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#### School on Synchrotron and Free-Electron-Laser Sources and their Multidisciplinary Applications

26 April - 7 May, 2010

Two-colour and photon correlation spectroscopy with lasers and FELs

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# Two-colour and photon correlation spectroscopy with lasers and FELs E **Fulvio** Parmigiani Department of Physics, University of Trieste and FERMI@elettra

The study of the electron dynamics relays on the ability to time-resolve the ultra-rapid scattering processes which result in energy and momentum relaxation, recombination and diffusion.

In typical experiments a short-pulsed (10-100 fs) laser can be used for photoemission experiments in the time-domain, whereas longer laser pulses (1-5 ps) provided by FT limited coherent sources can be used for photoemission experiments in the frequency (energy) domain with unrecorded resolving power.

Experimental techniques must be brought to bear in which bandstructure specificity are combined with time resolution. Angle resolved photoemission is particularly suited for such experiments.

### A glance on the photoemission



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TIME RESOLVED MULTI-PHOTON PHOTOEMISSION ( $h_V < \Phi$ )  $\longrightarrow$ band mapping of UNOCCUPIED STATES and ELECTRON SCATTERING PROCESSES mechanisms

#### **Polarization Coherence and selection rules**



### **Experimental Set-up**



## HGHG Free Electron Laser (FEL)

Ultra-fast laser pulse slicing of the synchrotron electron bunch

HHG in a hollow fiber yields a longer interaction length and "phase-matching."





#### **High Harmonic Generation laser source**





#### **Experimental Set-up**



Acceptance angle: ± 0.83° Energy resolution: 10 meV @ 2eV Detector noise: <10<sup>-4</sup> counts/s μ-metal UHV chamber

- residual magnetic field < 10 mG</p>
- Base pressure <2-10<sup>-10</sup> mbar
- photoemitted electrons detector: Time of Flight (ToF) spectrometer



G. Paolicelli et al. Surf. Rev. and Lett. 9, 541 (2002)

*Time-Resolved and Angle Resolved Photoelectron Spectroscopy – TR-ARPES and Time-Resolved and Spin Resolved ARPES – TR-SR-ARPES* (started 2006)



#### **ToF-SPIN DETECTOR**



#### Photoemission Spectra on Ag(100) single crystal



# NON-LINEAR PHOTOEMISSION on METALS and NON-EQUILIBRIUM ELECTRON DYNAMICS



Decay dynamics of non-equilibrium electron distribution in Au film:



W.S. Fann et al., Phys. Rev B 46, 13592 (1992).

#### **Detecting the unoccupied states**



G. Ferrini et al., Phys. Rev. B 67, 235407 (2003)

In most metals exists a gap in the bulk bands projection on the surface. When an electron is taken outside the solid it could be trapped between the Coulomb-like potential induced by the image charge into the solid, and the high reflectivity barrier due the band gap at the surface.







U. Hofer et al., *Science* **277**, 1480 (1997).

#### **Selection Rules**



#### • Polarization selection rules

IPSs can be excited only by the component of the electric field normal to the surface. Hence, no optical transitions are allowed by s-polarized light

## IPS k<sub>//</sub> Dispersion



### Test of the SR-TR ToF on the Au(111) surface states



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## Test of the SR-TR ToF on the Au(111) surface states



## Comparison between the 1kHz and 250kHz laser system



#### The space charge problem





FIG. 3. Space-charge effects on the photoemission spectrum of a polycrystalline gold sample under irradiation with  $\tau_0 = 60$  ps,  $\hbar\omega_0 = 34$  eV photon pulses. (a) Typical energy distribution curve with the Fermi cutoff at 29.38 eV (taken from Ref. 9). (b) Comparison between simulated and measured (Ref. 9) Fermi edge shifts and broadenings in the range of (0–2000)e<sup>-</sup> per pulse. Note the different scales of the vertical axes. Power-law fits to the simulated energy shift and broadening serve as guides for the eyes ( $\Delta E_{skift} \propto N_c^{1.1}$ ,  $\Delta E_{broad} \propto N_c^{0.83}$ ). The spot size is 0.43 × 0.3 mm<sup>2</sup> and the test electrons are emitted with an angle of 45°. In the simulation, a cosine distribution of the cloud electron emission angles is used.

