History of the manuscript CB10255/Toke Revised 04/06/2012

History of the manuscript CB10255/Toke			
1. Status as by February 27, 2012			
2.	Report by the first Referee		
3.	Reply to the first Referee	. 5	
4.	Report by the second Referee	. 7	
5.	Letter to the Editors regarding the report by the second Referee	. 9	
6.	The original reply to the report by the second Referee	12	
7.	Letter from the Editors regarding author's original reply to the second Referee	21	
8.	Letter to the editors explaining the intended function of the original reply to the		
	second Referee	22	
9.	Sanitized version of the reply to the second Referee	23	
10.	Letter from the Editors announcing the end of the review process	30	
11.	Report by the third referee	31	
12.	Letter to the Editors regarding the report by the third Referee	34	
13.	Follow-up letter to the Editors regarding the alleged plagiarism	39	
14.	Letter from Editors regarding initiation of the appeal process	41	
15.	Report by the Editorial Board Member	42	
16.	Letter by the Authors to prof. Seto and the Attachment to it (sent 03/08/2012 and		
left unanswered by prof. Seto)			
17. Letter from the Editors re: Letter to prof. Seto			

1. Status as by February 27, 2012

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2. Report by the first Referee

Report of the Referee -- CB10255/Toke

there is a lack of references to previous experimental and other theoretical works on this issue. The is a lot work from MSU, and more recently Natowitz from Texas and A&M has been looking at the mass dependence of the limiting temperature. There was a lot papers from GSI ALADIN group on measurements and interpretation.

Also Lee Sobotka and his group made basically similar calculations for finite nuclear Phys.Rev. C 73, 014609 (2006). Assuming a Fermi-gas like dependence of the entropy, allow for expansion and increased surface diffusion. Adjust these to maximized the entropy and get the equilibrium configuration. They allowed for the modifications in the effective nucleon mass with excitation energy what is not the case in presented paper.

These equilibrium solutions are static, but experimentally we know the systems are expanding when they decay. The authors could discuss assumptions of their model more deeply. From the experimental data it follows, that situation is more complex and require time scale arguments.

The paper is not clearly written. Particularly section IV. Eq 15 shouldn't have rho_0 in it, this is defined earlier as the saturation nuclear density. One cannot vary Rhalf and maintain saturation density if you are conserving nucleons.

3. Reply to the first Referee

Reply to the Referee's report on CB10255/Toke

While the Referee has correctly pointed out a typo in Eq. 15, the rest of the report dwells not on issues discussed in our manuscript but exclusively on research conducted by others. Unfortunately, the language of the report tends to create an impression that what we claim to be our discovery of a new type of thermodynamic instability is a well known and documented issue in the published literature. Nothing is further from truth.

Re 1st paragraph of the report:

We agree with the Referee's statement that there is a lack of references to previous experimental and other theoretical works on the issue. This is so, however, because no works were published so far where the issue of surface boiling is discussed – neither experimental, nor theoretical. As it is clear from the title, abstract, and the body of our manuscript, our manuscript is not about measuring limiting temperature or studying equilibrium configurations of nuclear systems. It is about a new phenomenon predicted based on statistical thermodynamics of interacting Fermi matter. This is the essence of our study and the issue under scrutiny. This issue is not addressed in any of the papers from GSI, MSU, and Texas A&M.

We do offer a reference to a paper by Natowitz et al., but only in the context of the surface boiling explaining experimental observations, to which no plausible explanation can be found in published literature.

Re 2nd paragraph.

We are aware of the calculations made by Lee Sobotka and his group. We reference the second paper by this group on the equilibrium configurations of finite nuclei, which uses the same generic formalism as the first paper quoted by the Referee. Now we add a reference to the first paper, but again, solely in the context of contributing to the construction of a general thermodynamic framework for understanding behavior of excited nuclear systems. Importantly, neither of these two papers discusses the onset of thermodynamic (spinodal) instability at the crux of our manuscript.

Re. 3rd paragraph

One section of our paper is devoted to theory we use. It contains all details of our calculations, including the pertinent equations and discusses all relevant assumptions. A reader familiar with statistical thermodynamics and the Fermi gas model should be able to replicate our results based on the information provided. We object to a general statement by the Referee that we could discuss the assumptions of our model more deeply. This kind of a statement can be easily applied to every single manuscript and is too polemical in nature to be even discussed. The suggestion that the situation may be more complex is again of a polemical nature not amenable to an intelligent discussion.

Re 4th paragraph.

We again object to a polemical statement that the paper is not clearly written. In our opinion it is written clearly.

Regarding Eq. 15, there is indeed, a typo in it – the overall normalization factor was left out. This is corrected in the revised manuscript.

To summarize, the presence of the research and papers alluded to by the Referee not only does not diminish the value of our findings. It, in fact, enhances it. The presence of many research programs studying experimentally the systematics of limiting temperature demonstrates the urgency of publishing our theoretical finding of an explanation for the existence of such limits.

4. Report by the second Referee

Report of the Second Referee -- CB10255/Toke

I cannot recommend the paper for publication.

In the abstract and introduction the authors state that they consider a microcanonical ensemble. Looking at the nomenclature and formulae of Sec. II, they start out with entropy S as function of energy E and (hidden in E_config) particle number A, as it should be.

But from Eq. (10) on everything looks like a grand canonical ensemble. There is a temperature and a chemical potential.

I do not understand Eqs. (11) and (12), and I also do not see a reference where they are explained. Even the dimensions are wrong. What is a_0?

I can not see how the schematic model proposed by the authors is thermodynamically consistent. Is for example the Hugenholtz-van Hove theorem fulfilled in the case of finite nuclei?

In Sec. III, the authors discuss at length in a complicated way what they call boiling. But this is no more than the discussion of the coexistence region, which in a mean-field picture shows up by spinodal instabilities.

Section IV for finite nuclei assumes a density profile, and thermodynamic consistency is lost. I also don't understand the meaning of a central pressure shown in Fig. 4. In equilibrium, the pressure and the chemical potential are uniform, otherwise the matter would start moving. Shouldn't therefore the central pressure be equal to the outside pressure which in that case is zero?

To summarize, the paper does not help in understanding the situation encountered in heavy-ion collisions, as claimed in the introduction. In collisions one never is in an equilibrium state, everything is transient. The nuclei do not boil in the sense that they form bubbles of gas in the interior or separate off little drops from the surface. Rather, the heavy-ion collisions explore available phase space and thus the data reflect the nuclear many-body level density and with that the microscopic entropy which is the logarithm of the level density. All microscopic dynamical calculations show that the outcoming fragments are preformed very early and are not much rearranged during the expansion phase, again indicating that the time is too short for equilibration. Nevertheless, the partition into fragments is governed by phase space and level density. One has to regard a mixture of collisions that are all at the same excitation energy as the microcanonical ensemble. This cannot be described by a mean field picture and/or instabilities in the mean field picture.

The authors should acknowledge that the particle numbers are simply too small to use macroscopic (or bulk matter) thermodynamic terminology. But when doing the sampling of events with few particles in a microcanonical sense one can nevertheless draw conclusions about the (in the nuclear case not accessible) macroscopic system. This has been shown with small grains and Ising models where the macroscopic systems are also available.

5. Letter to the Editors regarding the report by the second Referee

Dear Editors,

We have read the report by the second referee many times over but were not able to determine on what scientific grounds he/she has disqualified our manuscript from being published in PRC. The Report is an incoherent collection of non-scientific (cannot be debated in scientific terms) "musings" by a referee who appears to be a complete ignorant in the field of nuclear thermodynamics, thermodynamics in general, and Physics 101. How else can one characterize one who states categorically (in the last paragraph of the Report) that:

"The authors should acknowledge that the particle numbers are simply too small to use macroscopic (or bulk matter) thermodynamic terminology".

The above statement does not border with what is in everyday language called **foolish** (or worse). It is **foolish**, as the Referee proposes here to trash a whole field of nuclear thermodynamics. Here is what we write in our Reply in this respect:

"The statement is ambitious in that it would at the same time invalidate **all** nuclear thermodynamics of the last 60+ years, which uses routinely the purely thermodynamical notions of entropy, temperature, chemical potential, pressure, thermal energy, free energy, etc. with respect to finite nuclei. Everything would have to go on the Referee's say so, including but not limited to:

all statistical thermodynamical models of compound nuclear decays on the market since over sixty years, including well established evaporation and fission models (PACE, CASCADE, GEMINI), Weisskopf's approach, statistical fission theory, not to mention the somewhat controversial mainstream models of multifragmentation – Expanding Emitting Source Model (EESM), Statistical Multifragmentation Model (SMM), and Metropolis Monte Carlo Model (MMMC). This encompasses over sixty years of thermodynamical theory as applied with amazing success to finite nuclei, thousands and thousands of scientific papers – the work product of a large community of both, theoreticians and experimentalists, hundreds and hundreds of conferences and numerous textbooks. Here is just one quick example of the (lead) subject of the coming International Workshop on Multifragmentation (Caen, 11/02-05/2011) – "Thermodynamics of finite nuclei & nuclear matter".

Does the Referee really believe that all of this is invalid? Has this then been a wholesale scam by a large community of nuclear scientists involved?"

And, then, we were stunned to read in the note by the Editors that based, among other things, on this foolish comment by the Referee, the Editors unconditionally disqualify our discovery of surface boiling as being unworthy of publishing. Do the Editors really share the view expressed by the Referee that all of nuclear thermodynamics is invalid? Have the Editors never accepted any publication in the field of nuclear thermodynamics or do they contemplate not accepting any from this day on? If so, we will have no choice but to seek a redress of our grievance and fairness on other ways available to us.

