

## **Quantum control of molecular bond-forming catalytic surface reactions and of dissociation reactions in the liquid phase**

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Ever since their invention lasers were considered the ideal tool for microscopic control of selective cleavage and formation of chemical bonds. However, controlling chemical reactions with tailored femtosecond laser pulses, so far has been restricted to bond-cleavage of molecules in the gas phase, or condensed phase processes without a change of the atomic composition of the system under study (e.g. excitation, isomerization) [1,2]. The selective formation of chemical bonds with tailored fs laser pulses has not been shown up to now.

Therefore, we investigate the reaction of carbon monoxide and hydrogen on a Pd(100) single crystal surface at room temperature and under high vacuum conditions. Several product molecules are synthesized, among them also species for whose formation three particles are required to interact with the surface and the laser (e.g. CH<sub>3</sub><sup>+</sup> or H<sub>3</sub>O<sup>+</sup>). By applying closed-loop optimal control, we manipulate these reactions and selectively optimize the ratio of different bond-forming reaction channels, in contrast to previous quantum control experiments aiming at bond-cleavage. Further experiments disclose that the control mechanism is nontrivial and sensitive to the relative proportion of the two reactants.

We also investigate the photo-induced Wolff-Rearrangement reaction of 2-Diazo-1-naphthoquinone (DNQ) dissolved in methanol. The ketene photoproduct is detected by transient vibrational spectroscopy in the mid-infrared. This technique allows for the observation of structural changes during the photoreaction. Thus, it is very well suited for quantum control experiments in the liquid phase, where one or more photoproducts are formed. This is due to the clear distinction of different vibrational bands, in contrast to the strong overlap of the electronic bands in the UV-VIS spectral region. We present first results of the application of this technique on the photoreaction of DNQ. The photoproduct yield shows a strong dependence on the variation of chirp and intensity of the excitation laser pulses.

### References:

[1] T. Brixner and G. Gerber, *ChemPhysChem* 4, 418 (2003).

[2] P. Nuernberger, G. Vogt, T. Brixner and G. Gerber, *Phys. Chem. Chem. Phys.* 9, 2470-2497 (2007).