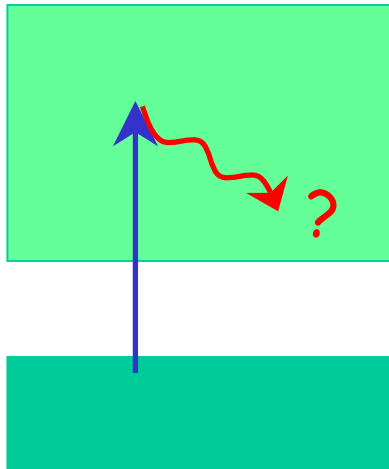


Des défauts ponctuels à l'endommagement: études résolues en temps dans les solides (diélectriques).

G. Geoffroy, J. Gaudin, G. Petite, S. Guizard.
Laboratoire des Solides Irradiés, CEA DRECAM/CNRS/X

A. Belsky, P. Martin.
Laboratoire CELIA, CNRS/Université de Bordeaux I/ CEA.



Aim: study the relaxation **dynamics** in solids after electronic excitation.

- energy distribution }
- density } → ρ (E)

cooling of excited carriers. . . : $\rho(E,t)$

- $e^- - e^-$
- $e^- - h^+$
- $e^- - \text{phonon}$

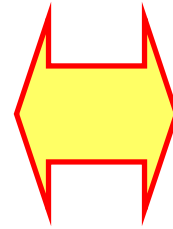


heating of lattice

- defect formation