(a) units) isity (arb. atul 88 90 Kinetic energy (eV) Number of electrons per pulse (exp.) (103) 200 400 600 (b) measured broadening (Pietzsch et al. measured shift 1.5+ + simulated broadening simulated shift 1.0 e) ų 0.5 50 100 150 200 Number of electrons per pulse (103)

FIG. 5. Space-charge effects on a W 4f core-level spectrum under irradiation with  $\tau_0$ =40 fs,  $\hbar\omega_0$ =118.5 eV photon pulses. (a) Typical energy distribution curve with photoemission peaks at kinetic energies of ~87 and ~89 eV (taken from Ref. 21). (b) Comparison between simulated and measured (Ref. 21) peak shifts and broadenings in the range of (0–200 000)e<sup>-</sup> per pulse. Note the different scales of the axes for the simulated (bottom axis) and measured data (top axis). Power-law fits to the simulated energy shift and broadening serve as guides for the eyes ( $\Delta E_{\rm shift}$  $\propto N_c^{0.98}$ ,  $\Delta E_{\rm broad} \propto N_c^{0.75}$ ). The spot size is 0.27×0.4 mm<sup>2</sup>. In the simulation, a cosine distribution of the cloud electron emission angles is used and the test electron acceptance angle is set to 13°.

#### The escape depth question





$$k_{out} = \sqrt{\frac{2m}{\hbar^2}} E_{kin}$$

$$k_{in} = \sqrt{\frac{2m}{\hbar^2}} (E_{kin} + V_0)$$

$$k_{out,\parallel} = k_{in,\parallel} \equiv k_{\parallel}$$

"Snell's Law"  
$$k_{\parallel} = \sin \theta_{out} \sqrt{\frac{2m}{\hbar^2} E_{kin}} = \sin \theta_{in} \sqrt{\frac{2m}{\hbar^2} (E_{kin} + V_0)}$$

Critical angle for emission

$$(\sin\theta_{out})_{\max} = \sqrt{\frac{E_{kin}}{E_{kin} + V_0}}$$





Fermi surface of BSCCO measured by <u>ARPES</u>. The experimental data shown as an intensity plot in yellow-red-black scale. Green dashed rectagle represents the <u>Brillouin</u> <u>zone</u> of the CuO2 plane of <u>BSCCO</u>.

## The escape depth question



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Panaccione et al. NIMA (2005) Torelli et al., Rev.Sci. Instr. 76, 023909 (2005)

High Resolution / High Flux beamline: ID16 @ ESRF

#### An outlook to the present and to the future

PRL 96, 017005 (2006)	PHYSICAL	REVIEW	LETTERS	13 JANUARY 2006
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#### Laser Based Angle-Resolved Photoemission, the Sudden Approximation, and Quasiparticle-Like Spectral Peaks in $Bi_2Sr_2CaCu_2O_{8+\delta}$

J. D. Koralek,<sup>1,2,\*</sup> J. F. Douglas,<sup>1</sup> N. C. Plumb,<sup>1</sup> Z. Sun,<sup>1,3</sup> A. V. Fedorov,<sup>3</sup> M. M. Murnane,<sup>1,2</sup> H. C. Kapteyn,<sup>1,2</sup> S. T. Cundiff,<sup>2</sup> Y. Aiura,<sup>4</sup> K. Oka,<sup>4</sup> H. Eisaki,<sup>4</sup> and D. S. Dessau<sup>1,2,†</sup>

PRL 97, 067402 (2006)	PHYSICAL	REVIEW	LETTERS	11 AUGUST 2006
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#### Time Evolution of the Electronic Structure of 1T-TaS<sub>2</sub> through the Insulator-Metal Transition

L. Perfetti,<sup>1</sup> P. A. Loukakos,<sup>1</sup> M. Lisowski,<sup>1</sup> U. Bovensiepen,<sup>1</sup> H. Berger,<sup>2</sup> S. Biermann,<sup>3</sup> P. S. Cornaglia,<sup>3</sup> A. Georges,<sup>3</sup> and M. Wolf<sup>1</sup>

