Probing Excitons in Time, Energy, Momentum, and Space by Photoelectron Momentum Microscopy

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Photoelectron Momentum Microscopy

Micron-scale

Nanoscale Dark-field Microscopy

3D ARPES

fs-Spectroscopy
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Measure:
- Kinetic energy
- Momentum
- Time dependence
- Real-space image
- Electron spin
- $h\nu$-dependence

Photoelectron Spectroscopy
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$\Delta t = -200$ fs
The Future

- Momentum Microscopy

https://scientaomicron.com

https://www.specs-group.com/
Electron microscope: electrostatic lenses image the electron beam
Momentum Microscope: Detection

- Energy dispersion: select photon energy range

https://scientaomicron.com
Momentum Microscope: Detection

- Time-of-flight detection: energy and 2D momentum for each electron

- Event-based detection: one electron per pulse

Keunecke et al., Time-resolved momentum microscopy with a 1 MHz high-harmonic extreme ultraviolet beamline, Rev. Sci. Instr. 91, 063905 (2020)
The photoelectron momentum microscope

- Light source: $\geq 500$ kHz HHG, 26 eV (20 – 70 eV)
- Pump pulses: OPA, 300 nm – 16 μm, 30 – 50 fs

Keunecke et al.,
*Time-resolved momentum microscopy with a 1 MHz high-harmonic extreme ultraviolet beamline*,
Into the third dimension
TMD Nanostructures

Nature 499, 419 (2013)

Physik Journal 18, 29 (2019).
Interlayer excitons: electron-hole separation at atomic scales

Control of opto-electronic properties through:
- Band alignment (W/M, S/Se)
- Interlayer twist
  - Moiré potential

Interlayer exciton promises:
- Enhanced lifetime
- Confined states
- Strongly-correlated phases
ARPES at the microscale?

- 2D materials are often exfoliated, and have $<100 \, \mu m$ size
- Layered structures are smaller, typically 10 $\mu m$

Sample by AbdulAziz AlMutairi and Stephan Hofmann, University of Cambridge
ARPES at the microscale?

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ARPES separates different excitons by energy & momentum
Direct access to dynamics

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Time-resolved μ-ARPES

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- Direct access to dynamics
- And excellent comparison with theory

ARPES separates different excitons by energy & momentum.

Direct access to dynamics.

And excellent comparison with theory.

microscopic modelling by Giuseppe Meneghini,
Samuel Brem, and Ermin Malic, University of Marburg
A unique momentum fingerprint

moiré superlattice hallmark!

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Threefold momentum signature:

- Umklapp scattering
  - Only for *interlayer exciton*
- Exciton confinement?
- Orbital hybridization?
  - Not expected for 10° twist

- Generalized interaction between lattices:
  - Coulomb (e-h) interaction
Exciton delocalization

- Plane wave model of photoemission:
  \[ I(k_\parallel) \propto |A \cdot k_f|^2 |\mathcal{F}(\psi)|^2 \times \delta(h\nu - E_B - \Phi - E_{\text{kin}}) \]

- Bohr radii:
  - Interlayer exciton:
    - 1.6 ± 0.2 nm
  - WSe$_2$ A-exciton:
    - 1.1 ± 0.1 nm

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Heterogeneity in 2D quantum materials:
- A major research challenge

Objective: Ultrafast nanoimaging of dark excitons

Solution: Dark field momentum microscopy
Suitable apertures in the momentum microscope select specific states:
- Exciton-specific imaging
Resolving nanoscale dynamics

Excitation of $A_W$

Formation of ILX

$\Sigma_W$

$A_W$

$WSe_2$

$MoS_2$

ILX formation map

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Correlating signatures

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Conclusions

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Spin

Upcoming!
3D orbital images

\[ I \propto |A \cdot k_f|^2 |\mathcal{F}(\psi)|^2 \times \delta(h\nu - E_B - \Phi - E_{kin}) \]

- Single photon energy: 2D information
- Many photon energies: 3D!
- HHG: ideal for 3D and fs time by time-compensating monochromation

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