Title	Comment on "Preserving the Boltzmann ensemble in replica-exchange molecular dynamics" [J. Chem. Phys.129, 164112 (2008)]
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Note	

EPAPS supplementary information for the JCP article "Comment on *Preserving the Boltzmann ensemble in replica-exchange molecular dynamics*"

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Note: A number in brackets in the section name designates the reference number indicated in the article.

A [3]. Simple overview of the criticisms raised in Reference 1

One set of questions raised in Ref. [1] is about the probabilistic description relevant to the Berendsen method [H. J. C. Berendsen et al., J. Chem. Phys. **81**, 3684 (1984)] and the behavior of the Nosé-Hoover (NH) equation in the case of a small system. Another set of queries pertains to general theoretical aspects of NH and related methods. The first set of questions has been discussed in many papers; these papers discussed solutions to the NH equation and the effectiveness of the NH and Berendsen methods. Therefore, in this study, I address the second set of queries.

B [11]. Technical terms in ergodic theory

Information on the following technical terms in ergodic theory is supplemented to make them more understandable.

- **Measure Preserving** (MP) [in Point (I)]: MP, compared with the term "ergodicity" itself, has not been stressed and may not be familiar in molecular dynamics studies. MP property $P(T^{-1}A) = P(A)$ says that by a transformation T (the inverse sign here is not essential in physics) every set A in phase space (PS) usually changes its shape but exactly retains its measure. Here the measure in the NH case is a volume endowed with a weight by the density ρ, viz. $P(A) = \int_A \rho dq dp d\zeta$. An interpretation of MP not by using a set in PS but by using simply a single trajectory in PS is also possible via, e.g., Eq. (2), which is a local expression of MP, indicating that the density change along the trajectory is evaluated by the Jacobian.

- **Invariant Set** [in *Point (II)* section]: A subset A is an invariant set if it confines every trajectory with an initial point in A. Ergodicity holds if any invariant set is trivial, i.e., either the entire set Ω or the empty set, by neglecting a subset of zero measure.

- **Condition B** [in *Point (II)* section] can be paraphrased into the following: A part that has a positive measure (i.e., that cannot be neglected) in B always reaches A after a certain time, even if the sets A and B are far apart at the beginning, as long as they have positive measures. Here we have imagined the evolution of only B while A to be fixed.

C [13]. Additional comments to the discussion in Reference 1

To constitute an invariant function for numerical checks, many numerical integrators (NIs) have been constructed not on Ω but on an extended space (e.g., $\Omega \times \mathbb{R}$), and MP is considered in this context; MP on $(\Omega, \mathcal{M}, P)$ for NI is ensured at least approximately in the sense that NIs are approximations of the exact flow, which is MP. The only point on which this paper agrees with Ref. [1] may be the importance of considering a NI that is exactly MP on $(\Omega, \mathcal{M}, P)$, although the discussion in Ref. [1] needs to be modified as pointed out below.

First, MP and volume preserving (VP) are not equivalent properties in general (they agree in a specific case where the measure has a unit density, as in a Hamiltonian system described in a whole PS). Second, considering VP in a space $\{(q, p)\}$, which is a projective space from the original PS $\{(q, p, \zeta)\}$, is meaningless (particularly from a global viewpoint), since each projective trajectory generally crosses itself and hence it no longer defines deterministic dynamics.

D [14]. Conclusive remark on point (III)

In summary, Lemma 5.4 (Ref. [1]) should be reconsidered. Its proof does not give a meaningful result if it implies (non)ergodicity on the whole PS with the Lebesgue measure. To make sense of the lemma, I have taken into account other possibilities in terms of the choice of measure, the use of a map (instead of a flow), and the set of assumptions. However, the lemma cannot be proved in a meaningful sense on the basis of the discussion in Ref. [1].

E [15]. Additional remark on the ergodicity

The current comment does not treat a conservation law, e.g., for (zero) total momentum under the assumption of zero total force, since Ref. [1] does not treat it. Although further discussions are needed when such an assumption is made, in general, if meaningful invariant sets are produced, e.g., by adding certain assumptions, it is natural to consider the ergodicity on each invariant set, provided that such a set and the induced invariant measure can be explicitly defined.

[1] B. Cooke and S. C. Schmidler, J. Chem. Phys. **129**, 164112 (2008).