temperature. Although all thermal effects are neglected in my study, I have mentioned this fact, in a few words, in the new version of the paper—see the paragraph where r_r is introduced, just before Eq. (3),

- the concept of stress-per-atom is beyond the scope of my study, which is essentially centered on the conformation tensor, *i.e.* a kinematic notion. However, I am well aware of the interest of this concept, to which I will certainly return in future studies. For the moment, I have only quoted and briefly commented the paper by Parthasarathy *et al* in the new version of the paper, in the last paragraph of Section 4.

Reviewer If an atom is in the neighborhood of a defect, it will have non-zero per-atom stress but the force on it will still be zero. On the basis of the above comments, the author should explain exactly what force is being referred to for determination of r_r . Also, if the definition of r_r is based on the net force experienced by an atom, it looks like the information contained in the conformation tensor will be not so different than the information contained in a local deformation gradient? For e.g. please check the following reference:

- Gullett, p. M., M. F. Horstemeyer, M. I. Baskes, and H. Fang: A deformation gradient tensor and strain tensors for atomistic simulations, *Modelling and Simulation in Materials Science and Engineering*, vol. 16, no. 1. p. 015001, (2008)

Author I have already explained how the characteristic length r_r is defined, I will not repeat it here. However, I insist on the fact that, if the net force resulting from all the forces acting on an atom, whether it is close to a defect or not, is necessarily zero, this does not mean that the tensor of the internal forces \widehat{F}^1 as it is defined in Section 3 of the paper, see Eq. (19) (and thus, in the continuous case, the stress tensor σ , the relation between \widehat{F}^1 and σ remaining however to be studied) is also zero. Similarly, in the continuous case, the fact that the stresses are necessarily balanced—*i.e.*, neglecting mass forces such as gravity, $\mathbf{div}_E \sigma = 0$ (where \mathbf{div}_E is the Eulerian divergence, not the Lagrangian one)—does not mean, in any way, that the stress tensor is itself zero.

The reference length is therefore not based on the net force resulting from all the forces exerted by its first neighbors on an atom but on the unique force existing between two atoms isolated from their environment (see the first part of my answer to comment 4). It is therefore not associated with a reference configuration, which, on the other hand, is essential for defining the transformation gradient. I therefore maintain that, in general, the information contained in the average conformation tensor Γ is different from that contained in the deformation gradient tensor T . The material derivative of Γ that I give in Section 5, see Eq. (37) in the current version, shows this clearly: it is not that of the transformation gradient (which is not symmetric, contrary to Γ), nor that of any deformation tensor. It is true, however, that in some special cases the conformation tensor does coincide with a strain tensor—the “small” strain tensor ϵ that appears in Section 6 (“An example of an elasticity model based on the conformation tensor”) of my paper is thus only an approximation of the Hencky strain tensor, which coincides with Γ in the simple tension case. But, I repeat one last time, what is true in this particular case is false in the general case.

Reviewer Line 1, Page 4: Please change “at the only atomic scale” and “at the only nanoscale” to “only at the atomic scale” and “only at the nanoscale”.

Author Both of these changes were made in the new version of the paper.

Reviewer Page 12: “In other words, the average conformation tensor is defined on the only current configuration of Δ —in the sense that it is not linked to any Lagrangian gradient.” Please change the position of the word “only” so that the meaning of this sentence is better understood.

Author This change was also made in the new version of the paper.

Reviewer Also, the above sentence needs to be explained with respect to comment 4.

Author I think I have already largely explained why the mean conformation tensor is not a deformation tensor in my previous answers to the reviewer’s questions, especially those I gave to comment 4. But I also understand that this very important point deserves to be well explained, in different ways and maybe even several times, especially for the future readers of the paper. In the new version, I have thus:

- recalled, in the introduction, how a deformation tensor is constructed and how it is related to the notion of Lagrangian gradient (paragraph following Eq. (2)),

- modified and completed, at the very end of Section 4, the sentence recalled above by the reviewer.