The above comment by the Referee is a blatant proof, that the Referee himself has never written anything in the field of nuclear thermodynamics, nor has he attended any conferences on related subject. So, we must ask, on what grounds was he appointed by the Editors to review our manuscript and why is he treated by the Editors as equal in expertise (our peer) with us who have a long record (dating back to the 1970s and 1980s) of publications and conference contributions in the field. It (the above comment) is also a blatant proof of a complete ignorance of thermodynamical theory by the Referee, reinforced further by his/her hollow "musings" in the remainder of the Report.

As we point out in our detailed analysis of the Referee's comments in our reply to these comments, there is not a single comment preceding the one (point 8) that is supposed to summarize the preceding "musings" (points 1 - 7) that makes sense when taken literally at their face value. When trying to second-guess what the Referee could have possibly had in mind one is invariably led to the conclusion that he/she is simply an ignorant. However, neither we nor the Editors have the right to second-guess the Referee and, therefore, the Report must be considered at its "face value" and therefore as hollow. The purportedly summary statement (point 8) itself is logically disjoint from the preceding statements and constitutes a textbook case of the Straw Man fallacy. It is, therefore, also invalid.

Importantly, the Referee never questions the validity of our claim of a discovery of a new phenomenon of surface boiling, neither does he/she question our assertion that this phenomenon explains experimental observations of limiting temperature, for which there was hitherto no meaningful explanation. He/she makes no mention whatsoever of the subject matter of our manuscript. To us, the reason for this is obvious – not understanding thermodynamics, he/she is incapable of comprehending what we try to explain to the reader familiar with thermodynamics.

In view of the above, we suggest that the Editors read our reply to the Referee and take a solid look at the "original comments" by the Referee to verify that what we say is, indeed, true. In our reply we tried to be as polite as possible, but here we will state that, e.g., the point 1 provides a testimony to the Referee's utter ignorance regarding thermodynamics generally, as it would be absolutely impossible to anyone familiar with thermodynamics to confuse the three approaches – microcanonical, canonical, and grand canonical, as the Referee does. It would be also impossible not to know that the temperature, pressure, and chemical potential are defined from the condition of microcanonical equilibrium and are, thus, naturally suited for the description of microcanonical systems, as they are also suited for the description of other ensembles. We can also state here that the point 7 provides a testimony to the ignorance of Physics 101 by the Referee. Does the Referee not know that, e.g., the atmospheric pressure is not uniform either (it decreases with altitude) and that the air masses can be in equilibrium, nevertheless, in contradiction to his/her remark?

And finally, we must note and complain that the publication of our important discovery has been already delayed by 5 months because of the Editors inability to tap a single referee familiar with

nuclear thermodynamics and capable of evaluating scientific merits of our claims (our peer, as in "peer review"), although we had supplied several names of individuals we consider competent theoreticians. Therefore, we would suggest that the Editors accept our manuscript for publication in Phys Rev. C, based on our long track record (dating back to at least 1981 – see the References section of our manuscript) of publications in the field of nuclear thermodynamics, none of which has been ever contested by our peers (i.e., scientists with expertise in the field in question), whether publicly or privately. It is our understanding that an inherent part of the editorial philosophy is that regarding claims of discoveries of new phenomena (ours has been vetted during well-attended scientific meetings – we will provide references upon request), it is safer to err on the side of publishing an erroneous paper rather than on the side of indefinitely delaying and, thus, suppressing publication of a valid discovery. One is truly tempted to wonder how could possibly, e.g., A. Einstein, get any of his great ideas published nowadays, with the present-day crop of mediocre references, immune to novelties.

We are attaching the revised version of our manuscript where:

- (i) We amended the notation in Eqs. 11 and 12, to make it consistent with the one used earlier.
- (ii) Added a sentence explaining the used notation.
- (iii) Prompted by a (hollow) remark by the Referee, and directly following Eqs. 10-11, we added a paragraph where we draw attention to the fact that in the simple model we use, the thesis of the Hugenholtz Van Hove theorem is fulfilled. We call the fact amusing, because it has no relevance to our conclusions, but may be of a potential interest to some readers.
- (iv) Added the reference (18) to the Hugenholtz Van Hove paper.

6. The original reply to the report by the second Referee

Reply to the Referee's Report

We are dismayed by the overly hostile tone of the Referee's Report and its highly prejudicial character, replete with entirely unsubstantiated claims, logical fallacies with a high potential to mislead the Editors, and gross misstatements of facts. For example, there is not a single piece of criticism in the report, preceding its summarizing paragraph which could possibly justify the unconditional or otherwise rejection of our manuscript. The summarizing paragraph itself is fallacious and constitutes a textbook example of the Straw Man fallacy. Because of this and this alone, the Referee's report is simply invalid regardless of the validity or lack of thereof of the preceding "criticism". Notably, and most importantly, the Referee never discusses nor disputes our findings and discoveries regarding the subject matter of our paper – the discovery of surface boiling and its importance for understanding certain experimental observations for which there was no explanation to this date. Furthermore, a complete lack of logical coherence in the report makes the task of intelligently replying to it very difficult. The difficulty arises from the fact that with the majority of statements in the Referee's Report being formally void of scientific message one is tempted to second-guess the Referee's intents, but no intelligent second-guessing actually leads to a sound statement. By an intelligent second guessing we understand the one, where we don't have to assume that the Referee does not understand the issues involved.

In spite of what is said above, we will try our best in terms of logic to answer the Referee's comments. We will begin with the last paragraph of the report, which, if true, would render all of the preceding text of the Report entirely moot, regardless of whether the latter contains valid points or not (in fact, not a single point is valid there).

In that last paragraph (assigned by us number 12, further below), the Referee states that:

"The authors should acknowledge that **the particle numbers are simply too small to use macroscopic (or bulk matter) thermodynamic terminology**."

Consistent with the lack of logical coherence of the Referee's report, noted in the first paragraph of our reply, this statement does not follow from anything that was said earlier in the Report. However, if this breathtakingly "ambitious" statement were true, our labor and discovery would be invalid indeed, regardless of anything else one could have conjured up – positive or negative - and said earlier. The statement is ambitious in that it would at the same time invalidate **all** nuclear thermodynamics of the last 60+ years, which uses routinely the purely thermodynamical notions of entropy, temperature, chemical potential, pressure, thermal energy, free energy, etc., with respect to finite nuclei. Major theoretical developments would have to be rescinded on the Referee's say so including, but not limited to:

all statistical thermodynamical models of compound nuclear decays on the market since over sixty years, including well established evaporation and fission simulation codes (PACE, CASCADE, GEMINI), Weisskopf's approach, statistical fission theory, not to mention the somewhat controversial mainstream models of multifragmentation – Expanding Emitting Source Model (EESM), Statistical Multifragmentation Model (SMM), and Metropolis Monte Carlo Model (MMMC). This encompasses over sixty years of thermodynamical theory as applied with amazing success to finite nuclei, thousands and thousands of scientific papers – the work product of a large community of both, theoreticians and experimentalists, hundreds and hundreds of conferences and numerous textbooks. Here is just one quick example of the (lead) subject of the coming International Workshop on Multifragmentation (Caen, 11/02-05/2011) – "Thermodynamics of finite nuclei & nuclear matter".

Does the Referee really believe that all of this is invalid? Has this then been a wholesale scam by a large community of nuclear scientists involved?

Less conspicuous, but equally breathtaking in its significance, if it were true, is the statement by the Referee (assigned by us number 11, further below) that:

"All microscopic dynamical calculations show that the outcoming fragments are preformed ..."

And, again, consistent with the lack of logical coherence, it is impossible to discern the relevance of this "ambitious" statement to our study of limits of thermodynamical stability of excited nuclear systems, unconcerned with a possible fragment formation at all. However, the statement is "ambitious", as (if true) it would disqualify wholesale all microscopic dynamical calculations for their implied inability to explain the most elementary types of collisions – elastic scattering, damped collisions, compound and fast fission, none of which involves early or otherwise fragment formation. Fortunately for the authors of these models, however, the statement is also blatantly untrue – all these models do describe the latter most common reaction modes. These reactions result in excited nuclear systems to be subsequently treated by thermodynamical models, such as ours. Contrary to the claim by the Referee, in all these calculations, no early or otherwise "preformation" of fragments ever occurs.

Here are our replies/comments to the specific points of, what is supposed to be criticism in the order in which they appear in the Referee's report:

1. "But from Eq. (10) on everything looks like a grand canonical ensemble. There is a temperature and there is a chemical potential".

We note that, as a matter of pure logic, nothing at all can possibly follow from the mere fact that to somebody – here to the Referee - something looks like something else, whether it were a monkey, an elephant, or a grand canonical ensemble. The statement is inherently non-debatable, bringing to mind the famous Latin maxim –*De gustibus non est discutandum*. Therefore, the statement is entirely hollow and unscientific. We note that in virtue of its strategic placement, right after the statement unconditionally disqualifying our findings, one would expect here a clearly stated criticism – a premise for the

preceding conclusion. This is what we label as logical incoherence, as it clearly qualifies as the logical fallacy known as *non sequitur*.

Regarding the second statement of the same paragraph, one would logically expect here a premise supporting the first sentence. Alas, it is logically disjoint from the first one (the logical fallacy of *non sequitur*), as all approaches (microcanonical, canonical and grand canonical) routinely use notions of temperature and chemical potential for the characterization of systems considered. In fact, both, temperature and chemical potential are defined from the conditions of microcanonical equilibrium, i.e., are in a natural way suited for the description of microcanonical ensembles. It is somewhat amusing that the Referee never disputes explicitly our approach being microcanonical, which it truly is.

