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Catalytic CO oxidation driven by ultrashort X-ray pulses

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Understanding the reaction paths and transition states of catalytic reactions is a key issue in order to design highly reactive and selective catalysts. Ultrashort free-electron laser (FEL) soft x-ray pulses are a novel tool ideally suited for investigating time-dependent surface processes, due to the femtosecond resolution and tunability of the excitation energy to specific adsorbate resonances. Here we show experimental evidence that oxidation of CO on a Ru(0001) surface can be driven by femtosecond x-ray pulses, by selectively exciting the adsorbed atomic O and detecting CO2 [1]. This opens a new route to controlling surface reactions, complementing the optical excitation mechanism which has previously been thoroughly investigated [2,3]. DFT calculations show that the valence hole state, which is formed after Auger decay of the core-ionized state, creates a strong repulsive force between the adsorbed O and the surface, giving enough energy into the system to drive the catalytic reaction.

[1] Schreck et al., submitted to JPC.
[2] Bonn et al, Science, 285, 1042 (1999).
[3] Öström et al., Science, 347, 978 (2015).

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