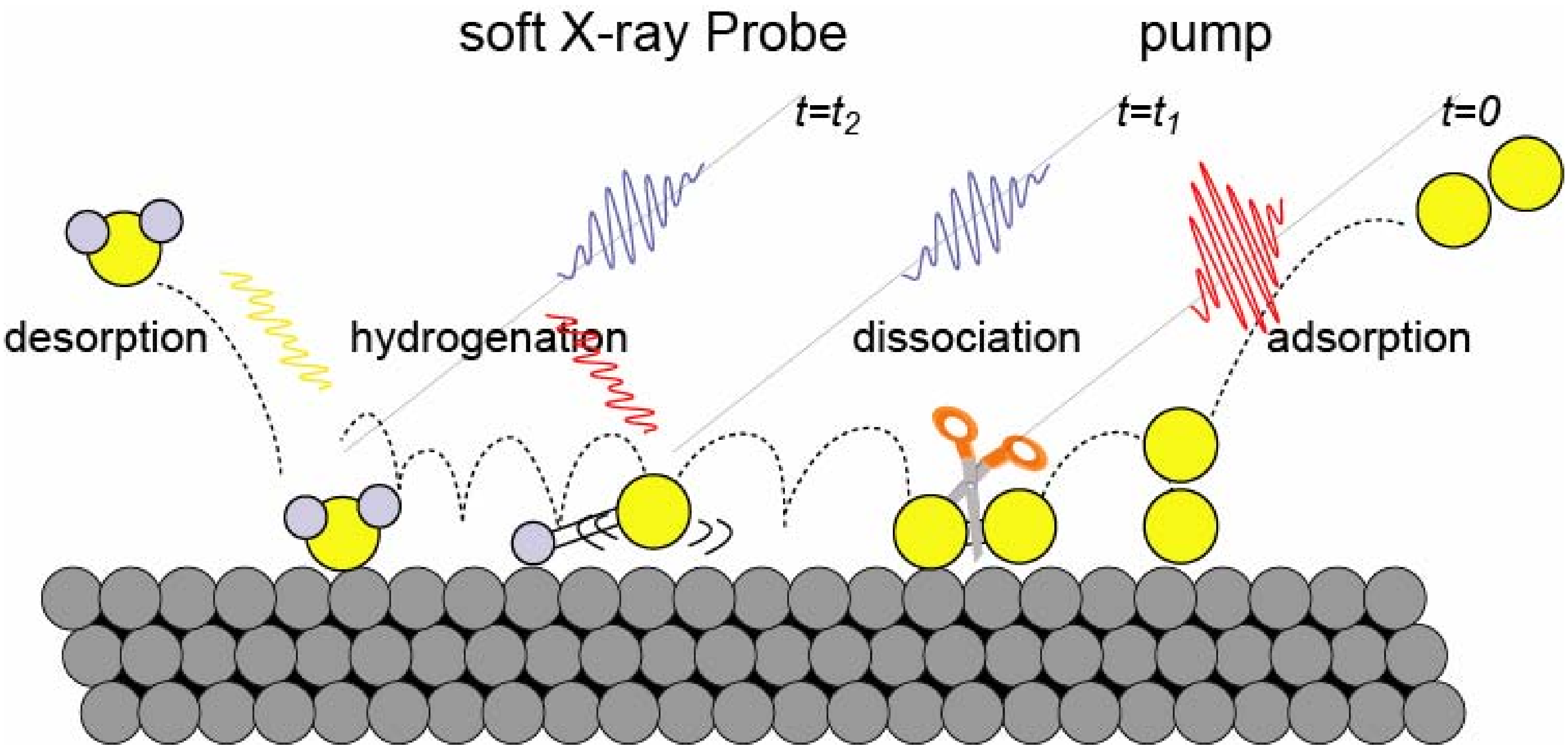
Coherent Control of Surface Reactions using THz Radiation

Hirohito Ogasawara Dennis Nordlund and Anders Nilsson,

Stanford Synchrotron Radiation Laboratory

- 80% of all important chemical reactions take place on interfaces
- Catalytic processes is the largest chemical industry
- Energy technology, fuel cells, splitting of water by solar
- Environmental science
- Semiconductor technology
- Biosurfaces

