nature portfolio

Peer Review File

Hyperforce balance via thermal Noether invariance of any observable

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The theoretical concepts developed in this work represent an important new building block in the construction of a modern view of statistical mechanics and of new pillars that constitute the derivation of exact sum rules. It promotes also the power of Noether's fundamental principle of translational invariance in general physics and here statistical physics. From this principle the authors are able to generate exact sum rules relating the covariance of various force and energy quantities to the mean gradient of an associated phase-space function. The simplest example is the Yvon-Born-Green relation relating the local force density to the gradient of the particle density (eq 1). This equation follows naturally from the Noether's principle (generalised to displacement fields) and appears as a particular and simple choice in the general equation 11 expressing the correlation of the local force with a general , momentum-independent observable A(r^N). Playing with the observable A leads to a multitude of known and unknown exact sum rules. These sum rules, as well as their convergence, are verified in simulation for a confined LJ fluid. They can be used either to optimize the statistical convergence or to assess thermodynamic equilibrium. I note that among the different possible ways of computing the one-particle density that emerge from the general theory (eqs 22-25), the already well-tested « force method » given by eq 1 apparently remains the most efficient and accurate one.
I have absolutely no doubt on the substance of the article and its suitability to Comm. Physics, and I strongly recommend its publication. I have just the minor remark that general equation 11 with an arbitrary observable A(r^N) should be balanced with with Coles et al, JCP 151, 064124 (2019) where it was used for particular A's based on single particle observables of the form sum_i delta(r - r_i) a_i.
Concerning the format, the paper is written in a format that I like where every equation is justified and follow logically from the preceding one. I am not sure though that it fits well to the format of the journal where the paper is recommended to begin with a result section where the outcomes are presented in the most clear and striking way, and end with a method section where the theoretical and computational methods are detailed and deepened. This might necessitate a little rearrangement. I leave this to the absolute discretion of the authors and to the recommendations of the editor.

Reviewer #2 (Remarks to the Author):

Report on Robitschko et.al

This is a very interesting paper that introduces important new relationships/identities in (classical) statistical mechanics. It is quite rare to see new fundamental and powerful formulae emerge in this field; current practitioners are usually concerned with *applications* in solid state and in soft matter.

The authors build upon earlier papers from their group refs[29-34] which showed that use of Noether's theorem leads to several exact identities involving force fluctuations. Here they present an impressive general treatment of the (deep) implications of Noether invariance for both global and locally resolved quantities. Many of the 'sum rules' they derive are remarkable in their simplicity and elegance and in the depth of physics they contain. The paper is well-written; it explains nicely the subtle issues associated with the different types of invariance as well as the importance of the new identities for assessing efficiency and accuracy of computer sampling schemes.

The authors choose to test the sum rules numerically via simulations of a (truncated) LJ fluid confined in a planar slit pore. This is a very sensible choice of model system and results are nicely presented.

Some comments:

1.The force balance equation (1) is indeed a statement of the 1st YBG equ. The latter is usually written as an integro-differential equation for the particular case of pairwise interparticle potentials, as in Ref.[1], Sec.4.2. For scholarly interest it would be useful to give a sentence or two of background. What did Yvon say?

2. In caption to Fig.1 (b) spell out that the wall separation is L.

3.The algebra involved in the various correlators and their manipulations is clearly presented and I followed this. (Indeed, I enjoyed checking several derivations.) Sometimes there is repetition in explaining the content of the sum rules but given their subtlety and generality I think there is no harm in this.

4.In the 1st derivation from (2) to (3) one might be pedantic and add 'noting that the partition sum is independent of eps_0'.

5.It would help the non-expert reader if the LJ potential $\phi(r)$ were spelled out in pg.5. This is important when it comes to the confining wall potential. As far as I can tell, this is not the sum of 9-3 potentials as is commonly used for confinement. The authors might write explicitly what is V_ext(z) for their confining potential. Also, it would assist the reader if they wrote an explicit formula for the average evaluated in (14) for the particular 1D geometry. The reader would then get a better sense of the physics and the import of this relation. After all the authors emphasize that (14) is 'remarkable' and later 'very counter-intuitive'-and the referee agrees. It would help to lay out precisely what is being calculated in the model system.

6. In the figures blue lines sometimes appear as blue dots. I was confused at first; the authors might add a sentence to the first caption explaining why there are blue dots.

7. Pg.6, 4 lines below (16) add 'which follows from (13) with operator A =1'. A pedantic point.

8.Eqs (22-24) are important. Eq. (24) should have z' in the final round bracket. Spell out what rho_0 is.

9.In the caption to Fig.5 the term ‘mirroring’ is introduced. This should be explained.

10. That results should not depend on the ensemble is mentioned on pg.5 Col.2 and again on pg.10 Col.1.At first, I was sceptical about this statement as the formal manipulations deriving the new identities are performed straightforwardly and naturally in the GCE. If the authors have shown that the manipulations remain valid in the CE this is important and worth emphasizing further. Of course, this also justifies employing Brownian Dynamics with fixed N to test the sum rules.

11. The use of the term ‘hyperforce’ might raise eyebrows. The hyper appears to arise from an analogy with Hirschfeld’s (1960) hyper virial theorem. Most readers will not be aware of the latter. I was not. Thus, the authors might like to clarify why (4) is the ‘global hyperforce identity’.

12. Sec.IV is a bit rambling but very thought provoking. The authors might like to review and re-draft to sharpen the messages. For example, it is well-known that the spatial derivative of (28) corresponds to the force balance eq. (1) and therefore it is clear what the physical significance is of rho(**r**)*gradc_(1)(**r**). The new thing is probing the derivative of F_int(**r**). Re-writing would clarify.

Summary.

This is a fine piece of work that makes a substantial and scholarly contribution to statistical physics. The sum rules/identities that are introduced are insightful and are likely to be valuable in applications across condensed matter physics and physical chemistry.

I recommend the paper be accepted for publication provided the authors address the suggestions above. Note: these are fairly minor.

Reviewer #3 (Remarks to the Author):

I have read the manuscript “Hyperforce Balance via Thermal Noether Invariance of Any Observable”. This manuscript describes an analysis technique, for computer simulations of particle dynamics, to relate correlation functions including forces to obtain local estimates of density gradients or force gradients, without computing the gradients. A rigorous derivation is provided for these relations for many-body particle systems at equilibrium. Applications are given and the method is tested on computer simulations of a confined liquid between two walls. The method is general and allows one to access a plethora of variables thanks to these force-based correlations. Calculating gradients of noisy data is a general challenge in small-scale physics. As such, the proposed method offers an interesting route to improve and complement existing analysis routines in computer simulations.

Given its potential broad impact, I believe the manuscript could be suitable for Communications Physics. In its current state, I find the manuscript hard to read, and the main results, as well as how the method compares to existing methods (in terms of accuracy and convergence speed) still remain somewhat elusive to me. The manuscript may contain a very important contribution to the field, however that contribution is lost within a vast array of results that are not necessarily sorted/hierarchized and the recurring use of jargon. In general, clarit could be greatly improved (a) by stating the main result in the introduction (b) by providing a table of derived examples and what information they provide (c) referencing a specific example in the abstract/intro (for instance calculating density gradients with a force-density correlation) (d) stating clearly how this method compares to existing method/how it might complement them. I provide specific point-by-point feedback below. Provided significant clarifying changes are made on the manuscript, I believe the manuscript will be suitable for Communications Physics.

