HHG in the condensed phase:

HHG spectroscopy on organic solids, PetaHertz currents in dielectrics, low energy scattering in liquids

Dr Mary Matthews

Imperial College

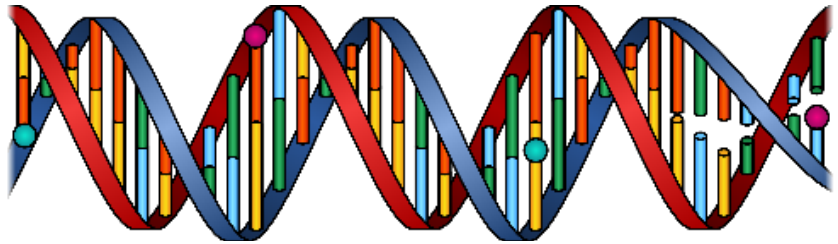
Why are ultrafast electonic and nuclear dynamics useful?

Basis of life:

- Ultrafast charge migration in organic systems

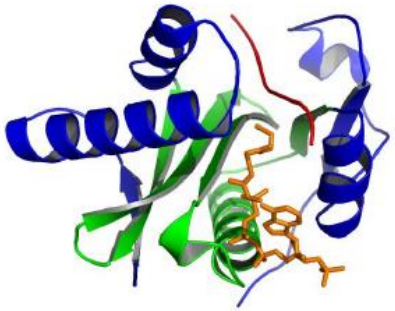
Photon therapies important part of health:

- Photo-activation of drugs
- Photo fragmentation and ionisation of large biomolecules:
  - Damage to DNA, ultrafast electron dynamics of primary and secondary radiation
  - Effects of laser mediated surgery with ultrafast lasers

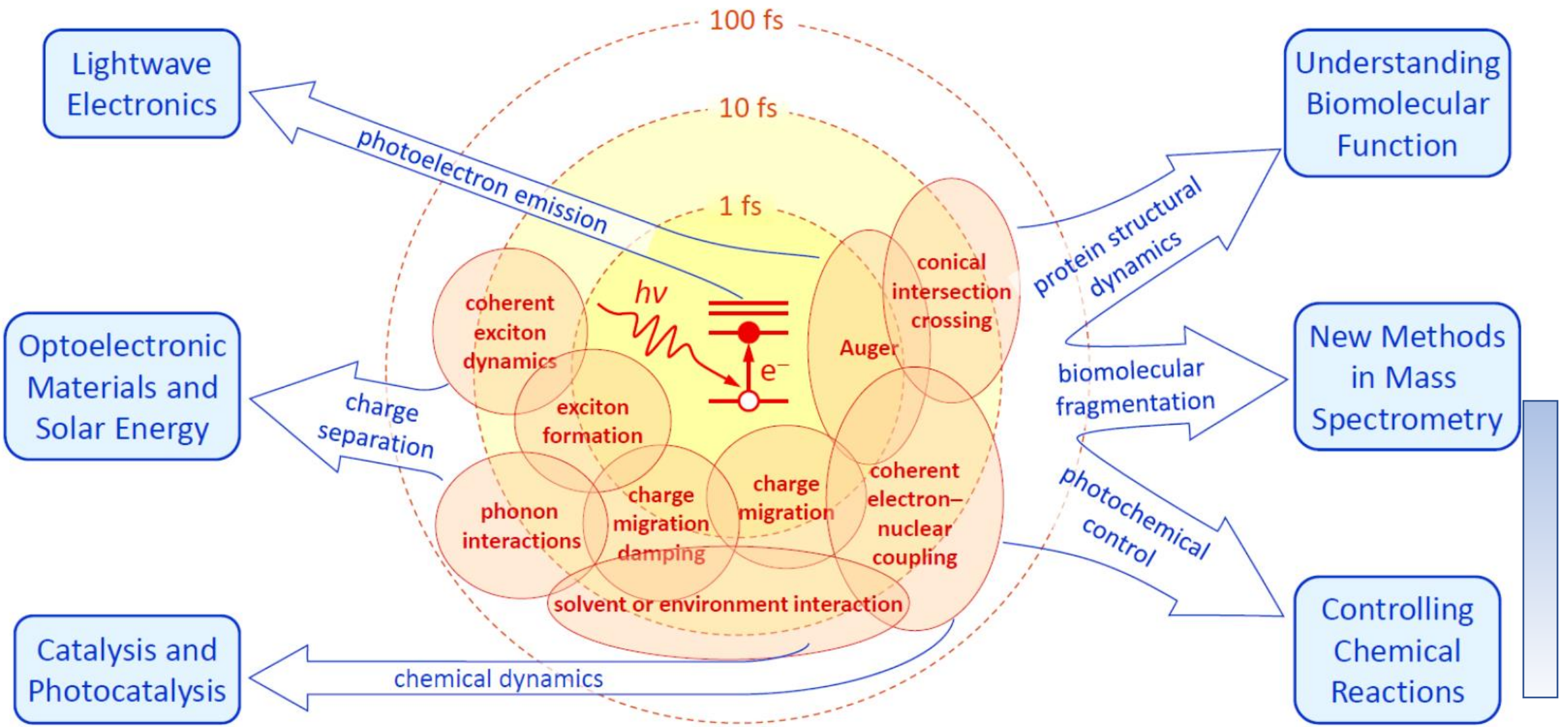


Photochemical reactions are essential for industries:

- Photolithography – basis of chip manufacture
- Photo-sensitive catalysts - plastic production