Elementary Surface Reactions

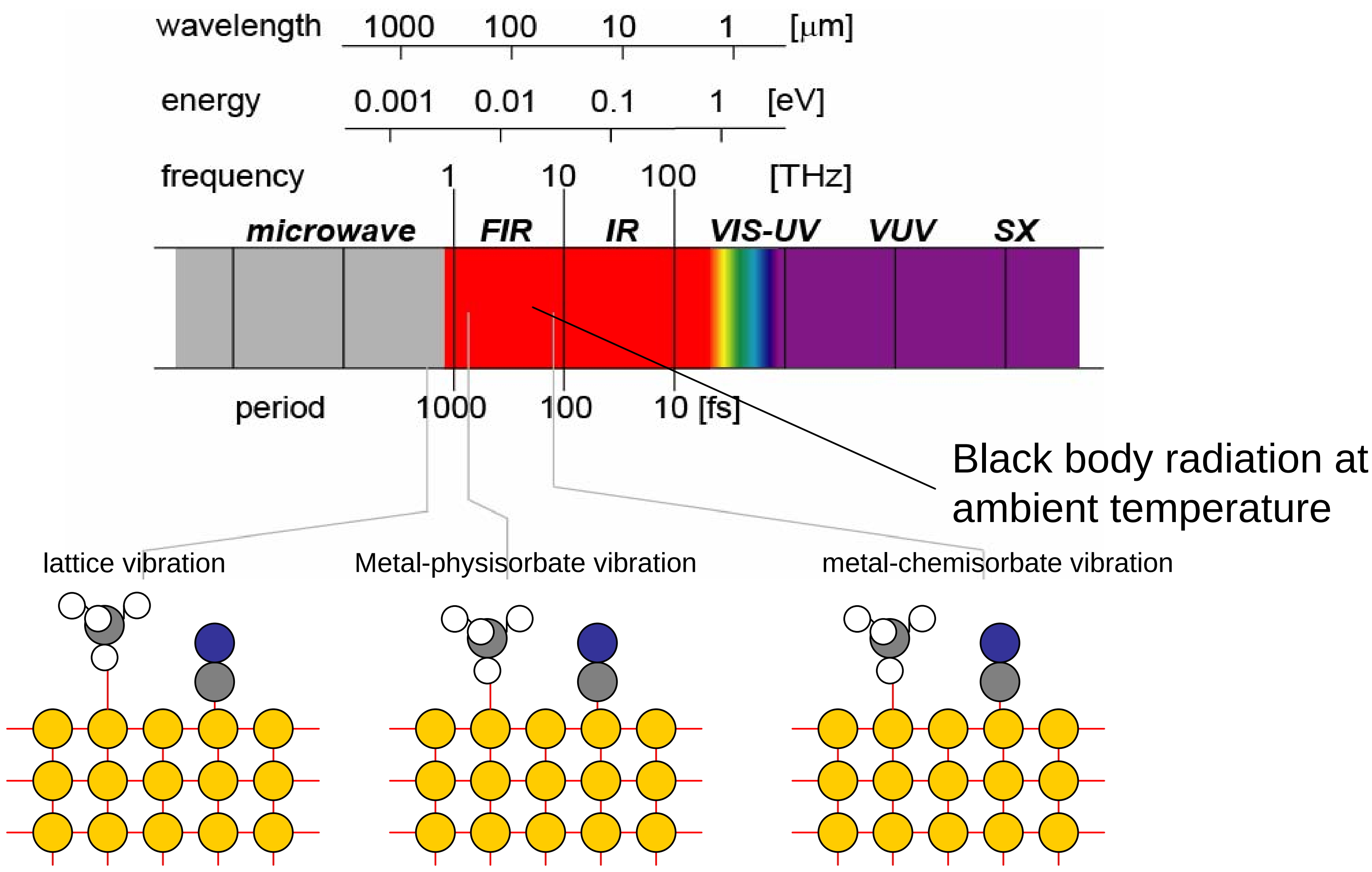


Challenges for Pump and Probe Experiments

- Most relevant chemical reactions are driven by temperature (kT) not optical excitations
- Synchronization between pump and probe
- Control of reaction coordinate system
- Selective detection of molecules on surfaces, differentiate between species

THz pump and X-ray probe

THz radiation and molecular vibration



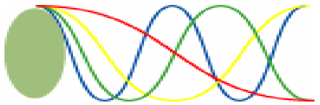
1-10 THz = Far-IR (~0.01[eV], ~30[μm]) can excite vibrations:
lattice vibration, adsorbate-metal vibration.

