From the Delay of the Photoelectron wavepacket to the Reconstruction of the Density Matrix

Eva Lindroth

Stockholm University

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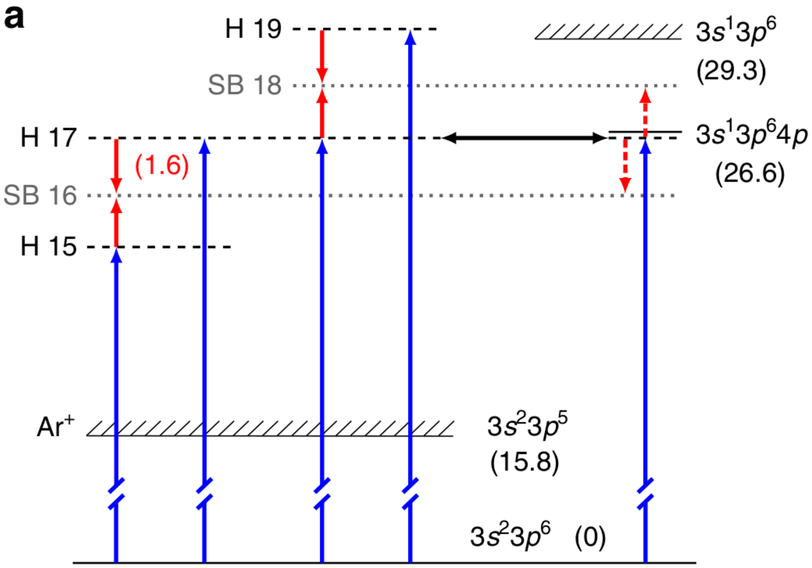
Overview

- Resonances: When the sideband symmetry breaks.
- the Kraken method - check the relation between the two-photon measurement and the desired one-photon information.

RABBIT over a resonance: ARGON example



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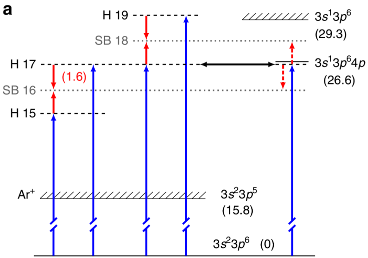

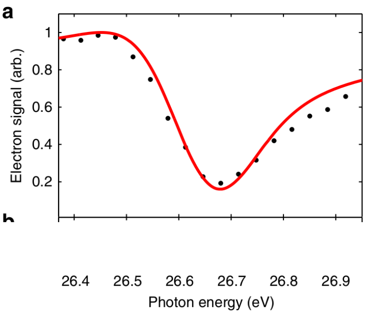

Kotur et al DOI: 10.1038/ncomms10566

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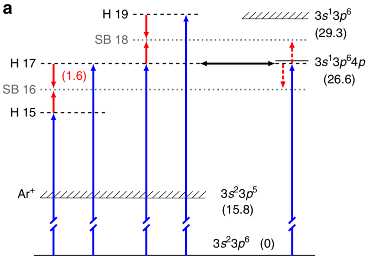

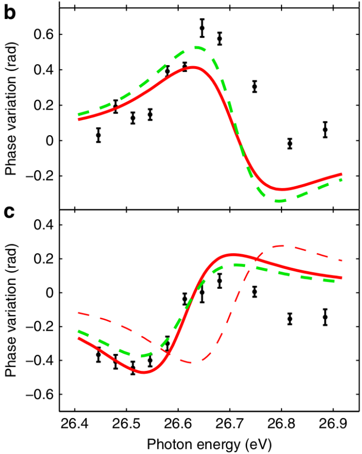

SB 16

SB 18

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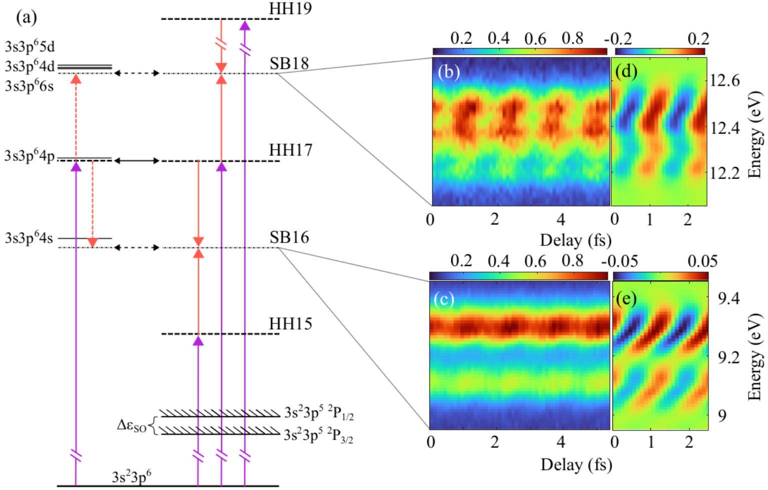
RABBIT over a resonance: ARGON example

rainbow RABBIT technique, narrow bandwidth probe (10 nmm)



Luo et al, to be published

RABBIT over a resonance: ARGON example

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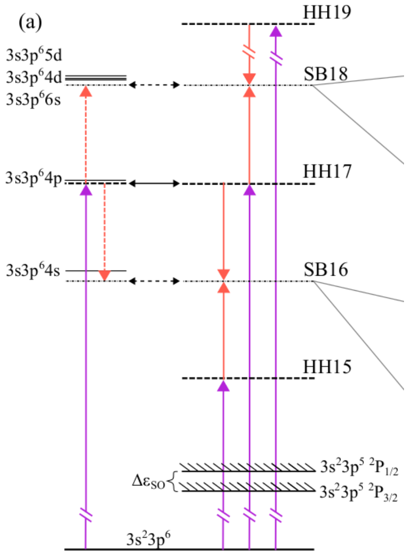
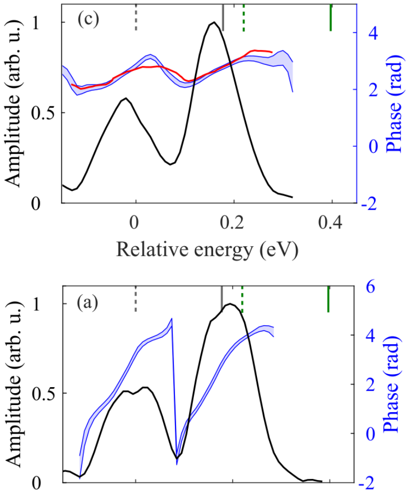




Why is the sideband symmetry broken?