Ultrafast electronic and structural dynamics

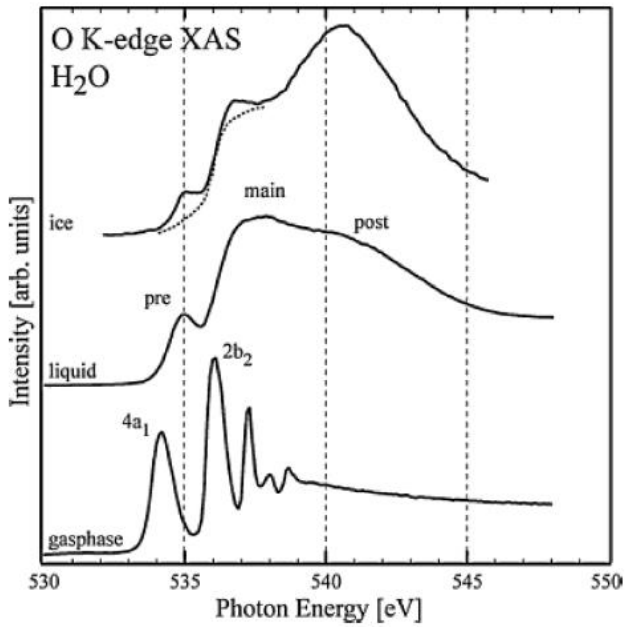


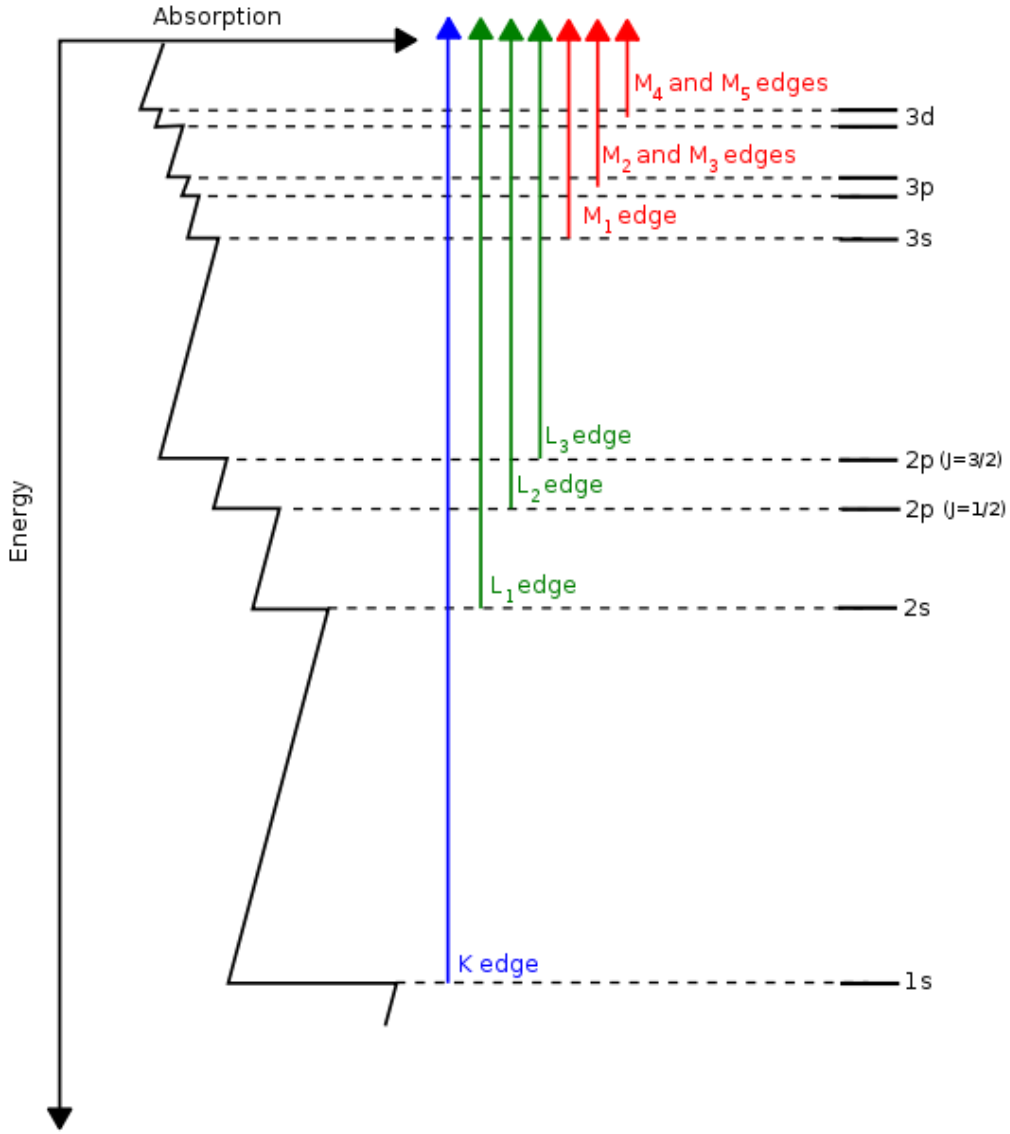
EUV as a spectroscopic tool

- Element specific:

  Core shells to valance band transitions, act like a fingerprint

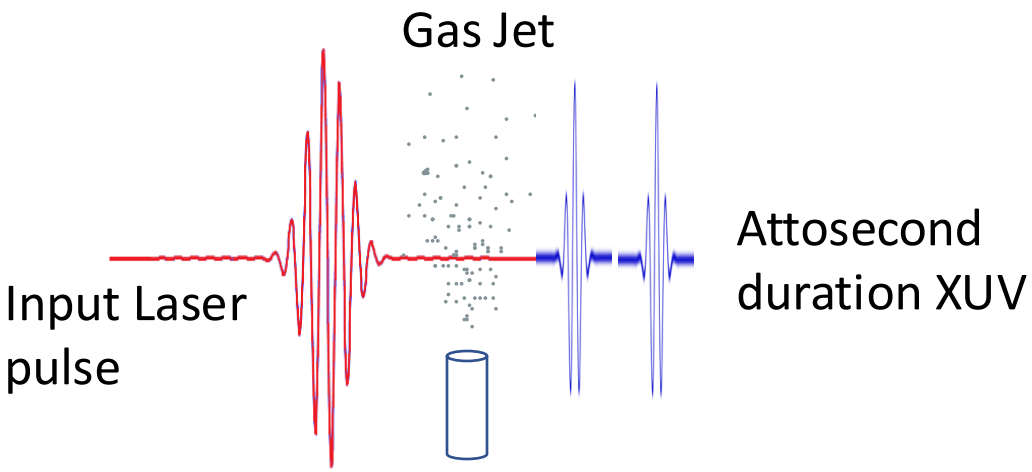
- Sensitive to local geometry, bonding and molecular structure