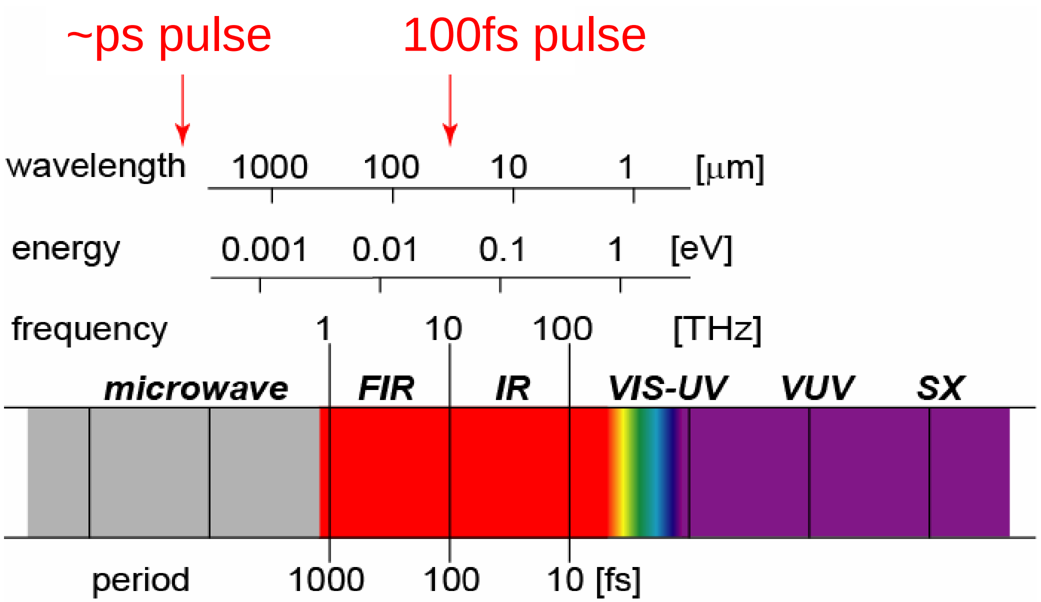
CSR from relativistic e-bunch

:bunch length

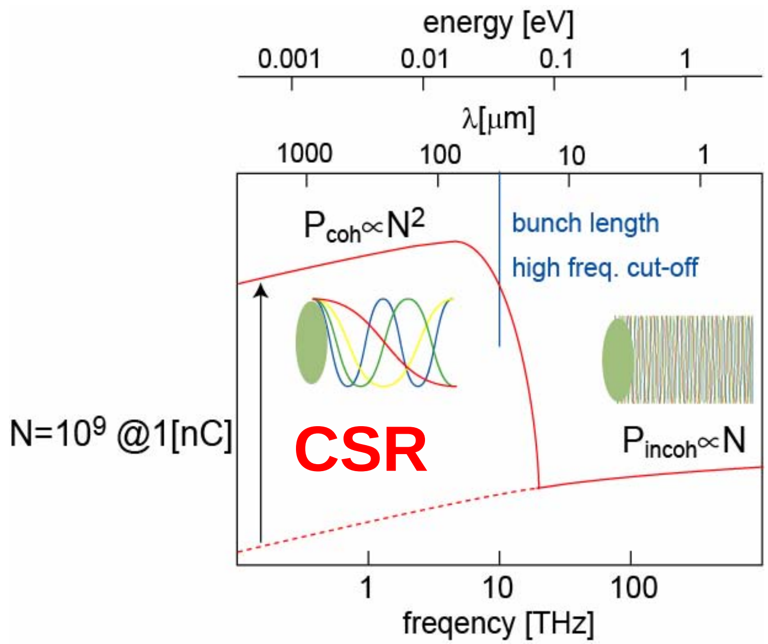
1) wavelength < bunch length ….. incoherent radiation