- The "usual" continuum-continuum path for the 2nd photon:

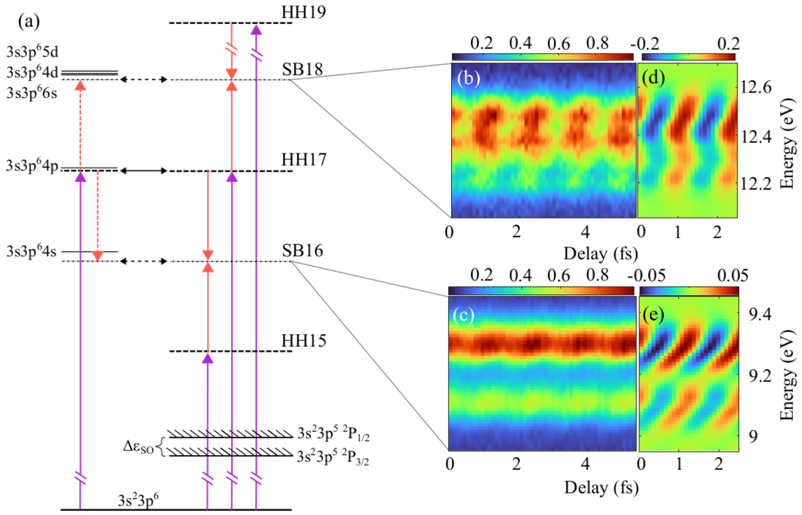
$$\text{ground state} + \hbar\Omega \rightarrow 3s^{-1}4p \rightarrow 3p^{-1}\,\epsilon s/d$$
$$+\hbar\omega \rightarrow 3p^{-1}\,\epsilon' p/f$$

- The "bound-bound" path:

$$\text{ground state} + \hbar\Omega \rightarrow 3s^{-1}4p$$
$$+\hbar\omega \rightarrow 3s^{-1}\,ns/nd \quad \rightarrow 3p^{-1}\,\epsilon' p/f$$

- does not have to be "on-shell"

Why is the sideband symmetry broken?



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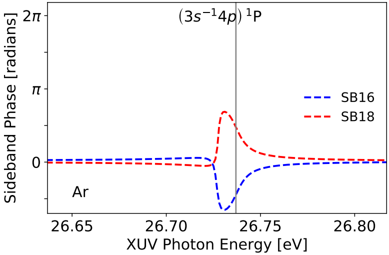

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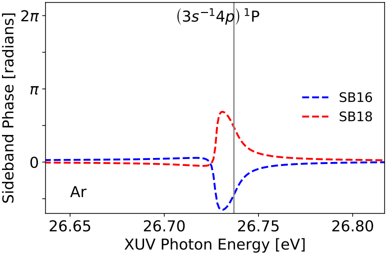
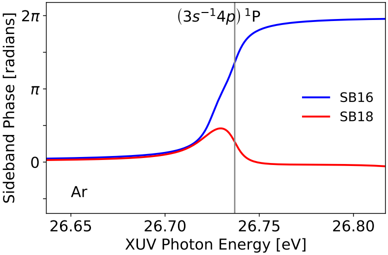


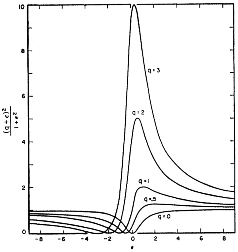


- The "usual" continuum-continuum path for the 2nd photon
- + the "bound-bound" part

Why is the sideband symmetry broken?

$$\sigma = \sigma_0 \frac{(\epsilon + q)^2}{1 + \epsilon^2} + \sigma_{bg}$$

$$\epsilon = \frac{E - E_r}{\Gamma/2}$$



- Fano PR 124, 1866 (1961)
- Asymmetric line profiles quantified by q

Why is the sideband symmetry broken?

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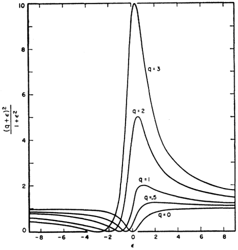
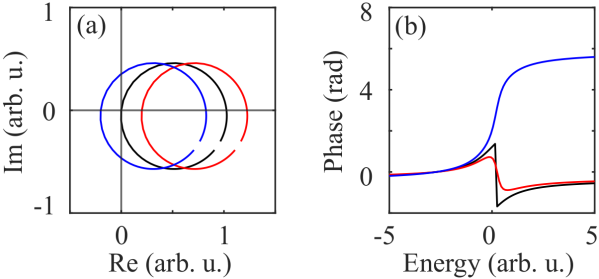


- Fano PR 124, 1866 (1961)
- Asymmetric line profiles quantified by q

$$A(\epsilon) = A_0 \frac{\epsilon + q}{\epsilon + i} + A_{bg}$$



Overview

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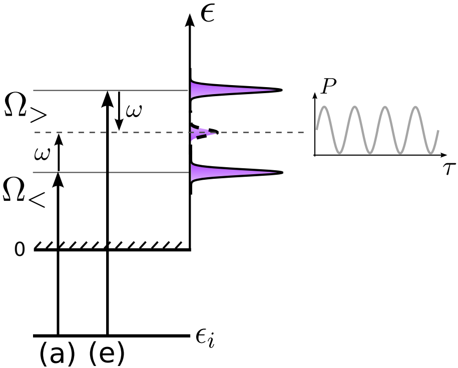
Photoionization delay

Calculations can disentangle the contribution from the second photon



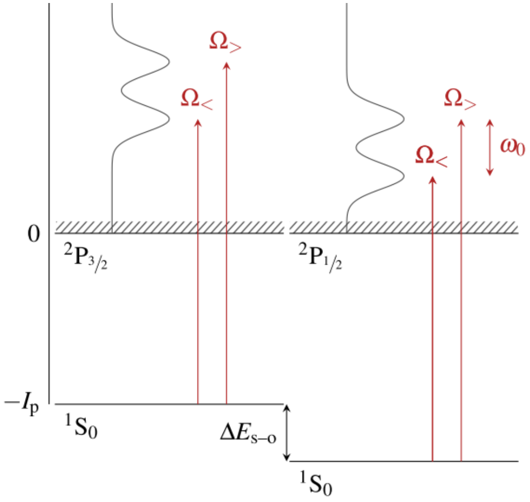
$$e^{i\eta_{emi}}e^{-i\eta_{abs}} =$$
$$e^{i2\omega(\eta_{emi}-\eta_{abs})/2\omega} =$$
$$e^{i2\omega(\hbar\Delta\eta/\Delta E)}$$
$$\rightarrow e^{i2\omega\tau_A}$$

Laser-induced sideband signal:

$$P \sim |\, M_{abs\,\omega} + M_{emi\,\omega}\,|^2 \sim A + B\cos[2\omega(\tau - \tau_{\rm GD} - \tau_A)]$$

where $\tau_{\rm GD} \approx (\phi_> - \phi_<)/2\omega$ is the group delay of the attopulse

But if we do not have pure quantum state?



from Carlström et al. J. Phys. B51,015201

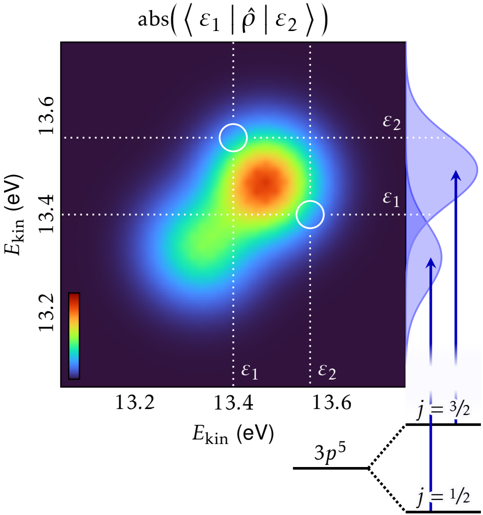
- A pure quantum state can be described by a state vector
- But often we have a mixed state

Because:

- experimental imperfections
- unobserved degrees of freedom

Measure the photoelectron

but not the ionic states (example: Ar)



The pure state:

$$| \Psi_{ion,e^-} \rangle = \sum_j^{ion} \int d\epsilon c_j(\epsilon) \, | j, \epsilon \rangle$$

Reduced density matrix:
Trace over ionic states,
and over photoelectron
angular momenta (ℓ_j).

$$\hat{\rho}(\epsilon_1, \epsilon_2) = \mathrm{tr}\left(| \Psi_{ion,e^-} \rangle \langle \Psi_{ion,e^-} | \right)$$

- Can $\hat{\rho}$ be reconstructed experimentally?