- abstract: “its negative correlation with the total force”: this is a statement I do not understand. The abstract is, in general, very hard to read/as well as the title, and does not provide the reader with an immediate understanding of what they might do with this technique.
- “The results clarify a very intimate link of global and locally resolved correlators and they suggest a very general statistical mechanical structure.” This is a vague statement that may be rendered clearer with a main result under the form of an equation. It would be really nice at this stage to already have an example, or to treat examples later on and say at the beginning what kind of examples will be investigated.
- “Our framework can be viewed as a generalization of the YBG equation”: I find this statement equally confusing. What is the added contribution/the things made possible by this work compared to previous works, beyond the conceptual and derivation aspects?
- “hyperforce” please define this word and say why it is meaningful in this context.
- I find it a bit challenging to understand why both the local and global identities are required and how they bring complementary information.
- In the derivation of local identities, the $\tau(r)$ component in Eq. (9) appears and did not appear previously in the global identities, please explain. maybe specify what is meant by the kinematic term — it’s unclear to me how this is linked to diffusion (see comment later).
- Steps leading to Eq. 10 are not crystal clear. Where has Fext gone? - I actually believe rather than using equation 9 here the right hand side of eq. 8 was just expanded.
- “For cases where the observable under consideration is independent of the momenta, i.e.” the derivations are full of small important statements like these which condition the assumptions under which the derivations/results provided are true. It would be useful to recapitulate (a) the results in a table but also (b) the list of these assumptions (including the fact that they are true at equilibrium).

- “also no global diffusive effect due to vanishing boundary terms.” I do not understand the reference to diffusion here.
- “As a further remark, by splitting off the kinetic term in Eq. (11), its left hand side can be re-written as” (there are too many words for similar but not the same things such as kinetic, kinematic, and diffusion…). The identity following this quoted sentence involving F_u for the first time is unclear to me – where had the τ contribution in Eq. (9) vanished?
- “Setting $\hat{A} = 1$ recovers the YBG equa- tion, as then then last term in Eq. (12) vanishes and the remaining terms constitute Eq. (1).” I have a hard time understanding this since the YBG is a local equality whereas this is integrated over the probability distribution function (true in average only). Is the YBG also supposed to have brackets? If so then this should be made clearer.
- “Paralleling the nam- ing convention of the hypervirial theorem, which gener- alizes the standard virial theorem to include a further observable, Eq. (12) attains the status of a hyper-YBG equation or hyperforce balance relationship.” This is an example of a jargon-full sentence. I personally do not retain concrete scientific content from this sentence. Maybe using simpler words, or adding a broader sentence for context, something such as “conceptually, this relation can be thought of as a … because…” might make things clearer. And also using references for e.g. “hypervirial theorem” and “to include a further observable”. Maybe the parallel between hypervirial/hyperforce could be made clear from the start. But the 1st occurrence of hyperforce is made without this parallel. In addition, the manuscript repeats content several times –this statement also appears in the introduction. Maybe these iterative statements could be avoided.
- “Besides the conceptual difference between correlation and covariance, in practi- cal sampling schemes it can be beneficial to work with covariances rather than correlations to reduce statistical noise, as we will demonstrate further below.” I found at this stage I was lacking a reason for “why” this was the case. It could be briefly mentioned now, I also could not find the answer to this question in the work below. This discussion and the one of the previous paragraph could be put in a “Section II. D. General discussion on the hyperforce relation”…
- “That Eq. (11) holds can again be verified a posteriori by phase space” the calculations in this paragraph are hard to follow.
- III. A. “We first consider the seemingly trivial case $\hat{A} = 1$,” – this case was considered before and I find it strange to mention it again, since the system is at equilibrium.
- “autocorrelation”(even “correlation”) I find this choice of word confusing since autocorrelation to me refers to me to a spatially or temporally dependent correlation function. Here this is not the case. The correlation functions are not really correlation functions so much as cross-averages/variances/moments of existing functions.
- “The result is the following global second order hyperforce” why second order?
- The identities in the text right above equation 14 relating derivatives of the density are used previously but not mentioned. Why is there no average on the right-hand side of eq. 14? Eq. 14 appears to be an example of relation to be put in a recapitulating table.
- “inverse” LMBW: why “inverse”? What this parallel brings to the general results is not clear to me.
- “Furthermore, by summing only over particle pairs with unequal indices we obtain the distinct identity $\langle F^0_{ext}\, \hat{\rho}(r)\rangle_{dist} = F_{int}(r)$” I found this derivation was not straightforward.
- What is “adaptive” BD? It seems that simple BD is used here which does not require an extra adjective.
- The methods for BD and MC are scattered in the middle of a text on applications. I would advise to split them apart with a “methods” subsection for clarity. what is the thermostat used? 10^8

steps for only 200 particles for thermalization sounds overdone - why was such a long relaxation time necessary? what is the “friction constant” and is it related to the thermostat?

- Figure 2: I’m not sure I understand why the green curve is related to the other plots in Fig. 2a ? Isn’t there a term missing for it to be equal to this? In Fig. 2d, isn’t there an inversion of the colors? Since the noisy blue curve would be expected to actually be the derivative. I am also confused about the results of Fig. 2d. They seem to hint that there is a non zero external force on the system. Is that the pressure from the walls? what is the ensemble in which these simulations are conducted?
- Which method is “better” to compute all these quantities from Fig. 2 is not clear to me. Please discuss.
- Why are these called Noether identities and not e.g. Green-Kubo? Green-Kubo imply time dependencies and hence I don’t think this is a good choice of words. However I found I could not say why they should be called Noether identities.
- In Fig. 3 the hyperforce relations seem to always lead to noisier results than standard methods. Please discuss.
- “Furthermore, increasing the spatial resolution and hence selecting $\hat{A} = \hat{F}(r)$ in Eq. (10) yields the recent Noether-constrained two-body force-correlation theory, which is discussed in detail in Refs. [34]. “ I could not understand this statement without delving into Ref. 34. Please simplify?
- “Here we generalize to higher orders by iteratively replacing”: this entire section seems ill-positioned and could be discussed in Sec. II as an extension of II-C, while section III is focused on tests/applications, etc. I found it hard to revert back to theoretical derivations at this point of the manuscript.
- Fig. 4: The new method (blue) converges the least fast. so how is the method better? The convergence time is not really commented on here… and could directly be studied instead. Is this due to the fact that there is just 1 run? And that statistics are not averaged over multiple independent runs?
- Fig. 5: It’s not clear to me what the conclusion of this plot is as discussed in the main text -- also the way this is plotted is such that it’s hard to tell which curve has the most noise?
- I got lost somewhere how can Eq. 23 and Eq. 24 both be true with respect to dimensional analysis?
- “Having such tools for quality control can be particularly useful” this seems a useful comment however, coming at the end of a long manuscript and without a practical step-by-step guideline on how and what to do this quality control check is unclear to me. How is this different from e.g. simply waiting for long enough times to see if this converges to a stationary solution?