However, we are truly puzzled as to where the Referee sees the resemblance of what we do to a grand canonical approach and why such a resemblance should matter. We submit that, for anybody familiar with thermodynamics, it is virtually impossible to confuse microcanonical approach with either the grand canonical one or the canonical one. We maximize the entropy at constant energy, volume, and number of particles and **all**, without exception, our conclusions are based solely on the functional dependence of the entropy on energy – clearly and indisputably the microcanonical approach. We note here that the microcanonical temperature we use represents simply the inverse of the first derivative of this entropy functional.

We note that in the grand canonical (*aka* macrocanoncal) approach, one would minimize the Landau potential (*aka* grand potential) at constant temperature, chemical potential, and volume - an entirely different "ball game". However, neither do we introduce and use the Landau potential, nor do we have the prerequisite thermostatic sink, nor do we allow the number of particles to float to keep the chemical potential equal to that of the sink.

We note that, regardless of the above, for the bulk matter the micro- *versus* macrocanonical "controversy" is a non-issue, as any approach, whether micro- or macrocanonical, or canonical yields exactly the same results in the domain where thermodynamics is applicable, i.e., up to the boiling point.

2. "I do not understand Eqs. (11) and (12), and I also do not see a reference where they are explained. Even the dimensions are wrong. What is a_0?"

This is again a textbook case of the logical fallacy of *non sequitur*, as nothing at all can possibly follow from the mere fact that somebody – here, the Referee – does not understand something or fails to see something. Regarding the dimensions, how possibly can the Referee state that the dimensions are wrong (they are, of course, correct) if he does not understand our notation. We do admit that after reviewing the notation, we found an inconsistency with the one we used earlier in the section. This we correct in the revised manuscript.

These equations are correct and have the correct dimensions $(MeV/fm^3 \text{ for the pressure} and MeV \text{ for the chemical potential})$, as a_0 – the level density parameter per nucleon is

in 1/MeV. The equations are so trivial that usually they are not even shown in published papers. Nevertheless, in the revised manuscript, we will add the (obvious) first steps in the derivations, i.e.,

$$T = 1/(\frac{\partial S}{\partial E})_{V,N} = \dots$$

$$p = T(\frac{\partial S}{\partial V})_{E,N} = \dots, \text{ and}$$

$$\mu = -T(\frac{\partial S}{\partial N})_{V,E} = \dots$$

We will also replace the symbol a_o with α_o , introduced earlier in the text for the level density parameter per nucleon.

At any rate, this is again a non-issue that would be easily resolvable, had there been a typo in our equations. It cannot possibly justify the unconditional rejection of our manuscript by the Referee, especially in view of the fact that our conclusions do not rely on these equations at all.

We are also somewhat puzzled about why the Referee did not just take the partial derivatives of the entropy function himself/herself – a rather trivial and fast task, indeed, to confirm his/her suspicions regarding the validity of our equations. For the sake of completeness, we wish to note here, that in grand canonical approach, one would have to take proper partial derivatives of the Landau potential, while in canonical approach, proper partial derivatives of the (Helmholtz) free energy.

3. "I can not see how the schematic model proposed by the authors is thermodynamically consistent.

Again, the statement is a clear-cut logical fallacy of *non sequitur*, as nothing at all can possibly follow from the mere fact that somebody – here, the Referee – fails to see something. The statement cannot be disputed as a matter of principle, simply because *De gustibus non est discutandum*. It is, thus, hollow.

The statement contains also a gross misstatement of facts. The fact is that we neither claim having proposed here any new schematic model nor do we propose one. We simply describe what we use – we do not add any new assumptions to the existing and well established model of the interacting Fermi gas and the well-established Thomas-Fermi approximation. All we do is that we consider explicitly the (obvious) degree of freedom of thermal expansion. This degree of freedom could be neglected at lower excitation energies, but cannot be neglected at elevated excitation energies. We do this strictly within the framework of microcanonical thermodynamics, relying solely on the well established notion of Boltzmann's entropy. In this respect, see, e.g., our earlier publications – dating back to 1981, as well as our references to publications 14, 15 and

16, where essentially the same approach was pursued in studying the response of the matter density profile to the excitation energy supplied.

Unfortunately, the above (hollow) statement by the Referee is also quite troubling to us, as it invokes a (to us) novel notion of "thermodynamic consistency". To our knowledge, such a notion has never been used with respect to microcanonical thermodynamics, which (to us) is inherently thermodynamically consistent. Therefore, we must consider for now that the said notion has been fabricated by the Referee *ad hoc*, for the purpose of his/her Report. And, therefore, we must kindly ask and insist that the Referee provides references to published work(s), where the term of "thermodynamic consistency" is defined (the relevant criteria are articulated) with respect to microcanonical models used in nuclear science and where any of the many models listed further above (third paragraph on page 1) is scrutinized in terms of such a consistency.

4. "Is for example the Hugenholtz-van Hove theorem fulfilled in the case of finite nuclei"?

This is yet another (multiple) logical fallacy and, thus, a hollow question.

Firstly, it is a typical *non sequitur*, as the statement is disjoint from the preceding (hollow) one, in spite of the "for example" clause. This is so, because the HvH theorem explicitly and specifically refers and (correctly) limits itself to the ground state energy, which does not involve any thermodynamical theory, consistent or otherwise, as a matter of principle.

Secondly, the question constitutes what is known as a fallacy of a *complex question*, a question that relies on a false premise being true. Here, the false premise in question is that the HvH theorem applies to non-extensive systems with finite-range interaction, while, in fact, it does not apply, similarly as it does not apply to excited systems. Therefore, the theorem in question cannot possibly be fulfilled, nor can it possibly be not fulfilled. All one can here state with certitude is that the case of finite nuclei does not contradict HvH theorem in the same fashion as it does not contradict, e.g., the Pythagorean Theorem – both statements being formally correct but of dubious value to the advancement of science.

We must also note that to our knowledge, the issue of HvH has never been invoked with respect to any of the many thermodynamical theories listed in our paragraph 3. Therefore, we must once again assume that, while being a sham issue, it has been fabricated by the Referee *ad hoc* for the purpose of his/her Report.

Furthermore, we wish to point out that our Eq. 12, while (admittedly) incomprehensible to the Referee, shows that for the bulk matter that can be characterized by an EOS and at equilibrium at T=0, the chemical potential is, indeed, equal to the average energy per nucleon, which is the thesis of the HvH theorem. We will point out this amusing fact in our revised manuscript.

5. "In Sec. III, the authors discuss at length in a complicated way what they call boiling. But this is no more than the discussion of the coexistence region, which in a mean-field picture shows up by spinodal instabilities".

Once again, *De gustibus non est discutandum*. Nothing at all can possibly follow (*non sequitur*) merely from the fact that something looks "at length" and "complicated" to somebody – in this case, the Referee. To us it is simple, and who can dispute that? Therefore, the Referee's statement is hollow.

We note that boiling is a phenomenon known from ancient, pre-science times and is not something that only we for some mysterious reason call boiling (as in Referee's somewhat contemptuous use of words "what they call boiling"). What we do, is that we first identify the boiling point of Fermi liquid on a Van-der-Waalsian plot of isotherms – this point does not belong to the coexistence region at all, contrary to what is claimed by the Referee. Then we try to explain the phenomenon in terms of a transport theory (via the behavior of chemical potential as a function of concentration). But the strictly microcanonical explanation lies in our Fig. 3, displaying the form of the entropy surface, featuring a saddle point.

Perhaps, the Referee could provide references to published works where the boiling of Fermi liquids is discussed in a way that (to him/her) appears substantially simpler than ours. We are unaware of such works and, therefore, we try to explain the boiling phenomenon in three different, but always simple (to us), ways.

6. "Section IV for finite nuclei assumes a density profile, and a thermodynamic consistency is lost".

The statement constitutes again a fine example of a multiple fallacy, consistent with the overall lack of logical coherence in the Referee's report, pointed out by us earlier. Firstly, the statement consists formally of two independent statements, which are logically disjoint in virtue of the use of a mere "and" (rather than an "and, therefore,") after the comma. Because of this, it makes no sense. Secondly, the first part of the statement is formally hollow because there is **always** a particular density profile involved of one kind or other, whether one wants it or does not want it.

One could be, perhaps, tempted here to second-guess the Referee that what he/her had in mind was that we have assumed a density profile with a diffuse surface domain, rather than one with a sharp cut-off profile, and that the second part of the statement was, indeed, intended as a conclusion to the premise expressed in such an "amended" first part. But, even were we to yield to such a temptation, the whole statement would still be wrong, as its first part would then constitute a gross misstatement of facts, a classical example of the *Straw Man* fallacy. This kind of a need for a second-guessing of the Referee's intent, which would still be insufficient to achieve logical coherence, explains our assertion in the first paragraph that it is very difficult to address intelligently the issues raised in the Referee's Report.

In fact, we do not assume any definite density profile at all. We try to account for the finite range of nuclear interaction – a commonly known fact and we do this in a way analogous to that employed, e.g., in BUU calculations. Accordingly, we allow the density profile to vary such as to result in maximum entropy (for finite excitation energy) or minimum energy for $E^*=0$.We note that the requirement of maximum entropy reduces, indeed, to the requirement of minimum energy, when the excitation energy is set to 0. Therefore, a density profile with a diffuse surface domain is not an assumption, but rather an obvious consequence of the finite range of nuclear interaction – a well established fact and not an assumption. It is a result of our having consistently applied microcanonical thermodynamics to a finite nucleus.

