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Reply to the comment on direct experimental evidence for a negative heat capacity in the liquid to gas phase transition in hydrogen cluster ions backbending of the caloric curve

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Reply to the Comment on "Direct experimental evidence for a negative heat capacity in the liquid-to-gas phase transition in hydrogen cluster ions: backbending of the caloric curve".

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This reply aims to clarify a key argument in two recent publications [1, 2] which has been criticized in the Comment of Chabot and Wohrer. The articles in question feature a novel method to derive a caloric curve from event-by-event ion decay data measured using multi-coincidence techniques after high-energy collisions (60 keV/amu) between mass selected hydrogen cluster ions ($H_3^+(H_2)_n, n = 6, 14$) and a helium target. Reactions corresponding to a given deposited energy Ed are selected and grouped into statistical sub-ensembles. A caloric curve can then be constructed by deriving corresponding temperatures for these microcanonical cluster ensembles. It is this method which is criticized by Chabot and Wohrer.

The essence of their criticism is expressed in the following extract from the Comment: "$\cdots$The authors assume that the internal energy E* is the energy deposited in the cluster by the collision, Ed, $\cdots$ The deposited energy Ed is the sum of the energy due to electronic excitation and, as a dominant contribution in their systems, to ionization. But the ionization energy should not be included in E*." Apparently, Chabot and Wohrer have misinterpreted the work; arguing along a single event consideration and not taking into account the statistical ensemble type situation which applies.

As explained in detail in [1, 2], the temperature derived from the data is that of a statistical ensemble comprising a large number of decaying $H_3^+(H_2)_n$ ions. Each sub-ensemble is characterized by the deposited energy and includes all of the processes induced by the high-energy collision (ionization, excitation, etc). This energy is deposited during a very short time (0.1 fs) and the system (n protons and n-1 electrons) is left isolated immediately after the collision. Therefore, the subsequent statistical analysis is carried out in the microcanonical frame [3, 4]. The results obtained are averages for each statistical ensemble comprising a large number of cluster ions with the same total amount of energy deposited, albeit distributed in different channels. The total energy (in the frame of reference of a single cluster ion) is equal to the sum of the internal energy before the collision and the energy deposited by the collision. It can be assumed that the internal energy before the collision is low in comparison with the deposited energy.

The temperature is derived from one observable; the size distribution of the largest fragment among residual cluster ions in the statistical ensemble. The residual cluster sizes are measured not only after the ejection of electrons (ionization) but also following the ejection of ions, atoms and molecules. The correlation between the temperature and the size distribution of the largest fragment has been previously demonstrated both in cluster physics [5] and in nuclear physics [6, 7]). Moreover, regarding the recent work of Thirring and co-workers [8](triggered by [1, 2]), it is worth noting that our measurements and analysis were carried out on a large number of cluster ions prepared at the same total energy, as opposed to observing the evolution of a single system over time (time averaged ergodic hypothesis). The key point being that, for each different sub-ensemble considered, we have explored all the available phase space. Finally, maintaining their non-statistical approach, Wohrer and Chabot argue that the fragmentation events should be grouped according to the internal energy of the multiply charged cluster ions after the electrons have been ejected. Besides not taking into account the many reactions where only excitation occurs (and no ionization), energy thus considered, far from equaling the total energy in the cluster ions, represents only the energy of a part of the system corresponding to an arbitrarily chosen intermediate state of the reaction process.

In conclusion, we cannot agree at all with the arguments given by Chabot and Wohrer. Besides containing some misleading and wrong statements, such as "but the number of neutral fragments as well as their individual masses are not identified experimentally" (the methods by which we were able to identify the neutral fragments are clearly stated in [1, 2] and references therein), their main point of criticism is based on a misconception of how to derive a temperature for a microcanonical ensemble of decaying cluster ions.

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[6] M. Belkacem et al., Phys. Rev., **C52** 271 (1995)
[7] J. Pochodzolla et al., Phys. Rev. Lett. **75** 1040 (1995) (see also references 15 et 16 in reference 2)
[8] W. Thirring et al., Phys Rev Lett. **91** 130601-1(2003)