Hyperforce balance via thermal Noether invariance of any observable
by S. Robitschko, F. Sammueller, M. Schmidt, and S. Hermann

Replies to Reviewers' comments:

Reply to Reviewer #1
==========

We cordially thank the Referee for deeply engaging with our work, for providing useful feedback, and for making a very favourable assessment. We are grateful for raising a specific comment, on which we comment below in detail.

> The theoretical concepts developed in this work represent an
> important new building block in the construction of a modern view
> of statistical mechanics and of new pillars that constitute the
> derivation of exact sum rules. It promotes also the power of
> Noether's fundamental principle of translational invariance in
> general physics and here statistical physics. From this principle
> the authors are able to generate exact sum rules relating the
> covariance of various force and energy quantities to the mean
> gradient of an associated phase-space function. The simplest
> example is the Yvon-Born-Green relation relating the local force
> density to the gradient of the particle density (eq 1). This
> equation follows naturally from the Noether's principle
> (generalised to displacement fields) and appears as a particular
> and simple choice in the general equation 11 expressing the
> correlation of the local force with a general,
> momentum-independent observable A(r^N).

> Playing with the observable A leads to a multitude of known and
> unknown exact sum rules. These sum rules, as well as their
> convergence, are verified in simulation for a confined LJ
> fluid. They can be used either to optimize the statistical
> convergence or to assess thermodynamic equilibrium. I note that
> among the different possible ways of computing the one-particle
> density that emerge from the general theory (eqs 22-25), the
> already well-tested « force method » given by eq 1 apparently
> remains the most efficient and accurate one.

> I have absolutely no doubt on the substance of the article and its
> suitability to Comm. Physics, and I strongly recommend its
> publication. I have just the minor remark that general equation 11
> with an arbitrary observable A(r^N) should be balanced with with
> Coles et al, JCP 151, 064124 (2019) where it was used for
> particular A's based on single particle observables of the form
> sum_i delta(r - r_i) a_i.

This is an excellent comment and we thank the Reviewer for pointing out the important connection to the work by Coles et al., which we had failed to recognize before. We now refer to their 2019 work in the following added discussion below Eq.(11):

"Equation (11) can be viewed as a generalization of the framework
developed by Coles et al. [17], where they consider observables of

the specific form A = \sum_i a_i delta(r-r_i), where a_i is a unique property of particle i only, such as e.g. its charge or, when taking orientational degrees of freedom into account, its polarization [17].

We also now refer to Ref.[17] several times in the manuscript in the context of generalized sampling techniques. We have updated our list of references to also include very recent work by this group [19].

Newly added references:

[17] S. W. Coles, D. Borgis, R. Vuilleumier, and B. Rotenberg, Computing three-dimensional densities from force densities improves statistical efficiency, J. Chem. Phys. 151, 064124 (2019).

[19] S. W. Coles, B. J. Morgan, and B. Rotenberg, RevelsMD: Reduced variance estimators of the local structure in Molecular Dynamics, arXiv:2310.06149.

> Concerning the format, the paper is written in a format that I
> like where every equation is justified and follow logically from
> the preceding one. I am not sure though that it fits well to the
> format of the journal where the paper is recommended to begin with
> a result section where the outcomes are presented in the most
> clear and striking way, and end with a method section where the
> theoretical and computational methods are detailed and
> deepened. This might necessitate a little rearrangement. I leave
> this to the absolute discretion of the authors and to the
> recommendations of the editor.

We thank the Referee for raising this point of presentation and for backing up our style of writing and developing the theoretical ideas. We are grateful for the positive feedback. Inline with the Editor's note on the possibility of an exception from the general stylistic guidelines for Communications Physics, where the "method" indeed is the research, we are very happy to be able to keep the structure of the paper intact.

We thank the Referee for the positive assessment and for making an important point in relation to the existing literature.

Reply to Reviewer #2
==========

We cordially thank the Referee for a critical and detailed reading of our paper, for the positive assessment and for making a number of highly useful remarks. We have addressed these comments as detailed in the following.

> Report on Robitschko et.al

> This is a very interesting paper that introduces important new

> relationships/identities in (classical) statistical mechanics. It
> is quite rare to see new fundamental and powerful formulae emerge
> in this field; current practitioners are usually concerned with
> applications in solid state and in soft matter.

> The authors build upon earlier papers from their group refs[29-34]
> which showed that use of Noether's theorem leads to several exact
> identities involving force fluctuations. Here they present an
> impressive general treatment of the (deep) implications of Noether
> invariance for both global and locally resolved quantities. Many
> of the 'sum rules' they derive are remarkable in their simplicity
> and elegance and in the depth of physics they contain. The paper
> is well-written; it explains nicely the subtle issues associated
> with the different types of invariance as well as the importance
> of the new identities for assessing efficiency and accuracy of
> computer sampling schemes. The authors choose to test the sum
> rules numerically via simulations of a (truncated) LJ fluid
> confined in a planar slit pore. This is a very sensible choice of
> model system and results are nicely presented.

> Some comments:

> 1.The force balance equation (1) is indeed a statement of the 1st
> YBG equ. The latter is usually written as an integro-differential
> equation for the particular case of pairwise interparticle
> potentials, as in Ref.[1], Sec.4.2. For scholarly interest it
> would be useful to give a sentence or two of background. What did
> Yvon say?

We have made our description in the text more accurate by adding the following text below Eq.(1).

"This is a classical result due to Yvon, Born and Green (YBG) [1], for particles that mutually interact only via a pair potential, the interparticle force density F_int(r) is expressed as an integral over the two-body density multiplied by the pair force [1]."

We also thank the Referee to triggering us to re-read the classical literature. This made us realize that giving a meaningful historical account, even a brief one, should not be taken light-heartedly, which is why we shy away in the present context and leave this to future work.

We have added two citations [61,62] to the classical works and are brushing up our skills in the beautiful French language to be able to take in Ref.[61] in its entirety. (Our specific copy of Ref.[61] is acquired via library loan from Stuttgart, where we gather it has been well-used over many years.)

[61] J. Yvon, La theorie statistique des fluides et l'equation d'etat (in French), Actualites scientifiques et industrielles No. 203 (Hermann, Paris, 1935).

[62] M. Born and H. Green, A general kinetic theory of liquids I. The molecular distribution functions, Proc. R. Soc. London, Ser. A 188, 10 (1946).

> 2.In caption to Fig.1 (b) spell out that the wall separation is L.

This is a good suggestion. We have appended to the caption to Fig.1(b):

"and L is the distance between the two walls."

> 3.The algebra involved in the various correlators and their
> manipulations is clearly presented and I followed this. (Indeed, I
> enjoyed checking several derivations.) Sometimes there is
> repetition in explaining the content of the sum rules but given
> their subtlety and generality I think there is no harm in this.

We have carefully checked whether the repetitions can make for tiresome reading. However, on balance we agree with the Reviewer's assessment that a certain level of redundancy helps in being precise in the description of the individual steps.

> 4.In the 1st derivation from (2) to (3) one might be pedantic and
> add 'noting that the partition sum is independent of eps_0'.