2) wavelength > bunch length …. coherent radiation





Spectrum



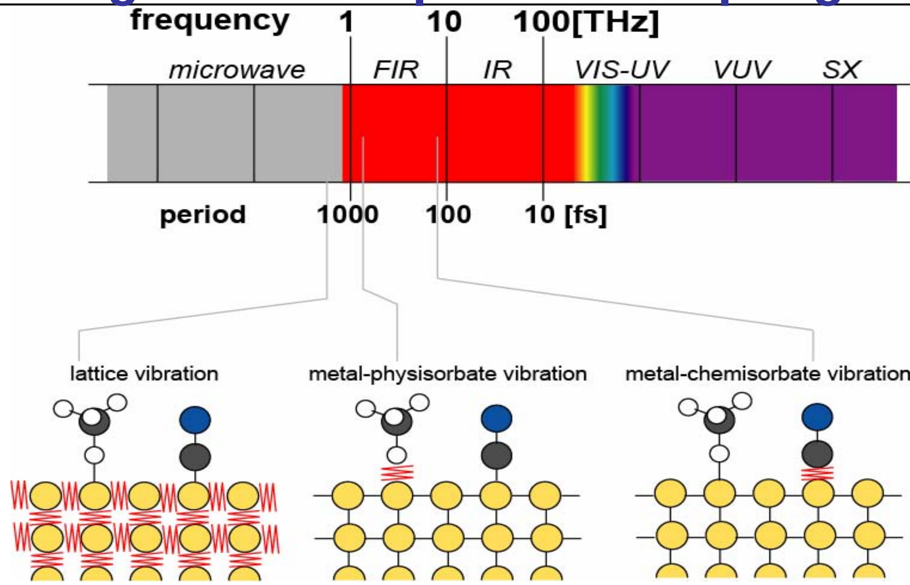
Ultra short electron bunch is necessary for coherent THz radiation.
Bunch length = **high frequency** cut-off

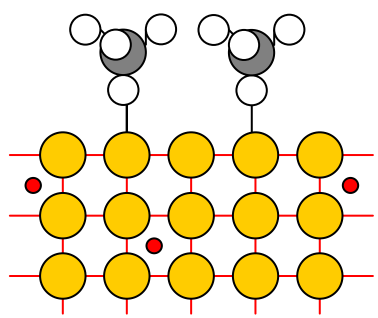
HIGH POWER and STRONG E-FIELD

Temperature jump:

vibrational excitation through electron-phonon coupling

Temperature jump
via
Electron-hole pair excitation,
Lattice vibration excitation,
….
required power: ~1-10 mJ/pulse





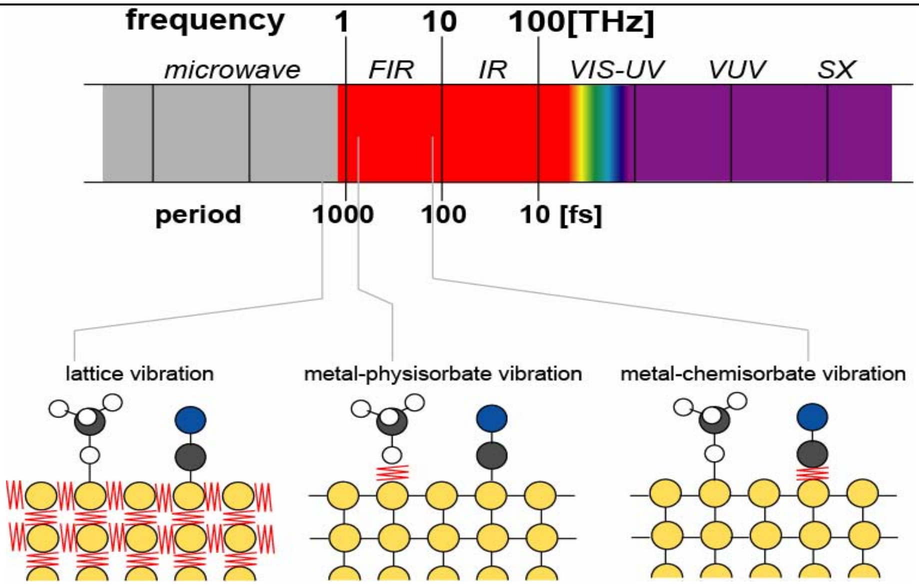
fs laser: hot electron problem

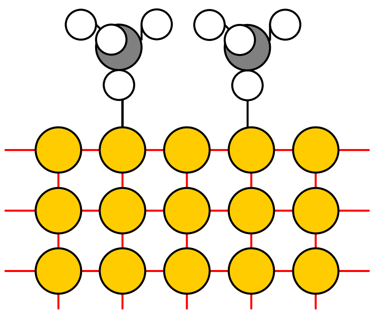
THz: NO hot electron

Temperature jump ensues the motion of adsorbate and stimulates surface chemical reactions.

