Reviewers' comments:

Reviewer #1 (Remarks to the Author):

In this Communication, Hermann and Schmidt report on an application of Noether's calculus in the framework of statistical-mechanical functional theories of equilibrium and out-of-equilibrium many-body systems. More specifically, they derive sum rules and hierarchies of equations that connect various static and dynamical quantities, based on the invariance properties of the equilibrium density functional and of the out-of-equilibrium power functional. Both translational and rotational invariances are considered, with an application to fluids made of anisotropic particles.

This is an interesting and well-written paper, reporting sound technical work. In my opinion, the calculations are rather straightforward, but many of them lead to novel results which should be of interest to members of the statistical-mechanical community and could be useful in the future. In this respect, the paper is manifestly devised as a resource for future work, condensing the technical information needed for developments to come. Here lies a weakness of the paper, in my opinion. Indeed, there is no guarantee that the beautiful formal results will not turn disappointing, being of little use in concrete cases. Actually, this is not an uncommon situation. Therefore, I missed a more detailed discussion of how this work could be put to good use, in order to answer actual physical questions. The discussion is full of promises, but it remains quite vague.

In conclusion, I think that this paper is suitable for publication in Communications Physics, in terms of soundness, novelty and interest in the field. But I believe that it would be very beneficial to its impact if the authors could provide stronger arguments with regards to the future usefulness of the reported results.

Reviewer #2 (Remarks to the Author):

See attached report

Reviewer #3 (Remarks to the Author):

Please find the report attached.

Reviewer #4 (Remarks to the Author):

This paper considers the application of Noether's theorem to the (classical) statistical mechanics of interacting particles. The generality of the approach, in the sense that it is applicable to a broad category of functionals, is exploited within the framework of DFT (equilibrium) and PFT (nonequilibrium). By applying Noether's concept of invariance with respect to various symmetry operations the Authors derive a large number of statistical mechanical sum-rules. Some of these are known from previous calculations and some are new.

On the one hand this is an elegant and clever way to derive sum-rules using a unified approach, on the other it makes clear the connection between each sum-rule and a symmetry of the many-body

system. This is a valuable contribution and not something I have seen or thought about before. The paper is well written and fairly easy to follow - although I admit that I found the nonequilibrium part much more difficult, but this is probably due to my being less familiar with the PFT approach.

Publication recommended.

The Authors may wish to address/comment on the following points which occurred to me while reading the manuscript:

(1) A detail- First sentence of section II. I don't think it is the Grand potential which is translated, but rather the coordinates.

(2) In the nonequilibrium section the Authors derive sum-rules involving derivatives of the excess part of the power functional. I recall that quite recently explicit approximations were developed for the excess power (Phys. Rev. Lett. 120, 028001 (2018)) - do these approximations satisfy the derived sum-rules?

(3) In the discussion: The second paragraph contains several very interesting but somewhat obscure comments. Perhaps this could be expanded a little to make these more explicit.

(4) Is it clear which of these sum-rules will hold for approximate functionals (as typically employed, e.g. in practical DFT calculations)?

Report for COMMSPHYS-21-0151-T
Noether's Theorem in Statistical Mechanics
Sophie Hermann and Matthias Schmidt

This work applied Noether's Theorem to a number of settings in statistical mechanics. As noted by the authors, Noether's Theorem is widely-used in a wide range of physical settings, in particular in classical and high-energy physics. Here, instead, the focus is on two formalisms for statistical physics, namely Density Functional Theory (DFT) and Power Functional Theory (PFT). Both approaches are of crucial interest to researchers in the field: DFT is widely-used and applicable in a wide range of equilibrium settings – it also has a dynamic, non-equilibrium counterpart called Dynamic DFT (DDFT); PFT unifies much of the work in DDFT and enhances the fundamental understanding of such systems.

A key aspect of DFT is the identification of 'sum rules', which are identities connecting correlation functionals. These are useful both (a) theoretically, for example, to enhance fundamental understanding and interpretation of these methodologies, or to perform analysis of particular systems, e.g., bifurcations/phase transitions, and (b) numerically to validate novel algorithms. The present work contributes to this area by (i) presenting novel derivations of a wide range of known sum rules through the application of Noether's Theorem; (ii) deriving novel sum rules for systems with anisotropic particles or memory. Step (i) is pedagogically interesting, and motivates the later derivations. In (ii), the treatment of anisotropic particles leads to (possibly unsurprising) spin-orbit coupling, whilst the treatment of memory effects appears to be entirely novel. The approach for dynamic systems seems to require the use of PFT, which appears to be the bottleneck preventing such results from being derived through 'classical' approaches.

In summary, I find the article to be instructive as to the use of Noether's Theorem for such problems, recovering many known results using a single approach, and demonstrating some novel applications of the methodology. However, I feel that the authors need to pay more attention to the motivation for, or importance of, the obtained results. For example, I do not think it is clear how the novel results obtained can be used in practice, or what the real benefit of the many-body hierarchies are. I am not claiming that these results are not of interest, simply that more effort should be given to explaining their importance. Relatedly, I think that the authors could be clearer about which results are novel (rather than re-derivation of existing results). I appreciate that citations are given in a number of places, but unfortunately my institution does not have access to digital copies of many of these so I could not check in detail and had to rely on memory of what is contained therein.