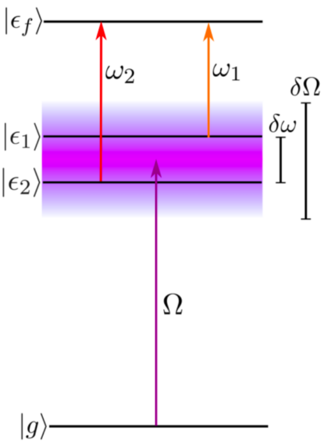
Attempts for continuum electron Quantum State Tomography

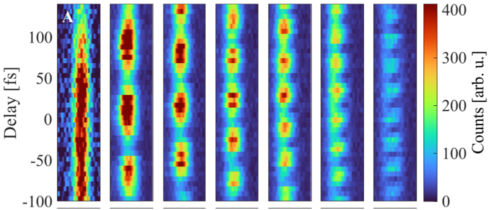
- Mixed-FROG: Bourassin-Bouchet et al, PRX 10, 031048 2020
- SQUIRRELS: Priebe et al, Nature Photonics 11,793, 2017
- KRAKEN

Kvanttillstånds tomog**R**afi av **A**ttose**K**und **E**lektro**N**vågpaket
(quantum state tomography of attosecond electron wave packets)

- Is it possible to reconstruct the density matrix?
- recall David Busto's discussion!
- How can theory contribute?

KRAKEN - THE EXPERIMENT





Photoelectron yield oscillates with $\delta\omega$ when τ is varied. $T = 100$ fs $\hbar\delta\omega = 41$ meV

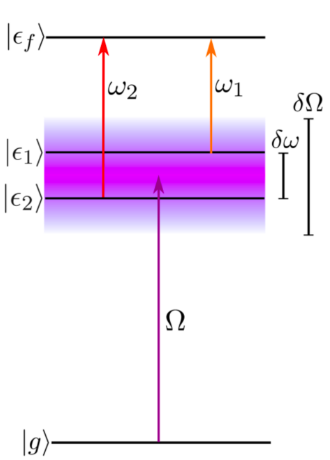
- $A_{\delta\omega}(\epsilon_f)$ and $\phi_{\delta\omega}(\epsilon_f)$ are extracted.

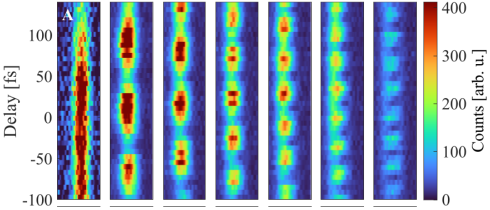
$$A_{\delta\omega}(\epsilon_f) \sim \mid \hat{\rho}\,(\epsilon_1, \epsilon_2) \mid$$

$$\phi_{\delta\omega}(\epsilon_f) \sim \arg\left[\hat{\rho}\,(\epsilon_1, \epsilon_2)\right]$$

Laurell et al arxiv.org: 2309.13945

Kraken - the Experiment





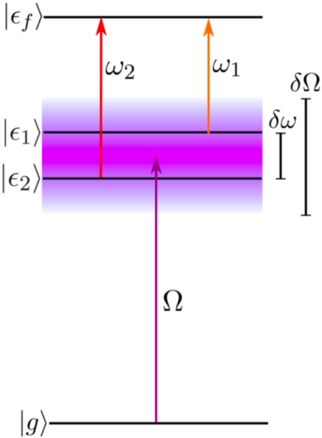
Photoelectron yield oscillates with $\delta\omega$ when τ is varied

- Obtain one photon information from a two photon measurement.

Laurell et al arxiv.org: 2309.13945

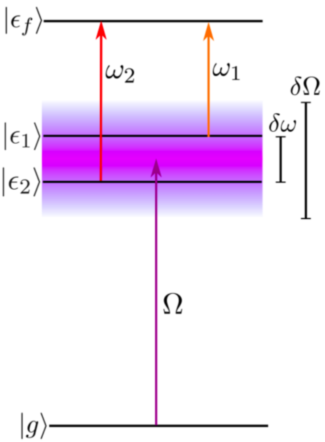
$$A_{\delta\omega}(\epsilon_f) \sim |\, \hat{\rho}\,(\epsilon_1, \epsilon_2)\,|$$

$$\phi_{\delta\omega}(\epsilon_f) \sim \arg\left[\hat{\rho}\,(\epsilon_1, \epsilon_2)\right]$$



- The density matrix after the XUV photon can be calculated

$$\hat{\rho}_{XUV}(\epsilon_1, \epsilon_2) \sim \sum_{I,\ell_j} \left\{ M^{(1)*}_{I,\epsilon_1,\ell_j} A^*_{XUV}(\Omega = E_I + \epsilon_1) \times M^{(1)}_{I,\epsilon_2,\ell_j} A_{XUV}(\Omega = E_I + \epsilon_2) \right\}$$

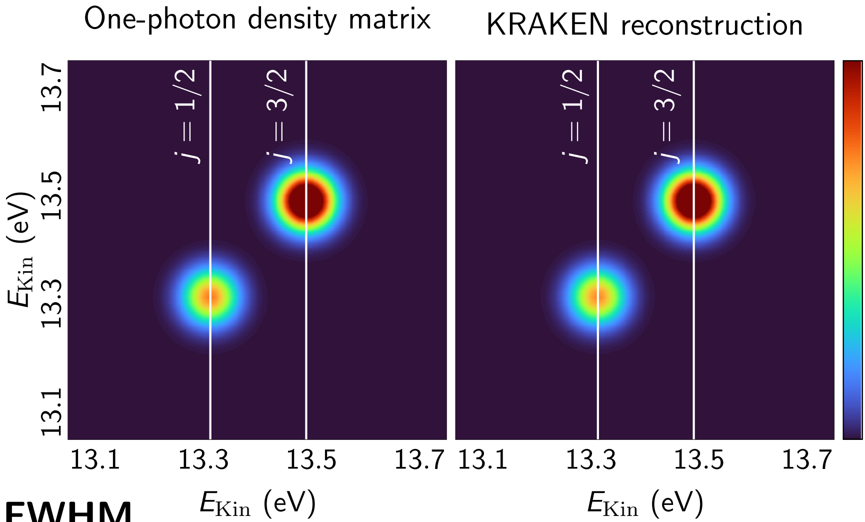


- The experimental reconstruction after the XUV photon can also be calculated

$$\hat{\rho}(\epsilon_1, \epsilon_2) \sim \sum_{I,\ell_j} \left\{ \left[\sum_{\ell'_{j'}} M^{(2)}_{\epsilon_1, \ell'_{j'} \to \epsilon_f, \ell_j} A_{XUV}(\Omega = E_I + \epsilon_1) \right]^* \times \left[\sum_{\ell'_{j'}} M^{(2)}_{\epsilon_2, \ell'_{j'} \to \epsilon_f, \ell_j} A_{XUV}(\Omega = E_I + \epsilon_2) \right] \right\}$$