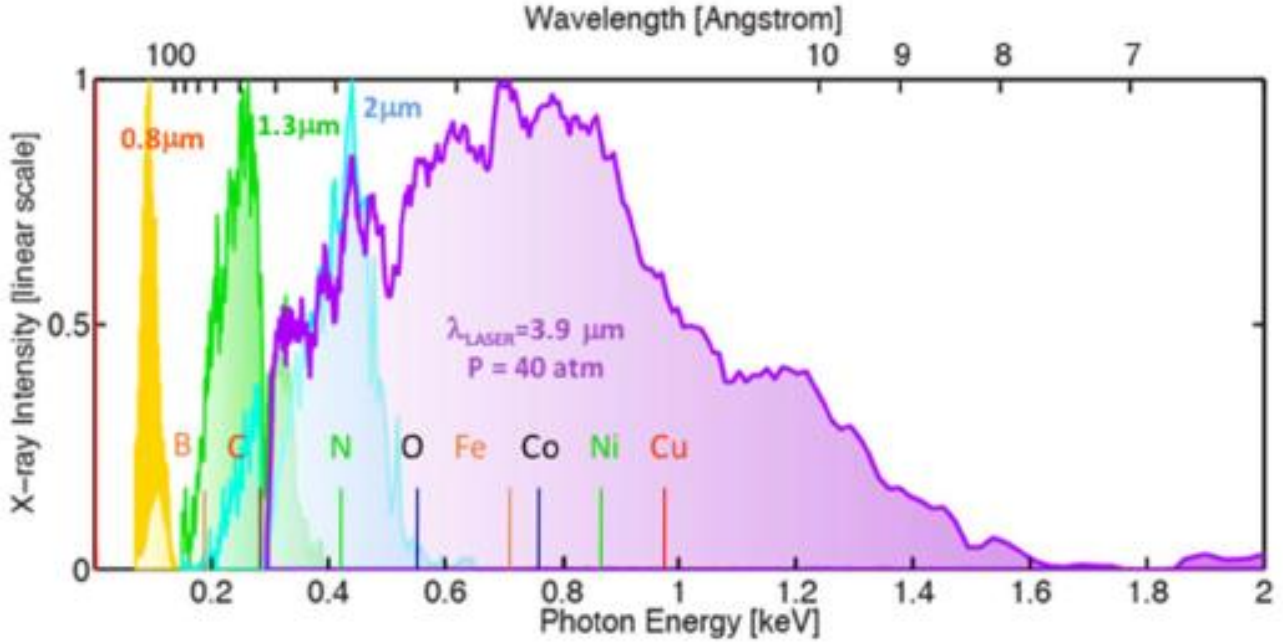




J. Phys. B: At. Mol. Opt. Phys. **51** (2018) 032003

EUV for time resolved spectroscopy





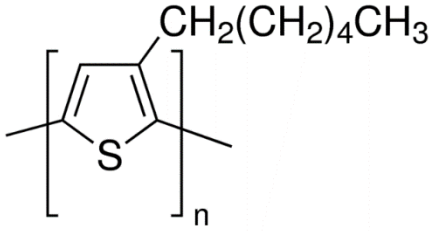
$\hbar\omega_{cutoff} = I_p + 3.17U_p$

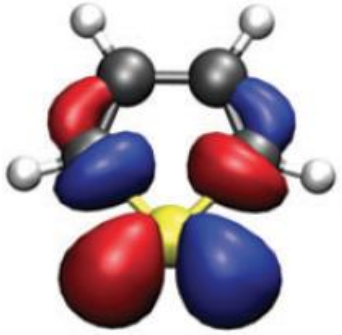
$$U_p = \frac{e^2 E_a^2}{4m\omega_0^2}$$

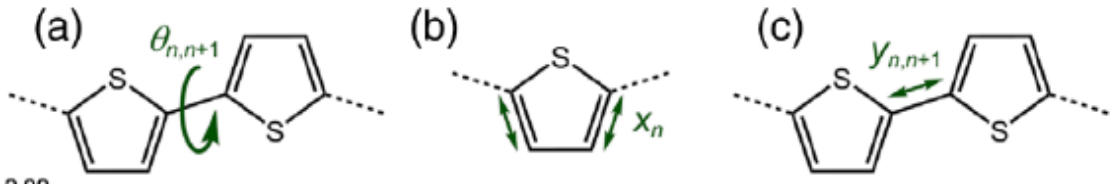
Carbon, Nitrogen and Oxygen K edges at 285eV, 410eV and 555eV

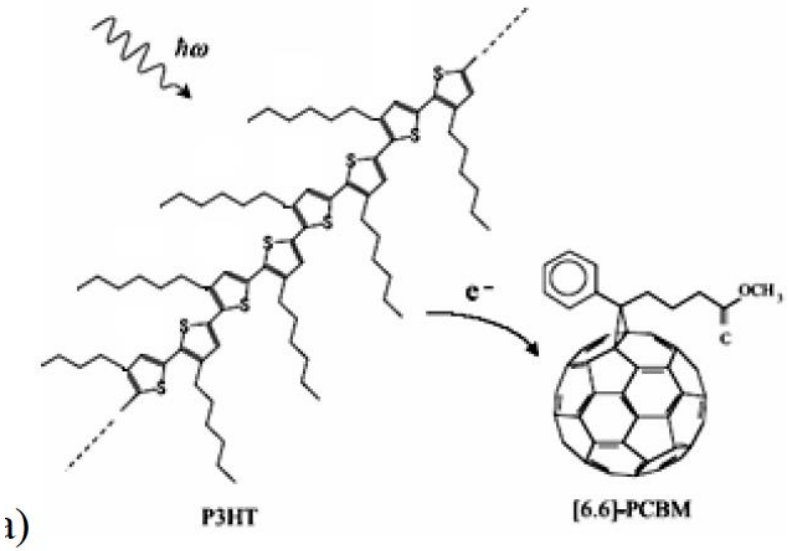
Long wavelength drivers and Higher field strengths
Gives access to *'water window' photons*

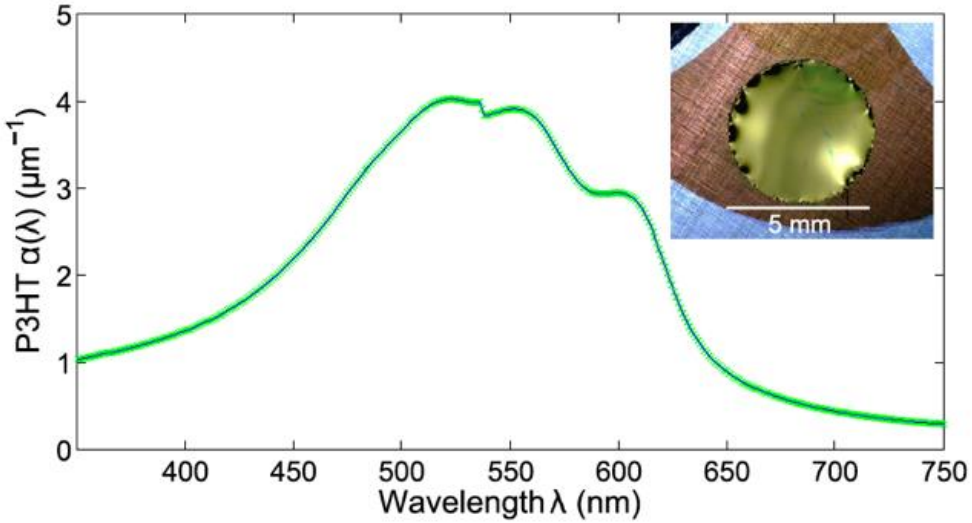
Time resolved transient absorption on organic P3HT:



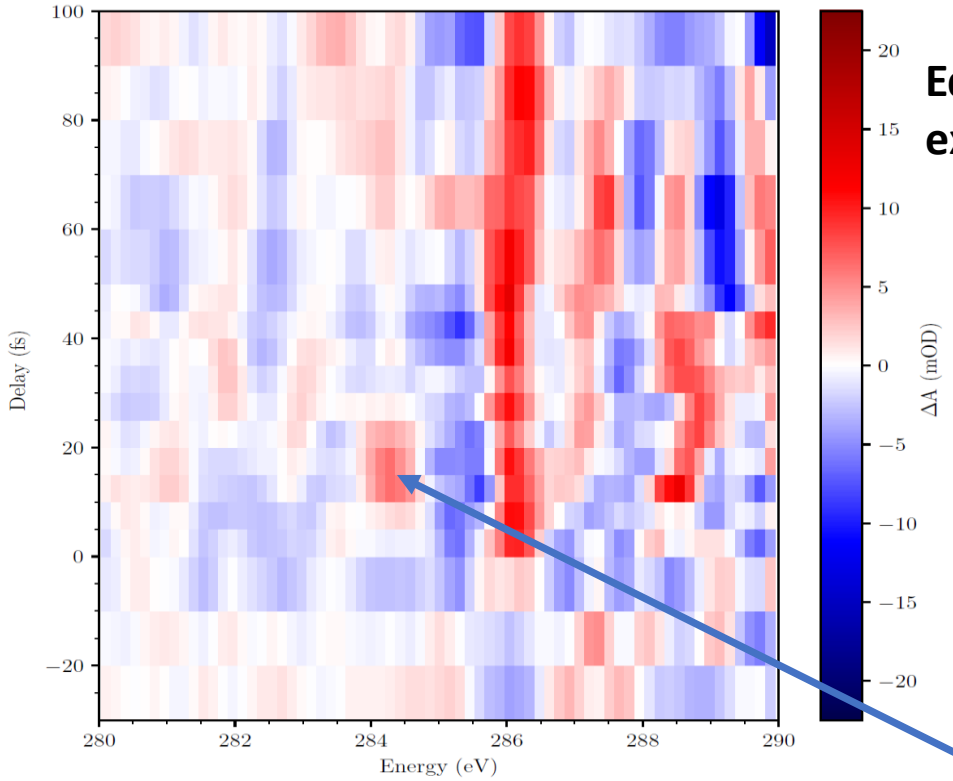




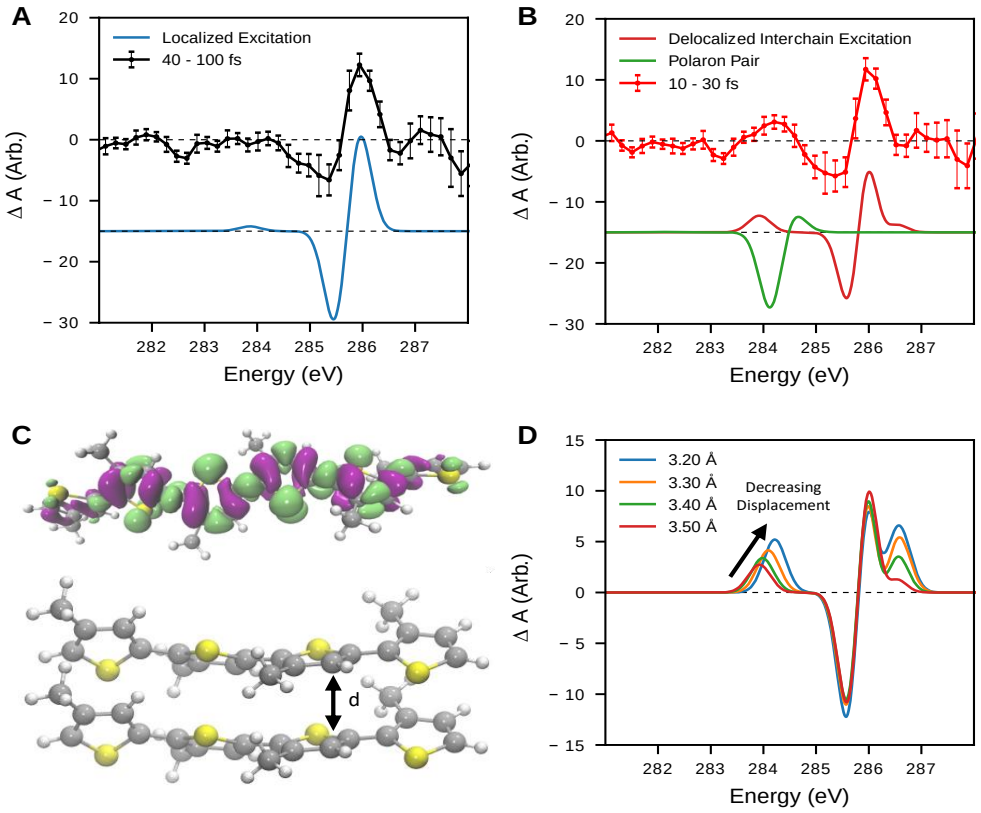




Direct probe of the initial localisation and cooling of the hot exciton formed by the pump