Additionally, I think that the following points should be addressed before the article could be accepted. Below are also some minor points/typos.

1. [As above] Motivation and importance of results. As a simple example, the authors present (11)/(12) without any significant discussion of its implications. However, at least one application is immediately demonstrated in the next section. I feel that, for a more general audience, these connections could be spelled out earlier in the manuscript. This is especially true for the novel results obtained.

2. [As above] Clearly indicate which results are novel, and which are 'only' novel derivations (these derivations are still interesting, in my opinion).

3. [p2] Can the authors expand on the connection (or lack of connection) between their results and the NOZ relations?

4. [p2] The authors state that they 'discuss illustrative applications, including phase coexistence' – could they clarify which other parts they consider to be applications?

5. [p3] As the authors note, the derivation of (3) and (4) requires integration by parts. They claim that the boundary terms vanish; presumably this follows from some assumption they are making as it is clearly not true in general. Can they please state this assumption more clearly? Also applies to the derivation of (5) on p4, and the discussion of open boundaries at the start of Section IV.

6. [p7, Section VII] Could the authors comment more on the difficulties of a general anisotropic set-up, rather than the uniaxial case considered here?

7. [p8/9] The authors assume that the shift vanishes at the time end points (which seems reasonable). Are there any restrictions on the regularity of the shift in time, e.g., does it need to be continuous, smooth, something in between?

8. [p9 after (28)] The authors assume that the second derivatives can be interchanged and note that his is a common assumption. Has this been shown to be true (or not) in any known cases? I assume that the authors are aware the the relevant results such as Schwarz's/Clairaut's theorem, or the result that the partial derivatives themselves being differentiable is sufficient for them to commute.

9. [p1/10] In the introduction, the authors state that they obtain novel identities for active fluids, but in the conclusions they identify self-motility as an open question. In fact, I could not see where the question of active particles was treated (explicitly?) in the manuscript. Could the authors please clarify this?

Minor points:

- [p3] Presumably 'functionally derive (2)' should be 'functionally differentiate (2)'.
- [p4, first full paragraph] I think it would be clearer to say 'globally translationally invariant'.
- [p4, before Section III] what is 'ii)' referring to?
- [p4, rhs above (11)] I think it would be helpful to remind the reader that the total internal force with $j \in V$ vanishes.
- [p10, start of Section IX] 'Known sum rules' rather than 'Known some rules'.

Report for manuscript COMMSPHYS-21-0151-T entitled "Noether's Theorem in Statistical Mechanics"

In this manuscript, the authors present an elegant method for deriving infinite hierarchies of correlation functions for many-body systems by utilising Noether's Theorem in variational-based classical field theoretical models. The method is based on exploiting simple symmetry properties of the fundamental functional of the system, thus resulting in global conservation laws, from which hierarchies of correlation functions can be generated. Interestingly, the Noether identities are different from the Ornstein-Zernike relations for both equilibrium and non-equilibrium scenarios.

My comments and questions are as follows.

- The title is way too general. The method is rather specific to variational-based classical density functional theories, a small piece of statistical physics. The present title would be a good title for a book, but here it promises too much.
- The work is quite elegant, but the results are very formal and their practical value is unclear. It would be nice to discuss the practical importance of the results.
- The structure of the manuscript is a bit hectic from section IV. Sections IV-VIII should be better positioned in the text. Those referring to equilibrium scenarios should be sub-sections of section II, and the others of section III.
- The enthusiasm of the authors is clear, but the manuscript is hard to read. Due to the lack of introduction into variational-based density functional theories makes the manuscript accessible to only a few DFT experts. The text is flooded with technical details rather than explanations of the main concepts and the results, which also does not help.
- There are minor mistakes in the manuscript, such as "functionally derive by" on page 3.

In summary, the manuscript presents very important results, but in its present form it is a highly technical document. To make it more accessible to the general reader, I recommend to remove the technical details and re-write the manuscript in a casual explanatory form. It would also be crucial to present and explain the basic concepts of DFT/PFT. Re-structuring the manuscript and putting the main results into contexts in regards of their practical importance would also increase the value of the manuscript.

25 May 2021

Reviewers' comments:

Reply to Reviewer #1 (*Original referee report in italics*)

We thank the Referee for a critical reading of our paper and for the positive assessment. We respond to the point of criticism below.

In this Communication, Hermann and Schmidt report on an application of Noether's calculus in the framework of statistical-mechanical functional theories of equilibrium and out-of-equilibrium many-body systems. More specifically, they derive sum rules and hierarchies of equations that connect various static and dynamical quantities, based on the invariance properties of the equilibrium density functional and of the out-of-equilibrium power functional. Both translational and rotational invariances are considered, with an application to fluids made of anisotropic particles.

