Mid-IR Pump, EUV Probe Femtosecond Time-and-Angle-Resolved Photoemission Spectroscopy

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Abstract: Using EUV high harmonic probe in time- and angle-resolved photoemission spectroscopy extends the energy and momentum observation window for studies of electron dynamics in condensed matter, while tunable mid-infrared pumping allows control of excitation mechanisms.

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1. Introduction

Novel materials such as topological insulators and graphene have great potential for use in optoelectronic or electronic applications. Using these materials as solar cells or laser materials requires understanding of ultrafast dynamics of the electron populations in response to incident light. Time- and angle-resolved photoemission spectroscopy (tr-ARPES) is a powerful technique for directly observing electron dynamics in condensed matter. By measuring the momenta of electrons ejected from a layered material by a pulse with photon energy higher than the work function plus binding energy, the energy and momenta of states occupied in the material can be retrieved.

Static ARPES is now a widely used technique on synchrotrons, where photon energies in the range 10-100 eV are typically used to be able to access states at the edges of the Brillouin zone. Laser based ARPES with 6 eV pulses has added temporal resolution to the technique [1], but the low photon energy limits the observation window in both energy and momentum space. Using EUV pulses produced by high harmonic generation as the probe overcomes this restriction [2,3].

In this paper, we describe a beamline for time-and-angle resolved photoelectron spectroscopy, with tuneable pumping from the UV to the mid-infrared, and sub-30 fs EUV probe pulses from 10-80 eV. Demonstration measurements are presented showing ARPES data from tantalum disulphide with infrared pump pulses from 1.3 to 6 μm and 21 eV EUV probe. The ability to tune the photon energy of the pump pulse adds an extra dimension to the physics accessible as it allows the excitation mechanism to be varied, enabling studies of population inversion [4] and charge multiplication [4, 5] in graphene.

2. EUV and mid-IR beamlines

The drive laser system for the beamline is a 1 kHz, 30 fs Ti:Sapphire laser system with an average power of 14 W. Of this, 8 W pumps an OPA system, giving 50 uJ per pulse at 3 μm wavelength and 20 uJ at 11 μm. The remaining laser energy is used to generate coherent EUV radiation through high harmonic generation. The EUV beamline enables single harmonics to be isolated using a monochromator based on a single grating used in the conical diffraction orientation, maintaining sub-30 fs pulse durations from 10 eV to 100 eV [6]. The EUV is focused on the sample with a gold-coated toroidal mirror at grazing incidence. The mid-IR pump pulse travels outside the vacuum beamline, in nitrogen-filled purge boxes to reduce absorption. It is focused by a spherical mirror and enters the toroidal mirror chamber through a ZnSe window. Spatial and temporal overlap of the nearly co-linear pump and probe beams is obtained first by overlapping the 800 nm zero-order light from the monochromator with the mid-IR pump. EUV and pump are then overlapped by imaging fluorescence from a Ce:YAG crystal inserted at the position of the sample.

3. UHV chamber for time- and angle-resolved photoemission

The ARPES chamber is a double-mu metal shielded UHV chamber (base pressure 8×10^{-11} mbar) equipped with a hemispherical electron analyser. Differential pumping along the beamline enables the pressure to stay below 3×10^{-10} mbar. A 5-axis liquid helium-cooled manipulator cooled allows sample temperatures to be controlled to below 10 K and stabilized to within 300mK. Samples are introduced to the chamber via a load-lock and can be characterized with LEED and a helium lamp. The electrons are detected on an imaging MCP and phosphor screen, with typically 3×10^4 counts s^{-1}. The energy resolution of the system is 130 meV.

4. ARPES measurements with mid-IR pump

The mid-IR pumping capability of the system was tested on tantalum disulphide, 1*T*-TaS_2. TaS_2 is a charge density wave material, which we have studied extensively in the same chamber with 800 nm pump and EUV probe. With 800 nm pumping, prompt collapse of the Mott and charge density wave gaps can be observed, followed by Mott gap recovery after a few hundred fs [7], demonstrating that we can observe dynamics on a 30 fs timescale with 150 meV energy resolution. To test the mid-IR pumping, we used the signal, idler and difference frequency ranges from the OPA, spanning the range from 1.3 µm to 4 µm, and looked for pump-probe signal using the laser-assisted photoelectric effect (LAPE) [8].

