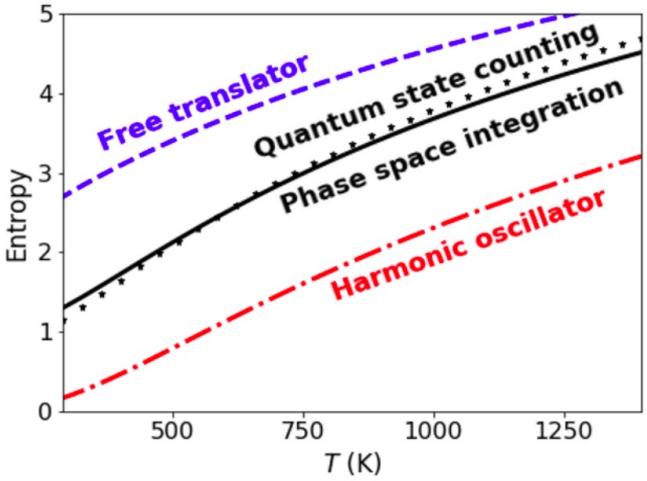
More Accurate Approach for Adsorbate Thermophysical Properties

Scientific Achievement

We found that contributions of translational anharmonicity are significant for H adsorbed on a Cu(111) surface, by computing thermodynamic properties via a phase space integration (PSI) approach. The approach is in excellent agreement with a quantum state counting benchmark over the temperature range of interest, especially compared to the most commonly used models, the harmonic oscillator (HO) and the free translator (FT). Obtaining more accurate adsorbate thermophysical properties favorably impacts microkinetic mechanisms for heterogeneous catalysis, as surface coverages depend heavily on these estimates.

Significance and Impact

The method is to be a part of the Sandia National Laboratories' ECC computational framework for automated chemistry, an effort to provide accurate microkinetic mechanisms. As we transition into the exascale, accurate methods that require computational power become relevant and useful tools to considerably improve chemical process predictions.



Katrín Blöndal, Khachik Sargsyan, David H. Bross, Branko Ruscic and C. Franklin Goldsmith, *The Journal of Physical Chemistry C*, doi: 10.1021/acs.jpcc.1c04009

Work was performed at Brown University, Sandia National Laboratories and Argonne National Laboratory

Research Details

- We produced training data for the potential energy surface by performing DFT calculations on the ANL Theta system.
- A minima-preserving neural network (MP-NN) is constructed and used within the PSI routine.
- Our method has been implemented in our open-source phase space integration code, AdTherm.