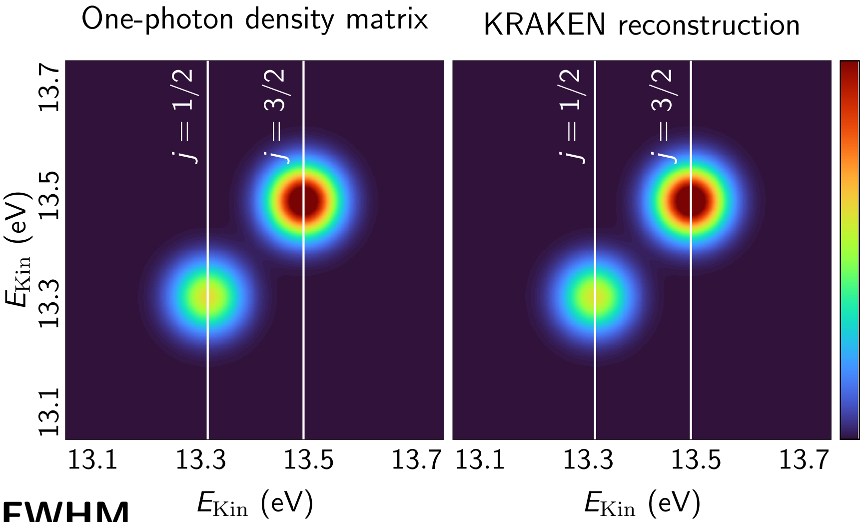
- and compared with $\hat{\rho}_{XUV}(\epsilon_1, \epsilon_2)$

The density matrix by FWHM (Ar FS=0.18 eV)



FWHM

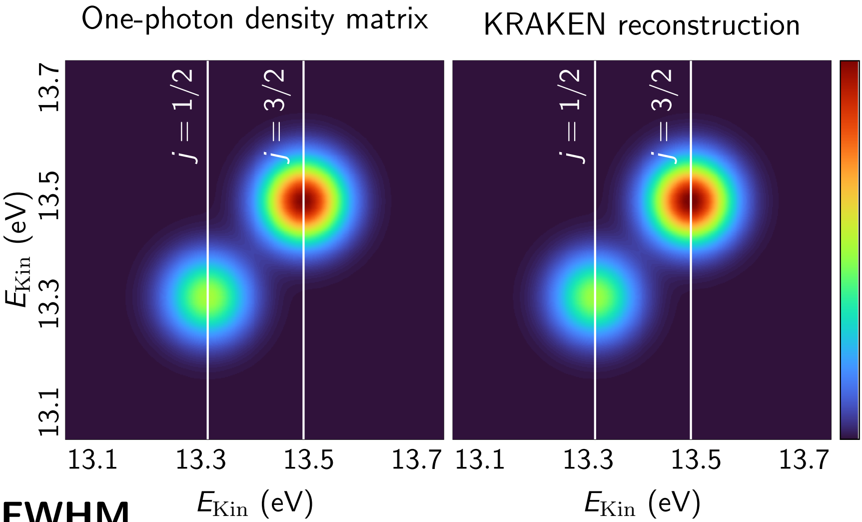
0.08eV


One-photon density matrix
KRAKEN reconstruction
$j = 1/2$
$j = 3/2$
$E_{\rm Kin}$ (eV)
13.1
13.3
13.5
13.7

FWHM

0.10eV

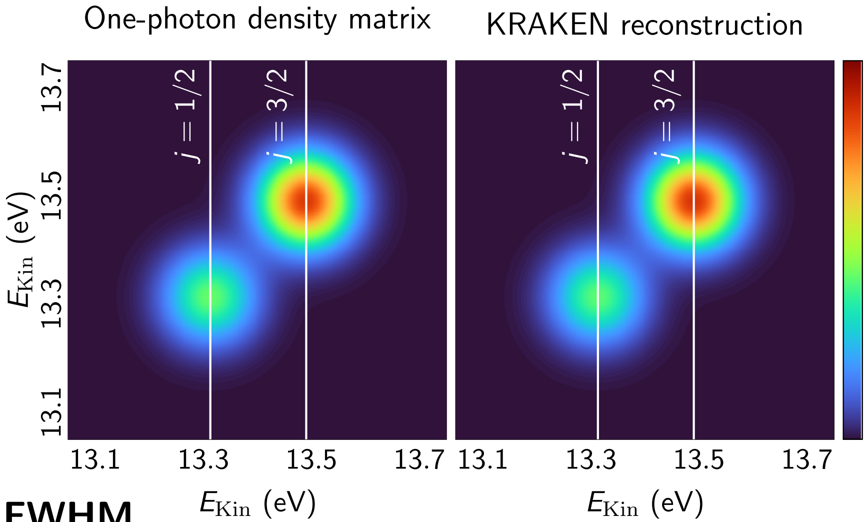
The density matrix by FWHM (Ar FS=0.18 eV)



FWHM

0.12eV

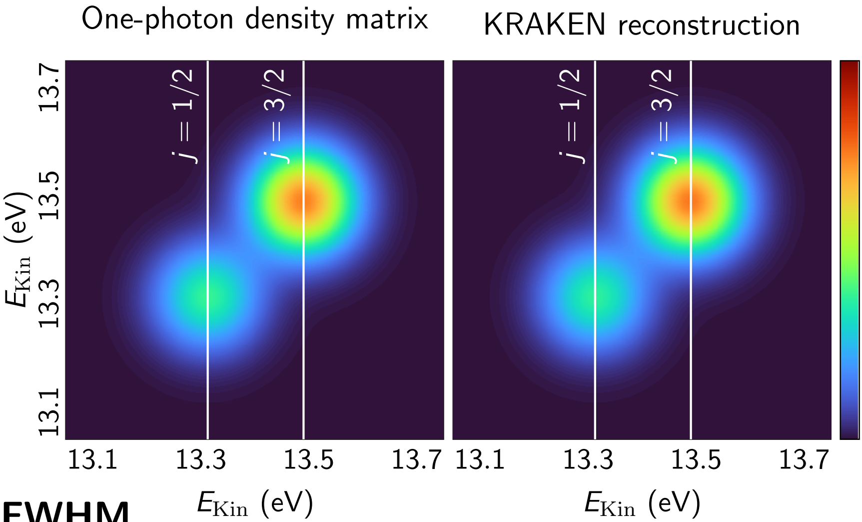
THE DENSITY MATRIX BY FWHM (AR FS=0.18 EV)