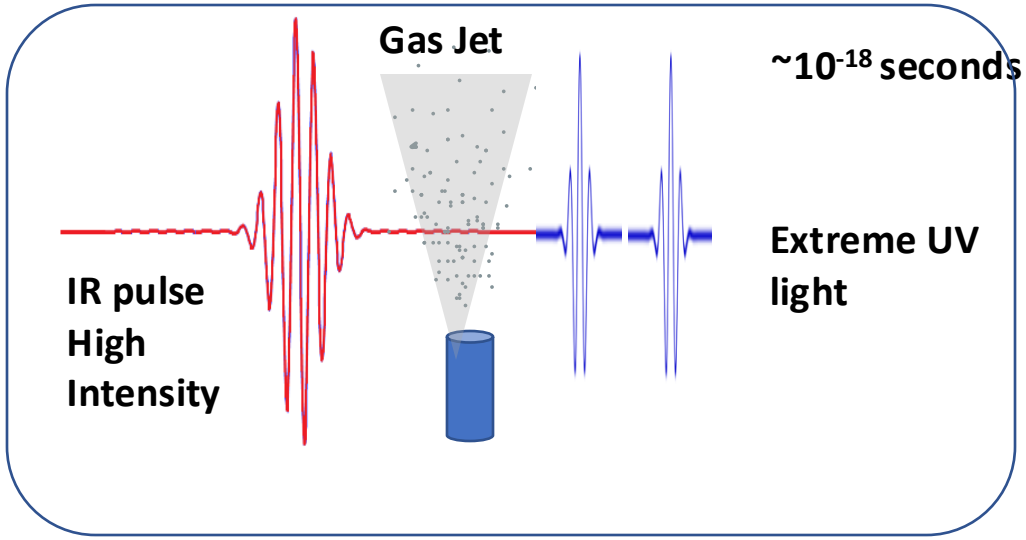
Edge shift following exciton creation



Direct observation of ultrafast exciton localization in an organic semiconductor with soft X-ray transient absorption spectroscopy

D. Garratt[1✉], L. Misiekis[1], D. Wood[1], E. W. Larsen[1], M. Matthews[1], O. Alexander[1], P. Ye[1], S. Jarosch[1], C. Ferchaud[1], C. Strüber[1], A. S. Johnson[1], A. A. Bakulin[2], T. J. Penfold[3] & J. P. Marangos[1]

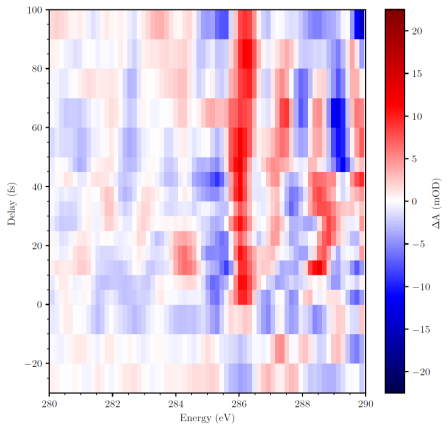
Transient feature – formation of hole state associated with exciton

