Thermal properties and dynamics of water cluster anions

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Thermal properties and dynamics of water cluster anions

Overview and Plans

Albert-Ludwigs-Universität Freiburg

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A. Piechaczek, C. Hock, C. Willibald, M. Hejduk, B. v. Issendorff

Clustertreffen 2015

Table of contents

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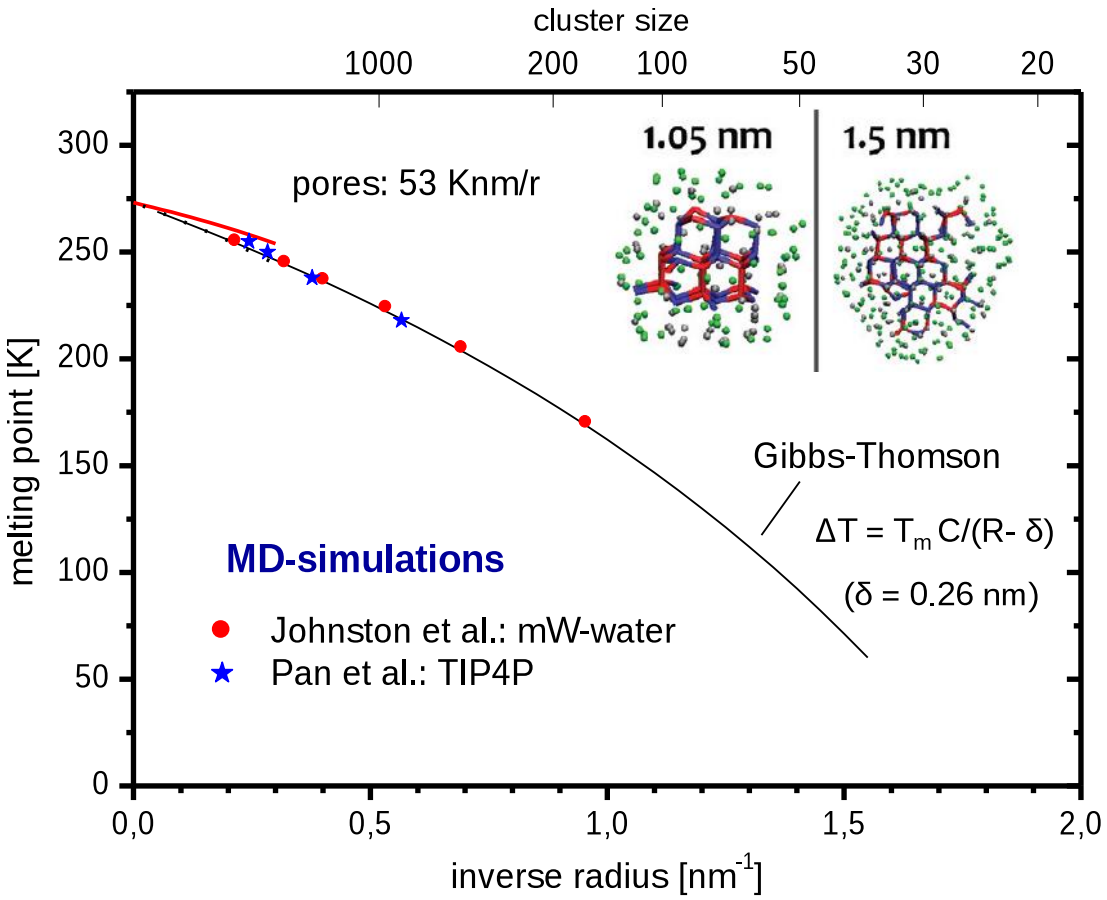
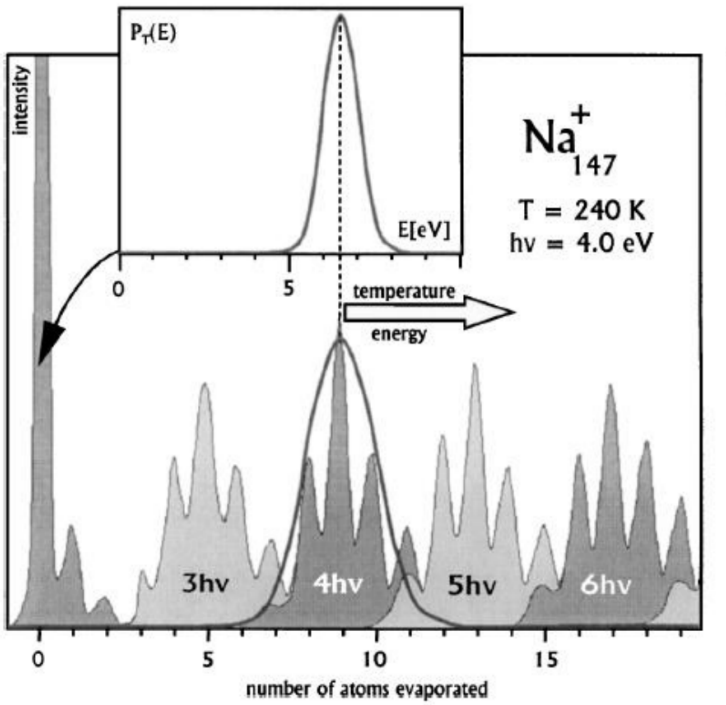
Prediction for neutral water clusters



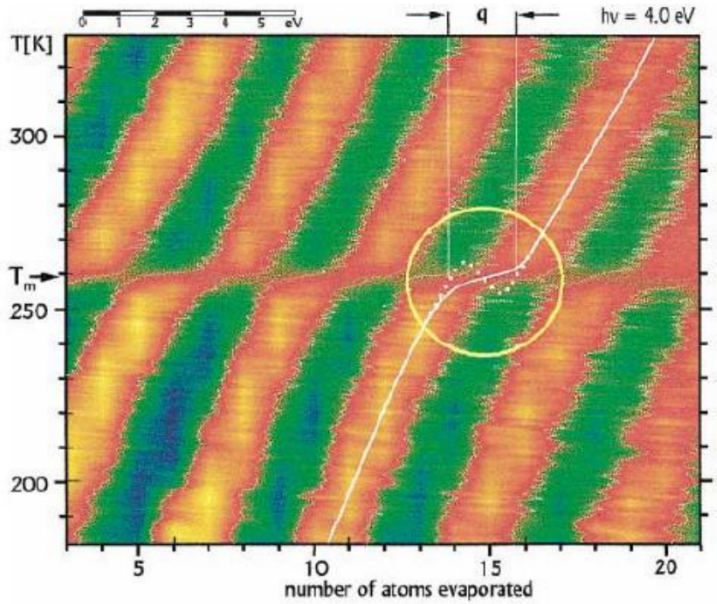
J.C. Johnston and V. Molinero, JACS 134, 6650 (2012)