This is again a good point. We have added this statement. The corresponding extended sentence reads as follows.

"[Carrying out the derivative is straightforward upon using the
Boltzmann form of the equilibrium distribution function] and noting
that the partition sum is independent of eps_0."

> 5.It would help the non-expert reader if the LJ potential $\phi(r)$
> were spelled out in pg.5. This is important when it comes to the
> confining wall potential. As far as I can tell, this is not the
> sum of 9-3 potentials as is commonly used for confinement. The
> authors might write explicitly what is V_ext(z) for their
> confining potential. Also, it would assist the reader if they
> wrote an explicit formula for the average evaluated in (14) for
> the particular 1D geometry. The reader would then get a better
> sense of the physics and the import of this relation. After all
> the authors emphasize that (14) is 'remarkable' and later 'very
> counter-intuitive'-and the referee agrees. It would help to lay
> out precisely what is being calculated in the model system.

We agree and we have added the requested clarifications. We give the formula for the LJ potential explicitly and also write out the wall potential. We emphasize in the accompanying text that our choice of wall potential is for convenience only. Please see the revised text (indicated in blue) on p.5.

We have also added a description of the sampling in planar geometry on p.6. Additional details are provided below Eq.(25).

Please see the revision indicated in blue in the paper.

> 6. In the figures blue lines sometimes appear as blue dots. I was

> confused at first; the authors might add a sentence to the first
> caption explaining why there are blue dots.

We are grateful to the Reviewer for pointing out the inconsistency in the graphics. Rather than adding an explanatory sentence we have modified the legend of the figure to now indicate a broken blue line (dots) rather than the previous full blue line. The legend and the graphs are now entirely consistent. This modification has affected the legends in Figs.2, 3, and 5.

> 7. Pg.6, 4 lines below (16) add 'which follows from (13) with
> operator A =1'. A pedantic point.

We do not think that the Referee's point is pedantic, but rather that it is both correct and helpful. We have added the statement as suggested.

> 8.Eqs (22-24) are important. Eq. (24) should have z' in the final
> round bracket. Spell out what rho_0 is.

Thank you for spotting the missing prime, which we have corrected. We clarify the meaning of rho_0 by the following added text:

"Here we choose the integration constant as rho_0=rho(0)=0 due to the divergent wall potential at z=0."

> 9.In the caption to Fig.5 the term 'mirroring' is introduced. This
> should be explained.

We have added the following description.

"[The results from sampling are symmetrized with respect to mirroring at the center of the pore,] i.e., via building the the arithmetic mean [F_int(z) - F_int(L-z)]/2 [...]"

> 10. That results should not depend on the ensemble is mentioned on
> pg.5 Col.2 and again on pg.10 Col.1.At first, I was sceptical
> about this statement as the formal manipulations deriving the new
> identities are performed straightforwardly and naturally in the
> GCE. If the authors have shown that the manipulations remain valid
> in the CE this is important and worth emphasizing further. Of
> course, this also justifies employing Brownian Dynamics with fixed
> N to test the sum rules.

This is a good suggestion. We have added a corresponding discussion to the conclusions. The added text reads as follows.

"In our theoretical derivations we have relied on the grand canonical ensemble, with fixed chemical potential mu and fluctuating number of particles N. Carrying out formal manipulations in this way is often more straightforward than working with fixed N, as is appropriate for a canonical treatment. (Temperature is constant in both ensembles.) A prominent

example is to obtain the density profile as a functional derivative rho(r)=d Omega/d V_ext(r) where crucially mu is kept fixed, rather than N, upon building the functional derivative. This prototypical example demonstrates the elegance of working grand canonically, and one could expect that a similar situation applies for the thermal Noether invariance. This, however, is not the case. Rather the phase space shifting transformation, whether global by a constant eps_0 or locally resolved in position via a three-dimensional vector field eps(r), is an entirely mechanical operation that applies equally well canonically. The shifting invariance gives a powerful new route to correlation functions and and their sum rules, alternative to the traditional method of integrating over degrees of freedom, as pioneered by Yvon [61] and Born and Green [62].

A detailed account of global shifting in the canonical ensemble is provided in Ref.[32]. The resulting Noether force identities are analogous in form to the results from a grand canonical treatment [31], with the sole (and trivial) difference of the definition of the respective ensemble averages. Here we find that the analogous situation holds for the hyperforce identities. As they originate from phase space transformations only, they are insensitive to the ensemble differences between the canonical and the grand ensemble. This theoretical fact is corroborated by our computer simulation results, where we have explicitly compared grand canonical Monte Carlo data and canonical results, with the latter obtained via sampling under adaptive BD time evolution [41]."

> 11. The use of the term ‘hyperforce’ might raise eyebrows. The
> hyper appears to arise from an analogy with Hirschfeld’s (1960)
> hyper virial theorem. Most readers will not be aware of the
> latter. I was not. Thus, the authors might like to clarify why (4)
> is the ‘global hyperforce identity’.

We certainly want to avoid raised eyebrows but also want to use terminology that reflects the essence of the developed concepts. Clearly we do not shy away from catchy language. As was detailed in the original paper and recognized by the Referee, it was Hirschfelder who introduced the generalization concept in his hypervirial theorem. In order to communicate this historical situation more directly, and also to give further exposure to work by Hirschfelder, we have added the following text to the abstract of our paper.

"Similar to Hirschfelder’s hypervirial theorem [J. Chem. Phys. 33, 1462 (1960)], hyperforce sum rules apply to arbitrary observables in equilibrium."

We also give further discussion about naming in the paragraph below Eq.(4).

"As announced in the introduction, Eq. (4) is similar to Hirschfelder’s hypervirial theorem [37] in the inclusion of the phase space function A(r^N, p^N) and our terminology of 'hyperforce' parallels his use of 'hypervirial'."

> 12. Sec.IV is a bit rambling but very thought provoking. The
> authors might like to review and re-draft to sharpen the
> messages. For example, it is well-known that the spatial
> derivative of (28) corresponds to the force balance eq. (1) and
> therefore it is clear what the physical significance is of
> rho(r)*grad c_(1)(r). The new thing is probing the derivative of
> F_int(r). Re-writing would clarify.

We accept the criticism. We have re-considered carefully the Conclusions and have made much more compact the description of possible links to force-DFT and to machine learning.

> Summary.

> This is a fine piece of work that makes a substantial and
> scholarly contribution to statistical physics. The sum
> rules/identities that are introduced are insightful and are likely
> to be valuable in applications across condensed matter physics and
> physical chemistry. I recommend the paper be accepted for
> publication provided the authors address the suggestions
> above. Note: these are fairly minor.

We thank the Referee for a wealth of constructive feedback, detailed criticism and useful suggestions for improvements. We hope that our revision meets the Referee's standards.

Reply to Reviewer #3
==========

We thank the Referee for a critical reading of our paper and for making a wealth of constructive and detailed comments. We appreciate the level of depth of engagement with our work. We have addressed the Referee's suggestions and criticisms as detailed in the following.