Now, the claim (in the second part of the statement in question) of thermodynamic consistency being lost is highly troubling to us as it is left entirely unsubstantiated by the Referee. As we stated earlier, we believe that such a claim cannot be substantiated, as the notion of "thermodynamic consistency" has (to our knowledge) never been defined, nor does it make (to us) any sense with respect to microcanonical thermodynamics. The statement is highly troubling, because it contains a scientific message that is potentially harmful to us – a message that we must consider for now as being fabricated by the Referee. Should we be right in our assessment here, the Referee would have had come dangerously close to what is definable as scientific misconduct (see, e.g., the definition of such in the Wikipedia). Therefore, we must kindly request and absolutely insist that the Referee provides references to published works where the notion of "thermodynamic consistency" is discussed, and the relevant criteria are articulated with respect to microcanonical nuclear thermodynamics and where it is actually proven that such is lost when finite range of nuclear interaction is accounted for, i.e., in all realistic cases.

Thirdly, the claim of thermodynamic consistency being lost, if true, would equally well apply to all thermodynamical models listed in the 3rd paragraph of this reply, as any practical model must account and, in fact, does account one way or other for the presence of the diffuse surface domain. The burden of proof would lie here with the Referee of why such consistency should be selectively considered with respect to our calculations and not others, using essentially the same concepts and approximations as we do.

7."I also don't understand the meaning of a central pressure. In equilibrium, the pressure and the chemical potential are uniform, otherwise the matter would start moving. Shouldn't therefore the central pressure be equal to the outside pressure which in that case is zero?"

This is once again a typical fallacy of *non sequitur*, as nothing at all can possibly follow from the mere fact that somebody – here, the Referee - does not understand something or somebody merely asks a question. It even would not have mattered if there were a simple answer to the question asked, as is the case here.

In fact, there is no mystery here whatsoever, as the condition for the dynamical equilibrium entails both, the pressure gradients and the mean-field gradients. These must be matched for the resulting forces to be zero. Only in the absence of mean-field

gradients, dynamical equilibrium requires the pressure to be uniform. However, there clearly is a gradient of the mean field at the surface of a finite nucleus (*vide* Saxon-Woods potential). This requires a matching gradient in the pressure and this is exactly what we get when maximizing the entropy – a non-zero pressure in the interior, a commonly known fact. The other way of looking at this is as the finite pressure in the interior resulting from the surface tension generated by the presence of the surface free energy. In terms of microcanonical thermodynamics, the finite range renders the finite system non-extensive (we discuss this point in our manuscript). For such systems, one cannot define a local pressure simply by taking the (local) partial derivative of entropy and, therefore, the uniformity of pressure is simply "not in the cards". This is why we show only the central pressure, as the latter refers to a virtually uniform density profile in the nuclear interior. We remind that we show the plot of the pressure for the sake of a better understanding by the reader, all our conclusions relying solely on the form of the (integral quantity) entropy as a function of (integral quantity) density profile – microcanonical approach.

8. "To summarize, the paper does not help in understanding the situation encountered in heavy-ion collisions, as claimed in the introduction."

Firstly, this crucial statement is a gross misrepresentation of what we claim in the Abstract and in the Introduction and of what we actually accomplish. This summary statement constitutes a textbook example of what is known as the *Straw Man* fallacy, as we neither claim, nor do we try to help understand what happens in heavy-ion collisions. Therefore, the summarizing statement is simply invalid and provides no grounds for the unconditional rejection of our paper.

Consistent with the overall lack of logical consistency of the Referee's Report (pointed out by us earlier), the statement is also a textbook example of the *non sequitur* fallacy. This is so, because it is completely disjoint from the preceding (sham) criticism. Had we been 100% right in our analysis of points 1-7, as we in fact are, we still would not have helped understand what happens in collisions, in agreement with what the Referee claims in point 8. This is not what the nuclear thermodynamics is, and has been, widely used for by others and by ourselves.

In contrast to all of the sham criticism discussed by us further above, nowhere in his/her report, does the Referee dispute our important claim of having discovered the phenomenon of surface boiling and that this interesting phenomenon, indeed, would explain the hitherto unexplained experimental observation of limiting temperature. The phenomenon of surface boiling is the topic of our study, stated in the title, the abstract, and throughout the manuscript, but the Referee makes no single mention of this.

The fact is that, with the exception of point 6, all points (1-5 and 7) preceding this fallacious summary statement are void of scientific message. Regarding point 6, we are unable to determine its scientific basis in the published literature and expect the Referee to provide the missing references and thus alleviate the impression that he/she has

fabricated the issue and the associated scientific messages involved *ad hoc* for the purpose of his/her report.

In view of the above, the Referee's report contains no justification whatsoever for the unconditional or otherwise rejection of our manuscript. We must again kindly ask, based exactly on which of the points 1 - 7 the Referee has made his/her determination that our study and discovery does not merit publication in Phys. Rev. C.

The Referee makes additionally several post-summary statements, two of which (11 and 12) are breathtakingly startling:

9. "In collisions one never is in an equilibrium state, everything is transient".

This is another textbook example of a *Straw Man* fallacy. We fully agree with this truism by the Referee, but nowhere have we made a statement to the contrary. Actually, we explain in quite a detail the meta-stable character of configurations we consider.

10. "The nuclei do not boil in the sense that they form bubbles of gas in the interior or separate off little drops from the surface".

We note that this is yet another example of the *Straw Man* fallacy. We fully agree, but we have never made a statement to the contrary. In fact, it is our study that explains for the first time, why there is no boiling of the bulk, interior matter – according to our study it is preempted by a large margin by the hitherto unknown phenomenon of surface boiling.

11. "All microscopic dynamical calculations show that the outcoming fragments are preformed ..."

This is example of another logical fallacy – *non sequitur*, as it has no relevance to our study that has nothing to do with reaction dynamics and fragment formation, early or otherwise. We discussed the dire implications of this statement (if true) for all microscopic dynamical calculations in the 4^{th} paragraph of our reply.

12. "The authors should acknowledge that the particle numbers are simply too small to use macroscopic (or bulk matter) thermodynamic terminology".

This is a truly startling statement that applies equally well to **all** thermodynamical models on market, as already noted and discussed in more depth in the third paragraph of this reply.

Once again, we kindly ask the Referee to be specific as to the scientific grounds on which he/she determines the unsuitability of our manuscript for publication. Also, as stated further above, we must absolutely insist on Referee providing meaningful references to published papers dealing with issues raised by him/her in points 3 and 6 with not enough specificity to determine the scientific basis for them.

7. Letter from the Editors regarding author's original reply to the second Referee

Re: CB10255

Surface boiling: A new type of instability of highly excited atomic nuclei by J. T = 0 ke and W. U. Schr\"{0}der

Dear Dr. Schroder,

I invite you to rethink your reply to the referee and formulate your response in less provocative terms. In many cases you seem to be attacking the referee rather than defending your point of view. I remind you that the fact that the referee does not understand a point in the paper is, indeed, meaningful and a reason for rejecting the paper since, if the referee doesn't understand a statement then the eventual reader likely will not either. Most authors, when confronted with this comment from the referee, simply go on to explain the point further. As for the point about there being too few particles to do thermodynamics, I have heard this objection before which has not stopped a large part of the community from brushing it aside and doing good work, as you say. In summary please try to make your response to the referee more neutral, you will have a much better chance of having the paper accepted.

Yours sincerely,

William R. Gibbs Associate Editor Physical Review C Email: prc@ridge.aps.org Fax: <u>631-591-4141</u> http://prc.aps.org/

8. Letter to the editors explaining the intended function of the original reply to the second Referee

Dear Editors,

We have decided to follow your most recent suggestion that we reformulate and tone down our reply to the Referee in order to "have a much better chance of having the paper accepted" by the Referee, as you say. We are attaching such a revised reply to the Referee's comments, while remaining highly skeptical that our technical explanations will be able to influence this particular individual.

We further wish to note that, while our original reply was formally addressed to the referee you have chosen to review our paper, it was factually also directed to you, the Editors of PRC, with the intent to prove that this particular referee does not meet the requirements for a peer reviewer. This referee has not only proven to be unfamiliar with nuclear thermodynamics, its issues, language, the dominating consensus as to the validity of mainstream approximations and models, etc., but has openly professed contempt for the whole field. Therefore, the attached reply should not be construed as lending legitimacy to the Referee as our peer, i.e., as a fellow (nuclear) scientist.

Sincerely,

W. Udo Schröder

Professor of Chemistry & Physics 466 Hutchison Hall University of Rochester (Chem.) Rochester, New York 14627-0216

9. Sanitized version of the reply to the second Referee

Reply to the Referee's Report

We have studied the Referee Report and were gratified to notice that the Referee does not question the validity of our main claim to have discovered a new type of thermodynamic instability of finite nuclei, which we call surface boiling. The Referee does not question that this kind of dynamical instability, following from the convexity of the entropy function S=S(E), does indeed explain the well-known phenomena of a limiting temperature and limits to the excitation energy that can be equilibrated in a compound nucleus, phenomena for which hitherto there was no explanation.

