

# fs-laser-driven dynamics of CO on Ru(0001)

a computational study using electronic friction (MDEF) and  
the generalized Langevin oscillator (GLO)

Robert Scholz

Institut für Chemie  
Universität Potsdam

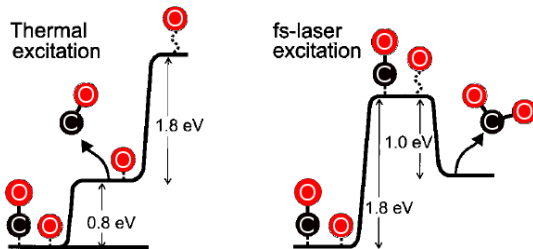
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# General motivation

## Why investigate fs-laser-driven surface dynamics?

- gain fundamental understanding of adsorbate bonding  
⇒ additional tool besides scattering experiments
- possible direct application in catalysis: “femtochemistry”  
⇒ new reaction pathways opened up by fs-lasers



CO/O-coadsorbate @ Ru(0001)

M. Bonn *et al.*, *Science* 1999

# Specific motivation for the CO/Ru-System

CO/Ru system important for catalysis

e. g. Fischer-Tropsch synthesis

Experimentally well studied system

- especially regarding fs-laser irradiation e.g.:  
Bonn *et al.*, *Science* 1999; Funk *et al.*, *JChemPhys* 2000  
(both Ertl group - chemistry Nobel prize 2007).
- recently, time resolved x-ray spectra (XAS and XES)  
⇒ “movie” of changes in orbital DOS

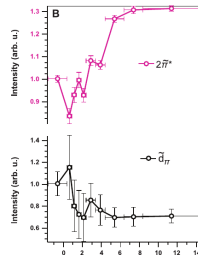
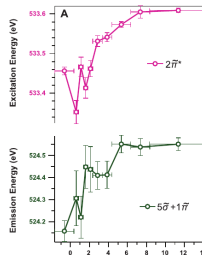
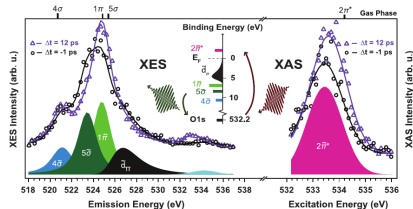
# Details of the time resolved x-ray experiment

## What was done?

- pump: *vis*-fs-laser
- probe: x-ray free electron laser (K edge of O-atom)

## What is observed?

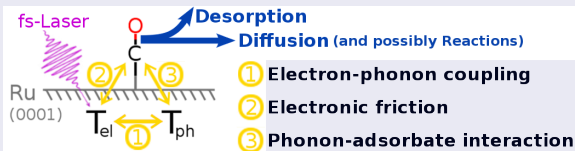
- orbital density of states
- energies shift towards gas-phase values of CO
- intensities change
  - $2\tilde{\pi}^*$  increase by  $\sim 30\%$
  - $\tilde{d}_{\pi}$  decrease by  $\sim 30\%$
  - Participator peak appears



# What happens after fs-laser excitation of the metal?

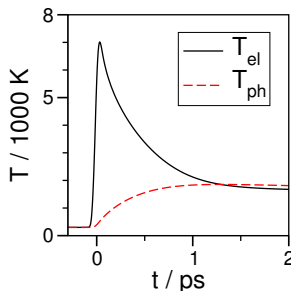
## Coupling between three different kinds of degrees of freedom:

- electron gas ( $T_{\text{el}}$ )
- lattice vibrations ( $T_{\text{ph}}$ )
- adsorbate movement ( $T_{\text{ads}}$ )



# Two-Temperature Model

$$C_{\text{el}} \frac{\partial T_{\text{el}}}{\partial t} = \frac{\partial}{\partial z} \kappa \frac{\partial}{\partial z} T_{\text{el}} - g(T_{\text{el}} - T_{\text{ph}}) + S(z, t),$$
$$C_{\text{ph}} \frac{\partial T_{\text{ph}}}{\partial t} = g(T_{\text{el}} - T_{\text{ph}}).$$



# Laser-Driven Diffusion