- phase transition
- ablation / breakdown

« microscopic » probe :

- measure *emitted particles*
- electrons
 - ions, atoms, molecules, clusters
 - photon



« macroscopic » probe :

- measure *change of optical properties*
- absorption, reflectivity, interferometry
 - diffraction (disorder/ phase transition)

- + highest sensitivity
- time resolution needs two pulse excitation

- lower sensitivity
- + time resolution by classical pump - probe

1+1 - photon photoemission

Interferometric probe of Δn

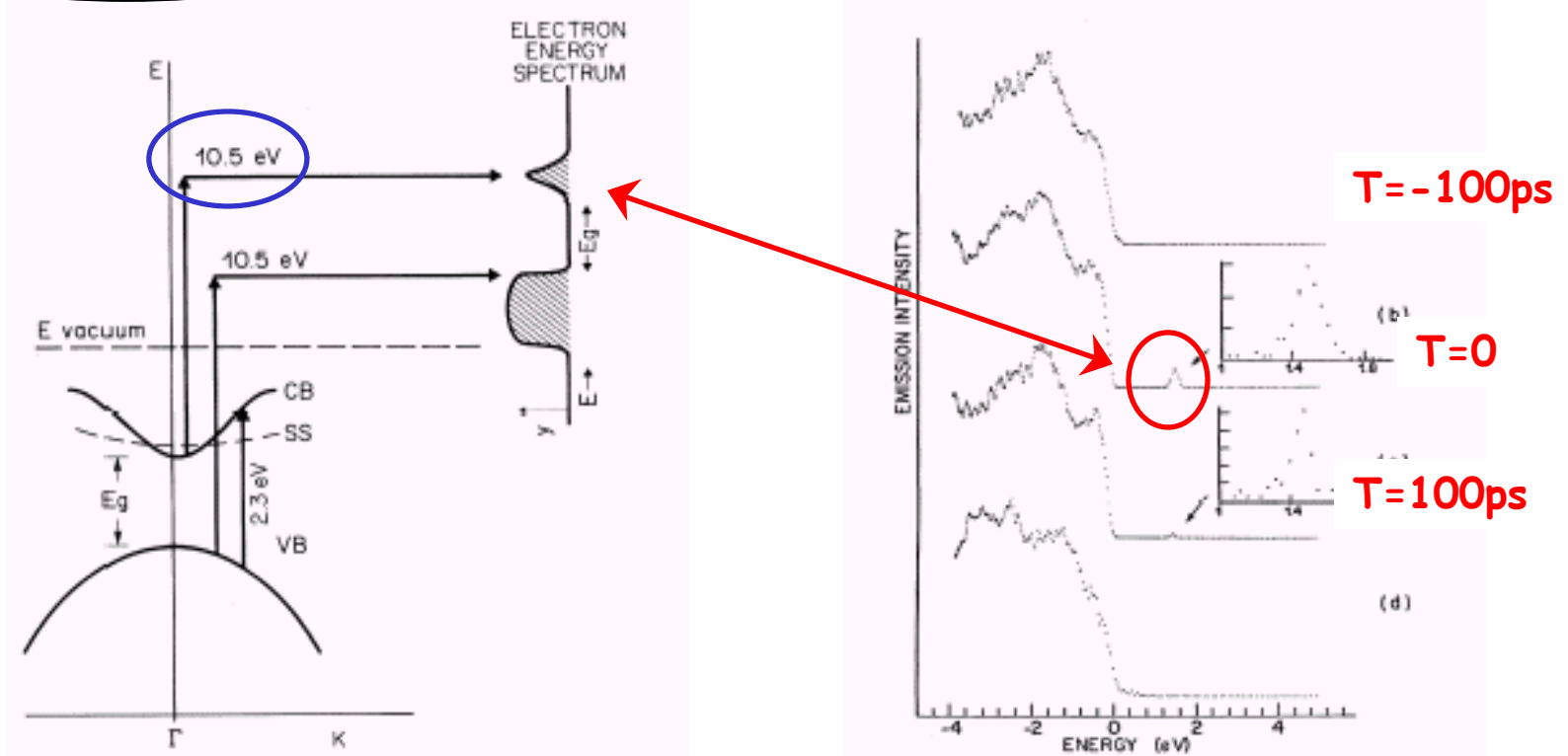
Picosecond Time-Resolved Photoemission Study of the InP(110) Surface