FWHM

0.14eV

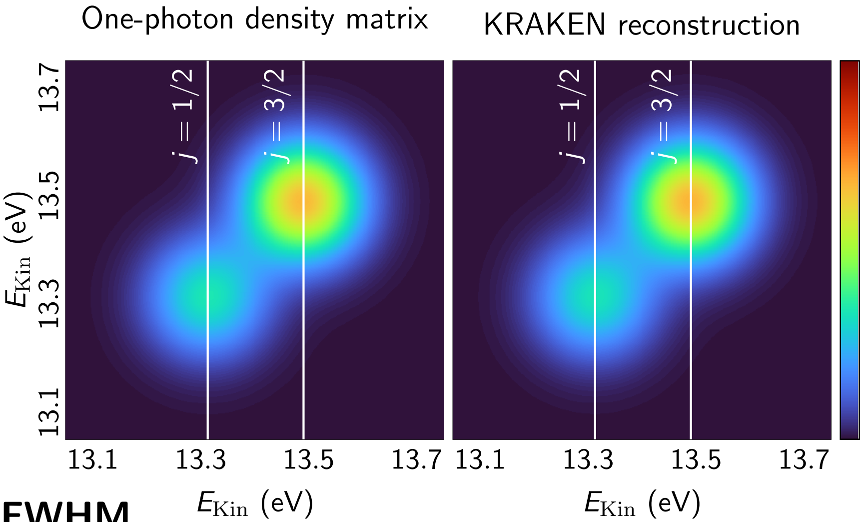
THE DENSITY MATRIX BY FWHM (AR FS=0.18 EV)



FWHM

0.16eV

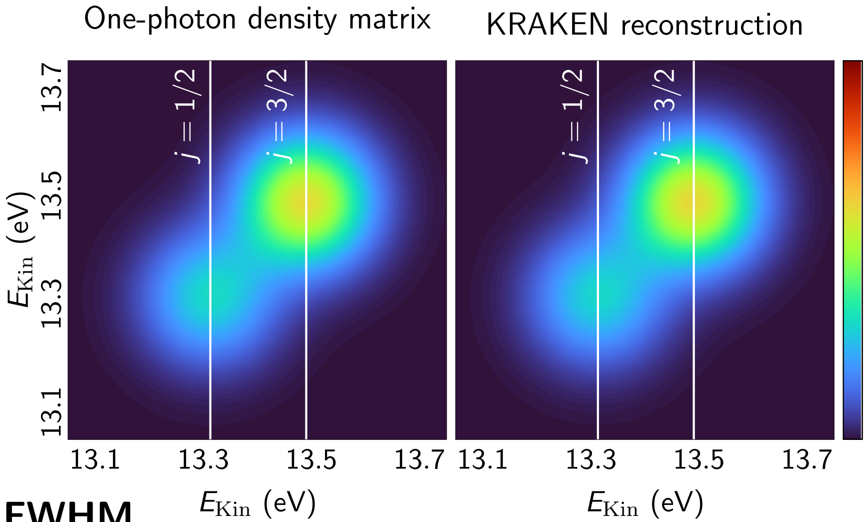
The density matrix by FWHM (Ar FS=0.18 eV)



FWHM

0.18eV

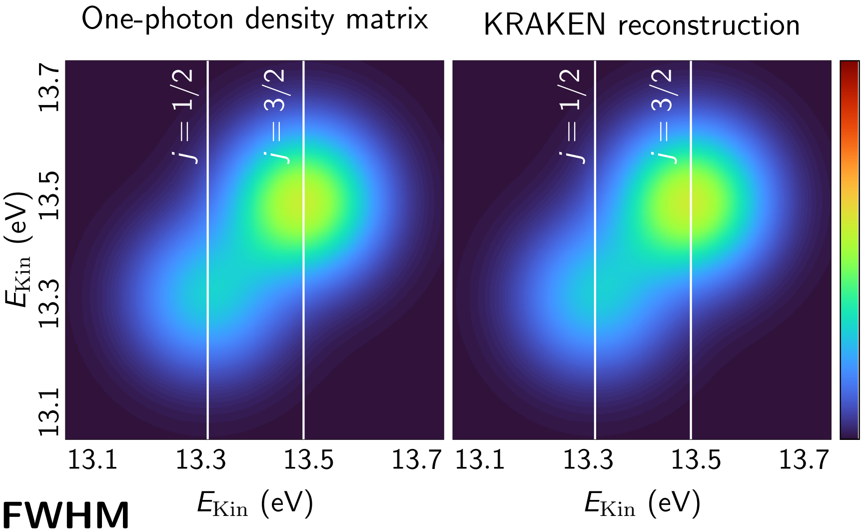
The density matrix by FWHM (Ar FS=0.18 eV)



FWHM

0.20eV

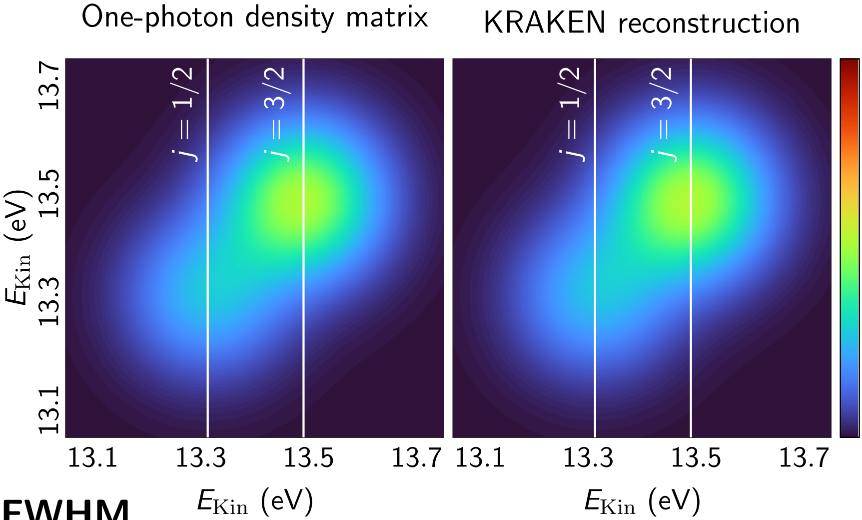
The density matrix by FWHM (Ar FS=0.18 eV)



FWHM

0.22eV

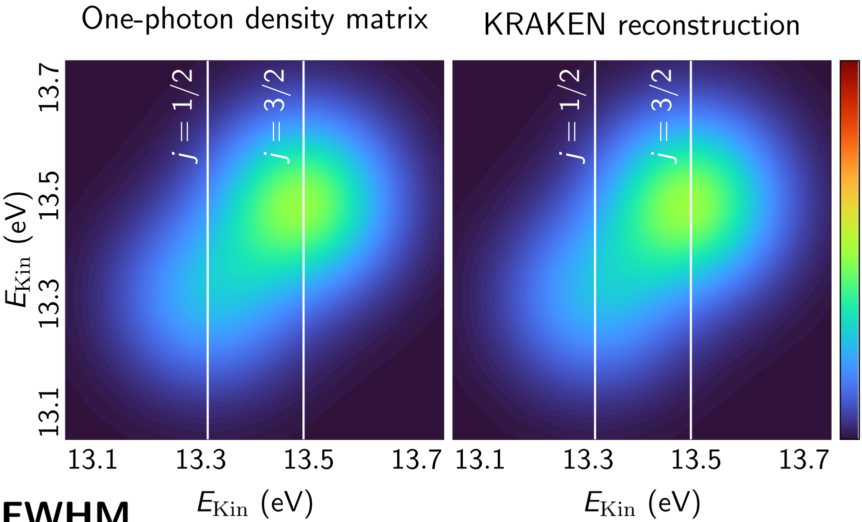
The density matrix by FWHM (Ar FS=0.18 eV)