This is an interesting and well-written paper, reporting sound technical work. In my opinion, the calculations are rather straightforward, but many of them lead to novel results which should be of interest to members of the statistical-mechanical community and could be useful in the future. In this respect, the paper is manifestly devised as a resource for future work, condensing the technical information needed for developments to come. Here lies a weakness of the paper, in my opinion. Indeed, there is no guarantee that the beautiful formal results will not turn disappointing, being of little use in concrete cases. Actually, this is not an uncommon situation. Therefore, I missed a more detailed discussion of how this work could be put to good use, in order to answer actual physical questions. The discussion is full of promises, but it remains quite vague.

Although we are somewhat more positive about the use of beautiful formal results, we do accept the criticism. It is of course a matter of judgement, but there is indeed a wealth of studies that rely heavily on exact sum rules, be it as mere consistency checks, but also to develop concrete theories. Whether sum rules have been exploited in a sufficient way or whether there is uncharted territory can be debated of course. We would not see a fundamental limitation of working with exact statement though. However, we do agree that one can easily envisage a much wider recognition and use of exact statements. Surely, such a prospect gave much motivation and impetus for carrying out the present work.

As a remedy to address the Referee's criticism, we have added two additional concrete example applications. Both of these examples address the behaviour of active Brownian particles, i.e. spheres that self-propel along an intrinsic orientation degree of freedom. As there is much current interest in the behaviour of active Brownian particles we deem this system to be a suitable test bed for the Noether formalism. (To illustrate the system for a general reader we have added Fig. 4 (b) and (c).)

The first example addresses the free interface between phase-separated states occurring in motility-induced phase separation (MIPS) of active Brownian particles. MIPS attracts much general interest in an ever growing range of systems and variants and there is much hot controversy about both the mechanisms for the nonequilibrium phase separation and in particular of the role of the interface for bulk

phase coexistence. One question is whether or not the interface is relevant for the bulk coexistence conditions. In equilibrium phase separation this is not the case, but the question is controversial for MIPS.

Here, using only the Noether identities, i.e. without having to approximate the coupled many-body effects, we are able to show that the active gas-liquid interface exerts no net force on the bulk phases, apart from a trivial polarization effect, which is by now well understood. This absence of interface-to-bulk coupling is hence analogous to the situation in equilibrium and here we provide the rigorous proof. We would hope that this exemplifies that while the exact identities indeed might be beauties, they can be put to use as work horses.

Secondly, we address the sedimentation behaviour of active Brownian particles under gravity. We treat the general case, i.e. time-dependent nonequilibrium with spatial inhomogeneity. We show that the global Noether force sum rules yield, despite the complexity of the situation, simple relationships between total quantities. In particular the active motion again creates no total net force on the system. Hence there is no effect caused by the swimming on the reading of a weighing scale. We added a new Fig. 5 to illustrate this physics.

Please see the revised manuscript (p.7-9) for the full text and all details of both applications. Any possible application of higher-order sum rules goes beyond the scope of the present study.

These changes also reflected in the revised abstract, where we have appended the following text.

> "When applied to active Brownian particles, the theorem clarifies the role of interfacial forces in motility-induced phase separation. For active sedimentation under gravity the global internal Noether sum rule constrains the motion of the center of mass."

Furthermore, we have added following description to the introduction (p.2).

> "We also consider the more general case of anisotropic interparticle interactions, as is relevant for the description of active Brownian particles. We address two prominent effects that occur in systems of active Brownian particles, namely motility induced phase separation and active sedimentation under gravity. In both cases the Noether sum rules shed new light on the nonequilibrium properties."

Furthermore, we mention some specific possible future applications in the conclusions (p.14) and added the following text.

> "It would be highly interesting to apply (49) to the recently obtained direct correlation function of the hard sphere crystal. This would allow to investigate whether Triezenberg and Zwanzig's concept that they originally developed for the free gas-liquid interface applies to the also self-sustained density inhomogeneity in a solid. [Furthermore, addressing further cases of self motility [42-44],] including active freezing [99,100], [as well as further types of time evolution, such as molecular dynamics or quantum mechanics should be

interesting.]"

In conclusion, I think that this paper is suitable for publication in Communications Physics, in terms of soundness, novelty and interest in the field. But I believe that it would be very beneficial to its impact if the authors could provide stronger arguments with regards to the future usefulness of the reported results.

We trust that the added examples demonstrate explicitly the usefulness. We cordially thank the Referee for a thought-provoking report.

Reply to Reviewer #2 (*Original referee report in italics*)

We thank the Referee for a detailed, substantial and constructive Report. We appreciate very much the efforts and time that went into its production. We respond to the points of criticism below.

This work applied Noether's Theorem to a number of settings in statistical mechanics. As noted by the authors, Noether's Theorem is widely-used in a wide range of physical settings, in particular in classical and high-energy physics. Here, instead, the focus is on two formalisms for statistical physics, namely Density Functional Theory (DFT) and Power Functional Theory (PFT). Both approaches are of crucial interest to researchers in the field: DFT is widely-used and applicable in a wide range of equilibrium settings - it also has a dynamic, non-equilibrium counterpart called Dynamic DFT (DDFT); PFT unifies much of the work in DDFT and enhances the fundamental understanding of such systems.