R. Haight, J. Bokor, J. Stark, and R. H. Storz
 AT&T Bell Laboratories, Holmdel, New Jersey 07733

and

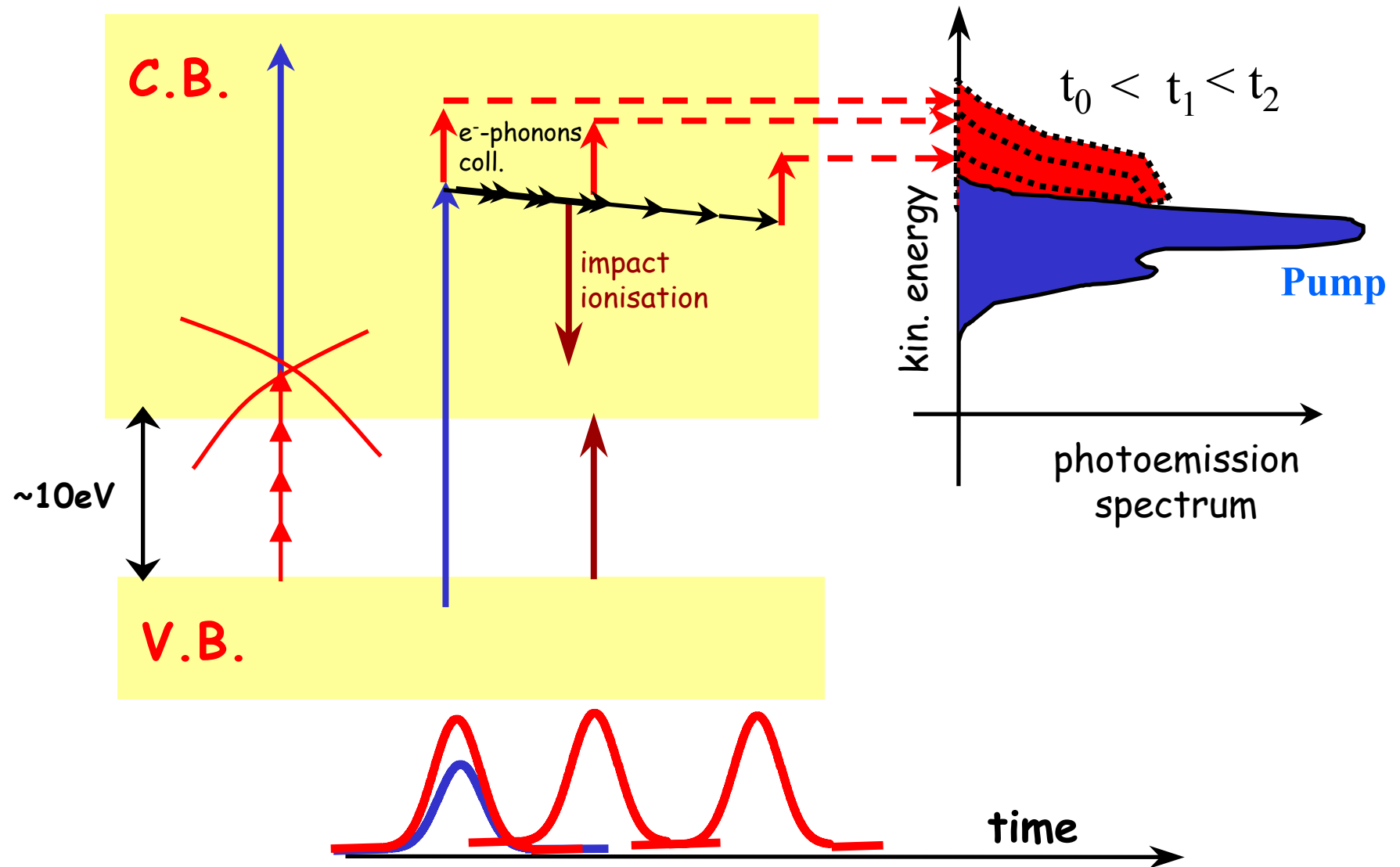
R. R. Freeman and P. H. Bucksbaum
 AT&T Bell Laboratories, Murray Hill, New Jersey 07974
 (Received 30 November 1984)

1985 : first two-colour
 time-resolved photoemission
 in the VUV

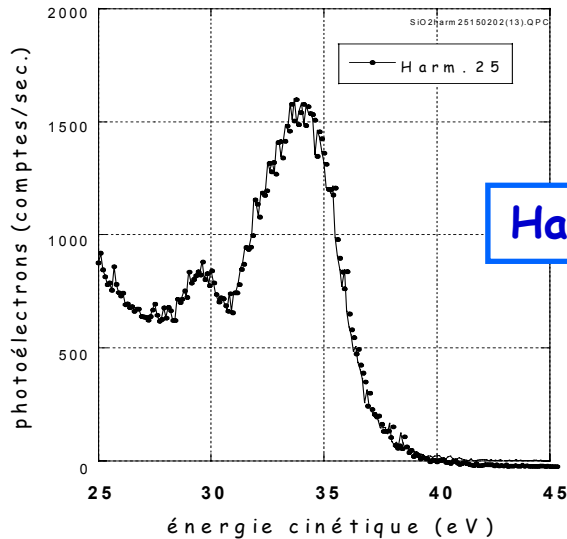


Third harmonic (in Xe) of the third harmonic of a ps-yag laser
 to probe surface state relaxation in InP.

our work : **LUCA** Saclay (B. Carré, H. Merdji, F. Quéré)
and **CELIA** Bordeaux (A. Belsky, P. Martin)



