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Velocity resolved kinetics for surface chemistry

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Knowledge of the rates of elementary gas phase reactions has contributed decisively to our understanding of important chemistry with great human impact. For example rate-constant data bases for atmospheric chemistry provide the essential input for computer models of urban air pollution, stratospheric ozone depletion and global climate change. Developing databases for elementary chemical reactions at surfaces could similarly have a powerful impact on our understanding of heterogeneous catalysis. Examples of possible impacts include: catalysts and electrocatalysts for producing cleaner fuels from fossil fuels or renewables (electricity or biomass), synthesis of ammonia and other commodity chemicals, cleaning automobile exhaust, converting carbon dioxide into fuels or chemicals, to name just a few. While there are databases already with numerous accurate experimental energies of adsorbed catalytic reaction intermediates on metal surfaces, there are relatively few known activation energies of elementary surface reactions, or the energies and entropies of their transition states. This is mainly due to limitations on our methods for measuring rate constant of elementary surface reactions. In this talk, I will present recent experimental advances in our ability to measure the rates of elementary reactions at surfaces, which rely on a stroboscopic pump-probe concept designed for neutral matter. In at least two examples, these measurements reveal data of sufficient quality to also reveal surface-site-specific kinetic information. This provides important benchmarks for testing first principles methods for calculating reaction rates, another potentially powerful tool with which to investigate heterogeneous catalysis. The time is ripe to expand efforts to understand the fundamentals of reaction rates in heterogeneous catalysis. New concepts for advanced experimental methods employing free electron lasers can enable this field.

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