D. Pan, L.-M.Liu, B. Slater, A. Michaelides, and E. Wang, ACS Nano 5, 4562 (2011)

Calorimetry



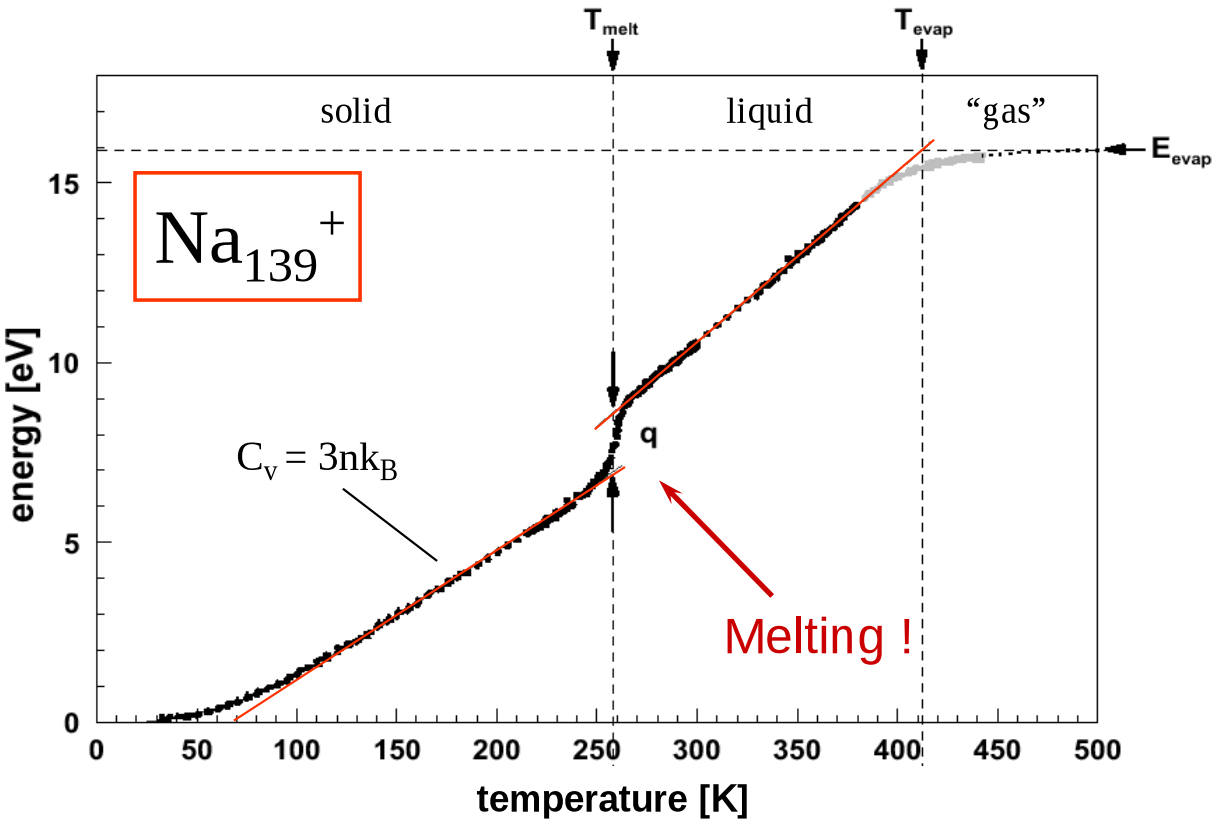
Photofragment distribution



Temperature dependence

PRL 79, 99 (1997)

Example: Free sodium clusters



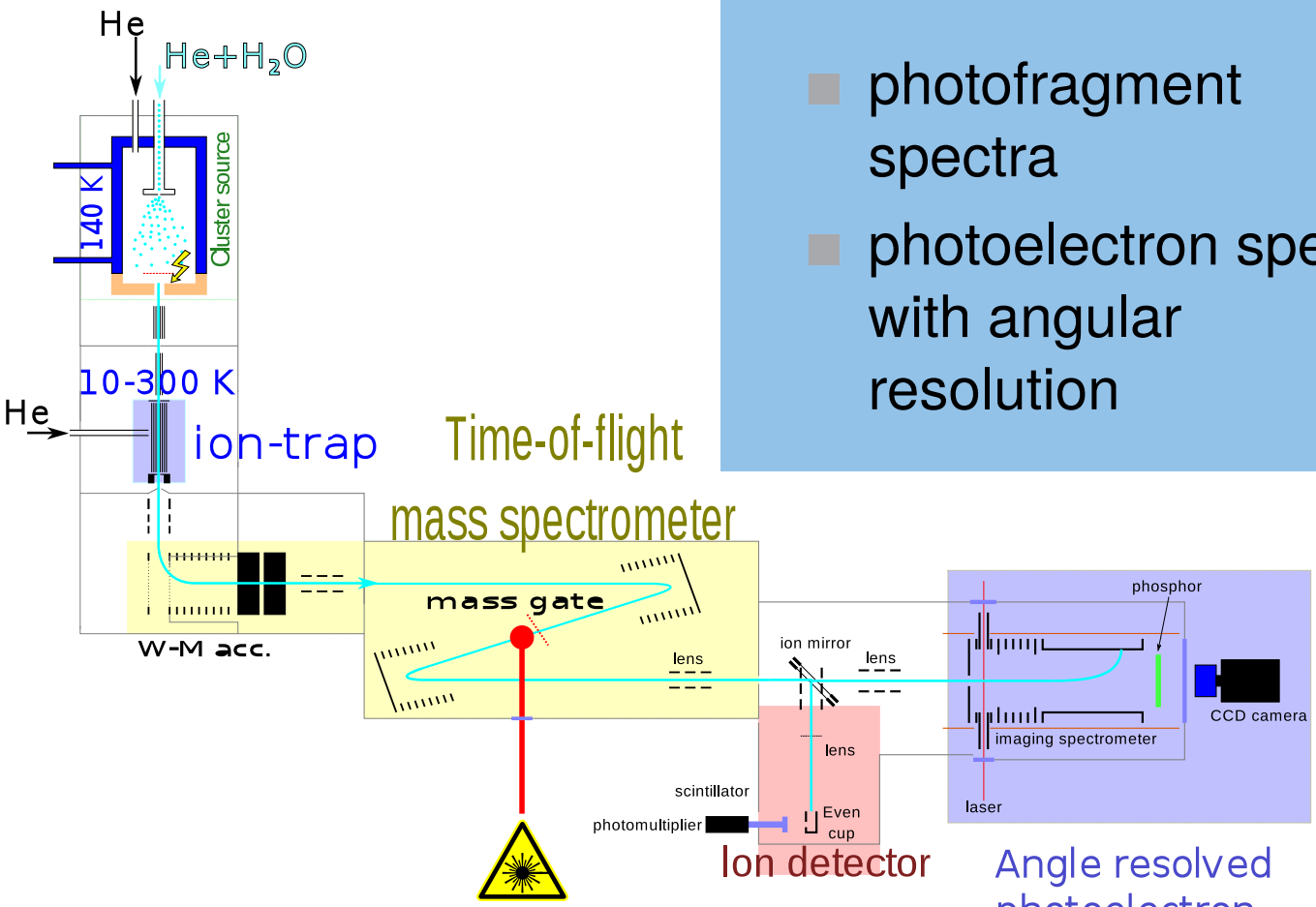
PRL 79, 99 (1997)

Nature 393, 238 (1998)