A key aspect of DFT is the identification of 'sum rules', which are identities connecting correlation functionals. These are useful both (a) theoretically, for example, to enhance fundamental understanding and interpretation of these methodologies, or to perform analysis of particular systems, e.g., bifurcations/phase transitions, and (b) numerically to validate novel algorithms. The present work contributes to this area by (i) presenting novel derivations of a wide range of known sum rules through the application of Noether's Theorem; (ii) deriving novel sum rules for systems with anisotropic particles or memory. Step (i) is pedagogically interesting, and motivates the later derivations. In (ii), the treatment of anisotropic particles leads to (possibly unsurprising) spin-orbit coupling, whilst the treatment of memory effects appears to be entirely novel. The approach for dynamic systems seems to require the use of PFT, which appears to be the bottleneck preventing such results from being derived through 'classical' approaches.

In summary, I find the article to be instructive as to the use of Noether's Theorem for such problems, recovering many known results using a single approach, and demonstrating some novel applications of the methodology. However, I feel that the authors need to pay more attention to the motivation for, or importance of, the obtained results. For example, I do not think it is clear how the novel results obtained can be used in practice, or what the real benefit of the many-body hierarchies are. I am not claiming that these results are not of interest, simply that more effort should be given to explaining their importance. Relatedly, I think that the authors could be clearer about which results are novel (rather than re-derivation of existing results). I

appreciate that citations are given in a number of places, but unfortunately my institution does not have access to digital copies of many of these so I could not check in detail and had to rely on memory of what is contained therein.

This is a valid point. In response we added an overview that summarizes which of the results are in the previous literature. (This information is also given in the text surrounding each of the relevant equations.) We also spell out explicitly which of the identities are new. Please see the section "Relationship to classical results" that we have added before the conclusions. Please see the revised manuscript for the full text, including references and equations. (This also addresses the point 2 below.)

As to the benefit of the many-body hierarchies, we can only speculate at this point. Most statistical/liquid state theories stop at the two-body level, so the Referee's point is certainly valid, given the current state of the art. (There are exceptions of course.) Still typical approximations at least implicitly imply higher-order correlations. This is particularly true in DFT, when starting from a functional approximation. Within e.g. fundamental-measure theory for hard sphere systems the higher-order correlators are surely non-trivial and unexplored to a large extent. We trust that if one wanted to make headway in this direction, then the Noether identities should be useful once one embarks on such (highly challenging) work.

We added a brief statement to the conclusions, which reads as follows.

> "We envisage that the higher than two-body Noether identities can facilitate the construction of advanced liquid state/density functional approximations. Such work should surely be highly challenging. In the context of fundamental measure theory (see e.g. Ref. [20] for an enlightening review) it is worth recalling that in Rosenfeld's original 1989 paper [93], he calculated the three-body direct correlation function from his then newly proposed functional. The result for the corresponding three-body pair correlations compared favourably against simulation data. Furthermore, the recent insights into two-body correlations in inhomogeneous liquids [94] and crystals [95] demonstrates that working with higher-body correlation functions is feasible."

Additionally, I think that the following points should be addressed before the article could be accepted. Below are also some minor points/typos.

1. [As above] Motivation and importance of results. As a simple example, the authors present (11)/(12) without any significant discussion of its implications. However, at least one application is immediately demonstrated in the next section. I feel that, for a more general audience, these connections could be spelled out earlier in the manuscript. This is especially true for the novel results obtained.

We accept this useful suggestion. We announce in section "Open boundaries" (p.6), that these considerations are relevant for phase separation.

> "For non-vanishing net boundary terms additional contributions arise in the above sum rules. These contributions occur if the system develops different (bulk) states, e.g. for $x \to \pm\infty$ as is relevant for bulk phase separation (see the section below)."

We have also added a brief statement at the end of the section "Open boundaries" (p.6).

> "We demonstrate in the following section the practical relevance of these considerations."

Furthermore, we have added two sections "Motility induced phase separation" (p.7) and "Active sedimentation" (p.8). These are two applications of the derived sum rules to motivate the importance of our results. Please see the revised manuscript for this added material. We have added the following text to complement the abstract and the summary of the content in the introduction (p.2).

> "When applied to active Brownian particles, the theorem clarifies the role of interfacial forces in motility-induced phase separation. For active sedimentation under gravity the global internal Noether sum rule constrains the motion of the center of mass."

> "We also consider the more general case of anisotropic interparticle interactions, as is relevant for the description of active Brownian particles. We address two prominent effects that occur in systems of active Brownian particles, namely motility induced phase separation and active sedimentation under gravity. In both cases the Noether sum rules shed new light on the nonequilibrium properties."

For a more general audience we have also included the figures 4(b), (c) and 5 to illustrate active Brownian particles and their physics in motility induced phase separation and under gravity.

As specific examples we mention the following possible applications in the conclusions (p.14).