> I have read the manuscript “Hyperforce Balance via Thermal Noether
> Invariance of Any Observable”. This manuscript describes an
> analysis technique, for computer simulations of particle dynamics,
> to relate correlation functions including forces to obtain local
> estimates of density gradients or force gradients, without
> computing the gradients. A rigorous derivation is provided for
> these relations for many-body particle systems at
> equilibrium. Applications are given and the method is tested on
> computer simulations of a confined liquid between two walls. The
> method is general and allows one to access a plethora of variables
> thanks to these force-based correlations. Calculating gradients of
> noisy data is a general challenge in small-scale physics. As such,
> the proposed method offers an interesting route to improve and
> complement existing analysis routines in computer simulations.

> Given its potential broad impact, I believe the manuscript could
> be suitable for Communications Physics. In its current state, I
> find the manuscript hard to read, and the main results, as well as

> how the method compares to existing methods (in terms of accuracy
> and convergence speed) still remain somewhat elusive to me. The
> manuscript may contain a very important contribution to the field,
> however that contribution is lost within a vast array of results
> that are not necessarily sorted/hierarchized and the recurring use
> of jargon. In general, clarity could be greatly improved (a) by
> stating the main result in the introduction (b) by providing a
> table of derived examples and what information they provide (c)
> referencing a specific example in the abstract/intro (for instance
> calculating density gradients with a force- density correlation)
> (d) stating clearly how this method compares to existing
> method/how it might complement them. I provide specific
> point-by-point feedback below. Provided significant clarifying
> changes are made on the manuscript, I believe the manuscript will
> be suitable for Communications Physics.

We comment as follows on the four points raised by the Referee.

> (a) by stating the main result in the introduction

Our main result is the first-principles construction of a general framework for correlation functions that systematically combine a given observable A with the relevant forces that act in the system. We give both global and position-resolved, local versions. The main results are summarized and anticipated in the following text at the end of Sec.I.

"Our framework can be viewed as a generalization of the YBG equation (1) to systematically include the dependence on a further given observable \hat A. The equilibrium force balance (1) itself is recovered for the trivial case \hat A=1. Such generalization is not uncommon in Statistical Mechanics. The relationship of our theory and the YBG force balance equation is akin Hirschfelder's hypervirial theorem [37] as a generalization of the standard virial theorem [1] to also invoke an additional dependence on a given phase space function \hat A. Our theory can hence be viewed as a hyperforce balance relationship, and we derive global and local variants below, see Eqs.(4) and (10). We further show that the local version simplifies further to Eq.(11) and more explicitly to Eq.(12) in case of hat A being independent of momenta."

We have appended (please also see the next point.)

"See Fig. 6 for an overview of results."

Figure 6a and b summarize the main results and they are shown as part of the Conclusions of the paper, but are anticipated in the Introduction by the above text.

> (b) by providing a table of derived examples and what information
> they provide

This is a good suggestion. We have provided an overview of the key results in the form of an added Fig.6. Thereby panel (a) shows the relevant general identities with a general operator \hat A. Panel (b) gives concrete examples for specific choices of \hat A. We have opted for figures rather than the suggested table format, as the latter proved impractical due to type-setting issues with formulae. Please see the manuscript for the newly added Fig.6 and its caption, which briefly describes the content and gives explicit references to the relevant equations in the main text.

> (c) referencing a specific example in the abstract/intro (for
> instance calculating density gradients with a force-density
> correlation)

We take the Referee's point and have added the specific example at the end of Sec.I.

"As a specific example, our methodology not only allows to sample density gradients, as is possible in force sampling schemes [13–19], but also to sample force density gradients."

> (d) stating clearly how this method compares to existing
> method/how it might complement them.

We have appended the following statement at the end of the introduction.

"The general method complements existing counting and force-sampling techniques and it gives much inspiration for rigorous statistical mechanical theories based on exact identities."

> 1 - abstract: “its negative correlation with the total force”: this
> is a statement I do not understand. The abstract is, in
> general, very hard to read/as well as the title, and does not
> provide the reader with an immediate understanding of what they
> might do with this technique.

The term correlation indeed has multiple meanings, first and maybe foremost as a complex physical phenomenon that is ubiquitous in condensed matter physics. Second, and much more specifically, in a stochastic terminology setting, which is surely not uncommon in physics, it is merely the average of the product of two random variables. It is the latter meaning that we intended to convey in our original abstract. In order to avoid any confusion we have re-phrased the sentence as follows.

"[Both global and locally resolved identities hold and they relate the mean gradient of a phasespace function to] its negative mean product with the total force."

We also clarify in the text as follows.

"[The sum rule (4) is striking, as it relates the correlation of \hat A with the external force operator (left hand side) to the mean negative global coordinate derivative of Aˆ (right hand side);] here we use the term “correlation” to imply the average of the product of two observables."

> 2 - “The results clarify a very intimate link of global and locally
> resolved correlators and they suggest a very general statistical
> mechanical structure.” This is a vague statement that may be
> rendered clearer with a main result under the form of an
> equation. It would be really nice at this stage to already have an
> example, or to treat examples later on and say at the beginning
> what kind of examples will be investigated.

The above modifications already cover these points. We do not accept the point that the quoted statement is vague. Rather it encapsulates, in compact form, the essence of the progress that we report. We have also shied away from adding an example at an early stage and rather point the Reader to the overview Fig.6.

> 3 - “Our framework can be viewed as a generalization of the YBG
> equation”: I find this statement equally confusing. What is the
> added contribution/the things made possible by this work compared
> to previous works, beyond the conceptual and derivation aspects?

The added contribution is the systematic incorporation of an arbitrary observable A, as emphasized throughout the manuscript, both in the text and -obviously- in the mathematics.

> 4 - “hyperforce” please define this word and say why it is meaningful
> in this context.

We accept this point. In order to be clearer about the terminology we have added the following statement to the abstract.

"Similar to Hirschfelder’s hypervirial theorem (J. Chem. Phys. 33, 1462 (1960)) hyperforce sum rule apply to arbitrary observables in equilibrium.:

> 5 - I find it a bit challenging to understand why both the local and
> global identities are required and how they bring complementary
> information.

Having resolution in position is crucial when investigating spatially inhomogeneous systems. Yet even in spatially inhomogeneous systems, well-defined global averages are meaningful. In our invariance theory the differences arise from either considering spatially resolved forces or global forces. Surely, having this variety is a strong feature of our treatment.

> 6 - In the derivation of local identities, the $\tau(r)$ component
> in Eq. (9) appears and did not appear previously in the global
> identities, please explain. maybe specify what is meant by the
> kinematic term — it’s unclear to me how this is linked to
> diffusion (see comment later).

These are all valid questions. However, this is standard material that is covered elsewhere in great detail. Our current paper is focused on a specific invariance property of general observables. It is impractical to cover the issues that the Referee raises. In order to address the comment we have added in parenthesis guidance for suggested further reading as follows.

"(The force density operator F(r) defined in (9) also arises as the time derivative of the one-body current operator [20]. In equilibrium the kinematic term in (9) reduces to a diffusive contribution: div <tau(r)> = -kT grad rho(r); we refer the Reader to Ref. [20] for details.)"

[20] M. Schmidt, Power functional theory for many-body dynamics, Rev. Mod. Phys. 94, 015007 (2022).