FWHM

0.24eV

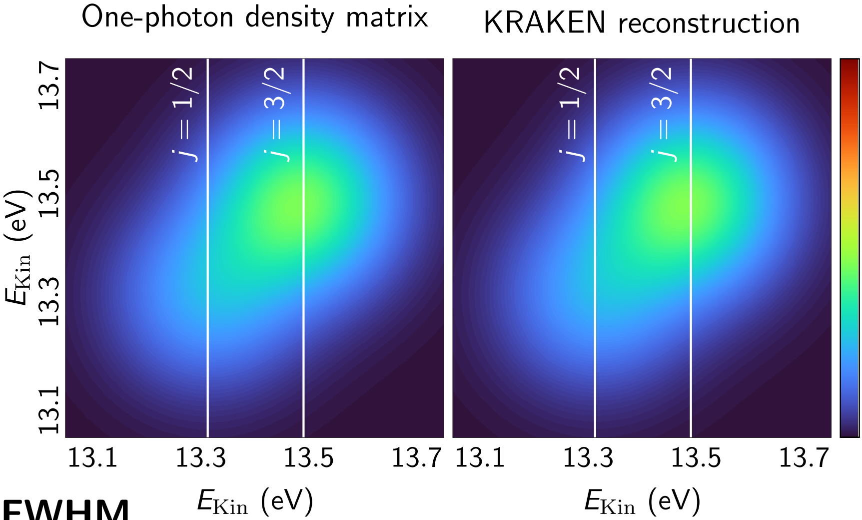
THE DENSITY MATRIX BY FWHM (AR FS=0.18 EV)



FWHM

0.26eV

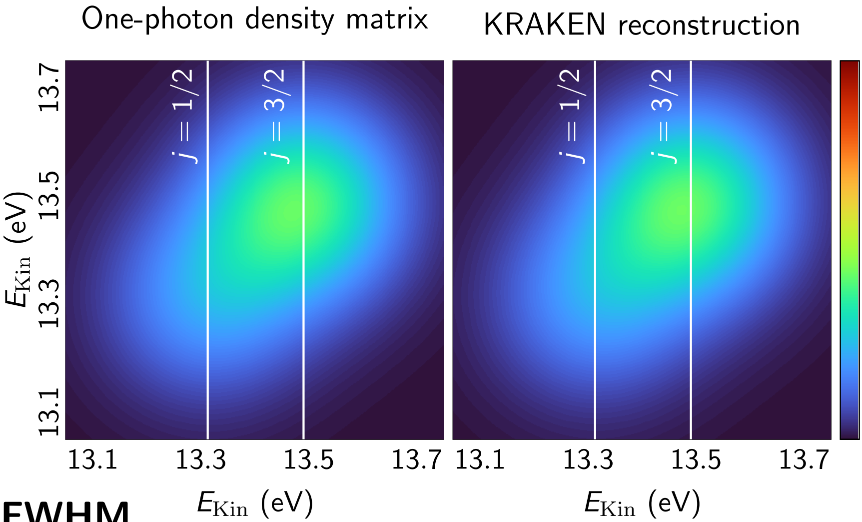
The density matrix by FWHM (Ar FS=0.18 eV)



FWHM

0.28eV

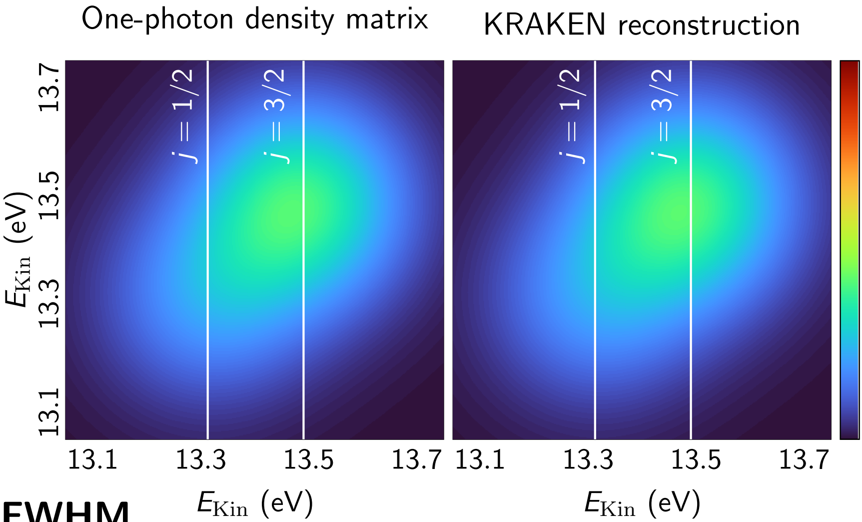
The density matrix by FWHM (Ar FS=0.18 eV)



FWHM

0.30eV

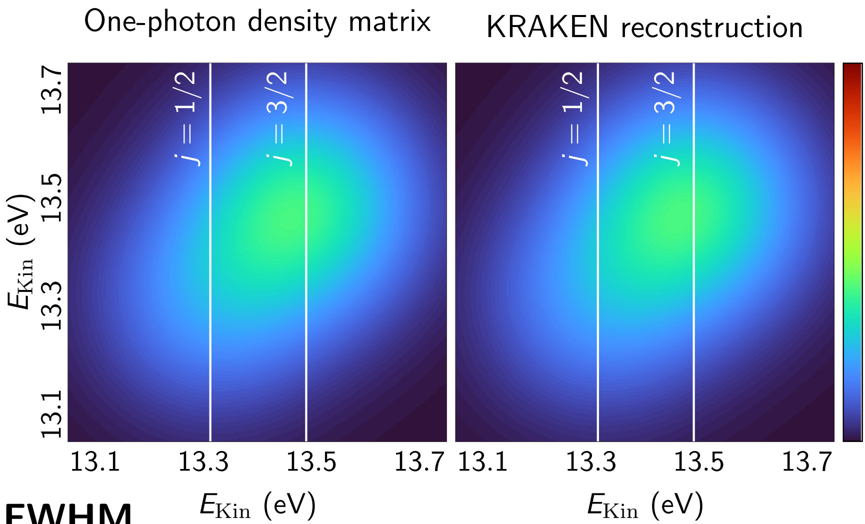
The density matrix by FWHM (Ar FS=0.18 eV)



FWHM

0.32eV

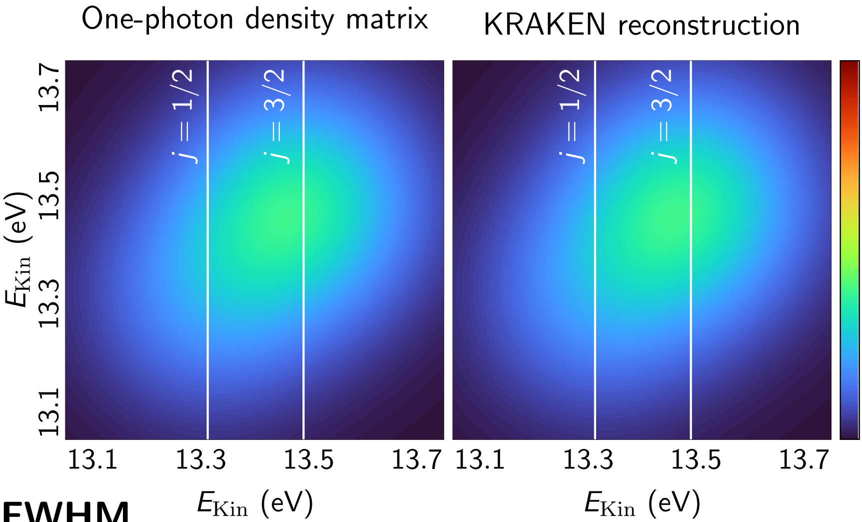
THE DENSITY MATRIX BY FWHM (AR FS=0.18 EV)



