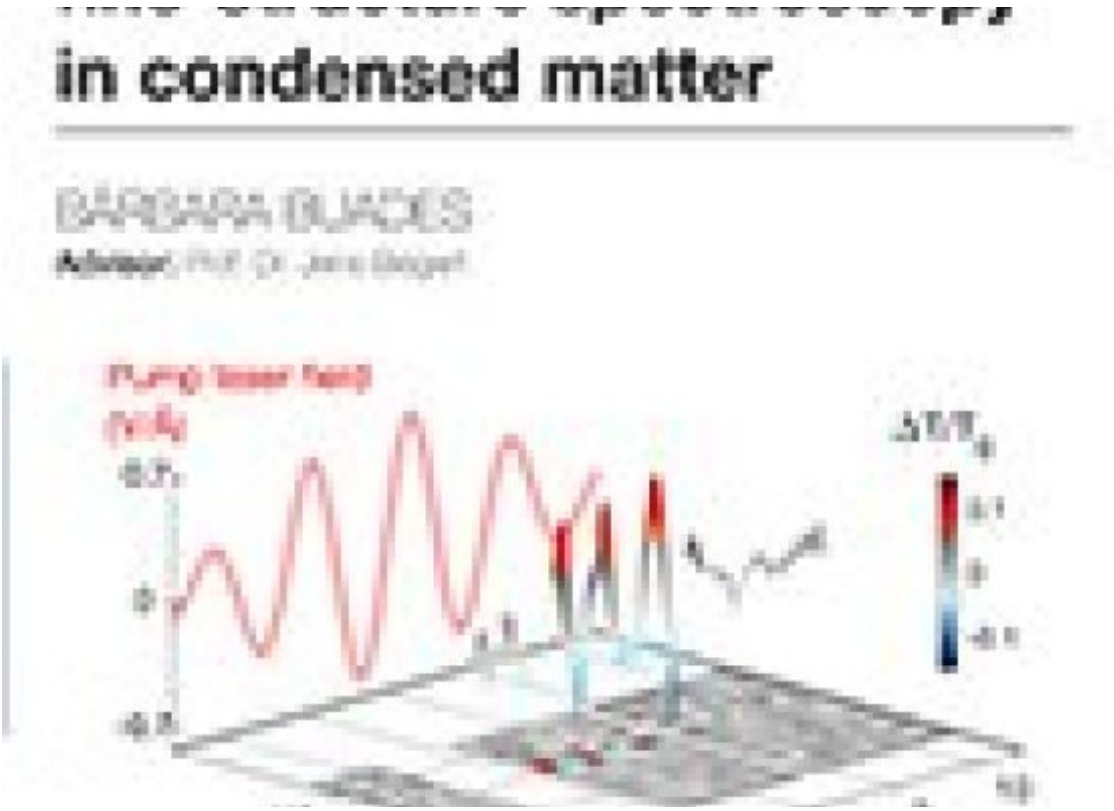

PhD Thesis Defense BARBARA BUADES 'Attosecond X-ray absorption fine-structure spectroscopy in condensed matter'

BARBARA BUADES

July 31, 2018

Thesis Defense, July 31, 2018, 11:00. ICFO Auditorium

BARBARA BUADES

Attoscience and Ultrafast Optics

ICFO-The Institute of Photonic Sciences

Attoscience aims to study electron dynamics in matter with unprecedented temporal resolution by using the shortest pulses generated on Earth. Currently, such resolution is only provided by sources that deliver attosecond pulses based on the high-harmonic generation

(HHG) process. In this thesis we make use of the demonstration of the generation of isolated attosecond pulses in the soft X-ray (SXR) regime covering the entire water window (284 eV to 543 eV) with pulse durations shorter than 300 as. Such a source is used to explore its own development, the spectroscopic capabilities of the pulses as well as the spectroscopic differences from existing X-ray sources, and finally to exploit the potential of the provided extraordinary temporal resolution.

We report on the ability to spectrally tune our source 150 eV across the water window by controlling the pressure during the HHG process and the HHG target position with respect to the focal plane of the driving laser pulse. We associate the changes in pressure and target position to a phase matching change between the driving laser pulse of the HHG process and the generated SXR radiation that is mainly caused by a change in the ionisation fraction. These phase matching changes are also compared to a carrier-to-envelope phase changes of the driving laser field.

The attosecond SXR source is used for X-ray absorption fine structure (XAFS) spectroscopy. Our XAFS studies enable the simultaneous probing of extended XAFS (EXFAS) and near edge XAFS (NEXAFS) in graphite, providing element specificity and orbital sensitivity with identification of the sigma* and pi* orbitals in synchronicity with the material?s four characteristic bonding distances. This illustrates the potential capability of correlating electron dynamics with structural dynamics with attosecond resolution being able to resolve charge migration, electron-phonon coupling and structural transitions. Our XAFS investigations also reveal spectral changes in graphite and TiS2 as a consequence of the shorter attosecond pulse compared to the longer picosecond pulse that are typically used in synchrotron facilities. An extended theory is still required to link Auger electron spectroscopy, total electron yield and XAFS using synchrotron radiation with attoXAFS to decouple the different electron dynamics involved on each of the measurements.

Finally, the attosecond pulses are employed to interrogate charge dynamics with unprecedented temporal resolution inside a compound quasi-2D material, TiS2. By synchronising a 1.85 m pump pulse with the probing attosecond SXR pulse, we observe tha the shape of the X-ray absorption line changes from a Lorentzian distribution to a Fano-typ distribution oscillating with twice the pump electric field frequency. The absorption change appear due to an acquired dipole phase response of the photo-excited core-level electro induced by the consecutive arrival of the infrared pump pulse. This demonstrates tha field-driven intra-band dynamics dominate over inter-band dynamics. SXR radiation als provides element specificity of attoXAFS which permits, in combination with theory, th visualisation of the flow of charge amongst the atoms inside the unit cell in real time. Th combined spatio-temporal capabilities of attosecond transient XAFS may prove decisive t

investigate the correlated motion of carriers in quantum materials such as phase-transition and superconductors.

Tuesday July 31, 11:00 h. ICFO Auditorium
Thesis Advisor: Prof Dr Jens Biegert

