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Ultrafast K-edge spectral evolution of physi- and chemisorbed carbon species

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A deeper understanding of catalytic activity, and in particular of catalytic selectivity, will help the development of more performant heterogeneous catalysts. The ability to discriminate competing reaction paths and to identify reaction intermediates will be crucial for gaining this insight. X-ray core-edge spectroscopy allows here to probe the electronic structure of catalyst and adsorbates with chemical species and site specificity. Moreover, the high repetition rate of free electron lasers (XFEL) allows for following the evolution of the electronic structure in catalytic processes in real time. In optical laser pump and x-ray probe experiments, the energy transfer from metal substrate electrons to phonons and adsorbate electronic and vibrational degrees of freedom can be observed with on the order of 100 fs temporal resolution via x-ray absorption and emission spectroscopy. Electronic structure simulations of heterogeneous catalytic reactions are here an indispensable tool to infer energy transfer and reaction mechanisms from the observed ultrafast x-ray spectral evolution.

We have followed the ultrafast K-edge spectral evolution of different carbon species (chemisorbed CO [1,2], atomic C [3], and physisorbed graphene [4]) in UV-vis pump + XFEL probe experiments and have used first-principles simulations to understand the underlying energy transfer mechanisms between metal substrate and adsorbate. For graphene, we show how excitation of strongly coupled optical phonon modes can explain the observed ¡ps spectral response. For chemisorbed atomic C, we found ultrafast electronic energy transfer to carbon before any excitations of C nuclear motion, and for chemisorbed CO, we found an ultrafast response of vibrational mode excitations. Such understanding of energy transfer from substrate electronic and phonon degrees of freedom to adsorbates will be important for gaining insight into the formation of catalytic precursor states.

[1] Diesen, E., et al., Phys. Rev. Lett. **127**, 016802 (2021).
[2] LaRue, J., et al., J. Chem. Phys. **157**, 164705 (2022).
[3] Schreck, S., et al., Phys. Rev. Lett. **129**, 276001 (2022).
[4] Ogasawara, H., et al., Phys. Rev. Materials **7**, 024005 (2023).

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