Experimental setup

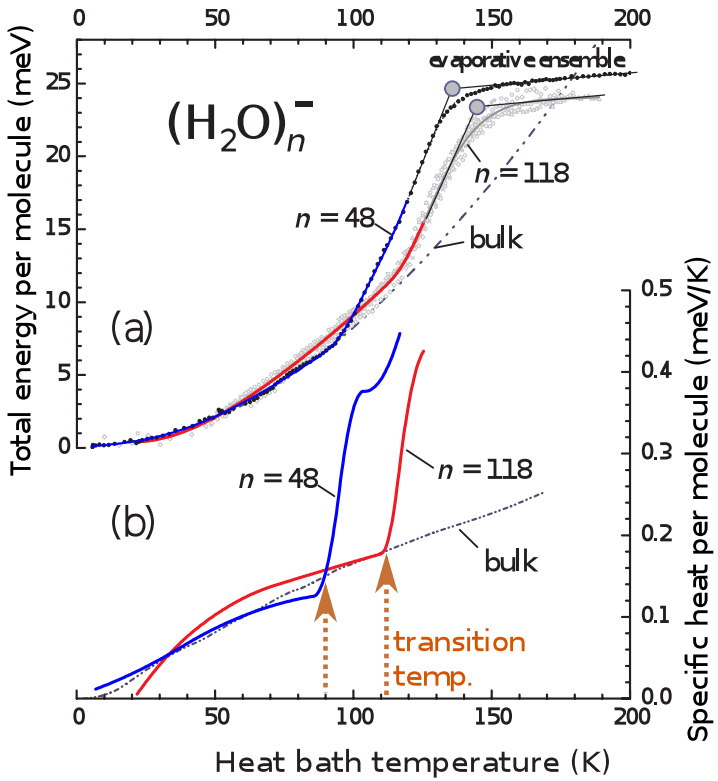
What we obtain

- photofragment spectra
- photoelectron spectra with angular resolution



Calory curves of water cluster anions

Thermal energy



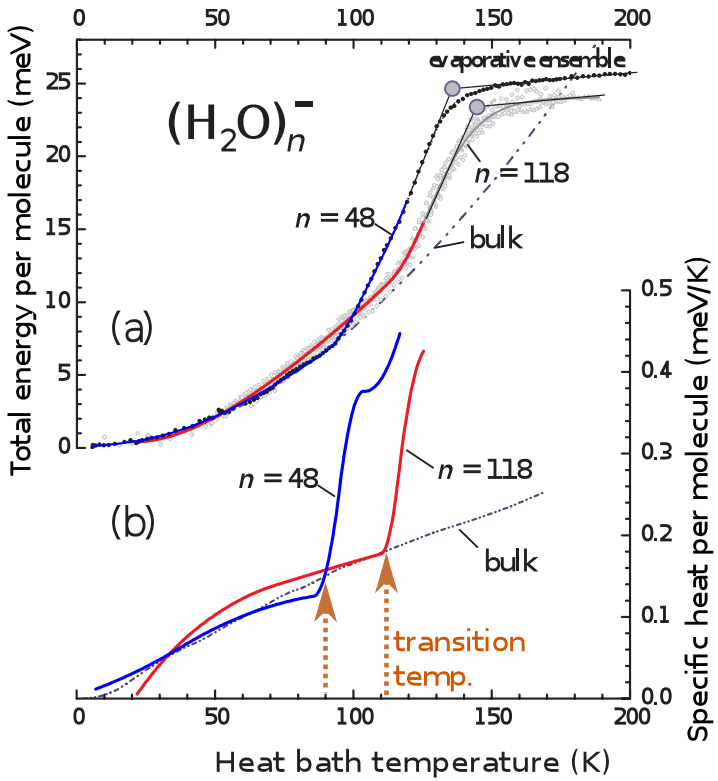
Heat capacity

Phys. Rev. Lett., 103, 073401

(2009)

Calory curves of water cluster anions

Thermal energy



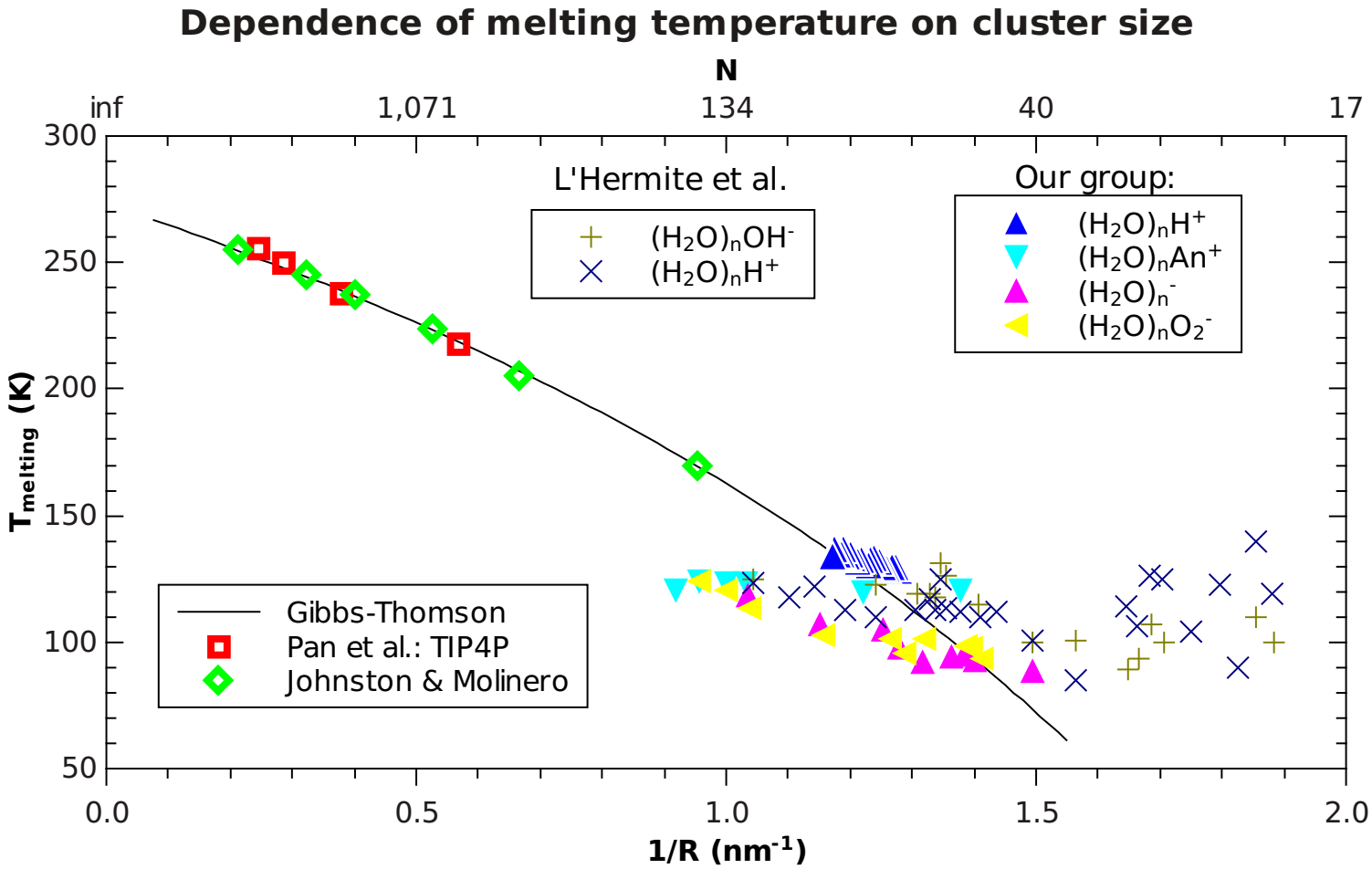
Heat capacity

Phys. Rev. Lett., 103, 073401

(2009)