> "It would be highly interesting to apply (49) to the recently obtained direct correlation function of the hard sphere crystal. This would allow to investigate whether Triezenberg and Zwanzig's concept that they originally developed for the free gas-liquid interface applies to the also self-sustained density inhomogeneity in a solid. [Furthermore, addressing further cases of self motility [42-44],] including active freezing [99,100], [as well as further types of time evolution, such as molecular dynamics or quantum mechanics should be interesting.]"

2. [As above] Clearly indicate which results are novel, and which are 'only' novel derivations (these derivations are still interesting, in my opinion).

As described above we have added an overview in section "Relationship to classical results" at the end of the paper (p.13) that precisely gives this information in compact form.

3. [p2] Can the authors expand on the connection (or lack of connection) between

their results and the NOZ relations?

This is a valid suggestion. We have added the following text on p.11.

> "Both the Ornstein-Zernicke (OZ) and NOZ relations are different from the Noether identities. The former relations are a direct consequence of the generality of the variational principle. Per se, neither the OZ nor the NOZ relations reflect the Noether symmetries."

4. [p2] The authors state that they 'discuss illustrative applications, including phase coexistence' - could they clarify which other parts they consider to be applications?

In the revision, we have added two further examples, apart from the already covered case of bulk phase separation in equilibrium. The new applications address the physics of active Brownian particles, namely the interface in motility-induced phase separation and nonequilibrium sedimentation under gravity. We have reworded the above statement in order to announce these further examples. The revised text reads as follows.

> "To illustrate the theory we apply it to both passive and active phase coexistence as well as to active sedimentation under gravity."

The new sections "Motility induced phase separation" (p.7) and "Active sedimentation" (p.8) describe these results. Please see the revised manuscript for the full text, including references and equations.

5. [p3] As the authors note, the derivation of (3) and (4) requires integration by parts. They claim that the boundary terms vanish; presumably this follows from some assumption they are making as it is clearly not true in general. Can they please state this assumption more clearly? Also applies to the derivation of (5) on p4, and the discussion of open boundaries at the start of Section IV.

We have clarified the situation. At the beginning of section "Adiabatic state" (p.2) we state more clearly that we assume the systems to be bounded by external walls and hence no boundary terms occur. The added text reads as follows.

> "For the moment we only examine systems completely bounded by external walls. Systems with open boundaries are considered below."

For (3) and (4) on p.3 we have explained more explicitly why boundary terms vanish.

> "[Recall that boundary terms vanish] as we only consider systems with impenetrable bounding walls."

For (5) (p.4) we remind the reader that boundary terms vanish.

> "As boundary terms vanish in the considered systems, [integration by parts yields,] [...]"

In the discussion of open boundaries (p.6) we now distinguish explicitly between net and no net boundary contributions. We added the following text.

> "In case that there are no net boundary contributions, all previous derived sum rules still hold. This includes e.g. an effectively one-dimensional system in planar geometry that evolves to the same bulk state at the left and right boundaries or if the boundary conditions are periodic. In both cases left and right boundary terms are equal up to a minus sign and hence cancel each other. This example can be generalized straightforwardly to more complex geometries.
> For non-vanishing net boundary terms additional contributions arise in the above sum rules. These contributions occur if the system develops different (bulk) states, e.g. for $x \to \pm\infty$ as is relevant for bulk phase separation (see the section below)."

6. [p7, Section VII] Could the authors comment more on the difficulties of a general anisotropic setup, rather than the uniaxial case considered here?

This is a good suggestion. We have performed this work and are covering now the general anisotropic case. In short, there are no surprises when going from a single unit vector to a pair of unit vectors (equivalent to the full Eulerian angles of general anisotropic bodies). Still this is a valid generalization and we are grateful to the Referee for providing the incentive.

The revised section "Anisotropic particles" (p.7) covers sum rules for translational symmetries and was generalized from uniaxial particles to anisotropic particles. See in particular the added paragraph at the end of this section.

For sum rules due to rotational symmetries in section "Orbital and spin coupling" we added a description how these can be generalized to anisotropic particles. An excerpt of the text on p.11 reads as follows.

> "So far we have restricted ourselves to uniaxial particles. […] The indices *n* and *m* belong to the number of functional derivatives with respect to **J** and **J**^ω as before."
> (Please see the revised manuscript for the full text, including all formulae.)

In the revised version of the paper we treat the rotational invariance of anisotropic particles later (p.10), below the section "Rotational invariance". We made corresponding changes to the section "Orbital and spin coupling". Please see the revised manuscript where these changes are indicated in color.

7. [p8/9] The authors assume that the shift vanishes at the time end points (which seems reasonable). Are there any restrictions on the regularity of the shift in time, e.g., does it need to be continuous, smooth, something in between?

This is a good question. We certainly have no answer that would satisfy a mathematician. At the heart of Noether's original work is analyticity. She assumed existence of all relevant derivatives etc. (There is of course a long literature of follow-

up work.) Our current work is carried out with an understanding of sufficiently smooth behaviour such that the formalism can be applied, representing e.g. jumps by sharp but continuous interpolation. Whether something genuinely interesting could happen for discontinuous/nonanalytical cases is beyond the scope of the present work.