We were also glad to notice that all specific concerns by the Referee appear to be of a "cosmetic" or semantic nature and can thus be readily answered. Before we do this, we wish to draw the Referee's attention to the following important facts:

- (i) We do not introduce any new model of ours. We rely on well-established models of the compound nucleus and the Fermi gas and use the well-established Thomas-Fermi approximation and a general expression for the entropy of a compound nucleus suggested by H. Bethe. As far as the evaluation of the binding energy is concerned, we use the same equation of state (EOS) and the same method used in the popular BUU or the droplet models. The Referee may wish to verify that an approach essentially identical to ours was used, e.g., in Refs. 15 and 16, and with the exception of accounting for the finite range of interaction, in our earlier publications (Refs. 7 14).
- (ii) Novel in our present study is that we notice that for finite nuclei there is a limiting energy beyond which no metastable equilibrium is possible. This is evidenced by the appearance of a negative heat capacity (convexity of the entropy function). The appearance of a negative heat capacity we identify with the onset of volume boiling, in the case of nuclear matter, and of surface boiling, in the case of finite nuclei.
- (iii) While for infinite systems (aka systems at the thermodynamic limit) all three approaches – microcanonical, canonical, and grand canonical lead to identical results, for non-extensive systems, only the microcanonical approach has been shown viable (see, e.g., the book by D.H.E. Gross on "Microcanonical Thermodynamics: Phase Transitions in 'Small' Systems", World Scientific Publishing Co., Singapore 2001).
- (iv) The microcanonical approach relies fully on the microcanonical partition function e^S, i.e., on the functional dependence of the entropy S on total energy E, volume V, and number N of particles. These are all integral or global observables and not local ones.
- All of our conclusions rely solely on the appearance of the microcanonical partition function. We use the local observables of pressure and chemical potential only to better characterize the system and to make it more intuitively

understandable to the reader. For non-extensive systems, no local observables can be reliably defined and, therefore, we limit ourselves to the central pressure that relates to a quasi-uniform matter distribution.

Following are our replies to the specific remarks by the Referee:

1. "But from Eq. (10) on everything looks like a grand canonical ensemble. There is a temperature and there is a chemical potential".

Our approach is microcanonical, as it relies on maximizing the microcanonical partition function or entropy at a fixed total energy, volume, and number of particles. In contrast, a grand canonical (aka macrocanonical) approach would minimize the Landau (aka grand) potential at fixed temperature, chemical potential, and volume. We note that temperature and chemical potential are defined from the condition of microcanonical equilibrium, via derivatives of the logarithm of the microcanonical partition function (the Boltzmann's entropy) and are, thus, natural observables for microcanonical systems at thermodynamic limits (infinite nuclear matter). They are of limited use in the case of non-extensive systems such as finite nuclei with diffuse surface domain, which is why we show only central pressure for such systems. To our knowledge, nobody has applied the grand canonical approach to non-extensive systems, like finite nuclei with finite-range interaction.

2. "I do not understand Eqs. (11) and (12), and I also do not see a reference where they are explained. Even the dimensions are wrong. What is a_0?"

These equations are correct and have correct dimensions (MeV/fm^3 for the pressure and MeV for the chemical potential), as a_0 – the level density parameter per nucleon- is in units of 1/MeV. The equations are so trivial that they are usually not shown in published papers. Nevertheless, in the revised manuscript, we will add the (obvious) first steps in the derivations, i.e.,

$$T = 1/(\frac{\partial S}{\partial E})_{V,N} = \dots$$

$$p = T(\frac{\partial S}{\partial V})_{E,N} = \dots, \text{ and}$$

$$\mu = -T(\frac{\partial S}{\partial N})_{V,E} = \dots$$

We noticed that the notation we used in these equations was not consistent with the one used by us earlier in the manuscript and that this might have caused confusion by the Referee. We correct the notation by replacing the symbol a_o with α_o , introduced earlier in the text for the level density parameter per nucleon and \epsilon_{EOS} by a similar symbol with a bar on top to indicate the energy per nucleon rather than per unit volume.

3. "I can not see how the schematic model proposed by the authors is thermodynamically consistent.

Microcanonical thermodynamics is inherently thermodynamically consistent, as it relies solely on the functional dependence of the microcanonical partition function on energy. It has its limits where it becomes inapplicable – the boiling point being one of such limits. It has also limits as to what one can do with this partition function. For example, one cannot define local observables of T, p, and chemical potential for non-extensive systems. Regarding the temperature, only a global microcanonical temperature can be defined.

To our knowledge, the issue of thermodynamic consistency has never been raised with respect to microcanonical thermodynamics and should the Referee wish a better explanation than the one we provided, we would kindly ask him/her to provide references to published works where the notion of thermodynamic consistency is defined and the criteria for such are articulated.

4. "Is for example the Hugenholtz-van Hove theorem fulfilled in the case of finite nuclei"?

The Hugenholtz-van Hove (HvH) theorem is merely a theorem and not a law of nature. As such, it is applicable only when the premise is met under which it has been proven true. Accordingly, it is inapplicable to excited (thermodynamical) systems and finite systems interacting via finite-range forces. Therefore, all we can say is that our case does not contradict the HvH theorem.

Furthermore, we wish to point out that our Eq. 12 shows that, for bulk matter that can be characterized by an EOS and at equilibrium at T=0, the chemical potential is, indeed, equal to the average energy per nucleon, same as the thesis of the HvH theorem. We will point out this amusing fact in our revised manuscript.

5. "In Sec. III, the authors discuss at length in a complicated way what they call boiling. But this is no more than the discussion of the coexistence region, which in a mean-field picture shows up by spinodal instabilities".

For infinite matter, our truly microcanonical explanation is based on Fig. 3 displaying the entropy surface which features a saddle point – indicating the onset of negative heat capacity. To our knowledge, the onset of boiling has never been discussed in terms of microcanonical thermodynamics and the Referee may wish to contradict us here by providing references to the contrary.

However, to allow the reader to understand the (hypothetical) volume boiling, we provide two more intuitive explanations. We note, that the boiling does not occur in systems enclosed in confinements, such as commonly studied theoretically and that, to our knowledge, it has not been discussed at length if discussed at all for nuclear matter. We would feel truly indebted to the Referee if he/she could provide references to works where boiling of Fermi liquid is discussed in simpler terms than ours. In that case, we could leave the offending "complicated" part of our explanation out and replace it with relevant references. We note that we are unaware of such works.

For finite system our conclusions are based solely on the appearance of the functional dependence of the first derivative of the entropy (microcanonical temperature) on total energy.

6. "Section IV for finite nuclei assumes a density profile, and a thermodynamic consistency is lost".

We note that we do not assume any specific density profile at all. We try to account for the finite range of nuclear interaction – a commonly known fact- and we do this in a way analogous to that employed, e.g., in BUU calculations or in the droplet model. Accordingly, we allow the density profile to vary such as to result in maximum entropy (for finite excitation energy) or minimum energy (for $E^*=0$). We note that the requirement of maximum entropy reduces, indeed, to the requirement of minimum energy, when excitation energy is set to zero. Therefore, a density profile with a diffuse surface domain is not an assumption of ours, but rather an obvious consequence of the finite range of nuclear interaction – a well established fact and not an assumption, either. It is result of a consistent application of microcanonical thermodynamics to a finite nucleus.

Regarding thermodynamic consistency being lost, we refer to our reply further above (point 3) and kindly ask the Referee to provide references to published works where the notion of thermodynamic consistency and the loss thereof are discussed and the relevant criteria are articulated for microcanonical systems. We are afraid that we must insist on receiving this additional information, in order to be able to discuss this point and possibly refute the criticism by the Referee in any more depth than we were able based on the bare (unsubstantiated) assertion by the Referee alone.

7."I also don't understand the meaning of a central pressure. In equilibrium, the pressure and the chemical potential are uniform; otherwise the matter would start moving. Shouldn't therefore the central pressure be equal to the outside pressure which in that case is zero?"

There is no mystery here whatsoever, as the condition for the dynamical equilibrium entails both, the pressure gradients and the mean-field gradients. These must be matched for the resulting forces to be zero. Only in the absence of mean-field gradients, dynamical equilibrium requires the pressure to be uniform. However, there clearly is a gradient of the mean field at the surface of a finite nucleus (*vide* Saxon-Woods potential). This requires a matching gradient in the pressure, and this is exactly what we get when maximizing the entropy – a non-zero pressure in the interior, a commonly known fact. Another way of looking at this is in terms of a finite pressure in the interior resulting from the surface tension generated by the presence of a surface free energy. We note that the pressure is directly related to the surface energy and its radius of curvature. In terms of microcanonical thermodynamics, the finite range renders the finite system nonextensive (we discuss this point in our manuscript and also further above in point 5). For such systems, one cannot define a local pressure simply by taking the (local) partial derivative of entropy and, therefore, uniformity of pressure is simply "not in the cards". This is why we show only the central pressure, as the latter refers to a virtually uniform density profile in the nuclear interior. We remind the Referee that we show the plot of the pressure for the sake of a better understanding by the reader, all of our conclusions rely solely on the form of the (integral quantity) entropy as a function of (integral quantity) density profile – clearly a microcanonical approach.

8. "To summarize, the paper does not help in understanding the situation encountered in heavy-ion collisions, as claimed in the introduction."

This statement is inaccurate as we do not claim that we explain what happens in collisions, in addition to our discovery of surface boiling which helps understanding the origin of a finite temperature observed in a host of experiments. The collision stage is addressed by a completely different set of theories implemented in dynamical models of nuclear reactions. It is known experimentally that, in the course of collisions, highly excited nuclear systems are formed such as, e.g., projectile- and target-like fragments, or fused compound nuclei. These systems are known (both experimentally and theoretically) to arrive at states of meta-stable equilibrium which then decay statistically. Such meta-stable systems are the subject of nuclear thermodynamics, as we know it since over 60 years, and also the subject of our present study.

The Referee may wish to verify what we actually say in our title, abstract, and the introduction (where we explicitly refer to highly excited systems produced in heavy-ion collisions and not to the collisions as such) and correct the above summary statement accordingly.