- No sharp transition
- Transition temperature varies with size

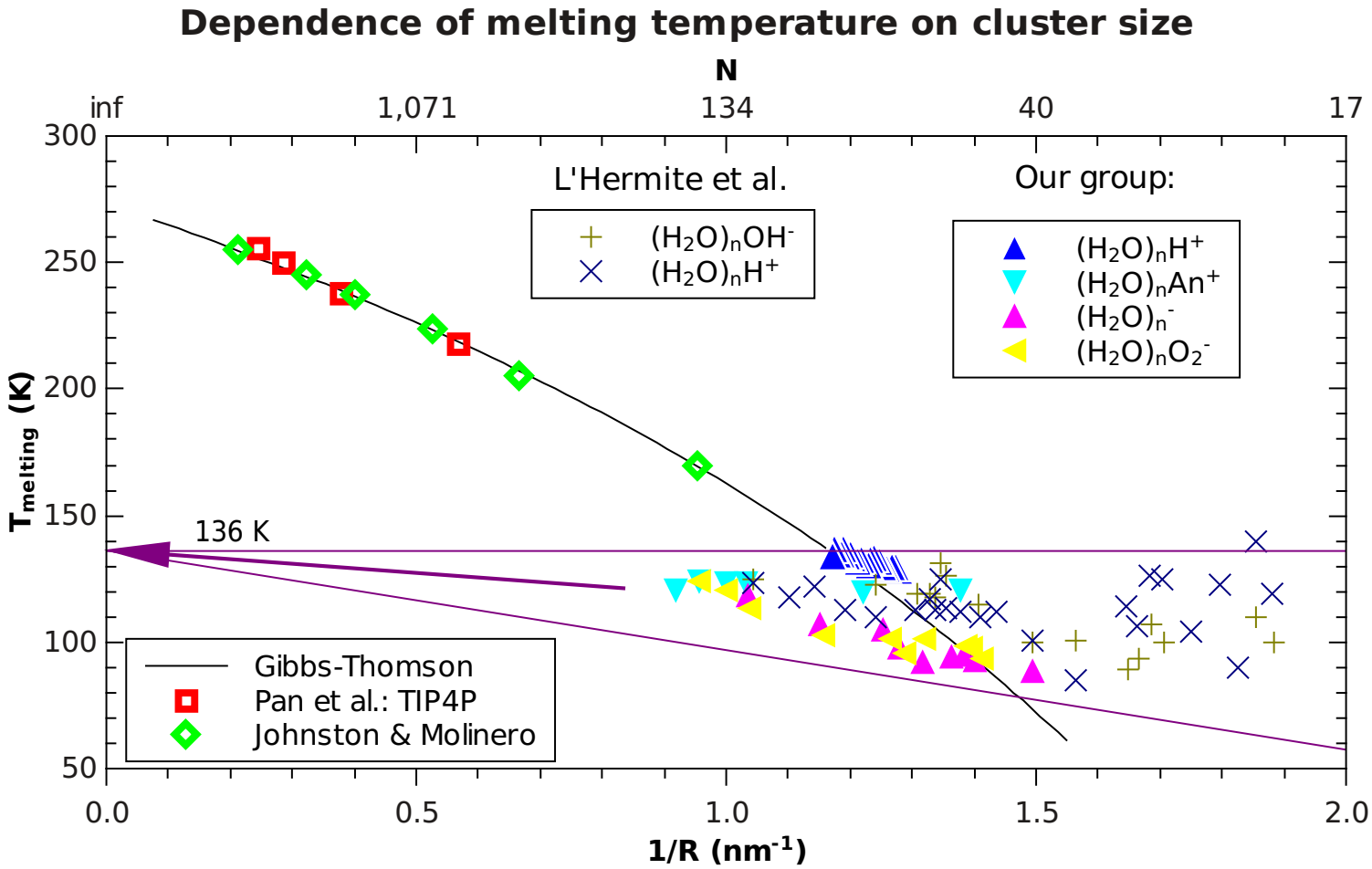
Dependence of transition temperature on cluster size (various dopants)



Julien Boulon, Isabelle Braud, Sébastien Zamith, Pierre Labastie, and Jean-Marc L'Hermite JCP 140, 164305 (2014)

M. Schmidt and B. von Issendorff, JCP 136, 164307 (2012)

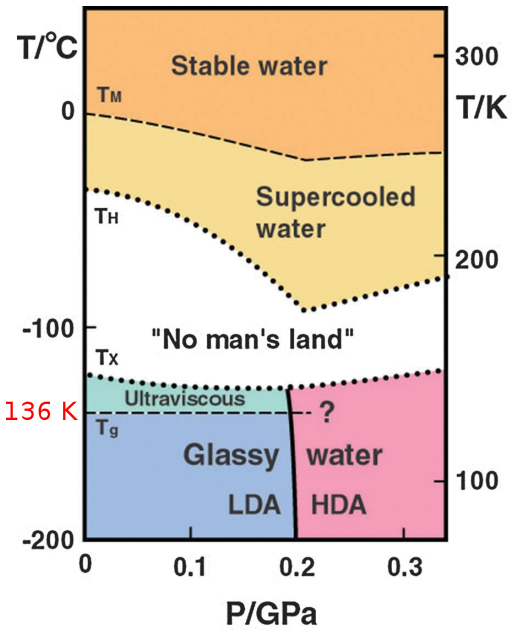
Dependence of transition temperature on cluster size (various dopants)



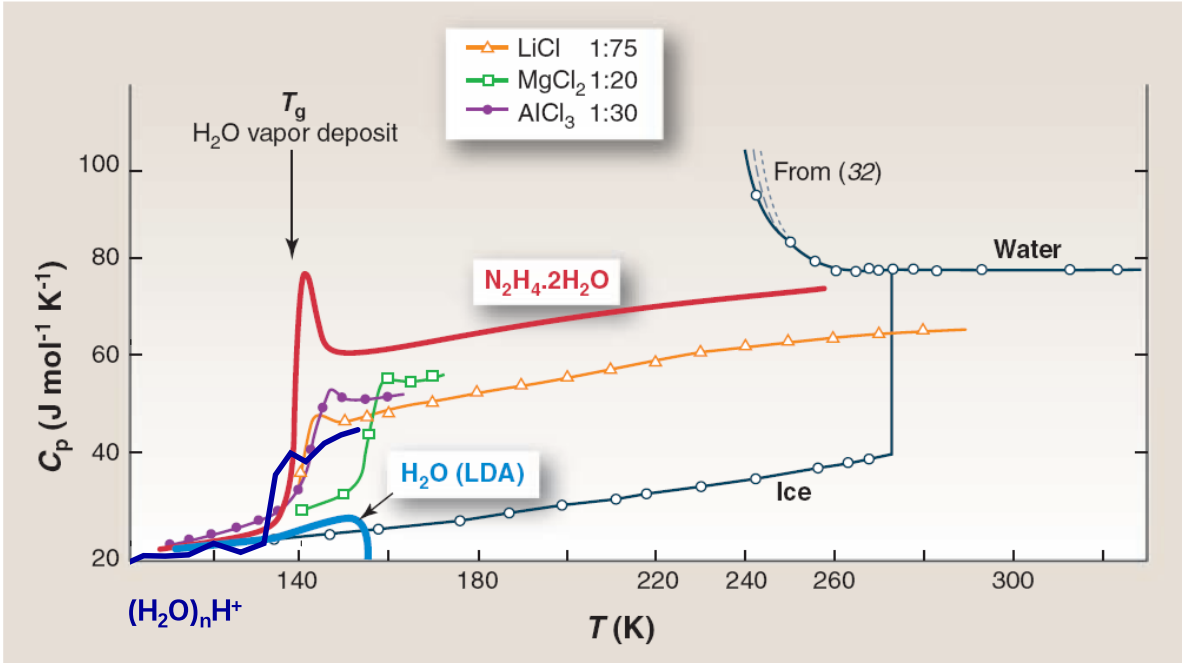
Julien Boulon, Isabelle Braud, Sébastien Zamith, Pierre Labastie, and Jean-Marc L'Hermite JCP 140, 164305 (2014)