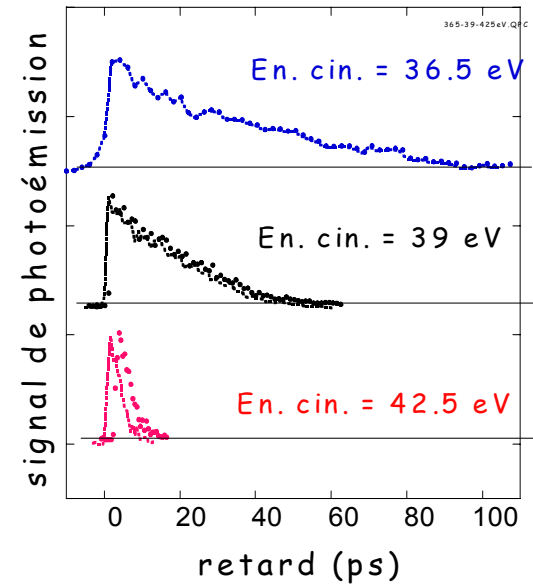
I/ photoelectron spectra with HOH



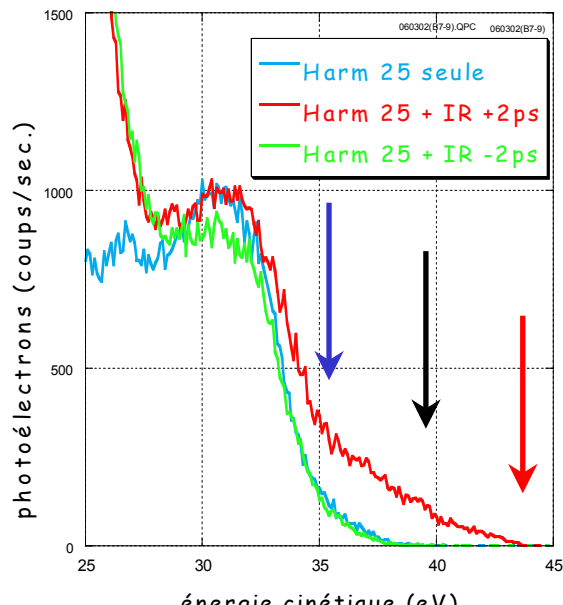
SiO₂

Harm. 25 (32nm, 39eV)

III/ measure pump-probe signal = f(t)



II/ photoelectron spectra with absorption of probe photon



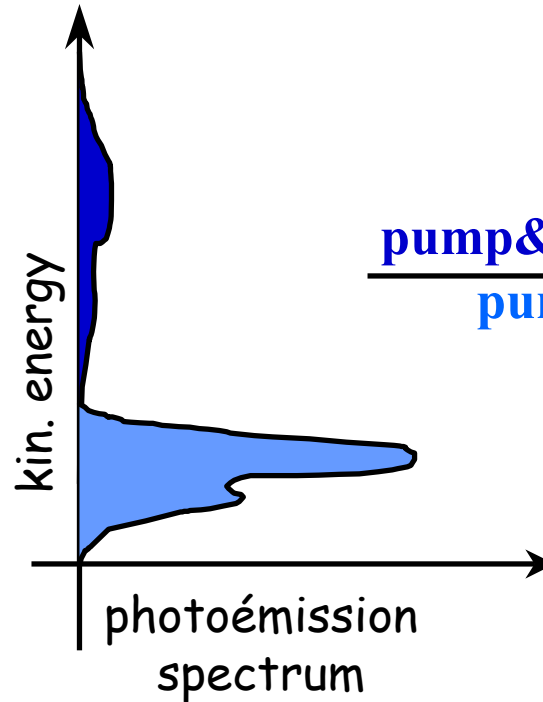
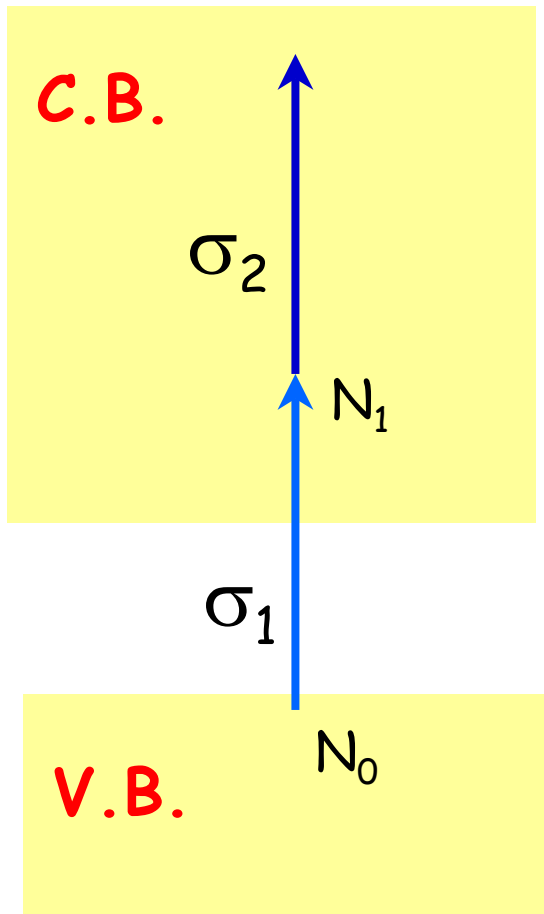
Summary :

