Velocity-resolved kinetics of elementary reactions in heterogeneous catalysis

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Knowledge of the rates of elementary gas phase reactions has contributed decisively to our understanding of important societal problems, for example stratospheric ozone chemistry. Growing our knowledge of rates of elementary chemical reactions at surfaces is crucially important to improving heterogeneous catalysis. In comparison to gas phase reactions, there are surprisingly few known activation energies of elementary surface reactions, nor knowledge of the energies and entropies of the reactions' transition states. This situation is a result of limitations on our methods for measuring rate constants of elementary surface reactions. Furthermore, first principles theories to predict surface reaction rates remain largely un-validated. In this talk, I will present recent experimental advances yielding the rates of elementary reactions at surfaces, which rely on a stroboscopic pump-probe concept designed for neutral matter. This method is also capable of revealing surface-site-specific kinetics information. Not only is site specific reactivity an essential aspect of surface reaction mechanisms, it is essential to provide benchmarks for testing first principles methods for calculating reaction rates, another potentially powerful tool with which to investigate heterogeneous catalysis.

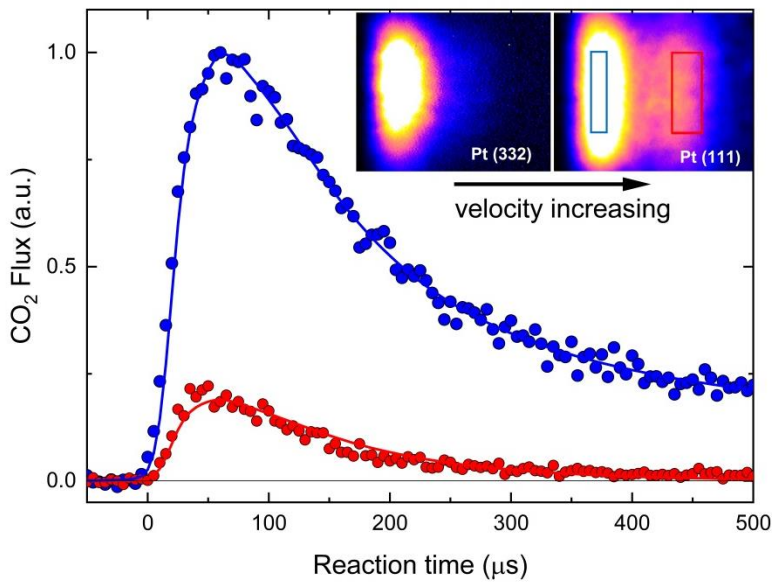

Figure 1: Site-specific CO oxidation producing. The insets show velocity space for CO oxidation on Pt(111) with 0.5% and Pt(332) with 16.7% step density, respectively, revealing a hyperthermal and a thermal channel. When velocity space integration is used to isolate the hyperthermal channel, the red kinetic trace is obtained. For the thermal channel, the blue trace is recorded. Under these conditions the 332 experiment has no O atoms at terrace sites, whereas the 111 experiment does. This shows clearly that the hyperthermal channel comes from terrace reactions while the thermal channel comes from step reactions.

References

[1] Neugebohren et al., Nature, **558**(7709), 280-3 (2018)

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POSITIONS

2010– Director, Max Planck Institute for Biophysical Chemistry, Göttingen Germany
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2010– Research Professor, Department of Chemistry and Biochemistry, University of California Santa Barbara, Santa Barbara CA USA
1988-2010 Professor, Department of Chemistry and Biochemistry, University of California Santa Barbara, Santa Barbara CA USA
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EDUCATION AND PROFESSIONAL PREPARATION

1986–8 Post-doc, Max Planck Institute for Fluid Dynamics, Göttingen, Germany
1986 Ph.D. Dept. of Chemistry, University of California, Berkeley, Berkeley CA, USA
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FELLOWSHIPS AND AWARDS

2010– Alexander von Humboldt Professorship
2009– Elected Fellow of the American Physical Society
2007– Elected Fellow of the American Association for the Advancement of Science
1998 Alexander von Humboldt Research Award to Senior US Scientists
1992 Alfred P. Sloan Research Fellow
1992 Camille and Henry Dreyfus Teacher Scholar Award
1989 National Science Foundation Presidential Young Investigator Award
1982 National Science Foundation Pre-doctoral Fellow

PUBLICATIONS

More than 230 research articles in peer reviewed Journals: 10 in Science, 4 in Nature or Nature daughters; 10 in Physical Review Letters, JACS, Angewandte Chemie - Int. Ed., Geophysical Research Letters or Accounts of Chemical Research. 'Google Scholar' reports 9020 cumulative citations, h-index=51; 'ISI-Web of Science' reports 7153 cumulative citations, h-index of 45