M. Schmidt and B. von Issendorff, JCP 136, 164307 (2012)

Amorphous ice and heat capacity

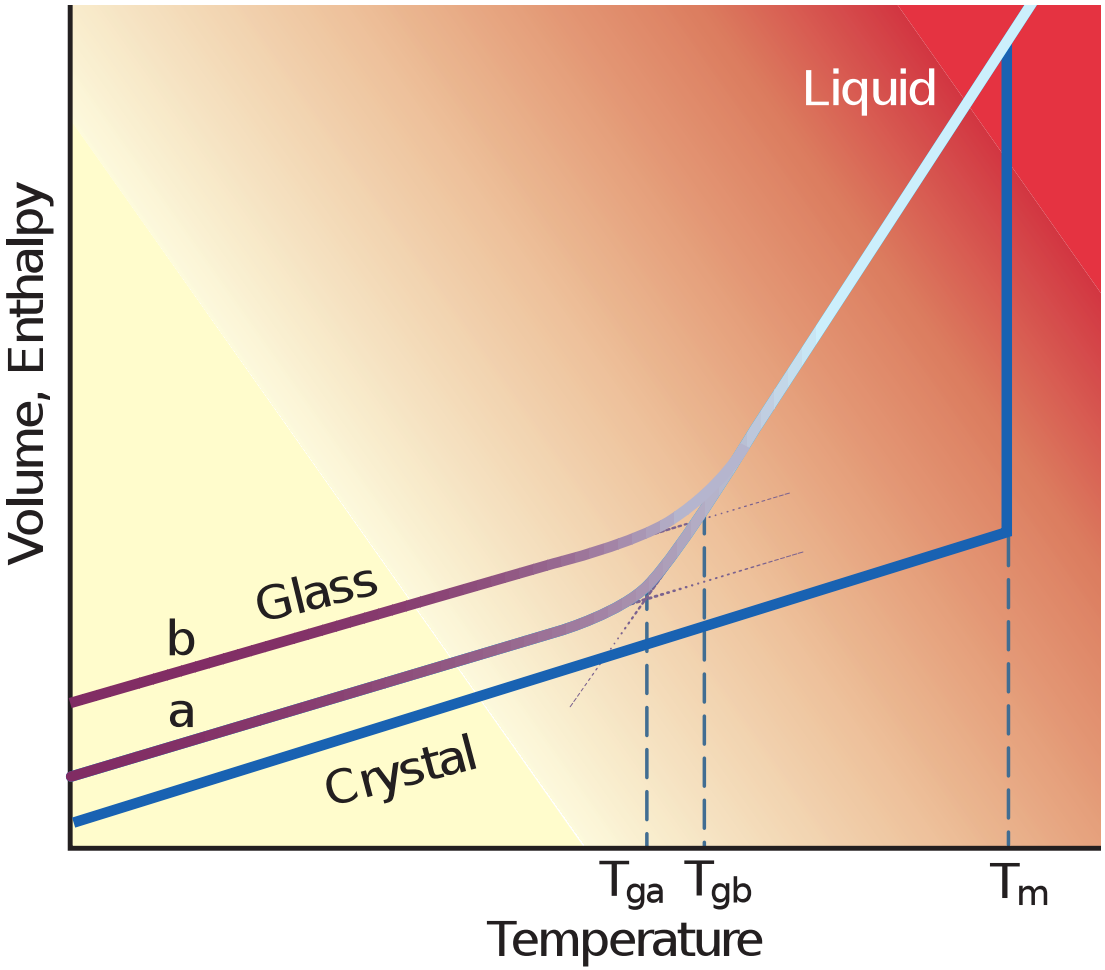


Loerting, PCCP 13, 8783 (2011)



Angell, Science 319, 582 (2008)

Glass transition

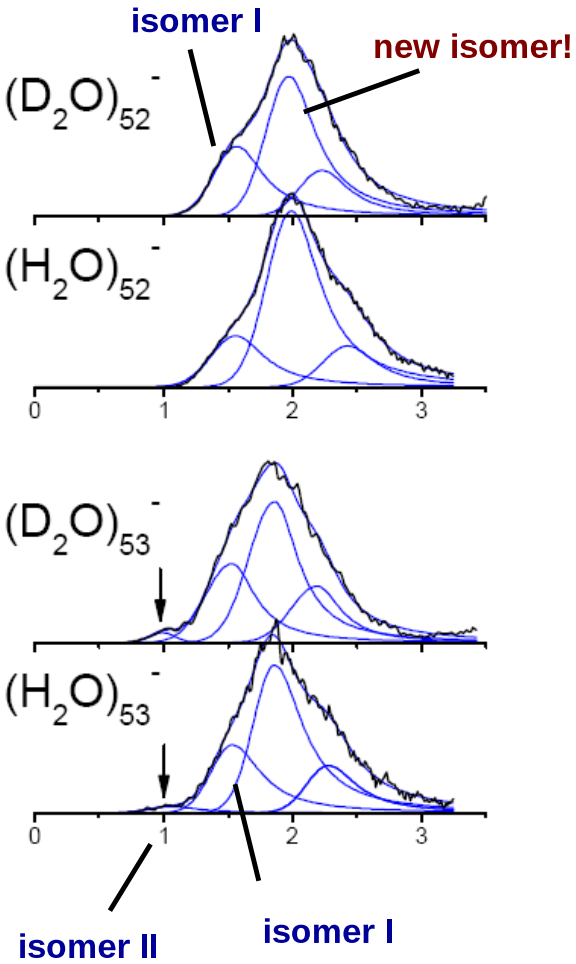


P. G. Debenedetti and F. H. Stillinger, Nature 410, 259-267 (2001).

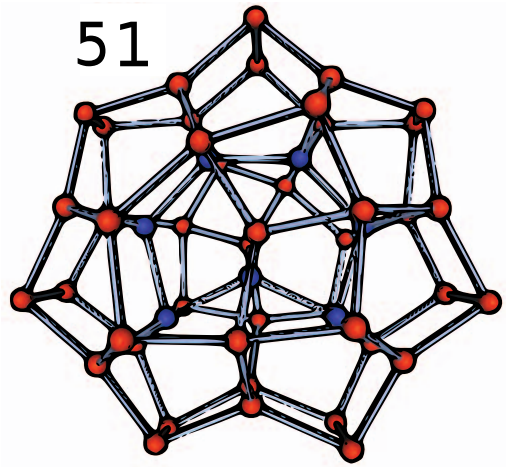
Glass transition

- infinite number of noncrystalline solid structures of different energy
- which structure is adopted depends on cooling rate

