UP 1: Multi-Electron Dynamics, Rearrangement and Fragmentation of Large Molecules and Clusters in Strong Laser Fields

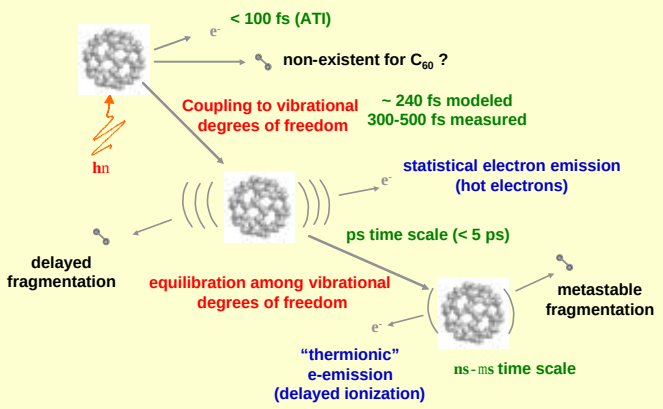
Max-Born-Institut

Cooperation: A4, A6, B2, C1, C2 and C6

Results (2001-2003): Control of Ionisation and Fragmentation by Selective Vibrational Excitation with Ultra-Short Pulses

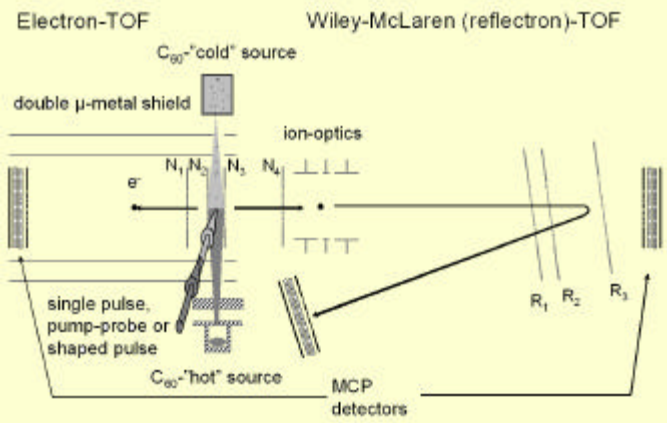
Motivation

Electron and Nuclear Dynamics of C_{60} – Analysis by Control



Experimental Setup

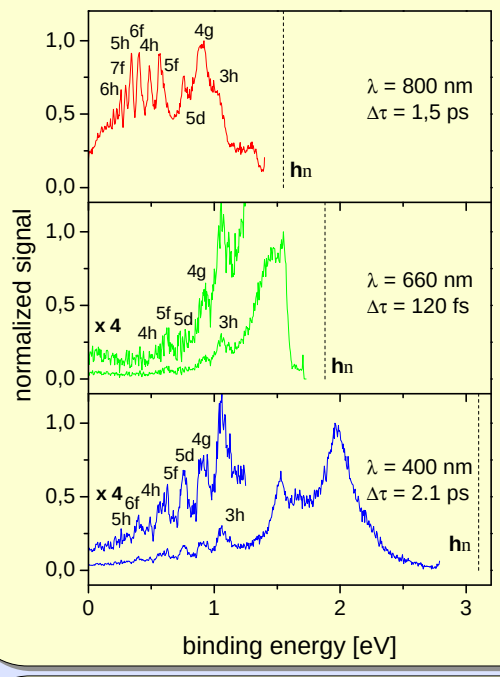
Ion an Electron Time of Flight



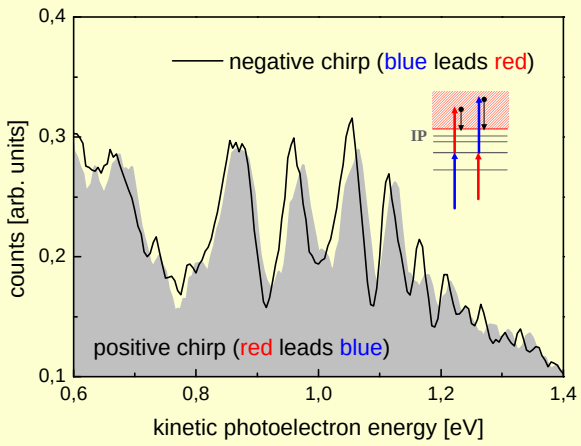
Single-Pulse Spectroscopy

Excitation of C_{60} Rydberg States Depending on:

Photon Energy

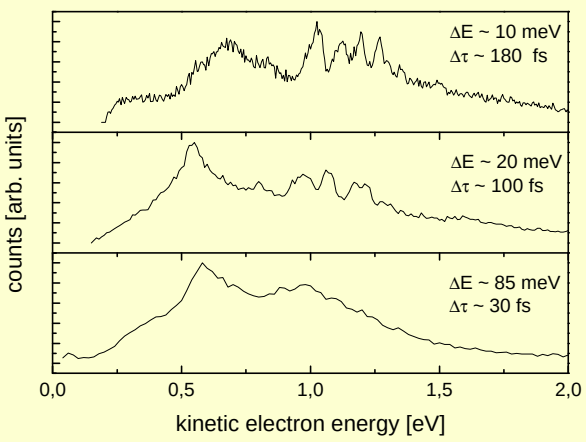


Chirp



- excitation on the rising pulse edge
- ionization in the latter half of the pulse due to one photon absorption

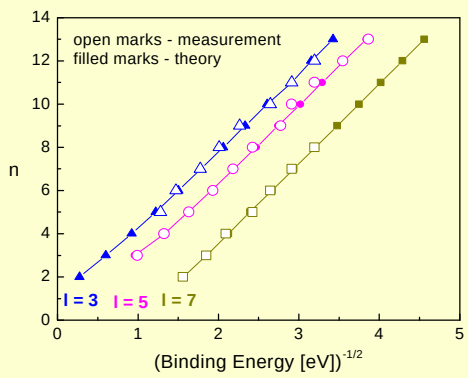
Pulse Duration

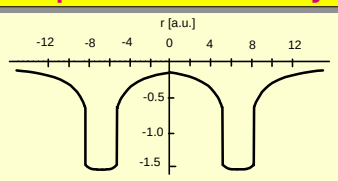


- fast population of Rydberg sates

Boyle et al., in preparation (2004)

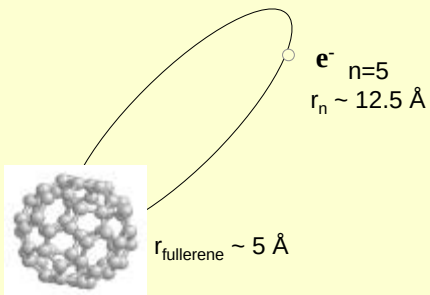
Comparison with Theory





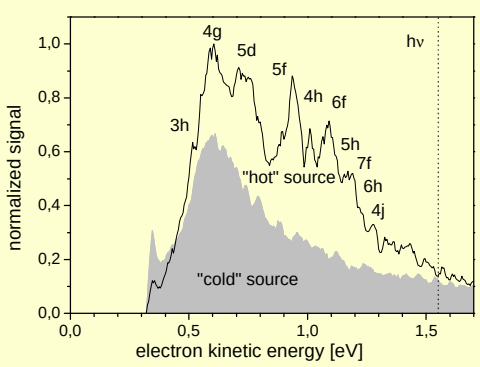
C_{60}-potential based on Puska and Nieminen Phys. Rev. A **47**, 1181 (1993)

- theoretically calculated binding energies in excellent agreement with experiments



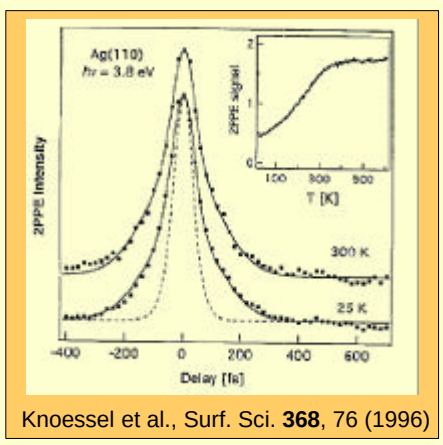
Boyle et al., Phys. Rev. Lett. **87**, 273401 (2001)

Hot and Cold C_{60}



- internal energy stored in vibrational modes effects population of Rydberg states
- similar to experiments on Ag(110) surfaces
- possible reason as in C_{60} with its "large" finite surface: higher population in vibrationally excited states enhances e-phonon coupling

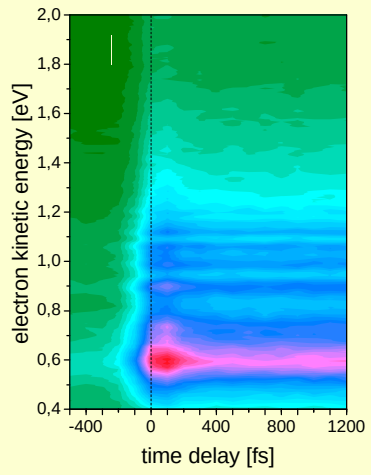
M. Boyle, et al., Two color pump probe study and internal energy dependence of Rydberg state excitation in C_{60}, Physical Review Letters (submitted).