> 7 - Steps leading to Eq. 10 are not crystal clear. Where has Fext
> gone? I actually believe rather than using equation 9 here the
> right hand side of eq. 8 was just expanded.

This calculation is straightforward. We provide more detail via the following additions in parenthesis:

"Equation (8) can then be written upon carrying out the functional derivatives and using Eq. (9) (on the left hand side) together with the chain rule (on the right hand side), as the following hyperforce sum rule [...]"

> 8 - "For cases where the observable under consideration is independent
> of the momenta, i.e." the derivations are full of small important
> statements like these which condition the assumptions under which
> the derivations/results provided are true. It would be useful to
> recapitulate (a) the results in a table but also (b) the list of
> these assumptions (including the fact that they are true at
> equilibrium).

All mathematical assumptions are provided in the text. Our entire theory covers equilibrium physics, as is clearly stated. Note that already our first sentence of the abstract starts with: "We demonstrate that *thermally averaged* classical phase space functions, [...]". A mere word count reveals that we use the word "equilibrium" circa 25 times throughout the paper. We can surely not be accused of being opaque about treating equilibrium physics. As detailed above, we have added Fig.6 to give a compact overview of results, as was sensibly raised by the Referee.

> 9 - "also no global diffusive effect due to vanishing boundary
> terms." I do not understand the reference to diffusion here.

We have clarified the connection to diffusion in point 6 above.

> 10 - "As a further remark, by splitting off the kinetic term in
> Eq. (11), its left hand side can be re-written as" (there are
> too many words for similar but not the same things such as
> kinetic, kinematic, and diffusion...). The identity following

> this quoted sentence involving F_U for the first time is
> unclear to me – where had the τ contribution in Eq. (9)
> vanished?

Again, this is covered in point 6 above.

> 11 - “Setting A = 1 recovers the YBG equation, as then then last
> term in Eq. (12) vanishes and the remaining terms constitute
> Eq. (1).” I have a hard time understanding this since the YBG
> is a local equality whereas this is integrated over the
> probability distribution function (true in average only). Is
> the YBG also supposed to have brackets? If so then this should
> be made clearer.

Inline with several standard works, we indeed refer to the YBG equation only in an averaged sense. In this context we have clarified that the specific contribution of YBG was to provide an integral equation hierarchy, where in particular the localized interparticle force density F_int(r) is obtained as an integral over r' of the two-body density multiplied with the pair force. We have added the following remark in parenthesis.

"Setting \hat A=1 recovers the YBG equation (which we take to imply thermal averages being taken), as then then last term [...]"

> 12 - “Paralleling the naming convention of the hypervirial theorem,
> which generalizes the standard virial theorem to include a
> further observable, Eq. (12) attains the status of a hyper-YBG
> equation or hyperforce balance relationship.” This is an
> example of a jargon-full sentence. I personally do not retain
> concrete scientific content from this sentence. Maybe using
> simpler words, or adding a broader sentence for context,
> something such as “conceptually, this relation can be thought
> of as a ... because...” might make things clearer. And also
> using references for e.g. “hypervirial theorem” and “to
> include a further observable”. Maybe the parallel between
> hypervirial/hyperforce could be made clear from the start. But
> the 1st occurrence of hyperforce is made without this
> parallel. In addition, the manuscript repeats content several
> times –this statement also appears in the introduction. Maybe
> these iterative statements could be avoided.

As detailed in point 4. above, we have clarified the naming convention in the revised abstract. We agree that there is some repetition, but we also think that the level of redundancy is acceptable and that it rather can help to get our point across.

> 13 - “Besides the conceptual difference between correlation and
> covariance, in practical sampling schemes it can be beneficial
> to work with covariances rather than correlations to reduce
> statistical noise, as we will demonstrate further below.” I
> found at this stage I was lacking a reason for “why” this was
> the case. It could be briefly mentioned now, I also could not

> find the answer to this question in the work below. This
> discussion and the one of the previous paragraph could be put
> in a “Section II. D. General discussion on the hyperforce
> relation”...

We have contemplated the Referee's suggestion but have decided to keep the present sectioning scheme, as it accurately and concisely structures the content of our paper. That covariances work better than correlations (understood as the mean of the product) is an empirical observation, which we attribute to a cancellation effect of statistical noise. We leave a more systematic investigation of the reasons to future work.

> 14 - “That Eq. (11) holds can again be verified a posteriori by
> phase space” the calculations in this paragraph are hard to
> follow.

In the interest of overall length of the paper we have kept these calculations brief. There is nothing of conceptual difficulty, so we deem it sufficient to only give a layout of the argumentation. Note that all results have been derived by the arguably more fundamental Noether invariance route earlier in the manuscript. Hence the phase space route plays a mere supporting role, which on the one hand we think is important to mention, as it is at face value independent of invariance and provides a check. On the other hand, we do not want to increase the length of the paper unnecessarily to not make our treatment look more complex than it actually is.

> 15 - III. A. “We first consider the seemingly trivial case A=1," I
> find it strange to mention it again, since the system is at
> equilibrium.

This is the first time in the manuscript that we treat \hat A=1 in the global context. We have commented on the sufficient clarification of "equilibrium" in point 8 above.

> 16 - “autocorrelation” (even “correlation”) I find this choice of
> word confusing since autocorrelation refers to me to a
> spatially or temporally dependent correlation function. Here
> this is not the case. The correlation functions are not really
> correlation functions so much as
> cross-averages/variances/moments of existing functions.

We have commented on our (standard) use of the word "correlation" above in point 1.

> 17 - “The result is the following global second order hyperforce”
> why second order?

The thermal Noether invariance, on which the present investigation is based, holds for finite displacements epsilon. Hence when Taylor expanding around the unshifted system epsilon=0, all orders in the series expansion need to vanish. Inspecting each order separately provides a classification scheme and the resulting identities have characteristic structure that reflects their order. In particular the second order provides variance (mean of the square) identities [34].

[34] S. Hermann and M. Schmidt, Variance of fluctuations from Noether invariance, Commun. Phys. 5, 276 (2022).

The situation here is similar as the external force operator appears as a product with itself on the right hand side of Eq.(13). We trust that our wording is sufficiently clear and we have not added further explanations in the interest of brevity.

> 18 - The identities in the text right above equation 14 relating
> derivatives of the density are used previously but not
> mentioned. Why is there no average on the right-hand side of
> eq. 14? Eq. 14 appears to be an example of relation to be put
> in a recapitulating table.

Surely the density profile rho(r) is an average! For Readers who have missed this very essential point, we have added the following clarification below Eq.(14).

"where we recall that on the right hand side rho(r)=<\hat rho(r)> is
the averaged density profile."

As the Referee suggested, we included Eq. (14) in the recapitulating summary (Fig.6).

> 19 - “inverse” LMBW: why “inverse”? What this parallel brings to
> the general results is not clear to me.

There are multiple sum rules associated with these authors, in particular involving direct correlation functions, so we find the naming to be useful. The equation itself is given explicitly, so we cannot see that any harm is done. We also make the origin of this result clear via citations of Refs.[39,40].

