Quantum dynamics at solid surfaces on attosecond timescales
Experimental project at Imperial
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This project explores the measurement and control of ultrafast photo-excitation of solids. The goal is to obtain new insight into the quantum dynamics of photoelectrons near the surfaces of solids on the attosecond time-scale and atomic length scale. These dynamics involve a number of complex and interconnected phenomena that are currently not well understood, but are fundamental to the function of current and future nano-scale quantum devices, solar cells, sensors and the catalytic action of surfaces. Some of the relevant processes and their characteristic timescales are: thermalisation of hot electrons at the Fermi edge (10s of fs), transport time for electron from delocalised and localised core-states to surface (<1fs), charge transfer of electrons from adsorbed atom to substrate (<1fs), charge screening and image charge creation (<1fs), direct and indirect electron scattering in clusters (<1fs to fs). Key questions that this project will address include:

(i) How do photoelectron wavepackets launched inside a metal or semiconductor propagate to the surface and on what timescales?
(ii) How are the photo-excitation and electron wavepacket propagation modified by adsorbate layers?
(iii) Can photoelectron emission and photocurrents from surfaces be controlled on the nanoscale and on the few femtosecond to attosecond timescales by tailoring both samples and optical waveforms?

This project is well-suited to a student looking for a challenging experimental PhD with a focus on cutting-edge attosecond science and its applications in ground-breaking quantum experiments. Experimental skills will be gained in the following areas: femtosecond high-intensity lasers, high harmonic generation, electron- and x-ray spectroscopy, attosecond-streaking, ultra-high vacuum and surface science techniques. There is also significant scope for computational work within the project, including development of control and data acquisition software, and writing computer simulations with the support of theorist.

6 month project

In the Attosecond Laboratory H007 in the Physics Department at Imperial College, we have recently developed the capability to temporally-resolve photoelectron wavepackets emitted from surfaces following photo-excitation by soft x-ray attosecond pulses using attosecond streaking (see Fig.1). We were the first group in the world to make such measurements from gold and semiconductor surfaces which are the basic ingredients of nanoplasmonic and optoelectronic devices.

Figure 1 A schematic of attosecond streaking at a surface. A photoelectron wavepacket is emitted through the interaction of an attosecond XUV pulse with a surface. The photoelectrons receive a momentum kick from a synchronised femtosecond NIR laser pulse which either increases or decreases the photoelectron energy depending on the instantaneous vector potential of the NIR pulse. The oscillations of the NIR field thus act as an attosecond resolution “clock”. By recording the photoelectron energy spectrum as a function of the time delay between the XUV and NIR pulses, a streaking trace is built up, from which the full photoelectron wavepacket (phase and amplitude) can be reconstructed. The inset on the right shows the retrieved photoelectron wavepacket we measured from a polycrystalline Au surface illuminated by a 250as XUV pulse with a central photon energy of 90eV. The broadening of the photoelectron wavepacket compared to the XUV pulse is attributed to a range of photoelectron emission depths near the surface.

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The goal of the 6 month project is to repeat our earlier photoemission measurements that used XUV (90eV) attosecond pulses, but now to temporally characterise the photoelectron wavepackets liberated by VUV attosecond pulses\(^2\) in the range 15-25 eV. These measurements (initially on Au and W/WO\(_3\) samples) will be the first of their kind and will shed new light on the photoelectron wavepacket dynamics in a photon-energy range where correlations and quantum effects will be more manifest, and where the electron dispersion relation can exhibit rapid variations in the group velocity with energy\(^3\). Theoretical support will be provided by our collaborator Professor Uwe Thumm\(^4\) at the University of Kansas, who is a world-leading authority in the quantum modelling of attosecond dynamics in solids.

**Full PhD Project**

The full project will extend the measurements to systems with chemisorbed and physisorbed adsorbate layers and epitaxially grown metal-metal and dielectric-metal interfaces to explore how photoelectron propagation dynamics are modified by adsorbates and how ultrafast dynamics in the adsorbates themselves can be extracted using attosecond streaking. Insights into the effect of adsorbates is relevant to photo-catalysis and solar-cells, e.g. metal nanoparticle plasmon sensitised semiconductors where electron transfer from the photoexcited plasmon into the semiconductor surface has to proceed within 20 fs to compete with plasmon relaxation\(^5\). In tandem, the project will also investigate, for the first time for surface streaking measurements, the use of longer wavelength MIR streaking fields to reduce the noise background from above threshold ionisation (ATI) electrons which presently hamper experiments. A natural extension of the research described above is the temporal control of photoemission on the few femtosecond to attosecond time scale. A variety of control schemes will be explored, including chirp control of the attosecond pulses and tailoring of the target composition and geometry.

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