8. [p9 after (28)] The authors assume that the second derivatives can be interchanged and note that this is a common assumption. Has this been shown to be true (or not) in any known cases? I assume that the authors are aware the the relevant results such as Schwarz's/Clairaut's theorem, or the result that the partial derivatives themselves being differentiable is sufficient for them to commute.

We thank the Referee for pointing out Schwarz's/Clairaut's theorem of which we are aware in elementary context. For the present functional setting we would rather be cautious and retain our notion of making an explicit assumption that the functional derivatives commute. While this point could be interesting from a rigorous mathematical point of view, we would not expect major relevance for the physics under consideration. As to the Referees specific question of what is known, we would expect that the functional derivatives commute in the standard cases. However, we are not aware that anybody targeted this question specifically in the literature.

9. [p1/10] In the introduction, the authors state that they obtain novel identities for active fluids, but in the conclusions they identify self-motility as an open question. In fact, I could not see where the question of active particles was treated (explicitly?) in the manuscript. Could the authors please clarify this?

The original text created indeed some confusion, which we have remedied. The revised paper contains two new applications for active particles, please see our responses to point 1 and 4.

Minor points:

** [p3] Presumably 'functionally derive (2)' should be 'functionally differentiate (2)'.*
OK

** [p4, first full paragraph] I think it would be clearer to say 'globally translationally invariant'.*
OK

** [p4, before Section III] what is 'ii)' referring to?*
We have deleted the numbering to avoid confusion.

** [p4, rhs above (11)] I think it would be helpful to remind the reader that the total internal force with j element of V vanishes.*
OK. We have added the following text.

> "The sum of all interactions between inside particles vanishes due to the global internal Noether sum rule (10). [For simplicity we restrict ourselves to systems that interact via short-ranged pairwise central forces, where **F**_*ij*

indicates the force on particle *i* exerted by particle *j*.] So only forces exerted from an inside to an outside particle contribute."

"The total internal force between particles inside of V, i.e. $i \in V$ and $j \in V$, vanishes due to (10)."

** [p10, start of Section IX] 'Known sum rules' rather than 'Known some rules'.*

Thanks for pointing this out. (We pondered whether we should keep that typo for shear beauty.) Corrected.

We thank the Referee for a multitude of useful points.

Reply to Reviewer #3 (*Original referee report in italics*)

We thank the Referee for the critical reading of our paper and for raising several constructive points of criticism.

In this manuscript, the authors present an elegant method for deriving infinite hierarchies of correlation functions for many-body systems by utilising Noether's Theorem in variational-based classical field theoretical models. The method is based on exploiting simple symmetry properties of the fundamental functional of the system, thus resulting in global conservation laws, from which hierarchies of correlation functions can be generated. Interestingly, the Noether identities are different from the Ornstein-Zernike relations for both equilibrium and non-equilibrium scenarios.

My comments and questions are as follows.

** The title is way too general. The method is rather specific to variational-based classical density functional theories, a small piece of statistical physics. The present title would be a good title for a book, but here it promises too much.*

We disagree and think that the title aptly describes the content of the paper. The material in the paper is by no way limited to DFT. Note that we start with symmetry properties of the partition sum in its elementary, introductory-course level form. No functional of density is involved at this stage, and still the symmetry properties of the partition sum (and hence the grand potential) yield exact identities for global force relationships. The elementary partition sum is viewed as a functional of the external potential, but Nother's Theorem is indeed one of functional calculus, so mentioning the Theorem in the title automatically implies a functional context. Moreover, we do treat both rotational and translational symmetries, and do so in equilibrium and out-of-equilibrium, which is reflected in the plain use of "Statistical Mechanics" in the title.

We hope that the Referee can accept that the title is not over-statement, but that is rather a compact, yet accurate description of the content of the article. (We are grateful for the implicit suggestion of writing a corresponding book; this is certainly an exciting prospect for the future.)

In order to stress the generality of our results, we highlight explicitly in the introduction, which of our results only require the partition sum, and not the density functional. The added text (p.2) read as follows.

> "[We obtain in each case a corresponding Noether Theorem that constitutes a global force identity.] Crucially, considering the symmetries of the partition sum does not require to engage with density functional concepts; the elementary definition suffices."

** The work is quite elegant, but the results are very formal and their practical value is unclear. It would be nice to discuss the practical importance of the results.*

We agree. We have added both a discussion section and have also added two concrete examples of the physics of active Brownian particles, i.e. the role of the interface in motility-induced bulk phase separation and the sedimentation behaviour under gravity. In the first case, the Noether identities allow us to make rigorous that there is no effect originating from the interparticle interactions that would couple from the interface to the bulk. This is an important point, as a variety of different corresponding theories have been formulated over the past few years. The Noether identities provide exact benchmarks.

Please see the new sections on "Motility induced phase separation" (p.7) and "Active sedimentation" (p.8) in the revised manuscript for the full text.

The additional content is also included in the revised abstract and in the introduction (p.2). We have added the following text.