PRL 99, 197001 (2007)	PHYSICAL	REVIEW	LETTERS	week ending 9 NOVEMBER 2007
FRL 33, 197001 (2007)				9 NOVEMBER 2

#### Ultrafast Electron Relaxation in Superconducting Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+δ</sub> by Time-Resolved Photoelectron Spectroscopy

L. Perfetti,<sup>1</sup> P. A. Loukakos,<sup>1</sup> M. Lisowski,<sup>1</sup> U. Bovensiepen,<sup>1</sup> H. Eisaki,<sup>2</sup> and M. Wolf<sup>1</sup> <sup>1</sup>Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany <sup>2</sup>AIST Tsukuba Central 2, 1-1-1 Umenzono, Tsukuba, Ibaraki 305-8568, Japan (Received 18 April 2007; published 9 November 2007)

PRL 100, 107002 (2008)	PHYSICAL	REVIEW	LETTERS	week ending 14 MARCH 2008
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#### Identification of a New Form of Electron Coupling in the Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub> Superconductor by Laser-Based Angle-Resolved Photoemission Spectroscopy

Wentao Zhang,<sup>1</sup> Guodong Liu,<sup>1</sup> Lin Zhao,<sup>1</sup> Haiyun Liu,<sup>1</sup> Jianqiao Meng,<sup>1</sup> Xiaoli Dong,<sup>1</sup> Wei Lu,<sup>1</sup> J. S. Wen,<sup>2</sup> Z. J. Xu,<sup>2</sup> G. D. Gu,<sup>2</sup> T. Sasagawa,<sup>3</sup> Guiling Wang,<sup>4</sup> Yong Zhu,<sup>5</sup> Hongbo Zhang,<sup>4</sup> Yong Zhou,<sup>4</sup> Xiaoyang Wang,<sup>5</sup> Zhongxian Zhao,<sup>1</sup> Chuangtian Chen,<sup>5</sup> Zuyan Xu,<sup>4</sup> and X. J. Zhou<sup>1,\*</sup>

### An outlook to the present and to the future



## **Photoemission on low density matter**



### **COHERENCE+TUNABILITY+POLARIZATION**

#### **Cluster and nanoparticle spectroscopy**

Spokespersons: **F. Stienkemeier**, (Univ. of Freiburg-D); **T. Moeller** (Frei University, Berlin) Co-proponents :K.Fauth (MPI- Stuttgart, D), M. Drabbels (EPFL- CH), M. Schmidt(CNRS –Orsay, Fr), U.Buck (MPI-Goettingen, D)

#### An outlook to the present and to the future



#### The 10<sup>-18</sup> s Challenge with 0.1-1 keV soft X-ray

#### Attosecond science | Nature Physics, June 2007

REVIEW

Philip H. Bucksbaum

Attosecond Spectroscopy

The motion of electrons on the atomic scale has been hidden from direct experimental access until recently. We review the revolution in technology that opened the door to real-time observation and time-domain control of atomic-scale electron dynamics, and address the expected implications of having the tools to monitor electrons with sub-atomic resolution in both space and time.



Excitation and relaxation. Electronic excitation and relaxation processes in atoms, molecules and solids, and possible ways of tracing these dynamics in real time. The labels 1+ and 2+ indicate single and double ionization.

#### ime.

#### Attoscience is the study of physical processes that occur in less than a fraction of a cycle of visible light, in times less than a quadrillionth of a second. The motion of electrons inside atoms and molecules that are undergoing photoionization or chemical change falls within this time scale, as does the plasma motion that causes the reflectivity of metals. The techniques to study motion on this scale are based on careful control of strong-field laser-atom interactions. These techniques and new research opportunities in attosecond spectroscopy are reviewed.

The Future of Attosecond Spectroscopy

#### 10 AUGUST 2007 VOL 317 SCIENCE



Fig. 2. (Left) Wave function simulating the most weakly bound electron in carbon dioxide, consisting of two p, orbitals separated by 2.32 Å. Red and blue parts of the wave function are  $\pi$  radians apart in quantum phase. (Right) Coherent scattering of a 25-eV electron from the carbon dioxide orbital out of which it was recently ejected by a strong laser field. The returning electron is incident from the right, and several views are shown over a 120-as time interval. The relative time (in attoseconds) is shown in the top right of the frames. Motion of the wave function is due to interference with the incoming electron.