FWHM

0.34eV

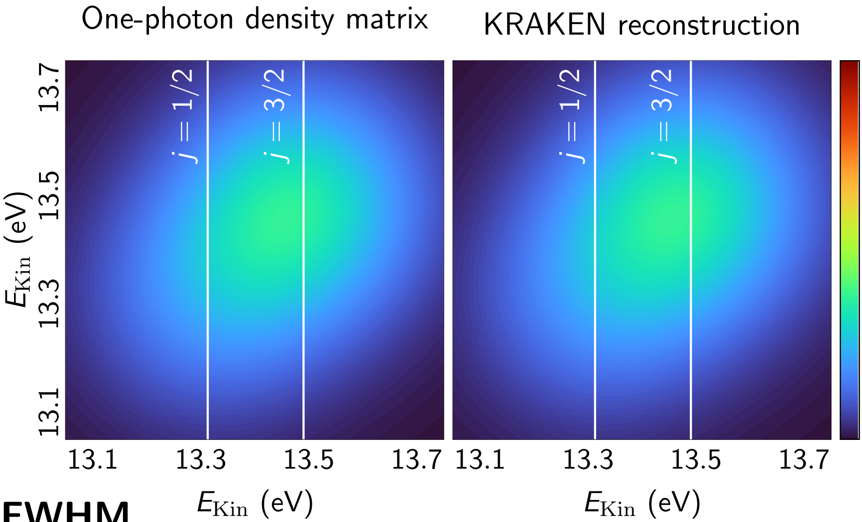
The density matrix by FWHM (Ar FS=0.18 eV)



FWHM

0.36eV

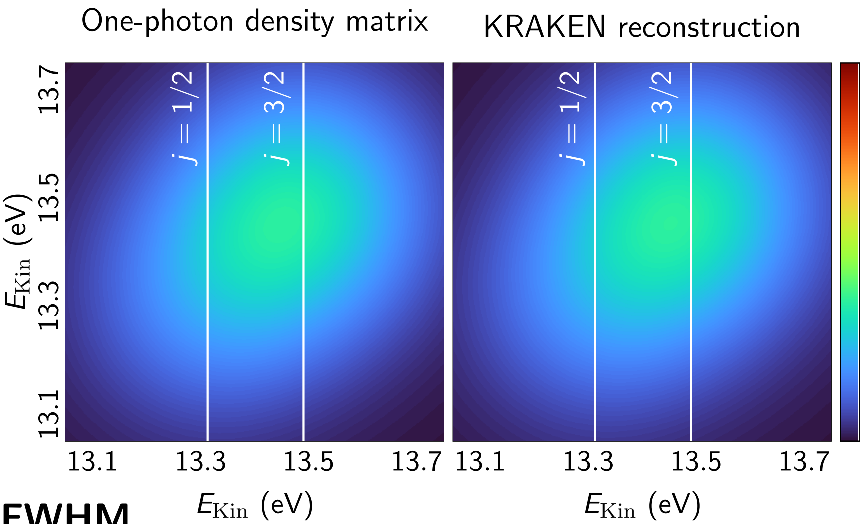
The density matrix by FWHM (Ar FS=0.18 eV)



FWHM

0.38eV

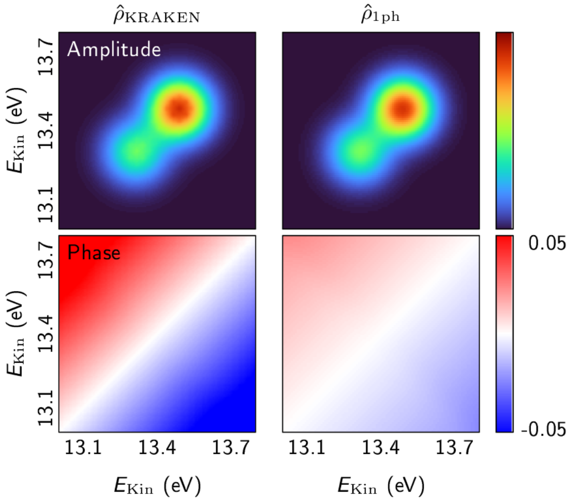
THE DENSITY MATRIX BY FWHM (AR FS=0.18 EV)



FWHM

0.40eV

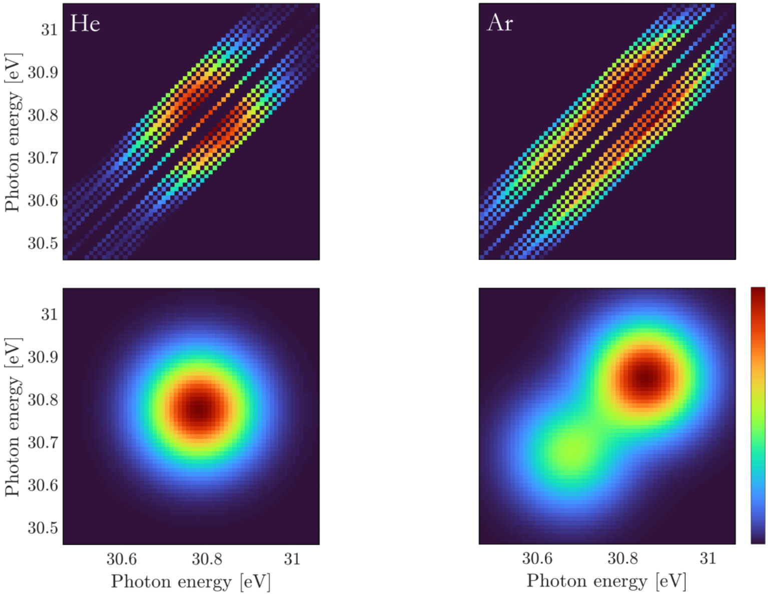
Kraken reconstruction versus One Photon



FWHM 0.14 eV, Argon

- flat part of the continuum
- 2nd photon contribution nearly constant over the interval
- 2nd photon contribution very similar in the two channels

Kraken reconstruction versus Experiment



Purity

$\hat{\rho} = A\hat{\rho}_{1/2} + B\hat{\rho}_{3/2}$
Normalization $A + B = 1$
Purity: $\gamma = \mathrm{tr}\left(\hat{\rho}^2\right)$
$\gamma = A^2 + B^2 + 2AB\mathrm{tr}\left(\hat{\rho}_{1/2}\hat{\rho}_{3/2}\right)$

- non-rel. limit $B = 2A$
- $FWHM \ll \Delta_{FS}$
  $\rightarrow \gamma = A^2 + B^2 = 5/9$