Temperature jump:
vibrational excitation

Temperature jump
via
Electron-hole pair excitation,
Lattice vibration excitation,
….
required power: ~1-10 mJ/pulse



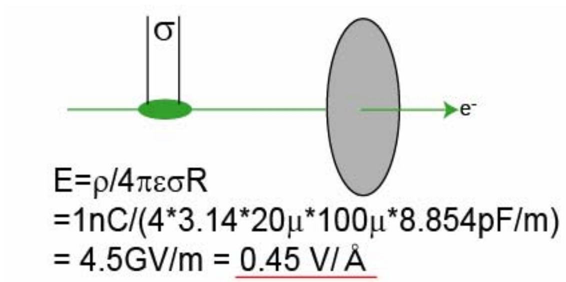


fs laser: hot electron problem

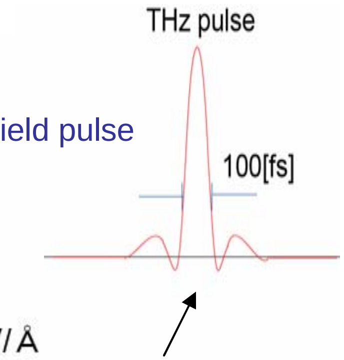
THz: NO hot electron

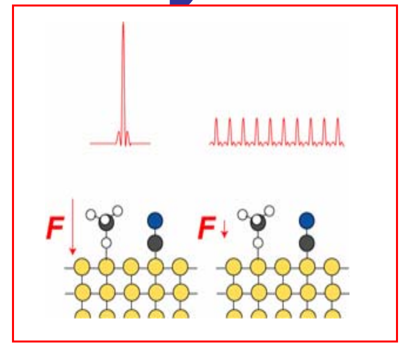
Temperature jump ensues the motion of adsorbate and stimulates surface chemical reactions.

