

FEMTOSECOND-LASER INDUCED DYNAMICS OF CO ON Ru(0001): NEW INSIGHTS FROM A HOT-ELECTRON, ELECTRONIC FRICTION MODEL INCLUDING SURFACE MOTION

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Introduction

Motivation

- research on small molecules adsorbed to metals is important for:
 - catalytic applications
 - fundamental understanding of bonding
- femtosecond(fs)-lasers are a valuable tool for such research as they
 - allow for investigations on small timescales
 - open up new processes compared to heating (femtophotochemistry)
 - may enable specific control over catalytic reactions (photocatalysis)

How does fs-laser-irradiation affect metal surfaces?

fs-Laser **Desorption**
Diffusion (and possibly Reactions)

Ru (0001)

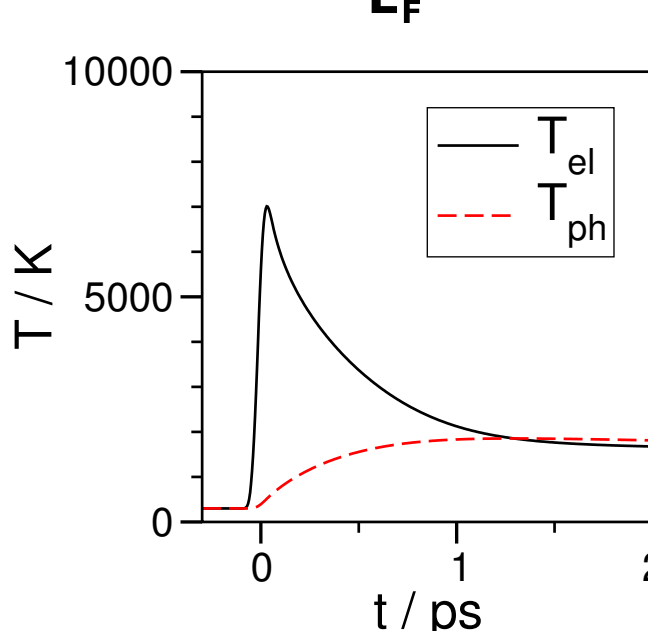
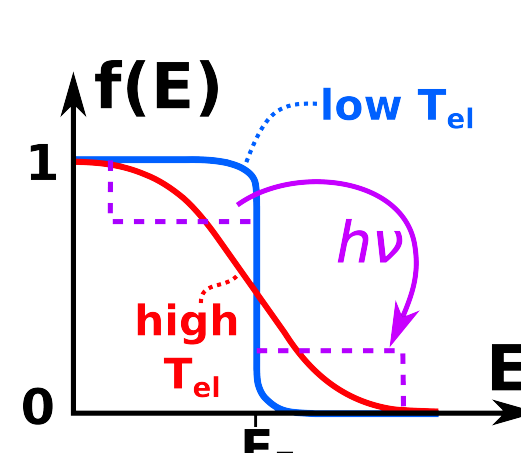
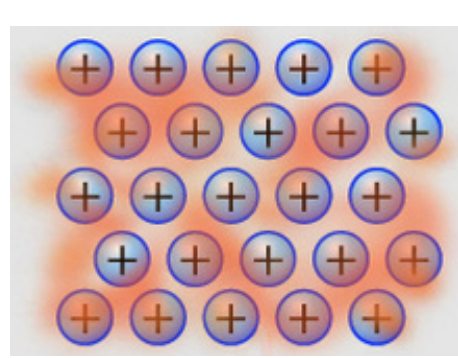
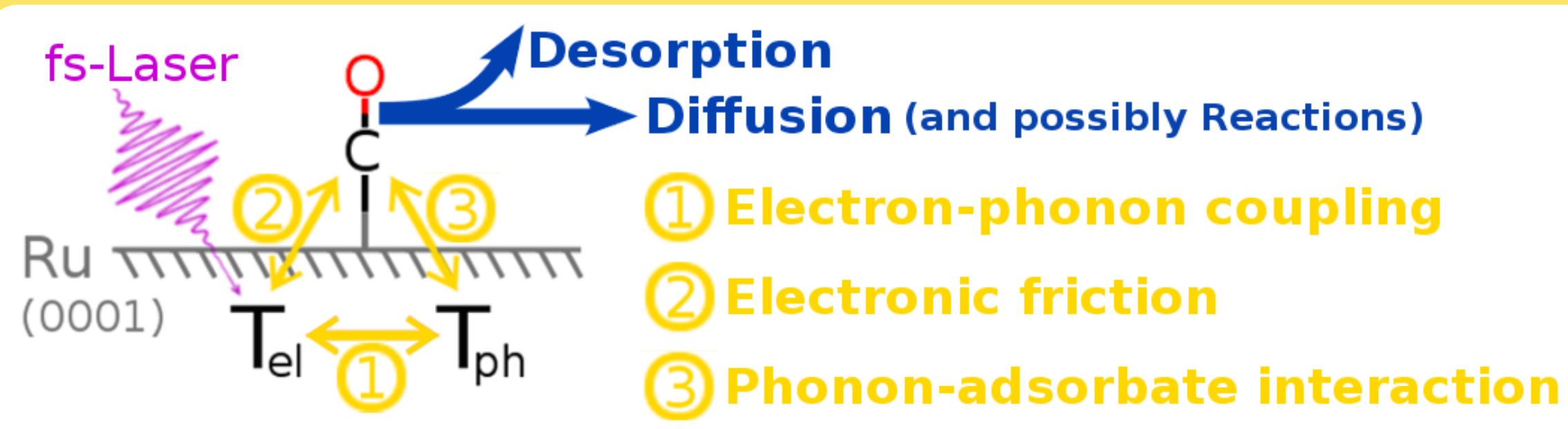
① **Electron-phonon coupling**
② **Electronic friction**
③ **Phonon-adsorbate interaction**