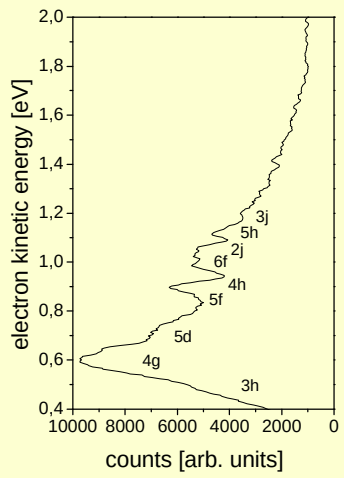


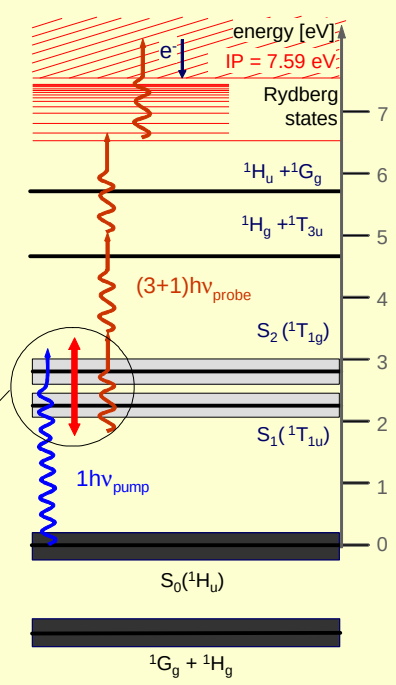
Knoessel et al., Surf. Sci. **368**, 76 (1996)

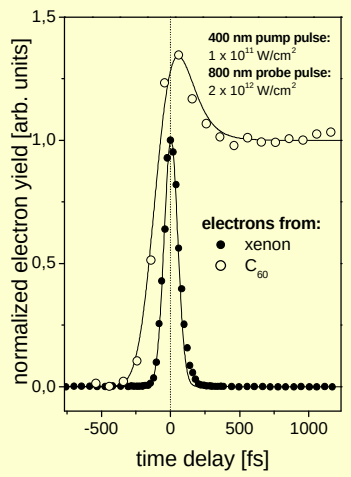
One- and Two-Colour Pump Probe Spectroscopy

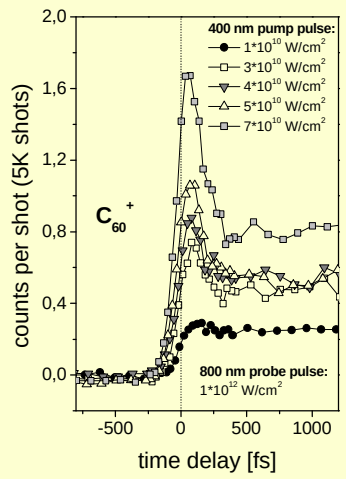
Nonadiabatic Multi-Electron Dynamics (NMED)

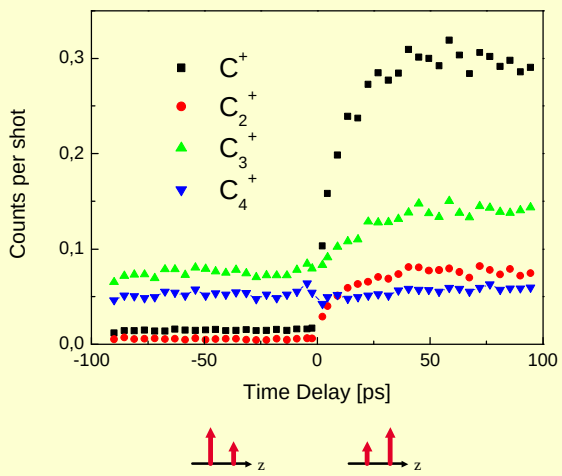




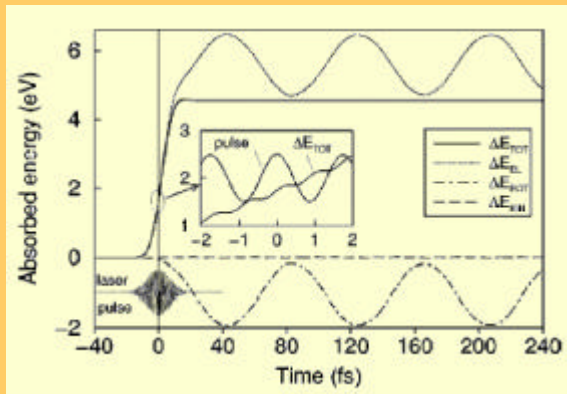








- evaporation of small neutral fragments on a ps time scale (800 nm, 120 fs, 5 and 1.8 x 10^{13} W/cm^2)