The Referee makes several additional post-summary comments:

9. "In collisions one never is in an equilibrium state, everything is transient".

This is true, but as we already stated in our reply to point 8, thermodynamics is of limited use in establishing what happens in collisions. For this purpose, there are numerous dynamical models on market. We do not attempt to describe collisions.

10. "The nuclei do not boil in the sense that they form bubbles of gas in the interior or separate off little drops from the surface".

This is true and we have not made a statement to the contrary. We note that it is our study that explains for the first time why there is no volume boiling in finite nuclei. According to our study this kind of boiling is being "pre-empted" by the surface boiling occurring at significantly lower excitation energies.

On the other hand, for hypothetical infinite matter (or matter interacting via zero-range forces), there is no reason why it should not boil in a similar fashion as we observe almost daily in our kitchen.

11. "All microscopic dynamical calculations show that the outcoming fragments are preformed ..."

This statement is somewhat inaccurate. What is relevant to our study is that all microscopic dynamical models confirm what is observed in experiments and what is observed is that, in the Fermi energy domain and at somewhat higher energies, most collisions proceed via a binary damped-collision mode and end up with two excited massive fragments. For more central collisions, these models confirm the experimentally proven existence of fusion reactions. This is where the dynamical model calculations come to a halt and where they "extend" an invitation to thermodynamic models to explain the further fate of these excited systems.

12. "The authors should acknowledge that the particle numbers are simply too small to use macroscopic (or bulk matter) thermodynamic terminology".

Nevertheless, there is an overwhelming consensus among nuclear scientists that thermodynamic language and method are well suited for the description of excited nuclei. This is evidenced by the over 60 years of nuclear thermodynamics research, which has resulted in tens of thousands of published works, in many books, in countless numbers of conferences and workshops, involving a very large community of scientists. What matters is not so much the number of particles but, rather, the number of micro-configurations involved which can easily assume values of 10 to the power of 100 (10**100) and higher. There is no hope for treating such huge systems quantitatively other than by invoking statistical methods. Here the statistical thermodynamics we use offers a proven method. We do not claim having invented it.

The revised manuscript includes the following changes – all on p. 7 of the revised manuscript:

- (v) We have amended the notation in Eqs. 11 and 12, to make it consistent with the one used earlier.
- (vi) In Eqs. 10-12, we added the first step leading to the final expressions.
- (vii) Added a sentence explaining the used notation.
- (viii) Prompted by a remark by the Referee, and directly following Eqs. 10-11, we added a paragraph where we draw attention to the fact that in the simple model we use, the thesis of the Hugenholtz Van Hove theorem is fulfilled. We call the fact amusing, because it has no relevance to our conclusions, but may be of a potential interest to some readers.
- (ix) Added the reference (18) to the Hugenholtz Van Hove paper.

We hope that the Referee accepts our explanations and, regarding points 3 and 6, supplies the additional information we would need to further address them in more depth, if required, and that he/she will now recommend our revised manuscript for publication in Phys Rev C.

10. Letter from the Editors announcing the end of the review process

Sept 16, 2011:

Re: CB10255

Surface boiling: A new type of instability of highly excited atomic nuclei by J. $T\$ and W. U. Schr\"{o}der

Dear Dr. Schroder,

The above manuscript has been reviewed by one of our referees. Comments from the report appear below.

We regret that in view of these comments we cannot accept the paper for publication in the Physical Review.

In accordance with our standard practice, this concludes our review of your manuscript. No further revisions of the manuscript can be considered.

Yours sincerely,

Benjamin F. Gibson Editor Physical Review C Email: prc@ridge.aps.org Fax: <u>631-591-4141</u> http://prc.aps.org/

11. Report by the third referee

Report of the Third Referee -- CB10255/Toke

I have carefully read the revised version of the manuscript and all the past correspondence. I agree with the main criticisms raised by the two previous referees and I do not recommend this work for publication.

* The main claim of this work is that a novel instability would have been discovered. What the authors actually do, is giving new names to well-known phenomenologies. Their volume boiling is nothing but the spinodal instability of homogeneous nuclear matter with respect to phase separation; their surface boiling is nothing but the manifestation of the same instability in finite nuclei. The boiling point is known in thermodynamics as the flashing point and as such has already been introduced in nuclear physics before [see for instance A. Rios, Nuclear Physics A 845, 58 (2010)]. The fact that surface effects lower the temperature and excitation energy of the instability with respect to the bulk limit is also well known and discussed in the literature, see P. Chomaz et al., Phys. Rep. 389, 263 (2004) and references therein.

* The authors recognize themselves their model is not new either. In this context, as it has already been stressed by the other referees, the total lack of reference to previous works cannot be accepted. In particular, X. Vinas and collaborators have been performing finite temperature Thomas-Fermi calculations for finite nuclei for years [see for instance PLB 638, 160 (2006) or PRC 75, 054608 (2007)]. Their treatment, including kinetic energy, correlations beyond mean field, deformations and expansion, seems to me superior to the present formalism, and I do not see what this paper brings more than the cited work.

* The authors claim that the novelty here is (1) the statement that for finite nuclei there is a limiting energy beyond which no metastable equilibrium is possible, (2) and that this is evidenced by the appearance of a negative heat capacity. Concerning (2), it is textbook knowledge that instabilities correspond to backbendings of the appropriate equations of state. Concerning (1), the existence of a limiting excitation for nuclei was discussed as early as in the 80's [Levit et al., NPA 436, 265 (2085)] in a mean-field model which is microscopic but conceptually similar to that of the authors'. The connection between this limiting excitation energy, the emission in the continuum with the associated onset of expansion, and the plateau in the caloric curve was advanced by many authors, namely J. Natowiz, the already mentioned work by X. Vinas et al. (with Thomas-Fermi arguments very similar to the present work), and also in the context of another model by C. Dorso PRC 58, 632 (1998).

* The other claim of originality comes from the use of the microcanonical ensemble. I do not think this statement is correct. The spinodal or "boiling" instability is present in homogeneous nuclear systems independent of the ensemble. The choice of the ensemble determines only the observables in which this instability will manifest.

Going to the observables presented by the authors, the decrease of the temperature with increasing energy can be very well obtained at the canonical level (T,V,N) if the average energy $\langle E \rangle = - partial lnZ(T,N,V)$ \partial \beta on the abscissa is not calculated with V=cte as it is the case at constant pressure. Indeed such canonical backbendings have been observed, see for instance Das et al., Phys. Rep. 406, 1 (2005). What is really specific in the microcanonical thermodynamics (as explained in the book by D. Gross the authors acknowledge in their answer to the referee but do not even cite) is that the convexities giving rise to backbendings can correspond to stable microcanonical states (while they are unstable in the canonical ensemble). In order to explore this ensemble inequivalence one needs a real microcanonical model considering all the possible microstates. A mean-field model like that of the authors' by construction does not include the multibody configurations and does not exhibit ensemble inequivalence. meaning that I do not see why, within these severe approximations, getting a temperature out of an energy would be better than the other way out.

All the previous criticisms concern the novelty of the work. In addition to that, I think this work is not entirely scientifically sound.

* My main criticism concern the thermodynamics of the model itself. Equations (10)-(12) are simply not correct. The entropy entering here should be S_system, obtained by summing up all the possible configurations, and not by isolating one single (mean field) configuration as the authors do.

* It is not true that the only "natural" pressure is zero (page 5): all self-bound systems including the nucleus correspond to finite pressure.

* It is not true that pressure and temperature cannot be controlled in infinite systems (page 5), to the contrary, they are the natural parameters for bulk calculations which are routinely done in astrophysical applications

* It is not true that an instability cannot be explored (page 6): first, the appearance of an instability is deeply linked to the weakness of the model itself, namely the mean-field approximation used which ignores all of the possible microcanonical microstates but one; second, whether an instability is actually explored or not needs time scale arguments, as already stated by the other referees.

12. Letter to the Editors regarding the report by the third Referee

10/17/2011

Dear Editors,

We are deeply shocked by the manifest hostility and the offensive language used by the third referee, similarly as by the two previous ones. It is our understanding, that the Editors have asked the third referee to review the comments by the first two referees and our replies. We also trust that the Editors did not solicit yet another, new batch of comments to which we are then not allowed to reply. However, instead of addressing the issues raised by previous referees in a professional and impartial manner, as would have been expected, the third referee has filed what appears a frivolous report. Like the reports by the previous referees this one, too, contains no single valid criticism of our paper and reflects serious ignorance in the field of thermodynamics, but raises at the same time some serious ethical and legal questions including the question of the referee's motivation.

The third referee states that he/she agrees with the "main criticism" in the previous two reports. Yet, it transpires from this report that the third referee actually does not dispute any of our replies to the comments by the first and second referees. Occasionally, the referee even explicitly contradicts the second referee, such as concerning the question of pressure. Accordingly, he/she does not identify what this "main criticism" may be – we understand that he/she simply is unable to. Nor have the Editors stated what the "main" criticism" is supposed to be according to the third referee?

It is more than just an ethical issue when the referee makes knowingly a false, frivolous, or even fraudulent statement to inflict damage on our reputation and our research program. We must therefore insist that the Editors ask the third referee in unambiguous terms to address each and every one of our 10 replies individually and to identify the "main criticism" which would warrant the rejection of our paper.

