Quantum mechanical studies on transition metal oxides - from gas phase clusters to solid catalysts

Joachim Sauer

Institut für Chemie, Humboldt-Universität zu Berlin, Unter den Linden 6, 10099 Berlin, Germany

For solid catalysts that do not rely on transition metal (oxide) reactivity such as acidic zeolites, predictions of barriers for elementary steps are now possible with near chemical accuracy using hybrid MP2:DFT techniques.[1]

Understanding the structure and reactivity of complex solid catalysts based on (supported) transition metal oxides[2-4] is more difficult and profits from a close coupling of experiment and quantum mechanical calculations. We discuss

- use of DFT with GGA functionals to understand ultra-high vacuum model systems (surface science) such as Au on ultrathin Al_2O_3 films[5]
- use of quantum mechanics and IR spectroscopy to analyse the differences and similarities of gas phase clusters and solid catalysts for (transition) metal oxides[3,6]
- the merits and limits of DFT, specifically the need for hybrid functionals, (V_2O_5)[7,8] (CeO_2)[9]
- role of different spin states and broken symmetry approaches[2,6] compared with results of wave-function based correlation methods[10,11]

1 S. Svelle, C. Tuma, X. Rozanska, T. Kerber and J. Sauer, *J. Am. Chem. Soc.*, 2009, **131**, 816.
2 J. Döbler, M. Pritzsche and J. Sauer, *J. Am. Chem. Soc.*, 2005, **127**, 10861.
3 X. Rozanska, R. Fortrie and J. Sauer, *J. Phys. Chem. C*, 2007, **111**, 6041.
4 X. Rozanska, E. V. Kondratenko and J. Sauer, *J. Catal.*, 2008, **256**, 84.
5 N. Nilius, M. V. Ganduglia-Pirovano, V. Brazdova, M. Kulawik, J. Sauer and H. J. Freund, *Phys. Rev. Lett.*, 2008, **100**, 096802.
6 S. Feyel, D. Schröder, X. Rozanska, J. Sauer and H. Schwarz, *Angew. Chem., Int. Ed.*, 2006, **45**, 4677.
7 J. Sauer and J. Döbler, *Dalton Trans.*, 2004, **19**, 3116.
8 K. R. Asmis, G. Santambrogio, M. Brümmer and J. Sauer, *Angew. Chem., Int. Ed.*, 2005, **44**, 3122-3125.
9 M. V. Ganduglia-Pirovano, J. L. F. Da Silva and J. Sauer, *Phys. Rev. Lett.*, 2009, **102**, 026101.
10 M. Pykavy, C. van Wüllen and J. Sauer, *J. Chem. Phys.*, 2004, **120**, 4207.
11 X. Rozanska and J. Sauer, *Int. J. Quantum Chem.*, 2008, **108**, 2223.