- CsI
 - SiO₂
 - CeF₃
 - Al₂O₃
 - diamond
- H15 à H35
(22 to 54 eV)
- relaxation rates
1 to 100 ps

- + good time resolution
- need large pump intensity

Objective for Arc En ciel : observe **2 photons** or **1+1 photon** excitation in the **VUV**:

- interesting for **dielectrics** and **metals**
- **non-linear** processes in VUV
- relaxation in case of **core level excitation**



pump&probe
pump

$$\propto \underbrace{\frac{N_1}{N_0}}_{\leq 10^{-3}} \times \underbrace{\frac{\sigma_2}{\sigma_1}}_{\ll 1 (?)}$$

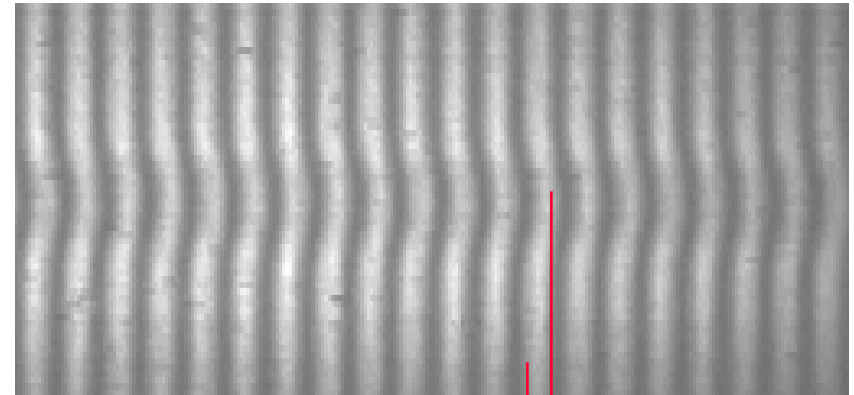
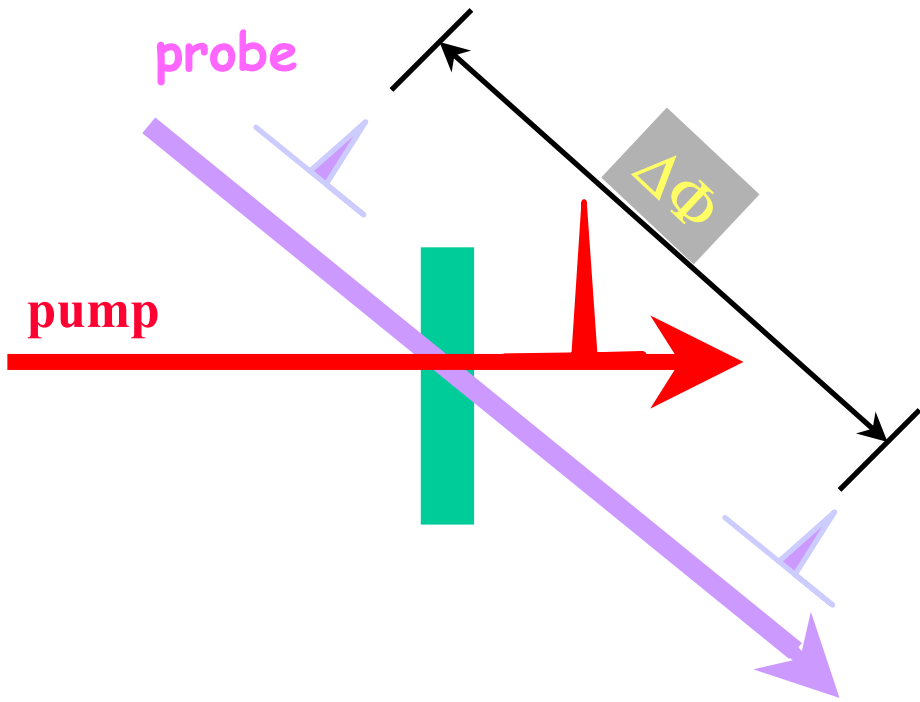
limited by :