Reviewer Last Line, Page 5: There is an additional assumption that the interaction potential chosen is a central potential.

Author In the new version of my paper, this assumption is clearly underlined in the paragraph where the interaction potential is introduced (see also my answer to the reviewer’s comment 4).

Reviewer Before Eq. (12), it must be mentioned whether U needs to be a pair-potential or it can also be a multi-body potential.

Author Eq. (12) is in Section 3 of the paper, devoted to the discrete case of an atom surrounded by N_l first neighbors. In this paragraph, energy is only discussed from Eq. (18), where the state potential of average free energy is denoted \mathcal{U} . But energy is also mentioned in Section 2 (case of two atoms), especially Eq. (11), where the interaction potential is noted u . So I am not sure what “potential” U the reviewer is talking about in his comment. The fact that he uses the term “multi-body potential” seems to indicate that it is \mathcal{U} . In either case, however, I can make the following two comments:

- if it is u , it can only be a pair potential for the reason I gave in the first part of my answer to the reviewer’s comment 4 and that to the previous remark (*Last Line, Page 5...*). It should also be noted that, in the new version of the paper, I have made it clear that u is a pair potential,
- if it is \mathcal{U} , it is indeed a multi-body potential since, via the elementary conformation tensors $C^{1,j+1}$, it involves the distances $r^{1,j+1}$ between atom 1 and its N_l first neighbors (the single non-zero eigenvalue of the elementary conformation tensor $C^{1,j+1}$ is indeed $\ln(\bar{r}^{1,j+1}) = \ln(r^{1,j+1}/r_r)$). However, it is interesting to note that these distances are not the only arguments that can be used in the expression of \mathcal{U} : since every elementary conformation is characterized by a tensor, \mathcal{U} can also depend on the crossed invariants of these tensors (since these tensors are objective, their crossed invariants are also objective). More precisely, the crossed invariant of the elementary conformation tensors $C^{1,p}$ and $C^{1,q}$ simply reads:

$$C^{1,p} : C^{1,p} = \ln(\bar{r}^{1,p}) \ln(\bar{r}^{1,q}) \quad N^{1,p} : N^{1,q} = \ln(\bar{r}^{1,p}) \ln(\bar{r}^{1,q}) \cos^2(\omega^{p,q}) \quad (4)$$

where $\omega^{p,q}$ is the angle between interatomic bonds $1-p$ and $1-q$. I have shown, in an unpublished paper available on HAL (“Approche énergétique de l’élasticité linéaire des cristaux à structure hexagonale compacte à l’échelle nanoscopique sur la base de la notion tensorielle de conformation : relation entre les descriptions discrète et continue.”; ref: hal-02052799), that it is crucial to take into account some of these crossed invariants—those of the “neighbouring” interatomic bonds, in a sense specified in the paper—in the expression of \mathcal{U} . For the moment, unfortunately, only a French version of this paper is available. I will propose an English version (which I will certainly submit for publication later) if the paper to which the previous “answers to the reviewer” are dedicated is finally accepted for publication.

Since crossed invariants can be important arguments of \mathcal{U} , I have added, in the new version of the paper, some words and an equation about them, just after Eq. (17).

Reviewer Please fix all spelling mistakes.

Author I think I have corrected all the reported mistakes. As for my responses to the reviewer’s remarks, I wrote them as clearly and accurately as I could. I hope that they will have convinced the reviewer of the validity of my approach.

Editor’s assessment (Lazslo TOTI)

The very first version of this article was refused for publication by the Associate Editor (AE) which was chosen by the Author. The Author then substantially clarified his paper and submitted it again by choosing another AE, who found a suitable expert for reviewing it. The Reviewer asked for major revision, however, the second AE became no longer available for processing the paper. Then the first AE accepted to continue the work and asked the Author to carry out the major modifications. The Author complied very well to the criticisms and reworked his manuscript substantially. Then the Reviewer accepted the new version with further minor

modifications for the final version, which was verified by the AE. In conclusion, the acceptance was based on one substantial external review, and on the evaluation by two AEs of the journal.

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