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Towards a Molecular Level Understanding of CO and H_2 Adsorption and Dissociation on Cobalt Nanoparticles

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— The development of sustainable energy technologies, including the production of synthetic fuels, is of global importance. Fischer-Tropsch synthesis (FTS) has recently gained increased attention as it involves the formation of hydrocarbons via the catalytic conversion of syngas (CO and H_2), which can be derived from renewable sources. FTS is often performed using Co-based catalysts that are greatly affected by the adsorption state of reactants, as well as nanoparticle shape and size. Here we have used low-temperature scanning tunneling microscopy (LT-STM) to study the interaction of syngas with well-defined Co nanoparticles grown onto Cu {111}, an inert metal for FTS. Hydrogen adsorbs dissociatively on the Co surfaces, resulting in three unique coverage-dependent phases. We demonstrate that these phases can resolve crystal packing ambiguities of the underlying Co nanoparticles, a question that has been debated in the literature. Simultaneous exposure of the Co to H_2 and CO results in segregated islands of the adsorbates on the nanoparticle surface at 80 K, and we propose that atomic H blocks CO adsorption, causing the build-up of CO at the nanoparticle step edges. With increasing CO coverage, a two-dimensional phase compression of H by CO is observed, providing *the first direct* visualization of this phenomenon in a catalytically relevant system. Our data suggest that FTS reactivity may be dominated by the interface length between the adsorbates and be subject to unforeseen kinetic restraints as a function of particle size.

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