- breakdown
- space charge

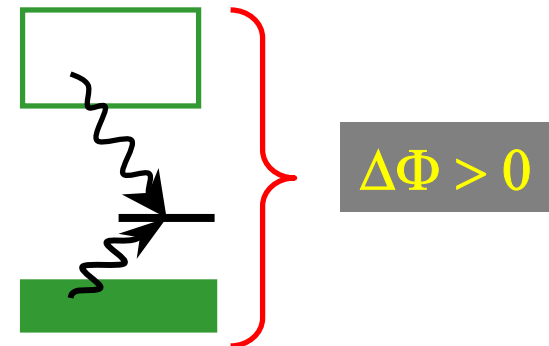
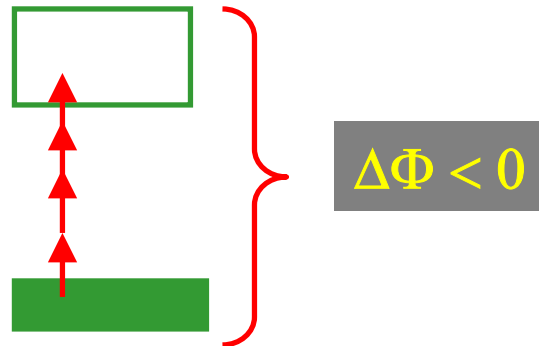
unknown : useful
for study of electronic

relaxation in solids with ultrashort Atto. pulses

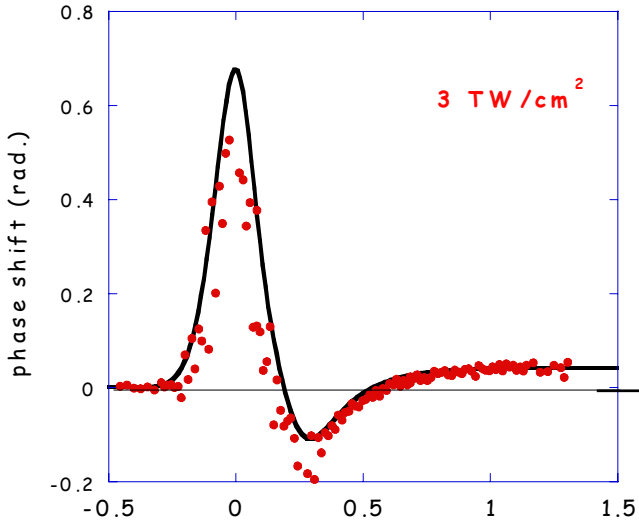
Probe electronic excitation/relaxation by time resolved interferometry :