P < 2 10<sup>-10</sup> mbar, T=300 K

120 fs; 1 KHz Rep. Rate  $\hbar\omega$ =3 – 5 eV ; F~100 µJ cm<sup>-2</sup>

High intensity (>GW cm<sup>-2</sup>), Spatially coherent light pulses Pulse duration (120fs) <<  $\pi^*$  excitation lifetime (ps)

ACCESS TO THREE IPS QUANTITIES : IPS PE YIELD - IPS LINEWIDTH - IPS EFFECTIVE MASS

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**ICTP-TRIESTE** 

# **BULK STRUCTURE of GRAPHITE**



Optically active in the 3-4 eV region, due to the  $\pi$  bands

van Hove singularity in the J-DOS due to the  $\pi$  bands Saddle point @ M point = HIGH ABSORPTION

Anisotropic: Surface excitations diffuse poorly in the

bulk

**IPS** band not fully studied with **NL-ARPES** 

Lehmann, PRB 60, 17037 (1999)

Layered: Possible High IPS-bulk coupling due to the presence of the Interlayer (IL) band

**IPS SENSIBLE TO BULK EXCITATIONS** 



## NON-LINEAR PHOTOEMISSION SPECTROSCOPY

INTENSE VIS / NEAR-UV LASER PULSES AS PROBE: MULTIPHOTON TRANSITIONS ( $hv < \Phi$ ) ACCESS TO EMPTY & EXCITED STATES



ABOVE TRESHOLD PHOTOEMISSION IN SOLIDS CONFIRMED USING 3.14 eV PULSES

# TWO FEATURES : IPS AND BULK $\pi^*$ SHOULDER





# MULTIPHOTON TRANSITIONS for IPS AND $\,\pi^*$

 $n^{-10^{20}} \text{ cm}^{-3} @ \text{F}=100 \ \mu\text{J cm}^{-2}$ 



HOW ABOUT  $\pi^*$  INTENSITY AND WIDTH?



# IPS YIELD AND LINEWIDTH vs. $\hbar\omega$

AT ħω=4.0 eV

PEAK IN THE IPS YIELD

STEP IN THE IPS FWHM of 60 meV

## INTENSITY INCREASE : EXPLAINED BY OPTICAL ABSORPTION + MPO CHANGE



HIGH IPS INTERACTION WITH BULK EXCITATIONS



# ANGLE RESOLVED SPECTRA: IPS EFFECTIVE MASS



COULD BE INDUCED BY THE TRANSIENT OPTICAL EXCITATION in  $\pi$  BANDS

# ROUGH, "SELF-ENERGY" APPROACH



# FITTING RESULTS

Previous results allows us to fit C-geometry (symmetric) measurements without further analysis

#### IPS EFFECTIVE MASS

**IPS FWHM** 



PEAK / STEP IN CORRESPONDENCE OF THE RENORMALIZED VAN HOVE SINGULARITY

IPS effective mass AND linewidth behaviour are linked by the model.

Requests for TR-PES -control of the photon density per pulse -control of the rr (from 10 MHz to kHz regime) -control of the polarization -photon energy between 10 eV and ~10 keV

## **Future Light Sources 1**





## **Future Light Sources 2**



12-2008 8777A29

X-ray Pulse Duration (ps RMS)

# Conclusions

- TR and SR PE-spectroscopy:
- 1. Control of the photon density in the probing pulse
- 2. Selective pumping excitation
- 3. Controlled repetition rate (from kHz uo to MHz)
- 4. Photon energy range from few eV up to few keV
- 5. Very low temperature sample holder (6-degrees freedom)

# • **Potential results:**

- 1. Dynamics of the Fermi surfaces
- 2. Electron-boson and electron-electron interactions in condensed matter (CDW, SDW and SC transition)
- 3. Electronic and magnetic structure of LDM
- 4. Dynamics of the VB and core elctrons

# **RESEARCH STAFF**



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