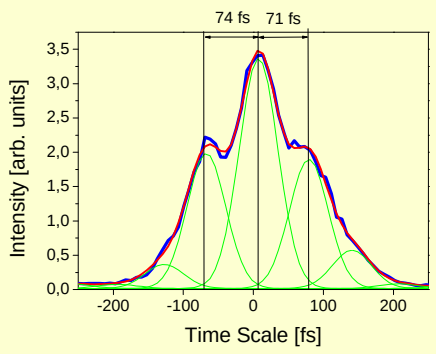
Zhang et al., Phys. Rev. B **68**, 152003 (2003)

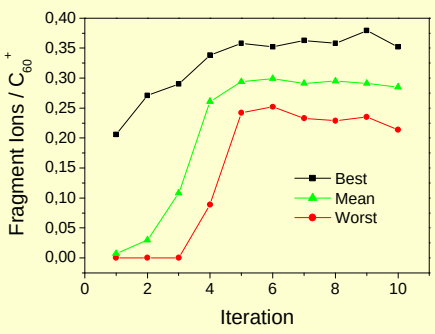
- multi-electron excitation of the S_2 "doorway state"
- thermalization within the hot electron cloud on a time-scale below 100 fs
- coupling to vibrational modes of C_{60}
- lifetime of excited doorway state: ~ few ps
- Rydberg state population by multiphoton absorption
- one-photon ionization from the excited state
- NMED is also active in multiple ionization fragmentation

M. Boyle, et al., Two color pump probe study and internal energy dependence of Rydberg state excitation in C_{60}, Physical Review Letters (submitted).

Program for 2004-2007:
Detailed Study of the Multi-Electron Dynamics, Rearrangement and Fragmentation of Large Molecules and Clusters in Strong Laser Fields

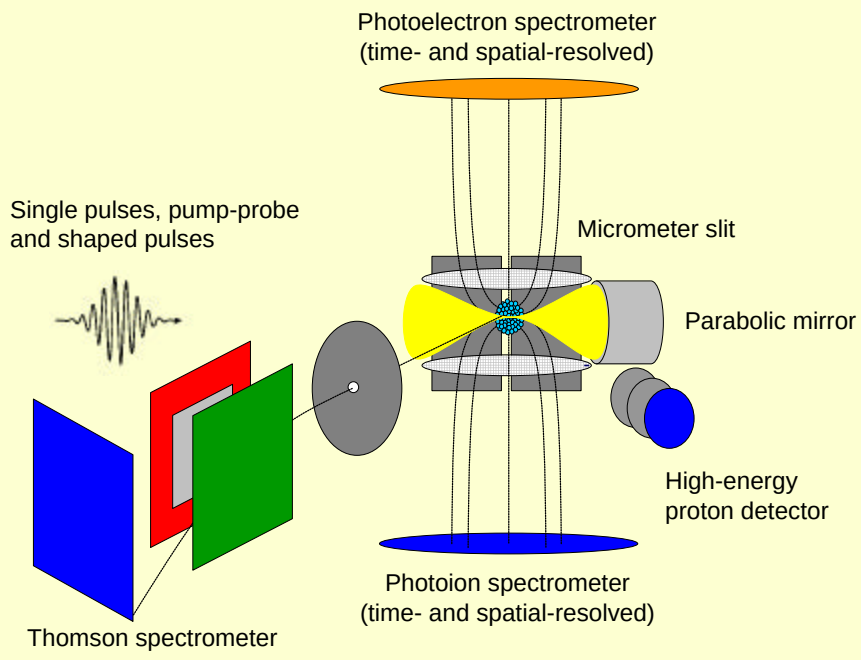
First Experiments - Optimal Control Feedback Loop





optimization of the fragmentation results in a pulse shape which reflects the importance of vibrational modes in the energy deposition process (A_{1g}: T =67 fs)

Reaction Microscope



- **optimization of different relaxation pathways – analysis by control of nonadiabatic multi-electron dynamics (NMED)**
- **photoelectron-photoion coincidence ("Reaction Microscope")**
- **experiments on $(H_2O)_n$-clusters – effect of electronic structure on NMED and optimization of a high-energy proton source**