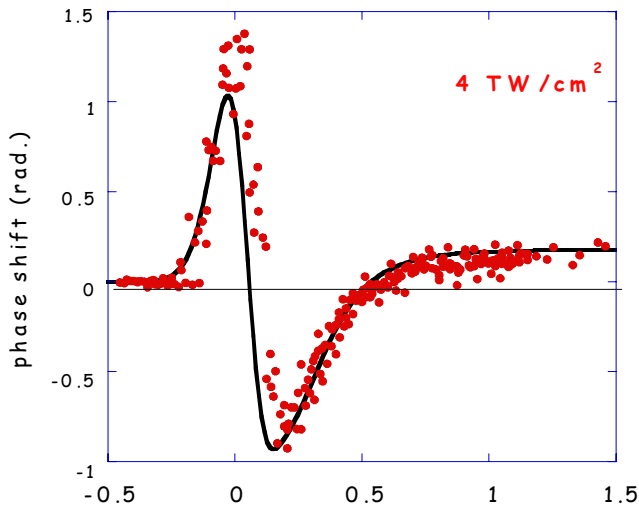
$$\Delta\Phi \propto \text{Re} [\Delta n(t)]$$



SiO₂

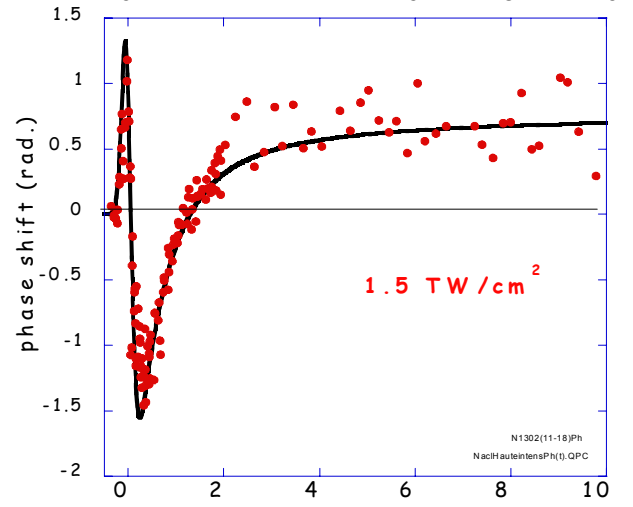
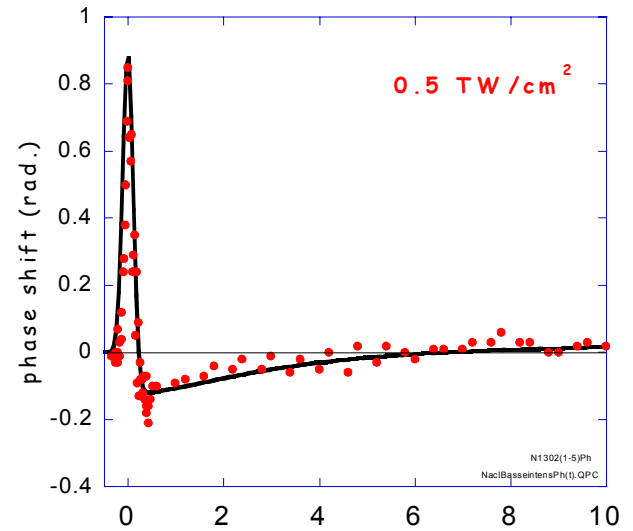


↑ e⁻ trapped
in bang gap
↓ e⁻ « free »
in C.B.