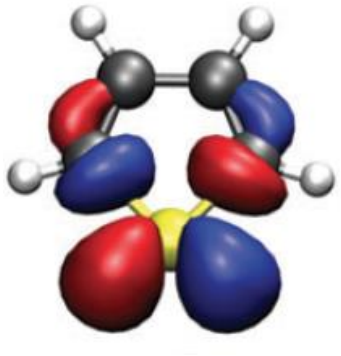


Summary

Ultrashort, long wavelength HHG emission can reach Nitrogen K edge

Tool to probe ultrafast electronic and structural changes

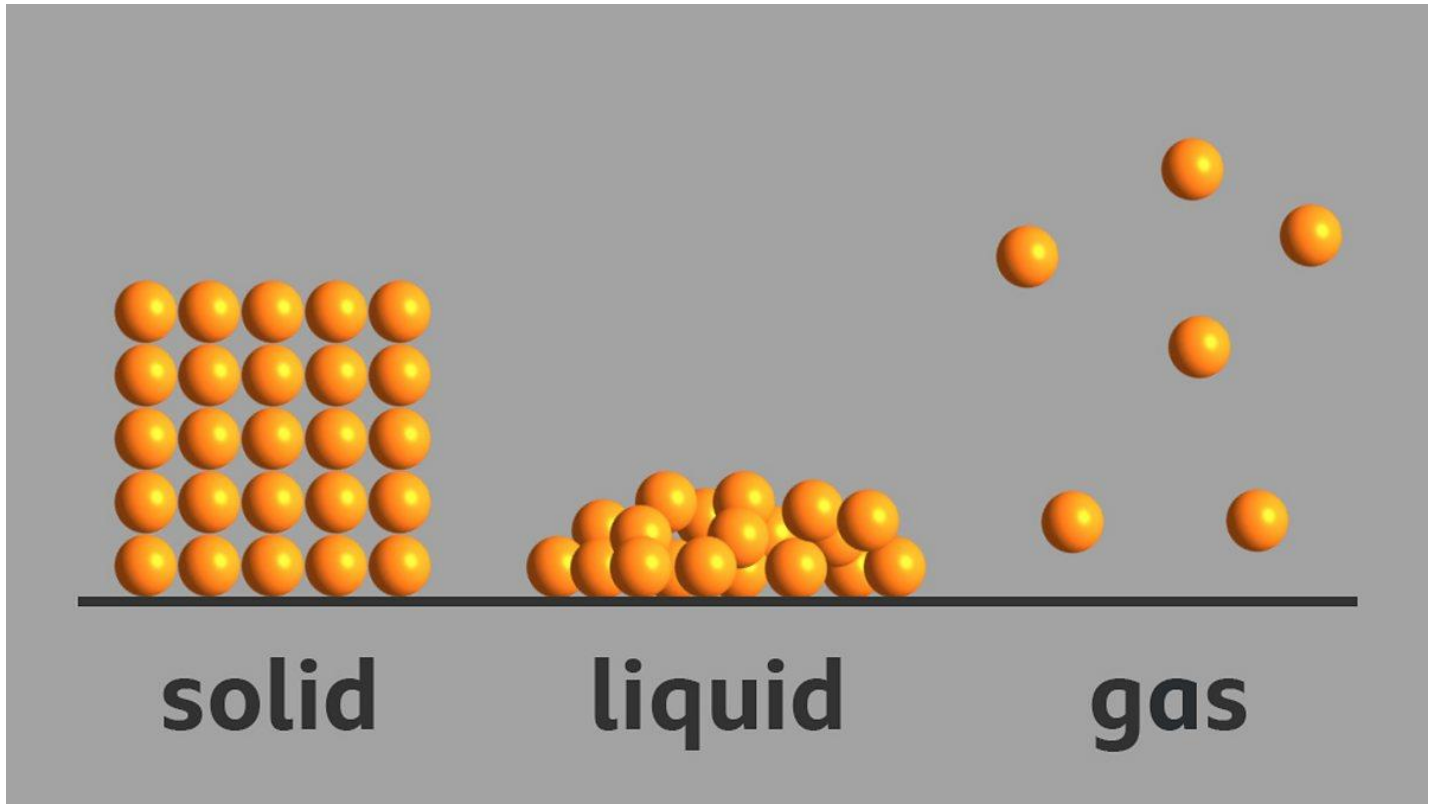




Provides insight into delocalisation of electrons in polymers

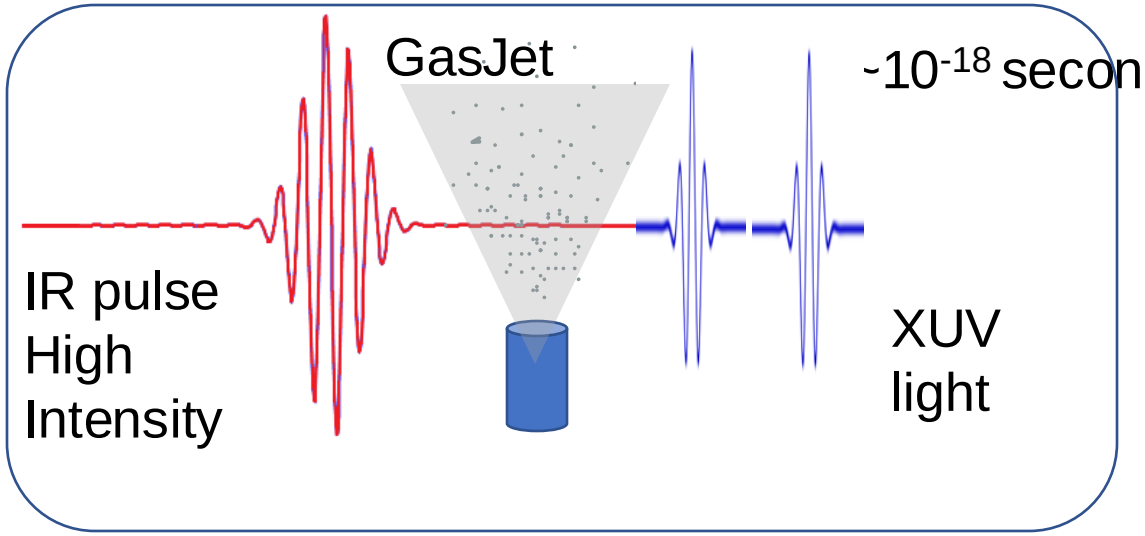
Sensitive to local geometry, bonding and molecular structure

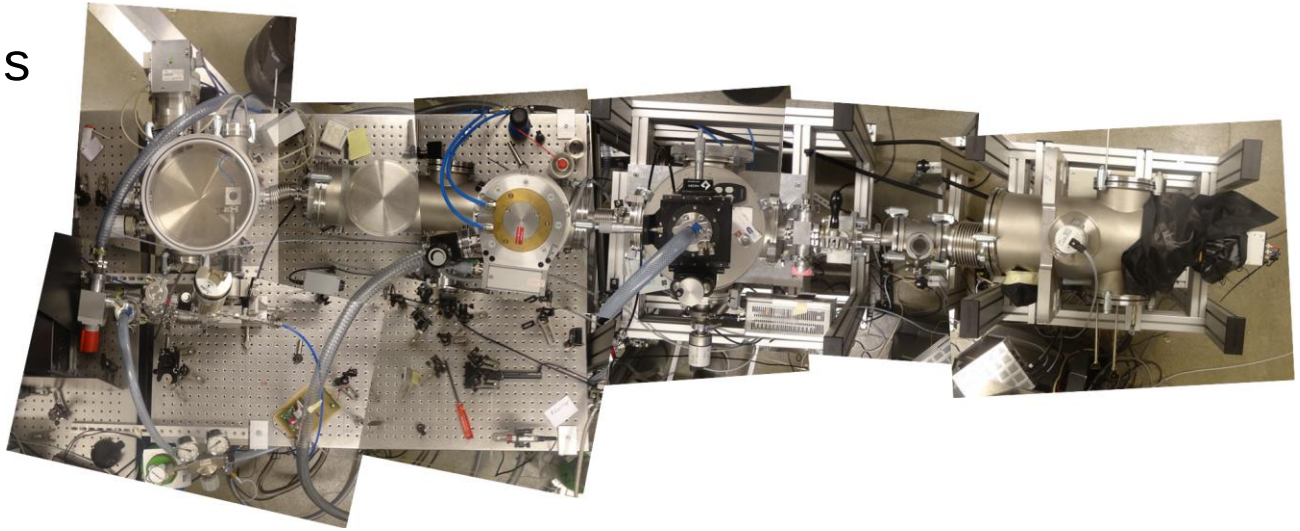
Can attosecond pulses be generated in different phases of matter?

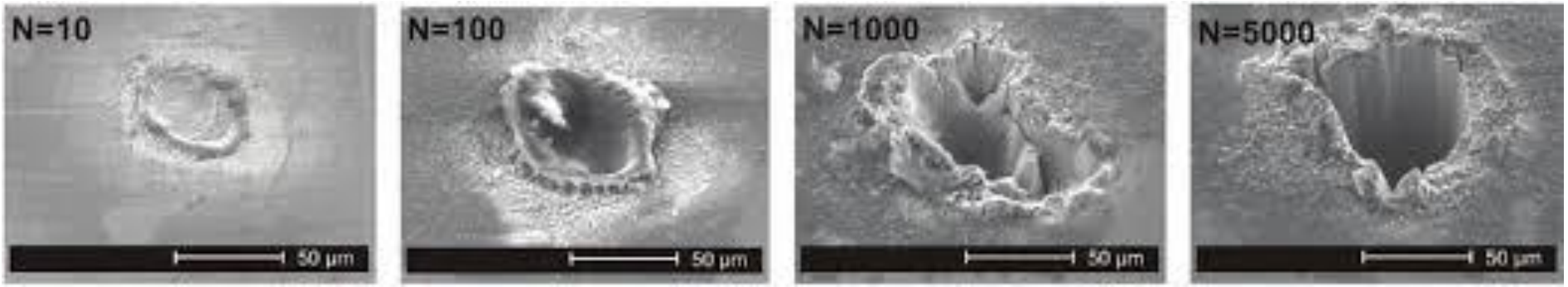


Attoseconds from solids?

