Two decades of studying non-linear phenomena during catalytic reactions – from spontaneous patterns to external forcing

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Over the past two decades, numerous heterogeneous catalytic systems have been found to exhibit a rich variety of nonlinear activity, including reaction rate oscillations and spatiotemporal pattern formation. As such, the nonlinear dynamics promoting this complex behavior have been the focus of many experimental and theoretical investigations, motivated as much by the goal of understanding the origins of complex behavior as by the potential of exploiting such behavior to improve system performance.

To that end, our present work is concerned with the manipulation and characterization of the dynamic behavior of two model surface reactions, CO oxidation and the reduction of NO by NH_3, both on Pt catalysts. While the dynamics of these systems have been investigated previously, the impact of our experimental work lies in the ability to address reaction dynamics on a local length scale, thus providing a unique perspective on various fundamental catalytic processes.

With regard to the control aspect of this topic, local manipulation of surface reactivity was accomplished using an external forcing technique designed to locally dose reactant onto the catalyst surface at variable frequencies and concentrations. This technique was further paired with imaging ellipsometry (EMSI) such that the surface response to forcing could be monitored under reaction conditions in real time.