E-field and surface chemistry



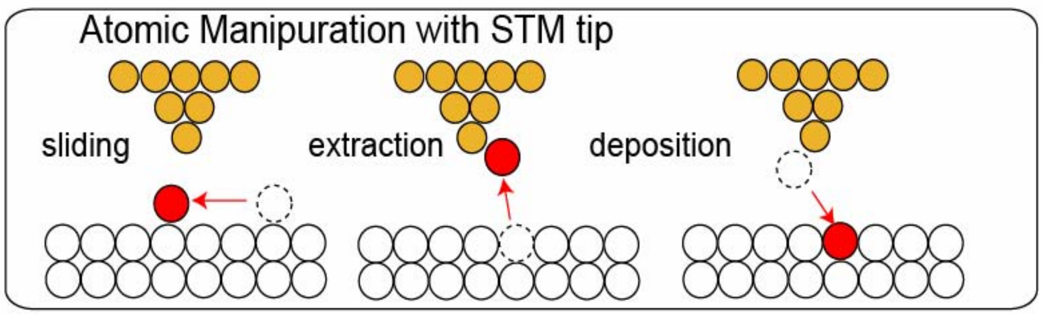
strong electric field pulse

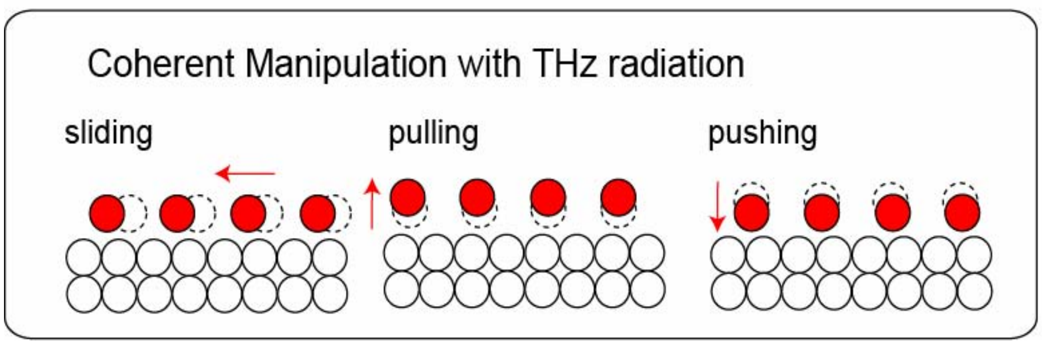


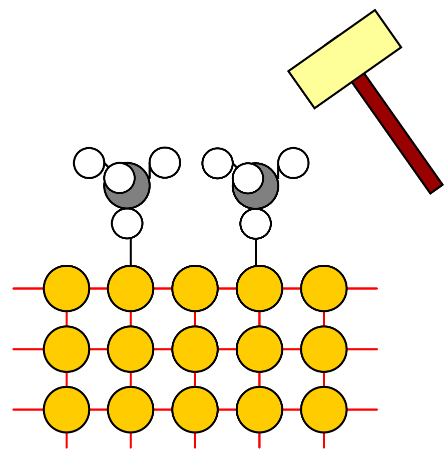


~ 0.1-0.3 V/Å

Pulse width ~ time scale of molecular vibration





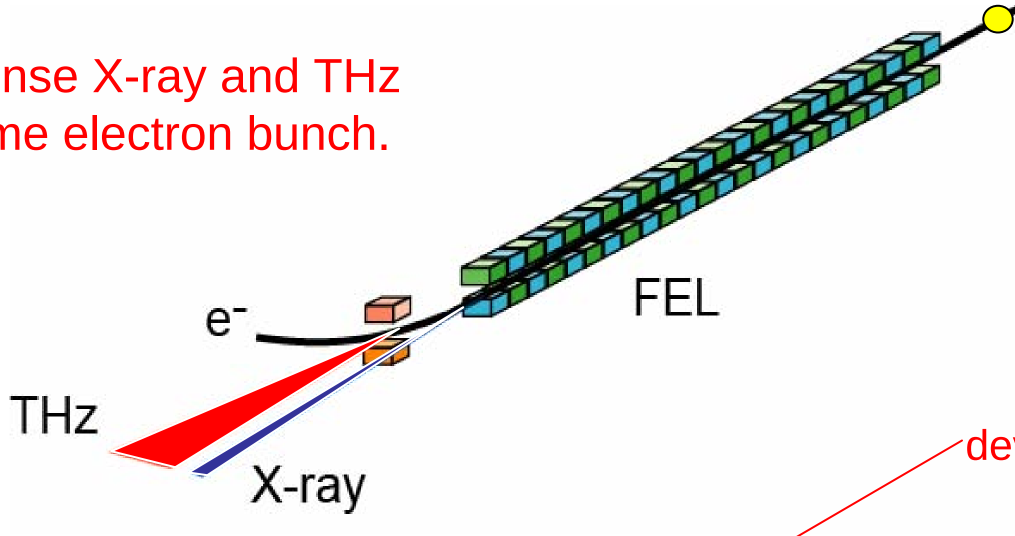


THz electric field ~ Coulomb force between e^- and the nuclei

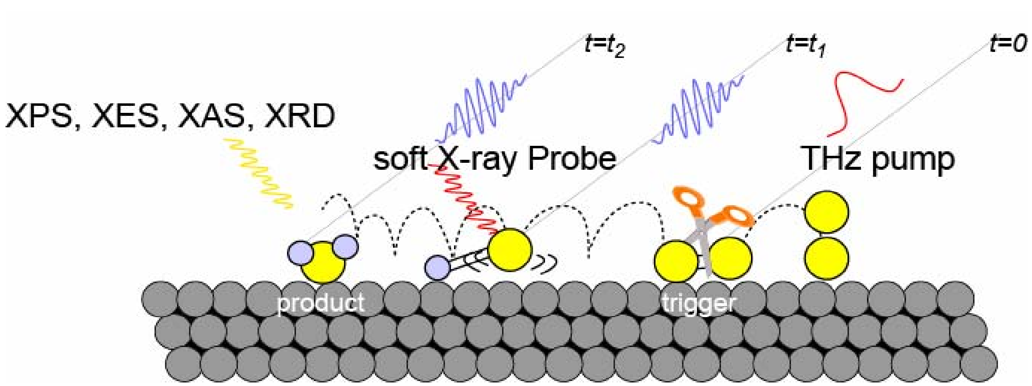
manipulation of molecule, coherent control molecular motion

How to probe THz induced process

FEL can produce intense X-ray and THz radiation from the same electron bunch.



develop at SABER



on-axis radiation, soft X-ray, hard X-ray, off-axis radiation: THz

Pump: THz, **Probe:** XPS, XES, XAS, XRD, IR

Summary THz generation

- THz region = far IR ~ *kT*
  lattice vibration, adsorbate-substrate vibration

- coherent radiation ~ 10^9 * incoherent radiation
- short electron bunch = high cut-off frequency
- coherent broad band = electric field pulse
- coherent atom manipulation on surfaces

Bunch length: ~100fs, Charge: ~nC

Experimental setup