Ultrafast & XUV Science

Ponderomotive acceleration of photoelectrons in pump-probe experiments

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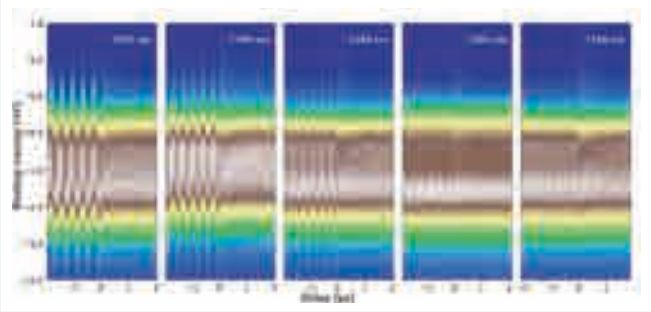
Photoemission is one of the most direct probes of the electronic band structure of a material. The application of femtosecond lasers in photoemission allows the technique to explore the dynamics of quasiparticles on their native timescales of femtoseconds to picoseconds. In some materials however, a complication can arise in photoemission spectra due to acceleration of the outgoing photoelectrons by the pump laser pulse as it reflects off the sample. This leads to an oscillation of the electron kinetic energy when the probe pulse precedes the pump. The effect is strongly wavelength dependent, increasing in amplitude as the wavelength increases into the infrared. These are the spectral regions increasingly of interest in condensed matter research, so it is important to understand the effect properly. In this work we demonstrate the influence the pump laser wavelength in pump-probe photoemission experiments on the W(110) surface and on the magnetization dynamics of Gd(0001). The effect has been observed previously on Gd and explained by Bovensiepen et al. [1] by ponderomotive acceleration of the outgoing photoelectrons by a transient optical grating formed by interference between the incoming and reflected laser pulse. We observe two significant differences from the published results. Firstly, the amplitude of the oscillations before time zero in our experiment is up to 50 times larger than observed previously, and secondly electrons excited from different initial states can show different oscillation amplitudes. The latter is surprising, as electrons should be screened within a fs and not react differently to a transient potential. We have extended the validity of the model of ponderomotive acceleration to correctly include the time when the pump and probe pulses overlap. This has allowed us to identify unequivocally a transient increase of the magnetizaton of gadolinium on the timescale of 100fs after excitation by a femtosecond laser pulse.



Oscillatory ponderomotive acceleration by the pump pulse of photoelectrons ejected from W(110) by XUV photons at 36eV. The pump wavelength decreases from left to right: 1610nm, 1450nm, 1384nm, 1300nm, 1148nm.

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Soft x-ray interferometry for pump-probe spectroscopy and hyperspectral imaging

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During high harmonic generation, laser-driven electrons may return to their parent ion and recombine along two predominant paths, known as the "long" and "short" quantum trajectories. The relative amplitude and phase of these trajectories plays a key role in the microscopic and macroscopic response of the generating medium to the driving laser field. Developing interferometric techniques capable of accessing the individual trajectory contributions to each harmonic is crucial for accessing the quantum dynamics inherent in high harmonic generation.

We report on an interferometer operating at extreme ultraviolet wavelengths in which trajectory-dependent interference can be identified. This interferometer is based on two longitudinally separated sources driven by the same laser pulse, such that it is inherently synchronized. The high stability of this technique, combined with sensitivity to quantum trajectories, offers a new route for the measurement and timing of ultrafast processes.

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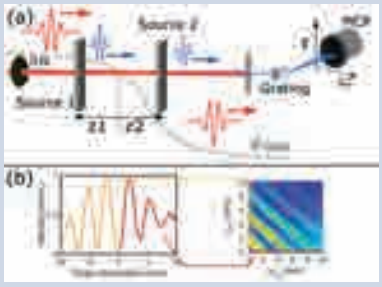
(a) Experimental setup. The spatially-resolved harmonic spectrum generated from two longitudinally separated gas targets is recorded as a function of the positions (z1,z2) of each gas cell. The Gouy phase shift across the focus and variation in the atomic dipole phase results in interference between the harmonics generated in each target. (b) Left: The on-axis harmonic interference resulting from moving a single cell about the focus while keeping the other static. Right: On-axis interferogram for q=15 resulting from moving both cells about the focus.