High harmonic emission in gases



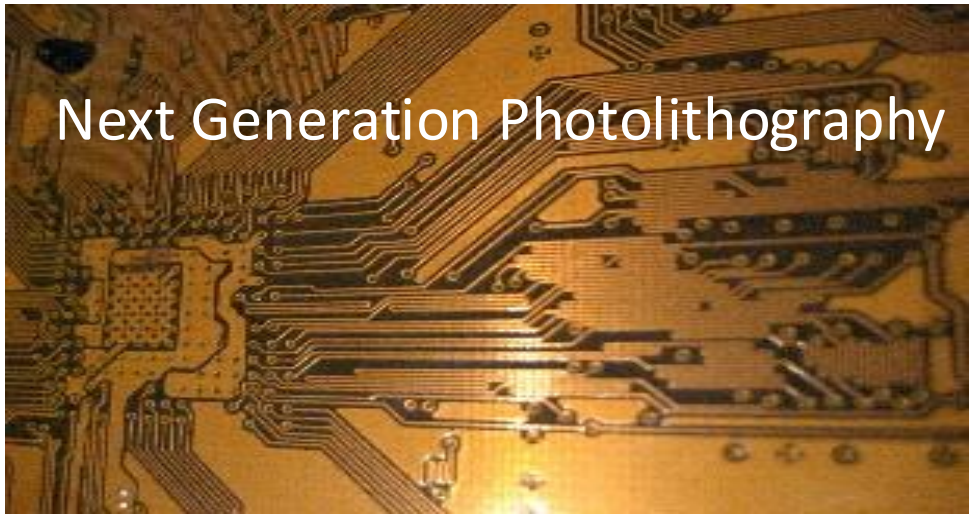


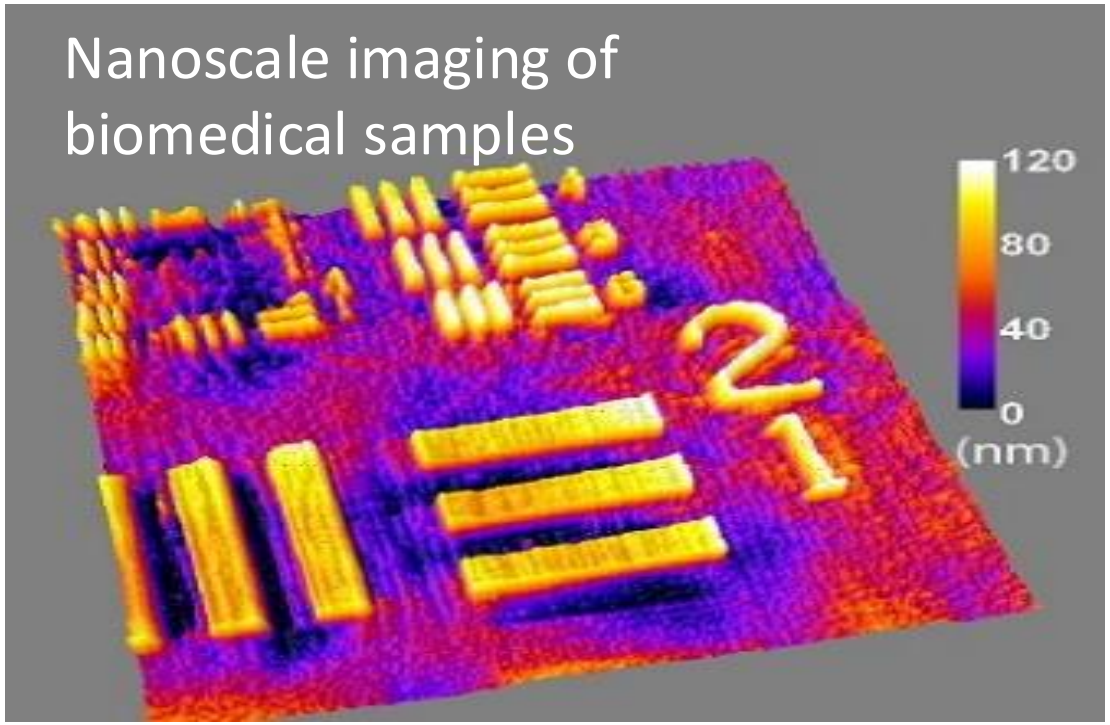


Strong field Physics in solids

Why do we need solid state attosecond sources?