T_{el} T_{ph}

- metals: ion lattice plus quasi-free electron gas
- visible light is absorbed only by the electrons
- produced electron hole pairs thermalize quickly
⇒ “hot” Fermi-Dirac-distribution (after ~ 10 fs)
- electrons transfer part of energy to ion lattice, via ① **electron-phonon coupling** (phonons = lattice vibrations; quasi-particles)
 - electrons couple to phonons as their fast movement causes “shockwaves” in ion lattice
 - equilibration process completes after ~ 1 ps

⇒ with fs-lasers, two different temperatures:

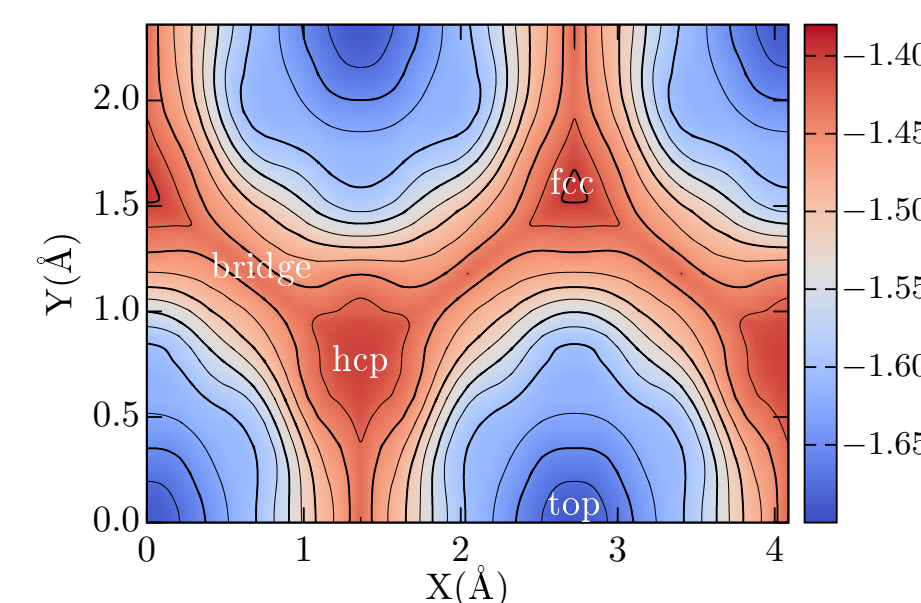
- T_{el} - electron temperature
- T_{ph} - phonon temperature

- time evolution simulated with a Two-Temperature Model (2TM) [1]


Models and Methods

Six-dimensional Potential Energy Surface (6D PES)

- Basis for dynamics: precomputed Potential Energy Surface (PES)
 - all 6 dimensions of the adsorbate
 - analytical PES and gradients ⇒ very fast
⇒ number and length of trajectories can be large
 - downside: surface atoms frozen ⇒ no phonons



Two-Temperature Model (2TM)

Electronic Friction: LDFA and Langevin Dynamics

Inclusion of Phonons: GLO-model

References

[1] S. I. Anisimov, B. L. Kapeliovich, and T. L. Perel'man, *Sov. Phys.-JETP* **39**, 375 (1974).

[2] M. Dell'Angela, T. Anniyev, M. Beye et al., *Science* **339**, 1302 (2013).