Abstract Submitted
for the MAR17 Meeting of
The American Physical Society

Importance of many-body dispersion and temperature effects on gas-phase gold cluster (meta)stability BRYAN R. GOLDSMITH, PHILIPP GRUENE, JONATHAN T. LYON, DAVID M. RAYNER, ANDRÉ FIELICKE, MATTHIAS SCHEFFLER, LUCA M. GHIRINGHELLI, Fritz Haber Institute of the Max Planck Society, Faradayweg 4-6, 14195 Berlin, Germany — Gold clusters in the gas phase exhibit many structural isomers that are shown to intercovert frequently, even at room temperature. We performed ab initio replica-exchange molecular dynamics (REMD) calculations on gold clusters (of sizes 5-14 atoms) to identify metastable states and their relative populations at finite temperature, as well as to examine the importance of temperature and van der Waals (vdW) on their isomer energetic ordering. Free energies of the gold cluster isomers are optimally estimated using the Multistate Bennett Acceptance Ratio. The distribution of bond coordination numbers and radius of gyration are used to address the challenge of discriminating isomers along their dynamical trajectories. Dispersion effects are important for stabilizing three-dimensional structures relative to planar structures and brings isomer energetic predictions to closer quantitative agreement compared with RPA@PBE calculations. We find that higher temperatures typically stabilize metastable three-dimensional structures relative to planar/quasiplanar structures. Computed IR spectra of low free energy Au_9, Au_{10}, and Au_{12} isomers are in agreement with experimental spectra obtained by far-IR multiple photon dissociation in a molecular beam at 100 K.

Bryan R. Goldsmith
Fritz Haber Institute of the Max Planck Society, Faradayweg 4-6, 14195 Berlin, Germany

Date submitted: 03 Nov 2016 Electronic form version 1.4