Ultrafast Multiphoton Photoelectron Circular Dichroism in alpha-Pinene

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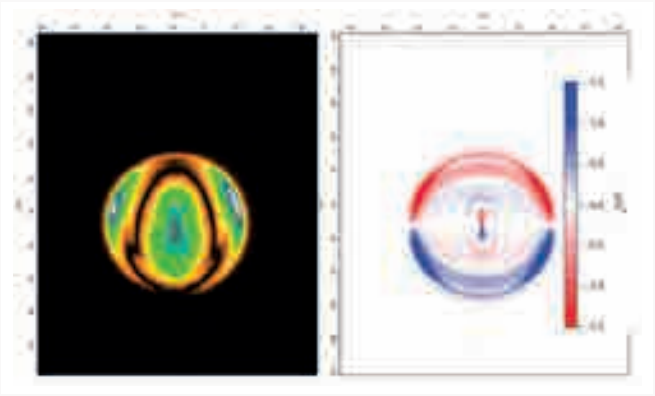
Photoelectron Circular Dichroism (PECD) was recorded by using velocity map imaging (VMI) to record the angular distributions of photoelectrons emitted upon ionization of a cooled supersonic beam of α-pinene enantiomers. As a chiral molecule, the distribution of photoelectrons has a forward-backward asymmetry (relative to the laser beam direction) allowing probing of the chirality. Measurements were made with a (2+1) resonant multiphoton ionization scheme at 400 nm, 370 nm, and with (1+1) ionization scheme at 200 nm. As can be seen in the 400 nm false colour-mapped images, very substantial chiral asymmetries (~10%) are recorded.



VMI dichroism images of R-α-pinene in a supersonic molecular beam, recorded with alternating left- and right- circularly polarised laser pulses at 396.52 nm. The laser beam propagates vertically in these figures. The left panel shows a raw 2D projection image, the right shows the antisymmetric Abel inverted 3D-slice. The red-blue colour coding of the latter indicates a strong (~10%) chiral asymmetry between electrons emitted up- and down- in the figure.

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Observation of Layer Pseudospin Interference in Bilayer MoS_2

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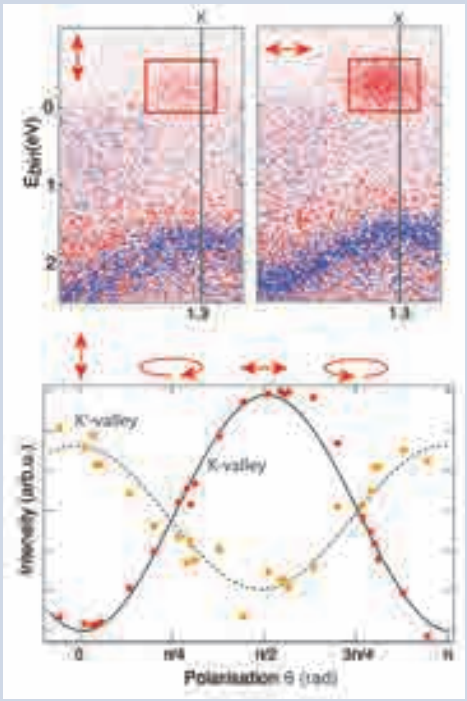
The semiconducting transition metal dichalcogenides, such MoS_2, comprise a class of materials where optical selection rules can be used to selectively excite electrons with opposite spin- and valley-quantum numbers. This dichroism effect, which is also referred to as the valley pseudospin, requires single-layer MoS_2, where the lattice inversion symmetry is inherently broken by the Mo and S atoms in the unit cell. This is not true for bilayer MoS_2 where the inversion symmetry leads to an absence of valley pseudospin polarisation. Here, we use elliptically polarized pump pulses followed by extreme ultraviolet (XUV) probe pulses to perform polarization-, time- and angle-resolved photoemission spectroscopy (TR-ARPES) to investigate the pseudospin associated with the two MoS_2 layers in a single-domain bilayer film. While the polarization of this layer pseudospin is minimal, we find an unexpected linear dichroism effect for the excited electrons in the conduction band states of the sample. A calculation of the optical transition probabilities reveals that this effect stems from an interference effect between the valleys of two MoS_2 layers. This constitutes the first observation of a layer pseudospin interference effect.



Observation of linear dichroism in bilayer MoS_2. The top panels display the response to an optical excitation with linear vertical and horizontal polarized pump pulses. In this case a larger response is seen in the case of linear horizontal polarization. The bottom graph displays the intensity of excited electrons for a range of polarization angles, exhibiting a clear polarization dependence that switches between the K and K' valleys of the material.

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