[39] R. A. Lovett, C. Y. Mou, and F. P. Buff, The structure of the
liquid-vapor interface, J. Chem. Phys. 65, 570 (1976).

[40] M. S. Wertheim, Correlations in the liquid-vapor interface,
J. Chem. Phys. 65, 2377 (1976).

> 20 - “Furthermore, by summing only over particle pairs with unequal
> indices we obtain the distinct identity ⟨F0ext ρ^(r)⟩dist =
> Fint (r)” I found this derivation was not straightforward.

We accept the point and have added the following description.

"The derivation is straightforward by starting from Eq. (14),
subtracting the self contribution which is the YBG equation (1)
in the form −\beta rho(r) nabla Vext(r) = −beta F_int(r) + nabla
rho(r), and dividing the result by beta."

> 21 - What is “adaptive” BD? It seems that simple BD is used here
> which does not require an extra adjective.

No, we indeed use adaptive BD [41] as stated clearly in the paper. We use this algorithm due to its tight error control on forces, which is not possible via the standard Euler-Maruyama BD scheme for the overdamped motion. In adapative BD the time step is automatically adjusted while keeping the

statistical properties of the white noise intact, which is nontrivial to achieve. The source code is available, in case anyone is tempted to give it a try.

[41] F. Sammuller and M. Schmidt, Adaptive Brownian dynamics,
J. Chem. Phys. 155, 134107 (2021).

> 22 - The methods for BD and MC are scattered in the middle of a
> text on applications. I would advise to split them apart with
> a “methods” subsection for clarity. what is the thermostat
> used? 10^8 steps for only 200 particles for thermalization
> sounds overdone - why was such a long relaxation time
> necessary? what is the “friction constant” and is it related
> to the thermostat?

Adaptive BD intrinsically operates at fixed temperature (which together with the friction constant controls the variance of the noise). We agree that the thermalization time is generous, but that creates no harm. The description of the simulation methods is rather compact, as we use standard techniques. We found that the present location in the text makes for easier reading rather than restructuring and as a result asking the Reader to jump around between different sections.

> 23 - Figure 2: I’m not sure I understand why the green curve is
> related to the other plots in Fig. 2a ? Isn’t there a term
> missing for it to be equal to this? In Fig. 2d, isn’t there an
> inversion of the colors? Since the noisy blue curve would be
> expected to actually be the derivative. I am also confused
> about the results of Fig. 2d. They seem to hint that there is
> a non zero external force on the system. Is that the pressure
> from the walls? what is the ensemble in which these
> simulations are conducted?

The GCMC simulations are grand canonical. The adaptive BD simulations are at fixed temperature and fixed number of particles, so they provide canonical averages. As we emphasize in the caption, the results shown in Fig.2 illustrate the sum rules (14), (15), and (16). We have checked that there are no terms missing, as is suggested by the Referee and that all information provided is correct and described accurately.

We have added a description of the validity of our theoretical results in the canonical ensemble to the conclusions.

"In our theoretical derivations we have relied on the grand canonical ensemble, with fixed chemical potential mu and fluctuating number of particles N. Carrying out formal manipulations in this way is often more straightforward than working with fixed N, as is appropriate for a canonical treatment. (Temperature is constant in both ensembles.) A prominent example is to obtain the density profile as a functional derivative rho(r)=d Omega/d V_ext(r) where crucially mu is kept fixed, rather than N, upon building the functional derivative. This prototypical example demonstrates the elegance of working grand canonically, and one could expect that a similar situation applies for the thermal Noether invariance. This, however, is not the case. Rather the

phase space shifting transformation, whether global by a constant eps_0 or locally resolved in position via a three-dimensional vector field eps(r), is an entirely mechanical operation that applies equally well canonically. The shifting invariance gives a powerful new route to correlation functions and and their sum rules, alternative to the traditional method of integrating over degrees of freedom, as pioneered by Yvon [61] and Born and Green [62].

A detailed account of global shifting in the canonical ensemble is provided in Ref.[32]. The resulting Noether force identities are analogous in form to the results from a grand canonical treatment [31], with the sole (and trivial) difference of the definition of the respective ensemble averages. Here we find that the analogous situation holds for the hyperforce identities. As they originate from phase space transformations only, they are insensitive to the ensemble differences between the canonical and the grand ensemble. This theoretical fact is corroborated by our computer simulation results, where we have explicitly compared grand canonical Monte Carlo data and canonical results, with the latter obtained via sampling under adaptive BD time evolution [41]."

> 24 - Which method is “better” to compute all these quantities from
> Fig. 2 is not clear to me. Please discuss.

This is a complex question for which we do not provide a definitive answer. Here we deliver new ways of sampling. The questions of their relative quality and how to put them to productive use remain to be assessed in future work. In the manuscript we are very clear about both the possibility of improved sampling but also that the hyperforce route can provide more "difficult" routes that enable one to judge quality of equilibration.

Please see Sec. III C ("Hyperforce sampling and equilibrium testing") where these matters are discussed in the manuscript.

> 25 - Why are these called Noether identities and not
> e.g. Green-Kubo? Green-Kubo imply time dependencies and hence
> I don’t think this is a good choice of words. However I found
> I could not say why they should be called Noether identities.

We certainly agree that "Green-Kubo" is not an appropriate classification. The new relationships come out of thermal Noether invariance, as we show in detail in the paper. Hence we find it fitting to relate to these equations as Noether identities. The need for having such terminology arises from the use of Noether's theorem in the statistical mechanical equilibrium context. In contrast, in e.g. classical mechanics we are used to application of the Noether theorem yielding conservation laws. The analogous results in the present context are the Noether identities.

> 26 - In Fig. 3 the hyperforce relations seem to always lead to
> noisier results than standard methods. Please discuss.

The observation is correct and the implications of using the presented hyperforce sum rules as alternative sampling schemes in simulations are discussed in Sec. III C

> 27 - “Furthermore, increasing the spatial resolution and hence
> selecting \hat A = \hat F(r) in Eq. (10) yields the recent
> Noether-constrained two-body force-correlation theory, which
> is discussed in detail in Refs. [34]. “ I could not understand
> this statement without delving into Ref. 34. Please simplify?

We are grateful to the Referee for delving into Ref.[34]. We have checked and the current presentation is indeed as simple as possible. Entirely leaving away the connection to Ref.[34] would surely be the wrong option, as this is a clear link of the present results to earlier work. We also do not find it appropriate to give an abridged version of what is contained in Ref.[34], as this is a very recent paper that Readers can access to their liking.

> 28 - “Here we generalize to higher orders by iteratively
> replacing”: this entire section seems ill- positioned and could
> be discussed in Sec. II as an extension of II-C, while section
> III is focused on tests/applications, etc. I found it hard to
> revert back to theoretical derivations at this point of the
> manuscript.

While we agree that we ask the Reader to follow a theoretical chain of arguments in Sec.III, we find the present structuring of the material to be the most natural one in terms of the flow of arguments. In general we agree that keeping "derivations" and "applications" more clinically separated can make much sense in a multitude of settings. Here our guiding principle is maximal clarity of argumentation, as we trust is provided with the present setup. Not all readers will react in the same way; some might appreciate having presented some concrete results in the form of plots before seeing how the concepts carry further; others might find refreshment in the theoretical derivations.