Indication of glass transition I: Isomers



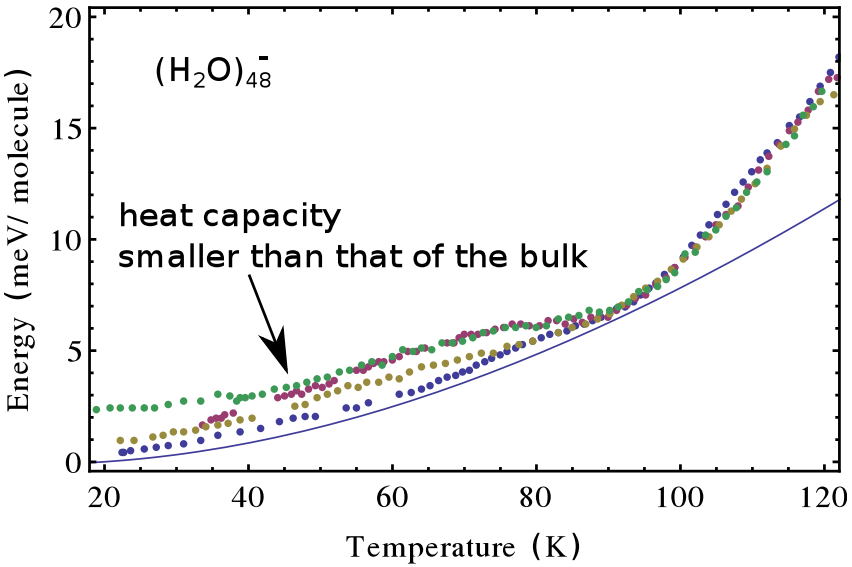
photoelectronspectra, isomer groups



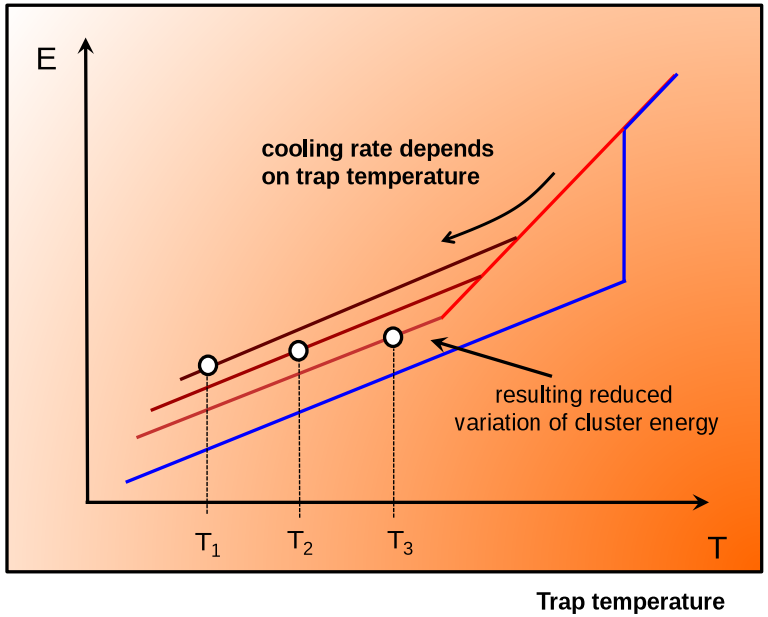
Sergey Kazachenko & Ajit J. Thakkar, JCP. 138, 194302 (2013)

Magic size 51: one way how to organize oxygen atoms, billion ways for hydrogen atoms

Indication of glass transition II: nonergodicity



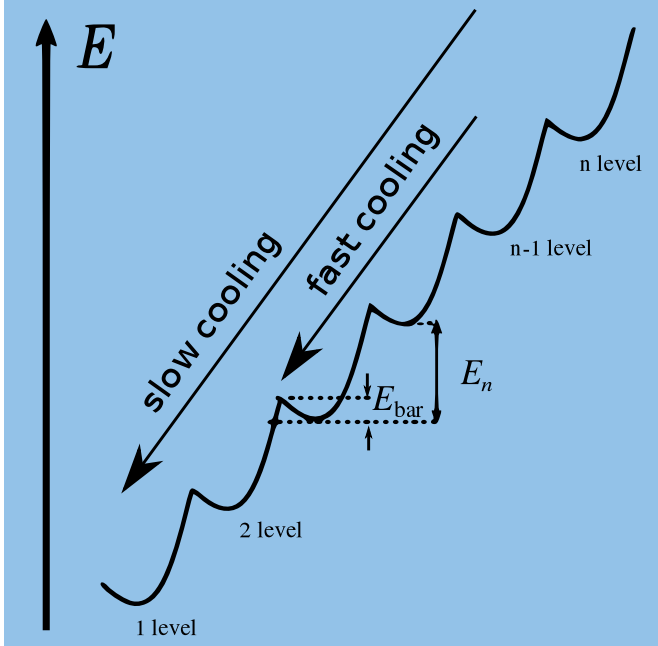
Different caloric curves on different days. Different cooling rates?



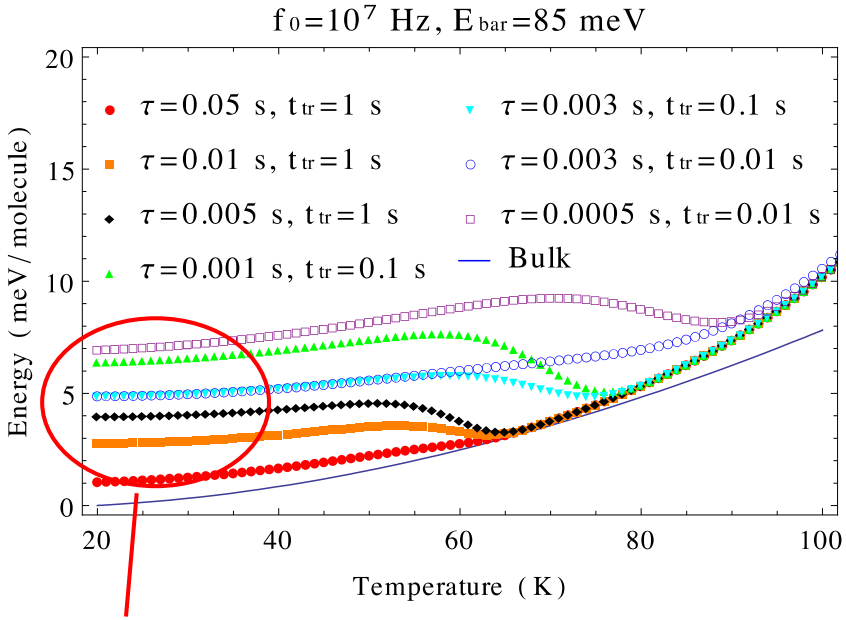
The cluster ends up in different isomer at different trap temperatures.

Cluster cooling model

Multilevel model



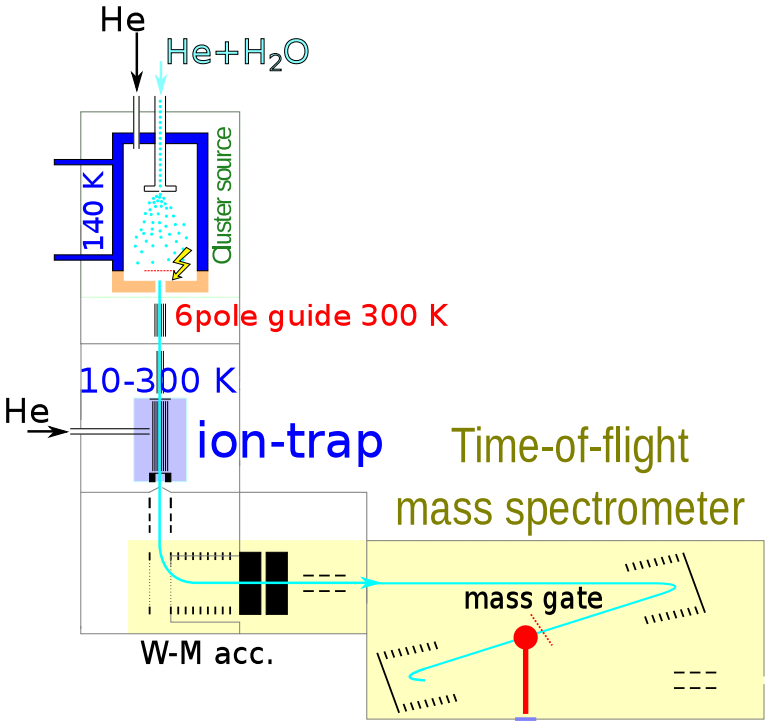
Thermalization scenarios

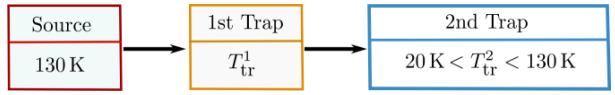


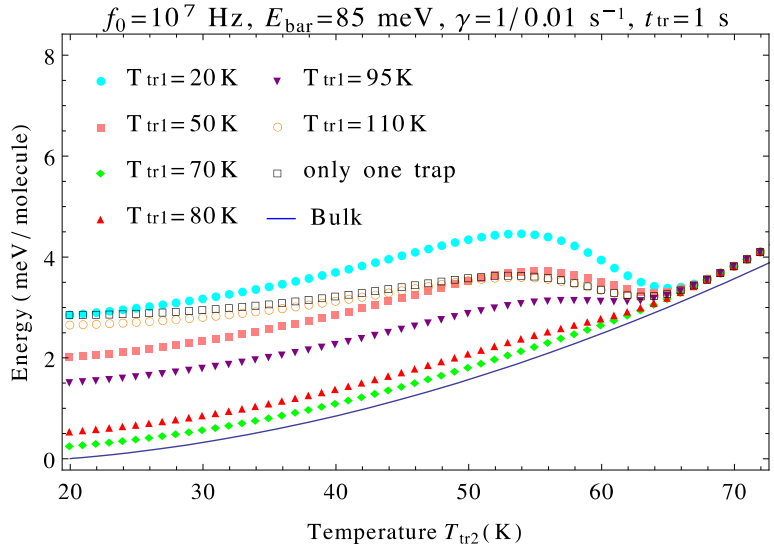
vanishing heat capacity!