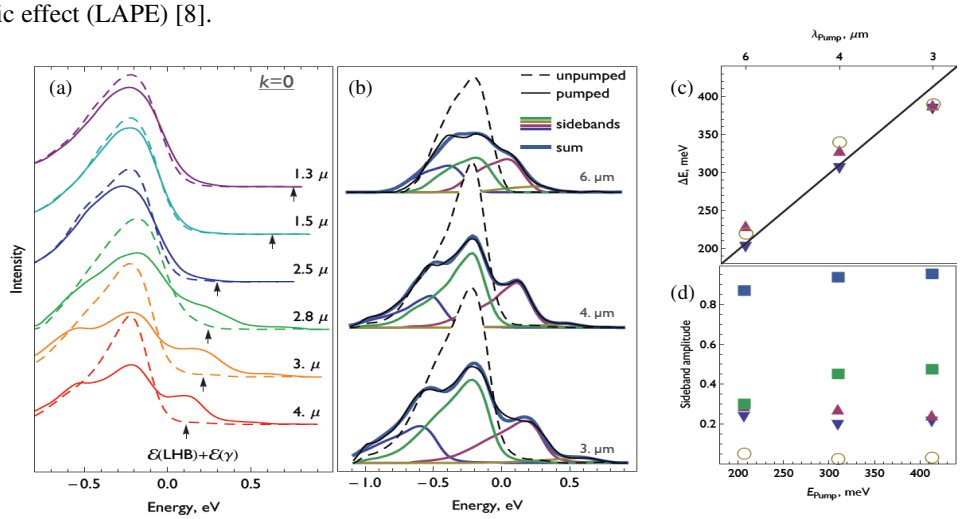

Figure 1. (a) Line-shapes in TaS_2 measured through ARPES at k=0 with EUV probe for a range of wavelengths in the infrared. The solid lines are the un-pumped signal, and the dashed lines are pumped. The arrows show the expected position of LAPE sideband. (b) Pumped signal fitted by summing discrete scaled copies of the un-pumped signal. (c) Fitted sideband offset versus pump photon energy and (d) sideband amplitudes.

Fig. 1a shows the energy distribution curves at k=0, for wavelengths from 1.3 µm to 4 µm. The arrows indicate the expected position of the LAPE sidebands, which are clearly visible with p-polarised 2.8 µm to 4 µm pump. Fig. 1b shows fits to the data. Discrete copies of the original line shape are each offset and scaled to fit their sum to the pump-induced spectrum. The sideband positions are one pump photon in energy from the main peak (Fig. 1c) and add to almost the original amplitude (Fig 1d), and so almost fully explain the effect of pumping. This confirms that ARPES measurements with mid-IR pump and EUV probe have been obtained.

5. Conclusion

We have demonstrated an experimental facility for ARPES measurements in condensed matter with mid-IR pump EUV probe. The mid-IR pumping allows the photon energy to be tuned to change the excitation mechanism of the sample, while the EUV probe allows dynamics throughout the Brillouin zone to be tracked. This capability, together with sub-30 fs EUV pulses and 130 meV energy resolution offer a unique capability for ARPES and photoelectron spectroscopy on liquid and gas-phase targets. The beamline has been used for experiments on a number of materials including graphene, where direct measurement of the electron dynamics enabled with EUV ARPES and mid-IR pump has allowed the observation of population inversion with a terahertz bandgap [4] and offered insights into charge multiplication [4, 5]. This illustrates the wide potential applicability of the technique for evaluating materials for novel applications in opto-electronics.

6. References

[1] L. Perfetti et al, "Ultrafast Electron Relaxation in Superconducting $Bi_2Sr_2CaCu_2O_{8+\delta}$ by Time-Resolved Photoelectron Spectroscopy", Phys Rev Lett **99** 197001 (2007).
[2] S. Mathias et al, Rev Sci Instrum **78** 083105 (2007).
[3] T. Rohwer et al, Nature **471** 490 (2011).
[4] I. Gierz et al, "Snapshots of non-equilibrium Dirac carrier distributions in graphene", Nature Materials **12** 1119 (2013).
[5] J.C. Johannsen et al, "Direct view of hot carrier dynamics in graphene", Phys Rev Lett **111** 027403 (2013).
[6] F Frassetto et al, "Single-grating monochromator for extreme-ultraviolet ultrashort pulses", Opt Express **19** 19169 (2011).
[7] J.C. Petersen et al, "Clocking the Melting Transition of Charge and Lattice Order in 1T-TaS2 with Ultrafast Extreme-Ultraviolet Angle-Resolved Photoemission Spectroscopy". Phys Rev Lett **107** 177402 (2011).
[8] L. Miaja-Avila et al, "Laser-Assisted Photoelectric Effect from Surfaces", Phys Rev Lett **97** 113604 (2006).