τ

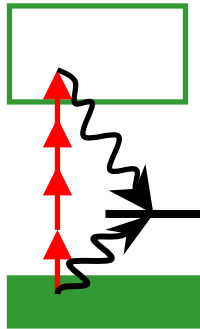
NaCl



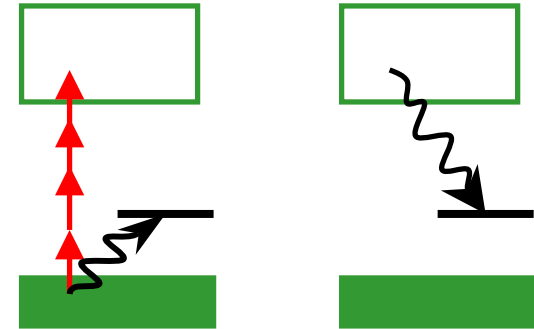
$\tau = f(\text{intensity/density})$



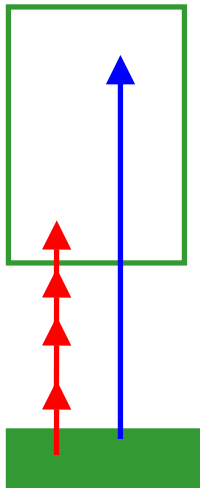
« 1 step » process :
e-h \rightarrow STE



3 steps process :
e - h \rightarrow hole trapping (Cl_2^-) \rightarrow STE



dynamics change with initial carrier density



- initial electron energy ?
- excitation of core levels ?

With HOH failed to observe signal :
modification of probe beam needs
« large » density of excited carriers !





30 nm, 1.1 mJ, 200fs, 85 μm



BREAKDOWN !!

Understand and compare VIS/ VUV interaction mechanisms at high intensity :

800 nm

- multiphoton excitation
- heating of electron in CB by sequence of (multi)photon absorption
- mechanisms:
 - single shot at threshold:
 ***coulomb explosion***
 - multiple shots or above threshold:
 ***thermal effect***
(melting and evaporation)

30 nm

- single photon excitation
- no heating of electron in CB (or very interesting)
- mechanisms:

? ?

Possible experiments: - photoelectron and photoions spectroscopy
- measure excited carrier density by :

- interferometric method
- reflectivity measurements...

Conclusion :

Arc En Ciel offers large broadening of experiments due to :

- - high intensity
- - wide tunability range
- - laser system synchronised with VUV light

Complementary from HOH source :

- - best time resolution
- - limited intensity and spectral range

New sources of HOH dedicated to the study of solid state recently appeared:
Germany: 2 (+1), Japan:1, Italy:1, USA:1, UK:1...

Europhysics letters 2002

Femtosecond time-resolved core-level photoelectron spectroscopy tracking surface photovoltage transients on *p*-GaAs, P. Siffalovic, M. Drescher and U. Heinzmann *Bielefeld, Germany*

Journal of Electron Spectroscopy and Related Phenomena 2002

Applicability of monochromatized high harmonic extended ultraviolet radiation for inner-shell photoelectron spectroscopy

M.Drescher,P.Siffalovic,M.Spieweck,U.Heinzmann *Fakultat fur Physik ,Universitat Bielefeld Germany*

Physical Review letters 2003

Time-Resolved Auger Decay in CsBr Using High Harmonics

Toshihiko Shimizu, Taro Sekikawa, Teruto Kanai, and Shuntaro Watanabe, Minoru Itoh
Institute for Solid State Physics, University of Tokyo, Japan

J. Opt. Soc. Am. B 2003

Laser based soft-x-ray pulses for photoelectronspectroscopy of surfaces

G. Tsilimis, C. Benesch, J. Kutzner, and H. Zacharias , *Münster, Germany*

Physical Review letters 2001

Direct Observation of Surface Chemistry Using Ultrafast Soft-X-Ray Pulses

M. Bauer, C. Lei, K. Read, R. Tobey, J. Gland, M. M. Murnane, and H. C. Kapteyn,
Boulder + Ann Arbor, USA

Optics and Lasers in Engineering 2002

Application of VUV laser harmonics radiation to the measurement of porous silicon dielectric fuction

F. De Filippo, C dE Lisio, P. Maddalena,S. Solimeno,G.Lérondel,C.Altucci *Naples + Potenza, ITALY*

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