On balance, while we appreciate the Referee's feedback, we have kept our form of presentation. We added a note of warning and now start the paragraph as follows.

"Reverting back to developing the general theory, [here we
generalize to higher orders by iteratively replacing]

> 29 - Fig. 4: The new method (blue) converges the least fast. so how
> is the method better? The convergence time is not really
> commented on here... and could directly be studied instead. Is
> this due to the fact that there is just 1 run? And that
> statistics are not averaged over multiple independent runs?

We deliberately show results from a single run in order to exemplify typical noise levels and statistical uncertainties. As we emphasize in the manuscript, the new ways of sampling can serve to assess the quality of the simulations, as more sensitive measures of accurate equilibration are provided than what was previously available.

> 30 - Fig. 5: It’s not clear to me what the conclusion of this plot
> is as discussed in the main text -- also the way this is
> plotted is such that it’s hard to tell which curve has the most
> noise?
> 31 - I got lost somewhere how can Eq. 23 and Eq. 24 both be
> true with respect to dimensional analysis?

The dimensional analysis of Eqs.(23) and (24) is valid. Both equations have units of the density profile, i.e. inverse length^3.

> 32 - “Having such tools for quality control can be particularly
> useful” this seems a useful comment however, coming at the end
> of a long manuscript and without a practical step-by-step
> guideline on how and what to do this quality control check is
> unclear to me. How is this different from e.g. simply waiting
> for long enough times to see if this converges to a stationary
> solution?

Sure, we do not question the validity of the common strategy to wait long enough in situations where this leads to convergence, which is often of course the case in small enough systems. In large systems, and in particular for conditions near (surface) phase transitions, it can be very hard to judge whether one has waited long enough or not. This is one of the primary reasons why we emphasize throughout our paper: Check the sum rules.

We thank the Referee for the wealth of feedback provided. While we were admittedly stubborn to an extent in certain matters of presentation, we have done our very best to implement all suggestions and criticisms that directly addressed the physics as well as the derivations at hand. We trust that this strategy is acceptable for the Referee.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

I am completely satisfied with the authors response letter, in particular their efforts to make their results even more clear and striking, and recommend publication as such.

Reviewer #2 (Remarks to the Author):

The authors have made a concerted and careful effort to address the points made in my original review. The revised version incorporates satisfactorily all the suggestions for improvement that I made.
I recommend strongly that the present version be accepted for publication.

Reviewer #3 (Remarks to the Author):

I would like to thank the authors for carefully addressing my comments and making an effort at clarifying some of their approach. I am happy to recommend this manuscript for publication in Communications Physics. I have a few remaining comments that I encourage the authors to address to their discretion.

- it could be specified for clarity that the correlation functions are taken at equal time.
- If someone uses standard BD or MD (with a non adaptive time step); should they expect something different in terms of noise on the measured quantities? It was unclear what whether the “tight error control on forces” would affect significantly the correlation measurements — they may likely reduce the errors on correlations, which might be worthwhile to say for people trying to sample with forces.
- In Fig 2 the method yielding the most noise is not always the same (force sampling or counting). Can the authors comment on that?
- I found the statement that force sampling could be used as another measure to check convergence of slowly converging systems very interesting. Unless I am mistaken this statement does not appear before section III C and could benefit from being stated in the intro or abstract in the clear way that the authors used in the response.

Hyperforce balance via thermal Noether invariance of any observable
by Silas Robitschko, Florian Sammüller, Matthias Schmidt, and Sophie Hermann

REPLIES TO REVIEWERS' COMMENTS:

Reply to Reviewer #1

> I am completely satisfied with the authors response letter, in
> particular their efforts to make their results even more clear and
> striking, and recommend publication as such.

We thank the Referee for the judgement and once more for the useful feedback in the previous round.

Reply to Reviewer #2

> The authors have made a concerted and careful effort to address the
> points made in my original review. The revised version incorporates
> satisfactorily all the suggestions for improvement that I made. I
> recommend strongly that the present version be accepted for
> publication.

We thank the Referee for the positive recommendation.

Reply to Reviewer #3

We thank the Referee for the positive assessment and for once again raising interesting points. We have considered these points and we respond in detail in the following.

> I would like to thank the authors for carefully addressing my
> comments and making an effort at clarifying some of their
> approach. I am happy to recommend this manuscript for publication
> in Communications Physics. I have a few remaining comments that I
> encourage the authors to address to their discretion.
> - it could be specified for clarity that the correlation functions
> are taken at equal time.

We accept the point and have added the following clarification to the Conclusions.

> "In our BD simulations, we have sampled all correlation functions at equal time as is the appropriate limit in equilibrium time evolution to recover static thermal ensemble averages. As an aside, Ref. [31] exploits Noether invariance in nonequilibrium dynamics."

> - If someone uses standard BD or MD (with a non adaptive time step);
> should they expect something different in terms of noise on the
> measured quantities? It was unclear what whether the “tight error
> control on forces” would affect significantly the correlation
> measurements — they may likely reduce the errors on correlations,
> which might be worthwhile to say for people trying to sample with
> forces.

In essence one is free to use any suitable algorithm to generate configurations to carry out the averaging. We are obviously fans of the adaptive BD method due to its multitude of virtues, but the validity of the hyperforce identities is naturally independent thereof. To emphasize this point we have added the following discussion.

"We have used overdamped Brownian time evolution as a means to sample in thermal equilibrium. We find the adaptive Brownian dynamics time stepping algorithm of Ref. [41] to be a convenient choice for our present purposes. The principle validity of the hyperforce sum rules is nevertheless independent thereof and we expect careful use of either the simpler Euler-Maruyama method [41, 63] or indeed Molecular Dynamics [63] to yield identical results."

Added reference.
[63] D. Frenkel and B. Smit, Understanding Molecular Simulation: From Algorithms to Applications, 3rd ed. (Academic Press, London, 2023).

> - In Fig 2 the method yielding the most noise is not always the
> same (force sampling or counting). Can the authors comment on
> that?

While we understand the Referee's impetus for pushing us to provide more detail, here we have to refrain from adding further discussion. It is not clear at this point to what degree the observed level of noise depends on the specific sampling method or on the details of the physical situation considered. As we cannot be more concrete about this point, we decided to not add any further discussion.

> - I found the statement that force sampling could be used as another
> measure to check convergence of slowly converging systems very
> interesting. Unless I am mistaken this statement does not appear
> before section III C and could benefit from being stated in the
> intro or abstract in the clear way that the authors used in the
> response.

This is a good suggestion and we have added the following text early in the paper, at the end of the introductory section.

"As we lay out, the degree of numerical accuracy to which the Noether sum rules are satisfied can serve as an estimator for sufficient equilibration of slowly converging systems."

We have also modified the last sentence of the abstract as follows.

"These local correlators quantify spatially inhomogeneous self-organization and their measurement allows for the development of stringent convergence tests and enhanced sampling schemes in complex systems."

FURTHER CHANGES TO THE PAPER:

We have implemented all requested changes as outlined in the Editorial Requests Table.