The second ethical issue with the report is its tacit endorsement, support or promotion of a factual (intentional or otherwise) serial plagiarism of our published findings by a group of other authors which are highly praised by the referee. The two papers referred to by the referee are:

(i). "Density reorganization in hot nuclei" by S.K. Samaddar, J.N. De, X. Vinas, and M. Centelles, PRC 75, 054608 (2007), and
(ii) "Nuclear expansion with excitation" by J.N. De, S.K. Samaddar, X. Vinas, and M. Centelles, Phys. Lett. B 638 (2006) 160.

We fully agree with the third referee that in our manuscript we use essentially the same methodology as applied in the above two papers by Samaddar, De et al. However, we have invented and publicized this methodology already back in 2002 where we used it

exactly for the purpose for which the above authors employ it. In 2003 we have then published the methodology in Phys. Rev. C (PRC 67, 034609 (2003); position 8 on our list of references).

Aside from the ethical issue associated with the fact that Samaddar, De, et al. do not cite our directly relevant prior work it is important to note that they publish findings identical to our earlier conclusions and call these findings interesting but begging further study. This is important for the present case, as it proves that findings presented in our manuscript are indeed new, not only as far as surface boiling is concerned, but also regarding the process of volume boiling. Apparently, the referee has chosen to ignore this "inconvenient" fact.

As evidence we attach copies of the paper (i) (PRC 75...) cited above and of our paper (PRC 67 ...)

Please, compare our Fig. 1 with Fig. 1 in the Samaddar paper. Both figures illustrate identical trends in the evolution of nuclear equilibrium density with excitation. They both refer to self-similar expansion (eq. 5 on our p. 1 and eq. 19 on their p. 4) and both were obtained using "our" condition of maximum entropy (zero pressure). Please, compare now our Fig. 2 (p. 3) with Figs. 5 and 6 on p. 8 in Samaddar's paper. They all illustrate nuclear caloric curves featuring negative heat capacity – something the third referee erroneously calls "textbook stuff." Please, read further (on p.8, last paragraph) what Samaddar et al. have to say about "their" finding of a negative heat capacity. They call that an "interesting feature" but do not provide any interpretation. We fully agree that it this feature is interesting indeed, as it does not appear in any of the three classical ensembles – micro-canonical, canonical, or grand-canonical. This behavior appears only in our microcanonical approach for self contained systems, used also by Samaddar, De et al.

In contrast, our present manuscript provides a full interpretation of that "interesting" feature in terms of boiling. Referring to our interpretation the referee calls this boiling instability incorrectly "spinodal instability." Obviously, Samaddar, De et al. know the language of thermodynamics and avoid such a misidentification. Why would it be interesting if it were the "textbook" spinodal? We note that the term spinodal is used to describe mechanical and not thermal instabilities. In fact, there can be no spinodal instability in microcanonical ensembles for a self-contained system for a trivial reason - the entropy here is not a function of volume.

What the example with the cited paper by Samaddar, De et al. tends to show is that:

- 1. The phenomenon of boiling of nuclear matter was not understood to this day and was not discussed earlier in the literature.
- 2. Our discovery is indeed new and merits prompt publication.
- 3. The criticism of our manuscript by the third referee for not providing references to a works that plagiarize our methods and findings is unwarranted and in a sense preposterous.

Now, regarding the serial character of factual plagiarism (likely unintentional) by Samaddar, De et al., these authors additionally "replicate" in their cited manuscripts two other discoveries reported by us in our earlier papers. These are the discoveries of the phenomenon of shape instability and the role of surface entropy in enhancing the fragment production. A minor difference consists in that they call "deformation entropy" what we call "surface entropy." The first of our papers Samaddar et al. fail to acknowledge is our PRL 82 (1999) 5008. This paper, "New type of shape instability of hot nuclei and nuclear fragmentation," describes the very mechanism which Samaddar, De et al. "re-discover" eight years later. The third referee of our present manuscript, however, has the references to our papers which we cite exactly in the context of continuing study and chose to ignore these altogether.

This brings the total of infringements on our intellectual property rights by these authors to four!!! These are (i) the methodology of microcanonical ensemble for self-confined systems, (ii) the observation of negative heat capacity in such systems, (iii) the discovery of shape instability, and (iii) the discovery of the role of surface entropy in fragment production. We are pursuing the issue of infringement of our intellectual property rights with the said authors independently of the present review process.

Plagiarism has not been a widespread phenomenon within our community and typically occurs unintentionally. This is good, but it is highly disturbing in the case at hand, as it begs a question as to how come that the referee centers his/her criticism of our omission to cite work by authors who have plagiarized our prior works reporting not one, but four of our discoveries? Could this be pure coincidence?

In summary, we are renewing our call for accepting our manuscript for publication in Physical Review C without further delay, based on the following observations:

- 1. The third referee failed to dispute explicitly any of our replies to the comments by previous referees.
- 2. The third referee failed to identify the criticism by previous referees that he/she considers major ("main") and therefore warranting rejection of our manuscript.
- 3. The third referee failed to substantiate a claim that we failed to cite works which needed to be cited.
- 4. A relatively recent paper has reconfirmed our prior observation of negative heat capacity within the special microcanonical approach that is uniquely suited to the description of atomic nuclei "suspended" in vacuum.
- 5. That relatively recent paper tacitly acknowledges that there has existed no known interpretation of the "interesting feature" of negative heat capacity.
- 6. The third referee filed an unprofessional report that reads like a personal vendetta against the present authors. He/she clearly does not act as an impartial referee.
- 7. The Editors may disregard the recommendations by referees and use their own judgment based on the entirety of circumstances (points 1-6).

Furthermore, we ask the Editors to request that the third referee address each and every one of our replies to previous referees individually, as he/she was supposed to do as a fair

referee, to begin with. We need the full account by the referee to determine if we might have a cause for initiating legal action against this individual.

Now, regarding the statements in the referee's report, to which we are supposedly not allowed to reply, we state the following:

1. Boiling is a commonly known term and not an invention of ours, contrary to what the referee has erroneously suggested. On the other hand, in thermodynamics textbooks one would search in vain for the term "flashing point," advertised by the referee as referring to boiling. It suffices to Google the term "flashing point" to verify this observation.

2. Surface boiling is a completely new type of instability referring to the (integral) stability of an isotropic density profile but not to the differential stability of bulk matter. In finite nuclei, the latter instability still exists but appears at much higher excitation energies.

3. The argument that discovery of surface boiling is not worth publication because it is "nothing more" but the manifestation of the same instability in finite nuclei is unbecoming of a physical scientist. This argument is simply absurd. By the same token, one may argue that finite size effects need not be studied at all, even if they happen to explain hitherto unexplained phenomena (as is the case with our finding). By the same token one may argue that the discovery of element 119 would not be worth publishing because it is the same as lighter elements, except for a few protons. Publication of every new discovery can be suppressed based on such arguments, as most discoveries are incremental – they describe mostly known features, except for the novelty.

4. Our model is not new, but it is our model, dating back to the year of 2002. It is therefore already nine years old and has instigated research by others, as demonstrated by a number of publications by other authors. We don't see why employing this model further should diminish the value of our present discovery, which is what the referee seems to suggest.

5. The novelty of our finding does not consist in the realization that there is a limiting excitation energy - this was indeed known. Rather, the novelty is that a particular kind of hitherto unknown instability explains experimental observations to which no plausible explanation was previously known.

6. We did not invent microcanonical ensemble as a statistical ensemble at constant energy and volume – i.e., the "textbook" microcanonical ensemble, but neither did D.H.E Gross in his textbook. In fact, this ensemble has been used from the dawn of nuclear thermodynamics. What we did back in 2002, we introduced the microcanonical ensemble uniquely suited for studying the thermodynamics of self-contained systems at zero pressure.

13. There is no boiling in canonical and grand canonical systems. Ensemble equivalence applies to systems at equilibrium. Instabilities – thermal, chemical, or

mechanical appear in some but not other ensembles. And so, there cannot be thermal instability in canonical and grand canonical ensembles for a simple reason – the relevant partition functions do not depend there on energy.

- 14. The equations (10)-(12) refer to a configuration and they are correct. Same as in every published study of instability of nuclear matter. Same as in the two papers cited earlier. It is absolutely impossible to evaluate the system entropy and it is absurd to suggest summing up all the possible 10^100 configurations.
- 15. Microcanonical pressure is zero because it is given by the first derivative of entropy over volume at constant energy. This derivative must be zero at equilibrium (maximum entropy).
- 16. In calculations one can control anything one wants. This, however, is not what we stated.
- 17. In a sense we agree, that the appearance of negative heat capacity reflects the incompleteness of the model and we have stated this in our paper. This is illustrated by our entropy surface. As one increases excitation energy, fluctuations in excitation energy distribution (density distribution) increase such that a uniform distribution becomes a poorer and poorer approximation to the system entropy. And the equilibrium thermodynamics becomes inapplicable past the boiling point.

13. Follow-up letter to the Editors regarding the alleged plagiarism

<u>10/26</u>

Dear Editors,

In our previous communication, we unfortunately forgot to attach the copies of the two papers discussed in our answer. We do it now and urge you to compare the following:

The first paragraph of our Section II reads:

"The present study assumes that an excited nuclear system expands in a self-similar fashion so as to reach a state of approximate thermodynamic equilibrium, where the entropy *S* is maximal for the given total excitation energy *Etot* * , i.e.," ...

This was novel back in 2002, when we submitted the original paper and was a cause of 8-month delay in publication because of the corresponding referee's inability to appreciate the meaning of maximum entropy.

Now, please read what Samaddar et al. say on p.2, last paragraph of their section II:

"In essence, this procedure aims at maximizing the entropy with respect to the collective coordinate that describes the expansion of the hot system under the constraint of constant

excitation energy. Along the process, there is a transfer from the initial thermal energy of the system to the expansion energy in search of maximal entropy."

