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Single atom catalysis: a surface science approach

Gareth S. PARKINSON*

Institute for Applied Physics, TU Wien, Austria

parkinson@iap.tuwien.ac.at

Single atom catalysis is a rapidly emerging but controversial area of catalysis research that aims to maximize the efficient usage of precious metals through the use of single atom active sites [1]. Although catalytic activity has been demonstrated for several single atom catalyst systems, the inability to accurately characterize a catalyst based on single atom active sites ensures that that the field remains controversial, and little is really known about how a single atom adsorbed on a metal oxide support can catalyze a chemical reaction. In this lecture, I will describe how we are addressing the crucial issues of stability and reaction mechanism using a surface science approach. The work is based on the magnetite (001) surface, which exhibits an unusual reconstruction based on subsurface cation vacancies [2]. A remarkable property of this reconstruction is that it stabilizes ordered arrays of metal adatoms (of almost any variety) with a nearest neighbor distance of 0.84 nm to temperatures as high as 700 K [3]. Crucially, because the geometry of the adatoms is uniform and precisely known, reactivity experiments are performed on a well-defined model system, and theoretical calculations can be performed to shed light on the mechanisms underlying catalytic activity and deactivation. Several examples of our recent work will be used to illustrate the trends we have discovered to date, including how strong CO adsorption destabilizes Pd and Pt adatoms leading to mobility and rapid sintering [4], and how extraction of lattice oxygen from the metal-oxide is central to catalytic activity in the CO oxidation reaction [5].

[1] X.-F. Yang, A. Wang, B. Qiao, J. Li, J. Liu, T. Zhang, Single-Atom Catalysts: A New Frontier in Heterogeneous Catalysis, Accounts of Chemical Research 46 (2013) 1740-1748.

[2] R. Bliem, E. McDermott, P. Ferstl, M. Setvin, O. Gamba, J. Pavelec, M.A. Schneider, M. Schmid, U. Diebold, P. Blaha, L. Hammer, G.S. Parkinson, Subsurface Cation Vacancy Stabilization of the Magnetite (001) Surface, Science 346 (2014) 1215-1218.

[3] G.S. Parkinson, Iron Oxide Surfaces, Surface Science Reports http://dx.doi.org/10.1016/j.surfrep.2016.02.001 (2016).

[4] G.S. Parkinson, Z. Novotny, G. Argentero, M. Schmid, J. Pavelec, R. Kosak, P. Blaha, U. Diebold, Carbon Monoxide-Induced Adatom Sintering in a Pd?Fe_3O_4 Model Catalyst, Nature Materials 12 (2013) 724-728.

[5] R. Bliem, J. van der Hoeven, A. Zavodny, O. Gamba, J. Pavelec, P.E. de Jongh, M. Schmid, U. Diebold, G.S. Parkinson, An Atomic-Scale View of CO and H_2 Oxidation on a Pt/Fe_3O_4 Model Catalyst, Angewandte Chemie International Edition 54 (2015) 13999?14002.