Abstract Submitted
for the MAR14 Meeting of
The American Physical Society

Stability and metastability of clusters in a reactive atmosphere: theoretical evidence for unexpected stoichiometries of Mg_MO_x
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In heterogeneous catalysis, materials function at finite temperature and in an atmosphere of reactive molecules at finite pressure. As a first step towards understanding the catalytic behavior of metal-oxide clusters, we study the (T, p) dependence of the composition, structure, and stability of the various isomers for each size M of Mg_MO_x clusters in an oxygen atmosphere. The calculations are performed via a massively parallel genetic algorithm in a cascade approach. With the term "cascade", we identify a multistep procedure in which successive steps employ higher levels of theory, with each next level using information obtained at the lower level. We find that small clusters ($M < 5$) are in thermodynamic equilibrium when $x > M$. The non-stoichiometric clusters exhibit peculiar magnetic behavior, suggesting the possibility of tuning magnetic properties by changing environmental pressure and temperature conditions. Furthermore, we show that density-functional theory (DFT) with a hybrid exchange-correlation (xc) functional is needed for predicting accurate phase diagrams of metal-oxide clusters. Neither a (sophisticated) force field nor DFT with (semi)local xc functionals are sufficient for even a qualitative prediction.

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Date submitted: 14 Nov 2013 Electronic form version 1.4