This is identical to our proposition, but the authors fail to cite our paper.

Now, please compare (i) our eq. 5 (another cause of a disagreement with our referees) with their identical eq. (19), (ii) our Fig.1 (showing thermal expansion – another source of disagreement with our referees) with "their" thermal expansion in Fig. 1, on p. 6. (iii) our Fig. 2 of a caloric curve featuring a domain of negative heat capacity with "their Figs. 5 and 6 showing an identical behavior.

Now, please read "their" characterization of the finding of negative heat capacity, on p. 8, last paragraph:

"The caloric curves show an interesting feature, namely, the occurrence of negative heat capacity, generally beyond an excitation

energy $E_*/A \sim 8$ MeV. Intuitively, one understands that if a system with

a given excitation expands, it does so at the cost of thermal energy and hence there may be a density region in which the temperature may decrease with increasing excitation if the system expands much in pursuit of maximum entropy."

There is nothing here about the effect being identical to a trivial spinodal instability, as claimed (erroneously) by the third referee. In addition, there is nothing about maximum temperature point being the "flashing point" (it is the boiling point according to our present understanding).

Our present manuscript offers a comprehensive explanation of what this behavior signifies in the case of bulk and finite matter. And this is stated in the title, abstract and throughout our manuscript. Yet, the third referee simply states that he/she does not see this explanation when he/she states that "... I do not see what this paper brings more than the cited work". Obviously, this individual does not understand our manuscript. This may be so because he/she lacks prerequisite familiarity with basic principles of thermodynamics and knowledge of the literature.

14. Letter from Editors regarding initiation of the appeal process

Re: CB10255

Surface boiling: A new type of instability of highly excited atomic nuclei

by J. T $\ensuremath{\sim}\ensuremath{\{0\}}\ensuremath{ke}$ and W. U. Schr $\"\{o\}\ensuremath{der}\$

Dear Dr. Schroder,

Thank you for your recent emails and the various attachments. I regret the delay in responding as I was away at the DNP Meeting.

I wish to confirm that we have received your emails. I thank you also for the copies of the published PRC papers, although a citation would have sufficed--we are the publishers and do have access to all papers published by the American Physical Society.

Aside from responding to the scientific concerns, you raise other concerns (alleged past plagiarism, general considerations about ethics in the editorial process, and a desire that the editors press one of the referees into responding to specific scientific issues). Because of the nature of your concerns, I will ask the editors to begin the appeal process right away.

However, it should be fairly obvious that we cannot transmit your recent letter to any of the referees, even if your appeal were granted--the manner in which you have written your response prohibits us from transmitting it beyond the editors or a member of the Editorial Board. Nevertheless, as mentioned above, we should not wait for a response from you that we can transmit.

Again, I will now ask the editors to begin the appeal process.

Yours sincerely,

Christopher Wesselborg Associate Editor Physical Review C Email: <u>prc@ridge.aps.org</u> Fax: 631-591-4141

15. Report by the Editorial Board Member

Report of the Editorial Board Member -- CB10255/Toke

I have read the paper, the reports of the three referees and the relevant correspondence. The authors have not made substantial changes in response to the criticisms made by the three referees. The process of review has been fair and appropriate .

Based on the reports of the referees and my own assessment, I recommend that this manuscript should not be published in Physical Review C.

Richard Seto Member, Physical Review C Editorial Board

16. Letter by the Authors to prof. Seto and the Attachment to it (sent 03/08/2012 and left unanswered by prof. Seto)

Dear Dr. Seto,

In the attachments we are sending you copies of a recent communication addressed to you, as a member of the Editorial Board, but sent via the editors of Physical Review C. The communication contained a letter to you, a history of the review of our manuscript so far, and references to invitations by the scientific advisory committees of two international conferences to one of us (J.T.) to present the work described in our manuscript.

We apologize if you have already received our letter and attached materials from the editors of Physical Review C.

Looking forward to hearing from you!

Sincerely,

W. U. Schroeder and J. Toke

The Attachment:

Dear Professor Seto,

We write you regarding your report on our manuscriptCB10255/TOKE, which we find quite troubling. This report tends to inflict damage to our reputation as scientists with 47 years of scientific and academic practice in the field of nuclear science, a long list of published papers in the field of nuclear thermodynamics, and a large number of lectures at conferences and workshop. It will also affect adversely our taxpayer-funded research program by further delaying the submission of papers discussing follow-up generalizations of our discoveries. Regarding the interest which the present findings have already generated in the nuclear science community, it suffices to mention that they have led to an invited lecture at the *International Workshop on Multifragmentation* in Caen (2011) and are subject of a further invited talk by J. Toke at the coming 11-th *International Conference on Nucleus-Nucleus Collisions* (San Antonio, TX, May 2012) - see, please, the attachment.

Importantly, and unfortunately, the said report is based on demonstrably false premises and must be then considered unfounded.

Please, consider this letter as being in lieu of a formal appeal, to which we are still entitled and consider the following material facts:

 Contrary to the claim in the first paragraph of your report, we have replied to each and every point of criticism raised by first two referees. We corrected some typos in the manuscript and added an immaterial, optional reference and a few optional statements.
 Not a single one of these replies has been disputed or refuted by the original referees or by the super-referee, in spite of our urgent requests for a transmission of any rebuttal addressed to the Editors.

3. There is no indication that the Editors have actually presented our replies to their respective addressees (the first and second referee) or to the super-referee. Please read the copy of our most recent communication to the editors regarding this point, you will find attached.

4. By any standards then, and certainly by legal standards, the criticism of our manuscript offered by the two first referees is "off the table" and must be tagged as fully resolved. And this is true regardless of the merits of the criticism and the actual merits of our replies. It certainly cannot serve now, to our surprise, as a basis for the rejection of our paper.

5. We were explicitly prohibited from responding to the "fusillade" of comments (all of which we found of dubious merits, some to the extent of being absurd) by the superreferee. Therefore, contrary to your assertion, these comments cannot serve as a basis for the rejection of our manuscript, either. Please, note that the super-referee does not dispute the validity and the adequacy of any of our replies to the first two referees, which would have been his statutory function in the review process. We find unacceptable such disregard by the super-referee of our explicit request for transmission of any response of the referees to our replies. One is led to conclude that rebuttals by the referees to our responses do not exist.

6. In spite of the prohibition by the Editors, we did reply to the most salient criticism by the third referee by pointing to the unfamiliarity of the referee with nuclear thermodynamics. We have heard of no rebuttal from him, either.

Furthermore, your assertion that a fair review process had been conducted appears again unfounded by a large margin. In our view, the process is in clear violation of the published rules of the PRC review process, as well as of rules of common sense. How can a review process be fair, in which our good-faith replies to the comments by the first referee remain unanswered, only to have the same sequence of events, or non-events, repeated in the case of the second referee appointed by the Editors? Then, having replied in good faith to the new comments by the second referee, again with no response, we had to see a super-referee appointed who gave us yet another batch of comments, to which we were not allowed to reply. We have received no communication from any referee that has addressed the substance of our responses, let alone a valid rebuttal. To us the process is in fact undistinguishable from one, in which the Editors simply disregarded our replies.

For your information, we attach a file documenting the history of the review process including, not only the criticism offered by the referees, but also our replies. You may wish to confirm with the Editors that these are, indeed, copies of the pertinent documents.

In summary, you may wish to reconsider your report, retract it and file a new report that would be consistent with the facts of the case. The most pertinent fact in this case is that there is no outstanding criticism on the table, that after 12 month worth of editorial process.

17. Letter from the Editors re: Letter to prof. Seto

Date: 05Mar2012-12:05:42 From: prc@aps.org Subject: Your_manuscript CB10255 Toke To: schroeder@chem.rochester.edu

Re: CB10255
Surface boiling: A new type of instability of highly excited atomic
nuclei
by J. Toke and W. U. Schroeder

Dear Dr. Schroder,

We have received your recent emails. I confirm that it contains a significant duplication of material that is already on record. For example, the list of events you obtained from the automatic manuscript information server is a direct extract of relevant events from our editorial database. It cannot contain additional information.

I will bring the additional voluminous material to the attention of Editor Dr. Gibson and Associate Editor Dr. Gibbs. If they have further information, then we will contact you again.

You wondered whether Dr. Seto, in his capacity as an Editorial Board of PRC, had received certain previous referee reports and your responses to it. I have just checked, and we sent Dr. Seto 235 pdf pages of editorial correspondence, in the form of referee reports and author responses, including duplicate previous material embedded by the parties via their email "reply" feature, referee responses to our reminder emails, and additional correspondence with the authors and their responses.

Our system of tracking previous correspondence is quite sophisticated. Creating a pdf package of previous correspondence is a meticulous process. As a matter of principle, we prefer to include everything short of automatic email acknowledgments.

As to the editorial process itself the published editorial policies state the following (see http://prc.aps.org/info/polprocc.html#appeal):

"The author of a paper that has been rejected subsequent to an Editorial Board review may request that the case be reviewed by the Editor in Chief of the APS. This request should be addressed to the editors, who will forward the entire file to the Editor in Chief. Such an appeal must be based on the fairness of the procedures followed, and must not be a request for another scientific review..."

Please state clearly whether or not you had intended to appeal to the Editor in Chief. Your previous correspondence will be available (*).

We are holding your manuscript in this office awaiting your response.

Yours sincerely,

Christopher Wesselborg Associate Editor Physical Review C Email: prc@ridge.aps.org Fax: 631-591-4141 http://prc.aps.org/