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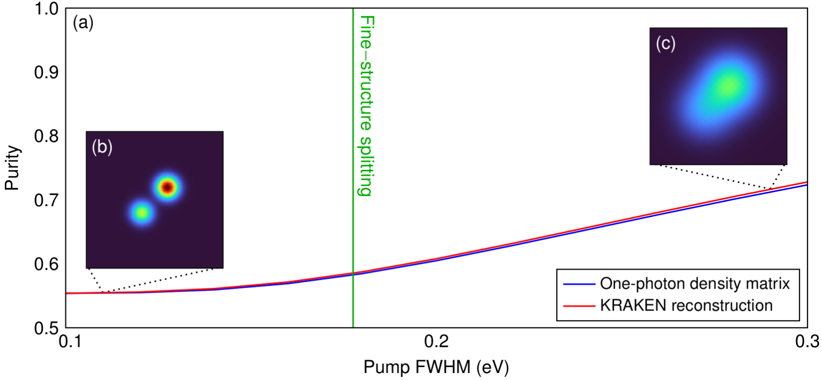

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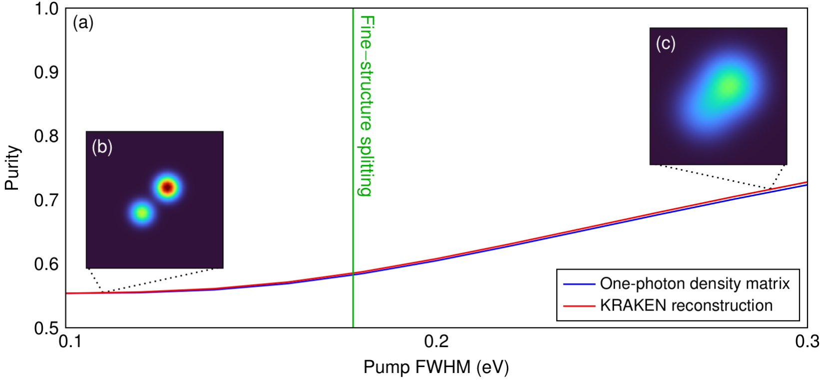


B ≈ 1.98 A, B ≈ 2.05 A

Purity

Purity (FWHM 0.2 meV)

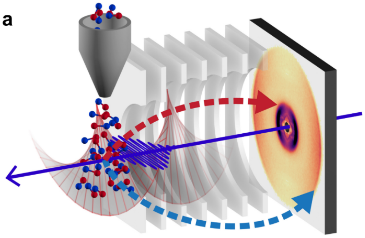
	Helium	Argon
Experiment	0.94 ± 0.06	0.65 ± 0.02
Theory	1.0	0.61

- How can the experimental purity be so high?
- compensate for detector response He purity: $0.87 \rightarrow 0.94$

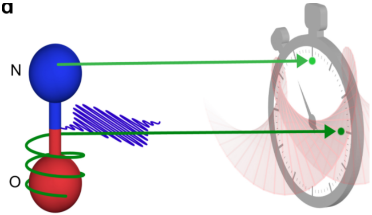
New Developments and Emerging Possibilities

X-ray Attosecond Delays

XFEL + IR-streaking laser



T. Driver et al

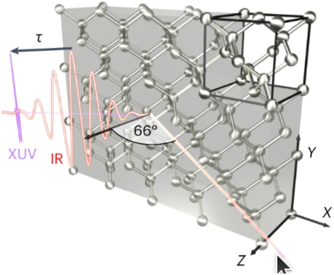


arXiv:2402.12764

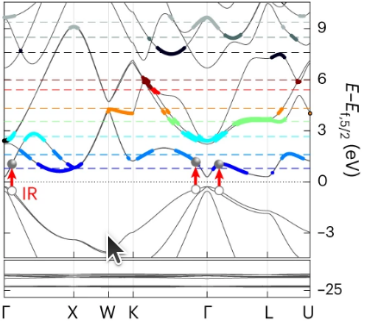
- Free-electron lasers: Higher (X-ray) energies, K-shell ionization
- Angular streaking
- Compare electrons from Oxygen($E_B \sim 500$ eV) and Nitrogen ($E_B \sim 400$ eV) in NO.
- up to 700 as delays in NO near O K-shell threshold

Solid state

light pulses to manipulate the electro-optical properties of a solid

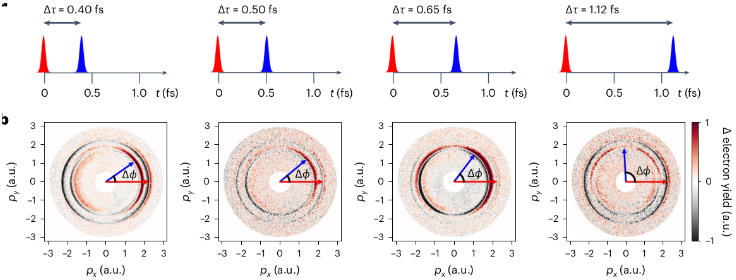


Inzani et al



Nature Photonics 17,1059 (2023)

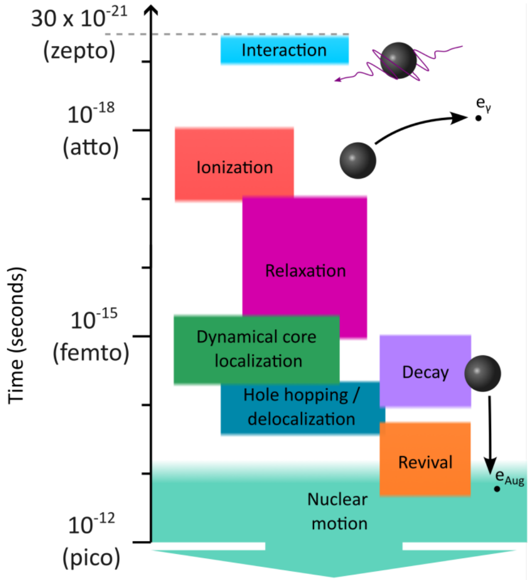
- Light-driven excitation in monocrystalline germanium
- Study of sub cycle response to intense (8 TW/cm^{-2}) IR pulse
- Probed by the attosecond pulse
- Ultrafast transient, as well as long-lasting, features observed.



Guo et al, Nature photonics
https://doi.org/10.1038/s41566-024-01419-w

- Two FEL pulses 370 eV and 740 eV. FWHM $\sim$ 1 fs
- $+$ Circularly polarized IR -field $\rightarrow$ Angular Streaking
- maps time to angle
- Looks at Carbon 1s ionization in several molecules.
- Post collisional effects

The Future

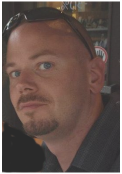


30 x 10^{-21}
(zepto)
10^{-18}
(atto)
10^{-15}
(femto)
10^{-12}
(pico)
Time (seconds)
Interaction
Ionization
Relaxation
Dynamical core localization
Hole hopping / delocalization
Decay
Revival
Nuclear motion
e_γ
e_{Aug}

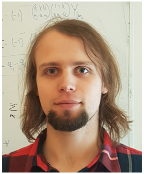
Acknowledgements

Marcus Dahlström

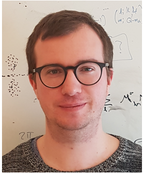
Anne l'Huillier and her group



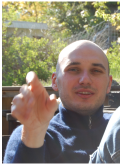
Jimmy Vinbladh



Johan Sörngård



Anton Ljungdahl



Luca Argenti, Orlando

Leon Pettersson

Soumyajit Saha