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Gold clusters at finite temperature: influence of fluxionality on ligand adsorption LUCA M. GHIRINGHELLI, ELIZABETH C. BERET, JÖRG MEYER, MATTHIAS SCHEFFLER, Fritz-Haber-Institut der MPG, Berlin, Germany — Metal clusters, in particular in relation with their catalytic properties, have been the object of intensive experimental and theoretical studies, in the recent years. A great deal of effort has been devoted by many theoretical groups to understanding the zero kelvin properties of such clusters. Here, by focusing on small gas phase An_N clusters ($3 \leq N \leq 20$) and their interaction with CO and O_2 as a showcase, we illustrate a methodology for the study of small clusters and their interaction with atoms and molecules at finite temperature. We combine all-electron density functional theory, including scf-density dependent van-der-Waals tail corrections, with finite temperature sampling techniques, like Biased MD and Parallel Tempered MD. We find an unusual flexibility of the clusters, at room and lower temperature. At certain sizes, Au_N clusters at room temperature are liquid droplets. This has an important implication, when accounting for the dynamics of ligand adsorption. One has to consider that the energy released by an exothermic ligand adsorption heats up the newly formed complex, and the equilibration with the environment is much longer than the typical timescale for conformational rearrangement. In this respect, the very concept of a preferred adsorption site in the bare cluster might be meaningless.

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