> "When applied to active Brownian particles, the theorem clarifies the role of interfacial forces in motility-induced phase separation. For active sedimentation under gravity the global internal Noether sum rule constrains the motion of the center of mass."

> "We also consider the more general case of anisotropic interparticle interactions, as is relevant for the description of active Brownian particles. We address two prominent effects that occur in systems of active Brownian particles, namely motility induced phase separation and active sedimentation under gravity. In both cases the Noether sum rules shed new light on the nonequilibrium properties."

To illustrate active Brownian particles as well as motility induced phase separation and active sedimentation we have added Fig. 4 (b) and (c) and Fig. 5.

In the conclusion (p.14) we added the following statement.

> "It would be highly interesting to apply (49) to the recently obtained direct correlation function of the hard sphere crystal. This would allow to investigate whether Triezenberg and Zwanzig's concept that they originally developed for the free gas-liquid interface applies to the also self-sustained density inhomogeneity in a solid. [Furthermore, addressing further cases of self motility [42-44],] including active freezing [99,100], [as well as further types of

time evolution, such as molecular dynamics or quantum mechanics should be interesting.]"

** The structure of the manuscript is a bit hectic from section IV. Sections IV-VIII should be better positioned in the text. Those referring to equilibrium scenarios should be sub-sections of section II, and the others of section III.*

We accept the criticism. As a remedy the original section "Anisotropic particles" (p.7) is split up into two sections in the revised manuscript. The first part "Anisotropic particles" now only includes sum rules due to translational symmetries and is placed after the section "Phase coexistence" (p.6). After that the new sections "Motility induced phase separation" (p.7) and "Active sedimentation" (p.8) were added. The new section "Orbital and spin coupling" which was originally part of the section "Anisotropic particles" is now placed below the section "Rotational invariance". We trust that these changes clarify the distinction between translational and rotational symmetries and improve the positioning of the sections in the text. Tiggered by the Editorial recommendation, we have also made the article conforming to the Journal style of Communications Physics, which uses no section numbering.

** The enthusiasm of the authors is clear, but the manuscript is hard to read. Due to the lack of introduction into variational-based density functional theories makes the manuscript accessible to only a few DFT experts. The text is flooded with technical details rather than explanations of the main concepts and the results, which also does not help.*

We thank the Referee for aptly identifying our enthusiasm. The paper presents our best efforts in making its content accessible to reader without compromising accuracy. We can see from the Referee's report that these efforts were not in vain, and we are delighted that the other three Referees could also follow our argumentations, even very deep into many details.

We do not think that the present paper presents a suitable platform for giving an introduction to "variational-based density functional theories" to non-experts. This would certainly require much space in order to be self-contained and accessible. A too compact version of such an introduction will again be accessible only to those readers who are already familiar with the material. Novices might even get confused by an abridged account. Note that while much of the functional background is relevant in a broader setting, for the specifics of the Noether identities, this material is not required.

Hence we have kept the present setup of the paper, which focuses on the genuinely new material. Note that our presentation starts with the grand potential, i.e. the logarithm of the partition sum, with which all readers will be familiar.

** There are minor mistakes in the manuscript, such as ”functionally derive by” on page 3.*

Corrected. Thanks.

In summary, the manuscript presents very important results, but in its present form it is a highly technical document. To make it more accessible to the general reader, I recommend to remove the technical details and re-write the manuscript in a casual explanatory form. It would also be crucial to present and explain the basic concepts of DFT/PFT. Re-structuring the manuscript and putting the main results into contexts in regards of their practical importance would also increase the value of the manuscript.

Similarly to the point two up, we trust that interested readers can cope with our style of presentation of the current material.

We thank the Referee for the report and hope to have made our reasons for the format of the paper understandable.

Reply to Reviewer #4 (*Original referee report in italics*)

We cordially thank the Referee for providing such a constructive and helpful report and for engaging with our work thoroughly. We are delighted that the Referee recommends publication and we respond to all raised points below.

This paper considers the application of Noether's theorem to the (classical) statistical mechanics of interacting particles. The generality of the approach, in the sense that it is applicable to a broad category of functionals, is exploited within the framework of DFT (equilibrium) and PFT (nonequilibrium). By applying Noether's concept of invariance with respect to various symmetry operations the Authors derive a large number of statistical mechanical sum-rules. Some of these are known from previous calculations and some are new.

On the one hand this is an elegant and clever way to derive sum-rules using a unified approach, on the other it makes clear the connection between each sum-rule and a symmetry of the many-body system. This is a valuable contribution and not something I have seen or thought about before. The paper is well written and fairly easy to follow - although I admit that I found the nonequilibrium part much more difficult, but this is probably due to my being less familiar with the PFT approach.

Publication recommended.

The Authors may wish to address/comment on the following points which occurred to me while reading the manuscript:

(1) A detail- First sentence of section II. I don't think it is the Grand potential which is translated, but rather the coordinates.

True. We have corrected the wording. The revised text on p.2 is as follows.

> "[We start with an initial illustration of Noether's concept] as applied to the grand potential Ω. We consider spatial translations of the position coordinate **r** [at fixed chemical potential μ and fixed temperature *T*.]"