τ: characteristic time of thermalization; t_{tr}: trapping time

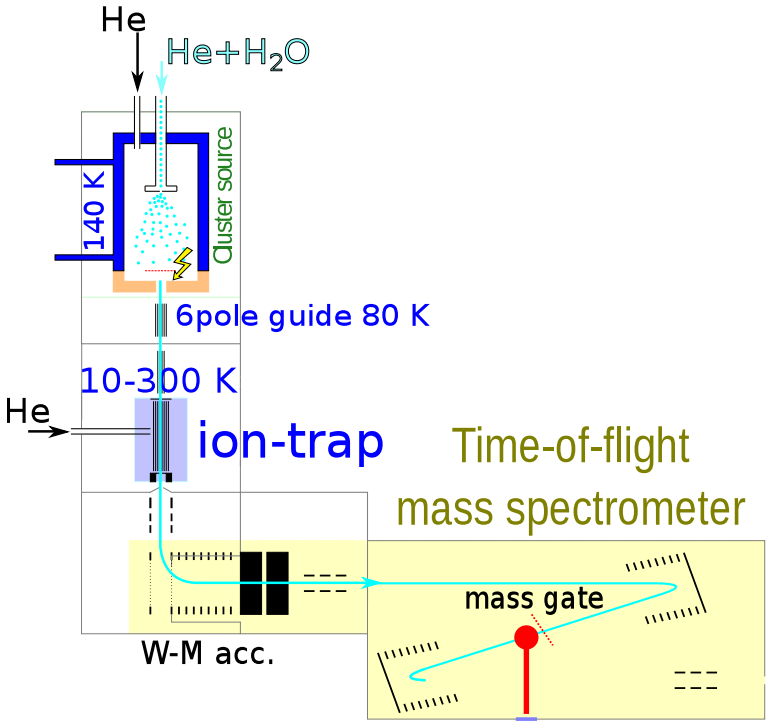
Precooling measure

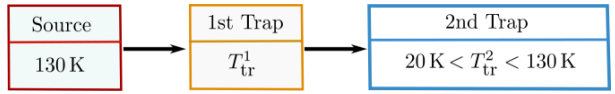


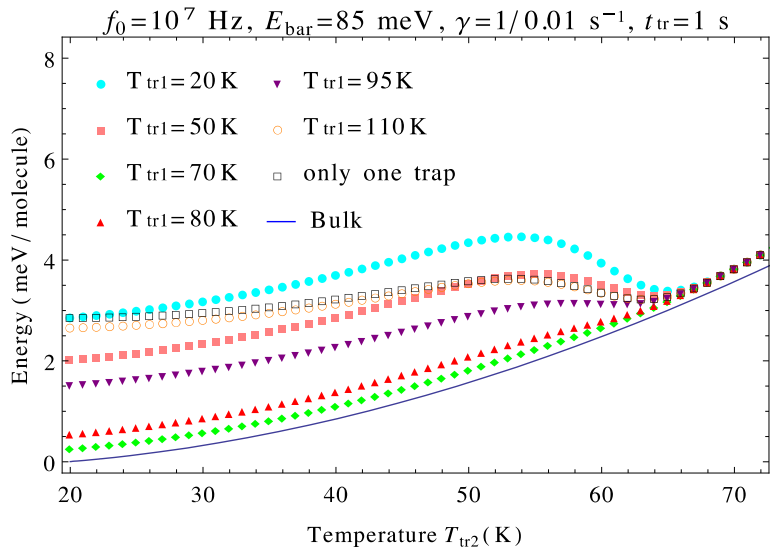




Precooling measure

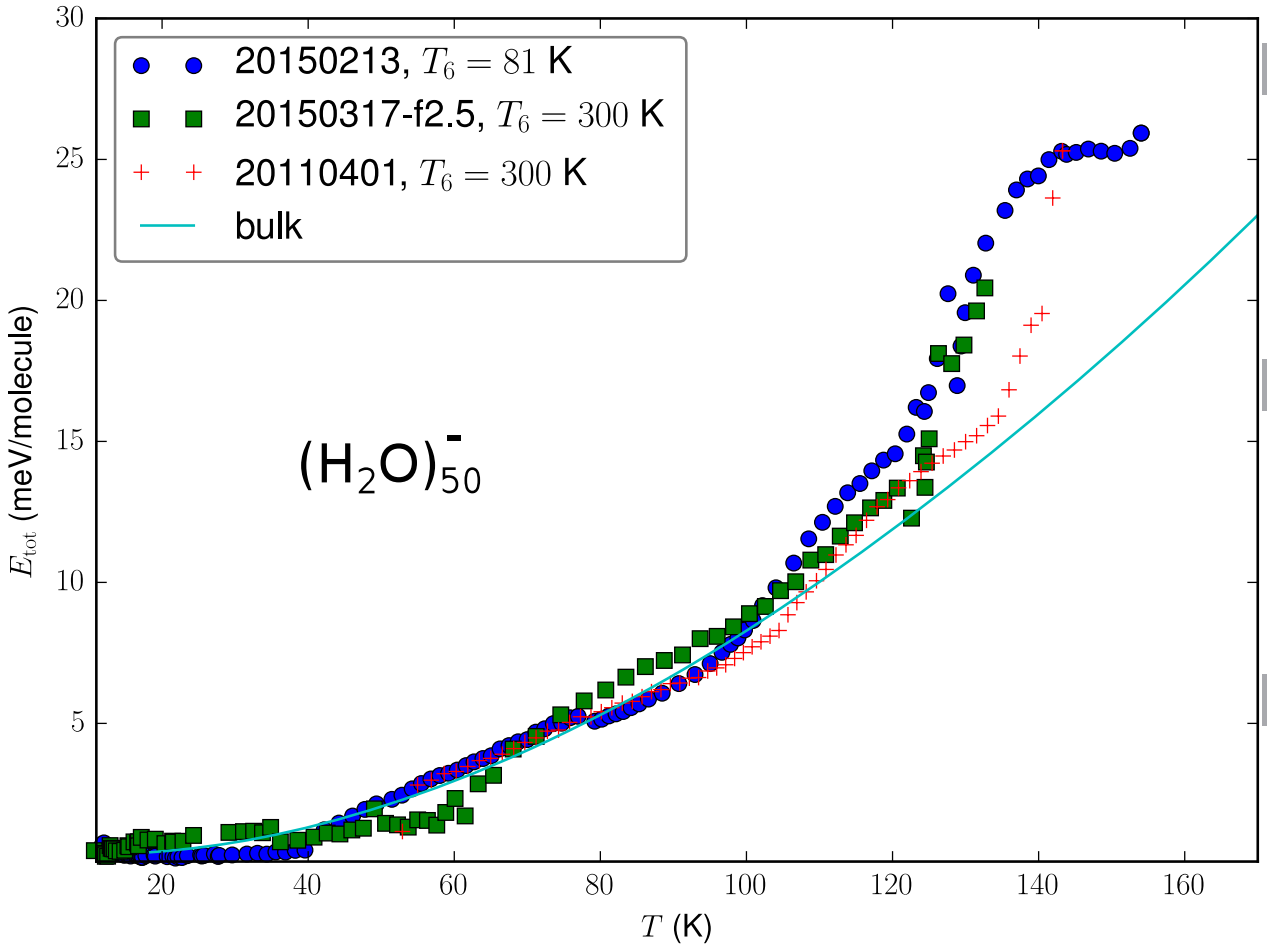






Preliminary results

Caloric curves for different temperatures of the 6pole guide

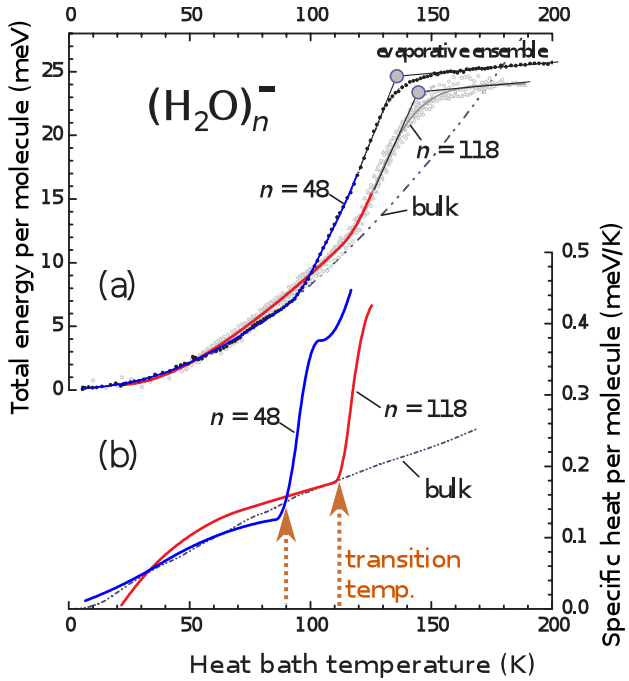


1. No significant difference between two caloric curves
2. Not enough collisions cluster↔6pole gas?
3. Trapping in 6pole needed.

Summary

1. Water cluster anions/cations undergo “softening” transition.

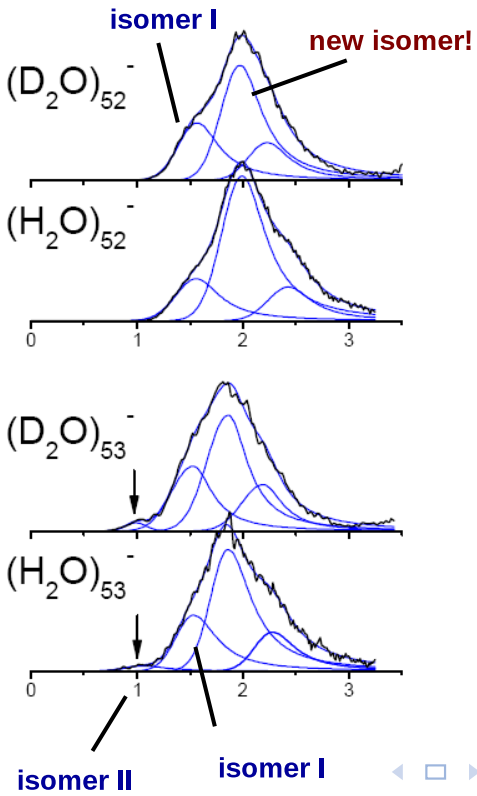
Thermal energy



Heat capacity

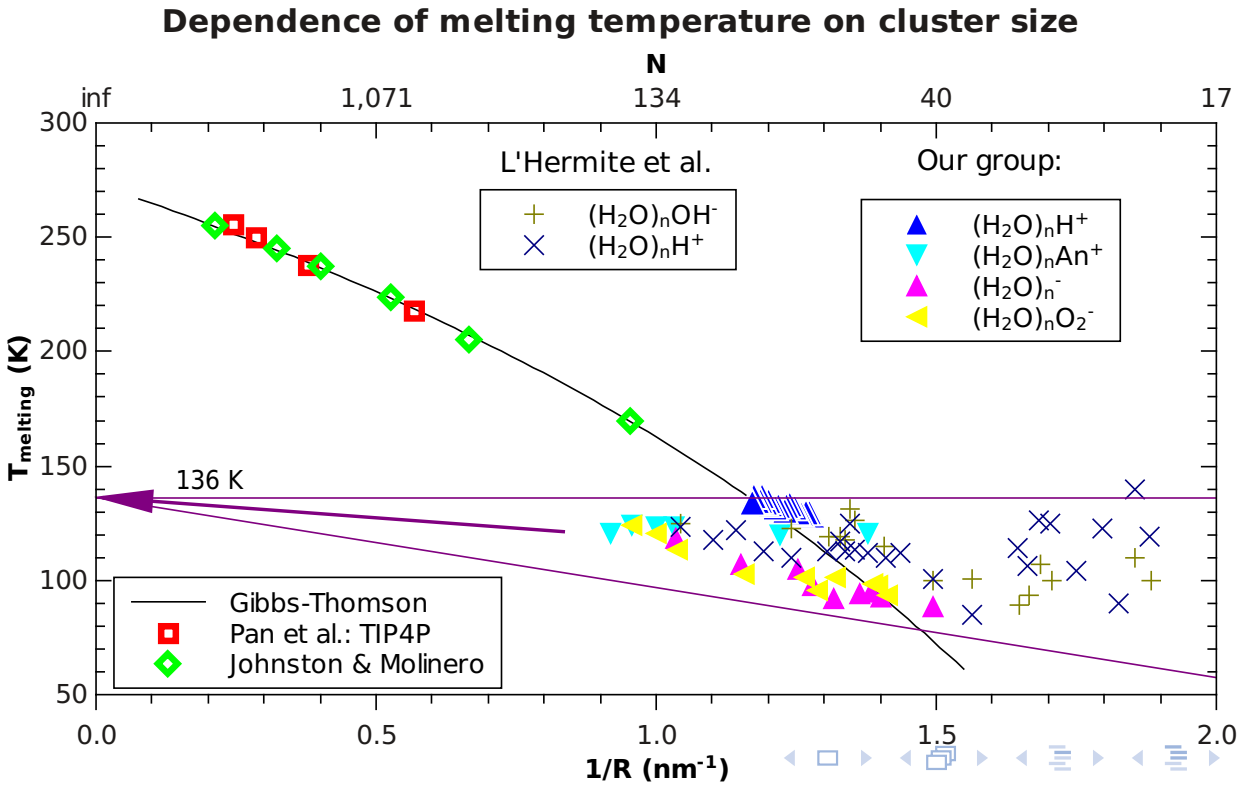
Summary

1. Water cluster anions/cations undergo “softening” transition.
2. Water clusters exist in many isomers.



Summary

1. Water cluster anions/cations undergo “softening” transition.
2. Water clusters exist in many isomers.
3. Size dependence of the transition temperature converges towards bulk glass transition temperature.



Summary

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1. Water cluster anions/cations undergo “softening” transition.
2. Water clusters exist in many isomers.
3. Size dependence of the transition temperature converges towards bulk glass transition temperature.

Hypothesis

Small clusters are glass like and undergo glass transition.

Future

Confirmation/refutation of multi-step cooling model by experiment