(2) In the nonequilibrium section the Authors derive sum-rules involving derivatives of the excess part of the power functional. I recall that quite recently explicit approximations were developed for the excess power (Phys. Rev. Lett. 120, 028001 (2018)) - do these approximations satisfy the derived sum-rules?

Yes, the velocity gradient form of the power functional (quoted above) does satisfy all three dynamical translational invariances. We added the following text on p.14.

> "The sum rules imposed by the three types of dynamical displacements are satisfied within the velocity gradient form of the power functional [54, 56]. It is straightforward to see that the functional is independent of the coordinate origin (static shifting). For the cases of dynamical shifting, the invariance of the functional stems from invariance of the velocity field against shifting. For both instantaneous and memory shifting, the velocity gradient remains invariant under the displacement."

(3) In the discussion: The second paragraph contains several very interesting but somewhat obscure comments. Perhaps this could be expanded a little to make these more explicit.

We agree and we have revised this paragraph. The new version (p.14) is as follows.

> "Crucially, the Noether sum rules are different from the variational principle, as embodied in the respective Euler-Lagrange equation. On the formal level, the difference is that the Euler-Lagrange equation (both of DFT and of PFT) is a formally closed equation on the one-body level. In contrast, the Noether rules couple n- and $(n+1)$-body correlation functions, hence they are of genuine hierarchical nature. They also describe different physics, as the Euler-Lagrange equations express a chemical potential equilibrium in DFT and the local force balance relationship in PFT. In contrast, the Noether identities stem from the symmetry properties of the respective underlying physical system."

We also added the section "Relationship to classical results" (p.13) which contains a discussion on the relationship of our results with the literature. See the revised manuscript for the full text that gives further description of the above points.

(4) Is it clear which of these sum-rules will hold for approximate functionals (as typically employed, e.g. in practical DFT calculations)?

This is a very good point. We have addressed the validity of approximate DFTs, and we are able to give a green light. We added the following text (p.14).

> "The standard DFT approximations, ranging from simple local, square-gradient, and mean-field functional to more sophisticated weighted-density-schemes including fundamental measure theory satisfy the internal force relationships. This can be seen straightforwardly by observing that these

functionals do satisfy global translation invariance. (The value of the free energy is independent of the choice of coordinate origin.) All higher-order Noether identities are then automatically satisfied, as these inherit the correct symmetry properties from the generating (excess free energy) functional. Our formalism hence provides a concrete reason, over mere empirical experience, why the practitioners' choices for approximate functionals are sound. The situation for more complex DFT schemes could potentially be different though. As soon as, say, self-consistency of some form is imposed, or coupling to auxiliary field comes into play, it is easy to imagine that the Noether identities help in restricting choices in the construction of such approximation schemes."

Further changes to the manuscript

We added equation (16) and the surrounding text for clarification.

Further minor changes and corrections have been performed throughout the manuscript.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

In this second version, the Authors have fully addressed the criticisms/scepticism expressed in my first report. The new applications greatly enhance the interest of the paper.

I now recommend publication as is.

Reviewer #2 (Remarks to the Author):

I thank the authors for carefully addressing all of my previous points. I am happy with their revisions; in particular, I feel that the paper benefits from the addition of the extra examples, and also from more careful explanations in places. I recommend acceptance for publication/

One small suggestion:

[p11; last paragraph before Memory invariance] \omega' would be clearer as \bar\omega as this is how it then seems to be used.

Reviewer #3 (Remarks to the Author):

I accept the response of the authors and I'm satisfied with the changes they have made. Re-structuring the manuscript made it more readable and extending it with applications clearly indicates the practical value of the formal results, and makes them more relevant for a broader audience. I recommend to publish the manuscript in Communications Physics.

Reviewer #4 (Remarks to the Author):

The authors have addressed all of the points I raised and I am happy to recommend publication.

Reviewers' comments:

Reply to Reviewer #1:

>In this second version, the Authors have fully addressed the
>criticisms/scepticism expressed in my first report. The new
>applications greatly enhance the interest of the paper.
>
>I now recommend publication as is.
>

We are delighted that the Referee recommends publication.

Reply to Reviewer #2:

>I thank the authors for carefully addressing all of my previous points. I
am happy with their revisions;
>in particular, I feel that the paper benefits from the addition of the
extra examples, and also from more
>careful explanations in places. I recommend acceptance for publication/
>
>One small suggestion:
>
>[p11; last paragraph before Memory invariance] \omega' would be clearer as
\bar\omega as this is how it then seems to be used.
>

We thank the Referee for pointing out this minor point. Corrected.
We are delighted that the Referee recommends publication.

Reply to Reviewer #3:

>I accept the response of the authors and I'm satisfied with the
>changes they have made. Re-structuring the manuscript made it
>more readable and extending it with applications clearly indicates
>the practical value of the formal results, and makes them more
>relevant for a broader audience. I recommend to publish the
>manuscript in Communications Physics.
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We are delighted that the Referee recommends publication.

Reviewer #4 (Remarks to the Author):

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We are delighted that the Referee recommends publication.