Compact, robust, extreme ultra-violet light source





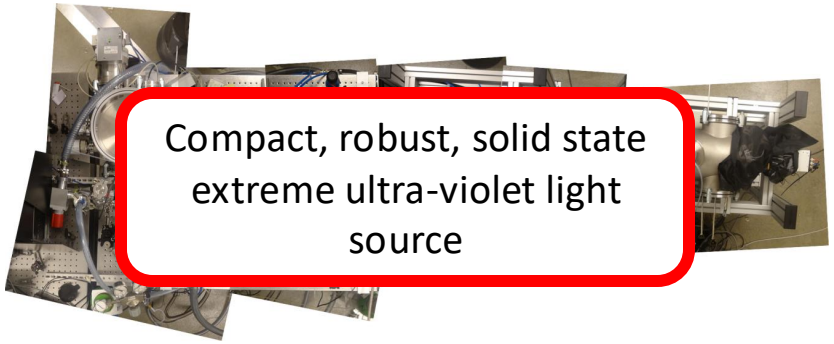
Attosecond Nanophotonics

High harmonic emission in solids

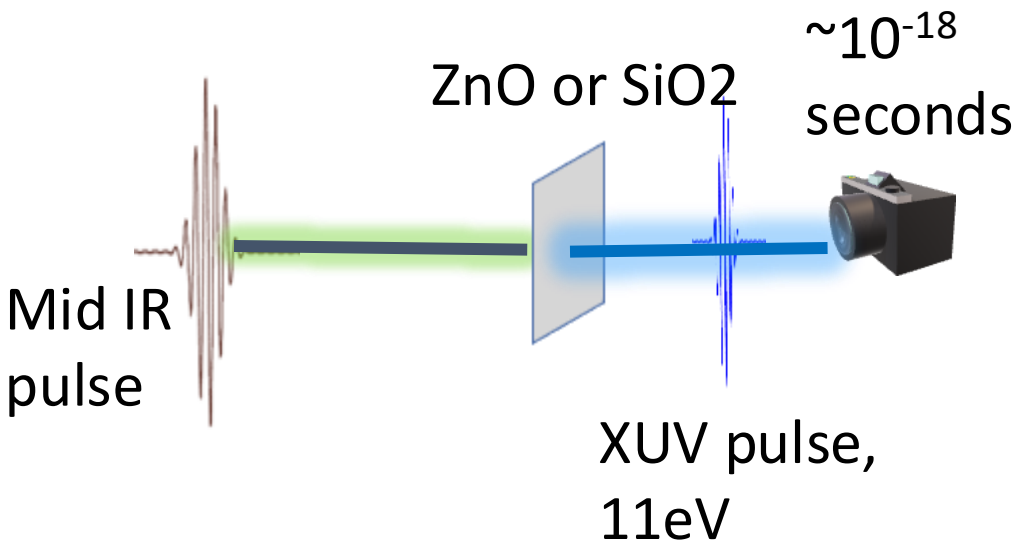
LETTERS
PUBLISHED ONLINE: 5 DECEMBER 2010 | DOI: 10.1038/NPHYS1847
nature physics

Observation of high-order harmonic generation in a bulk crystal

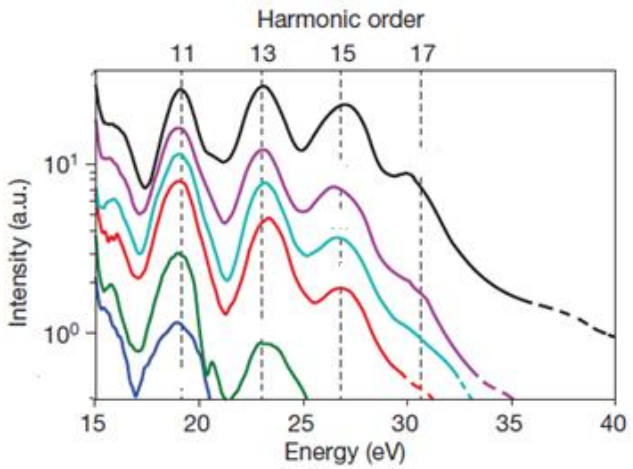
Shambhu Ghimire[1], Anthony D. DiChiara[2], Emily Sistrunk[2], Pierre Agostini[2], Louis F. DiMauro[2] and David A. Reis[1,3]*



Compact, robust, solid state extreme ultra-violet light source



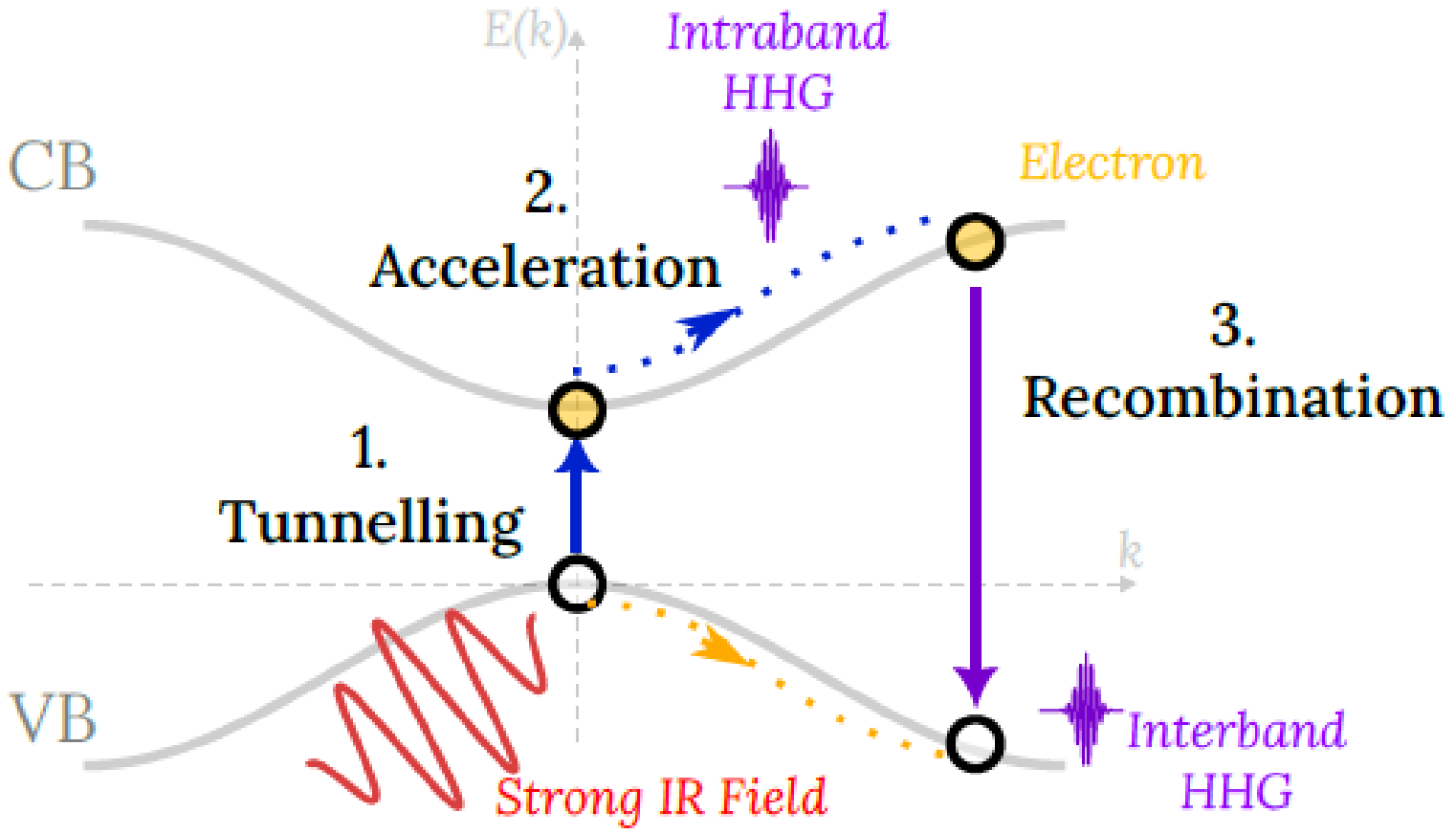
Strong field Physics



Strongly driven electron dynamics = deep insight into material band structure

PetaHertz electronics Driven by light fields

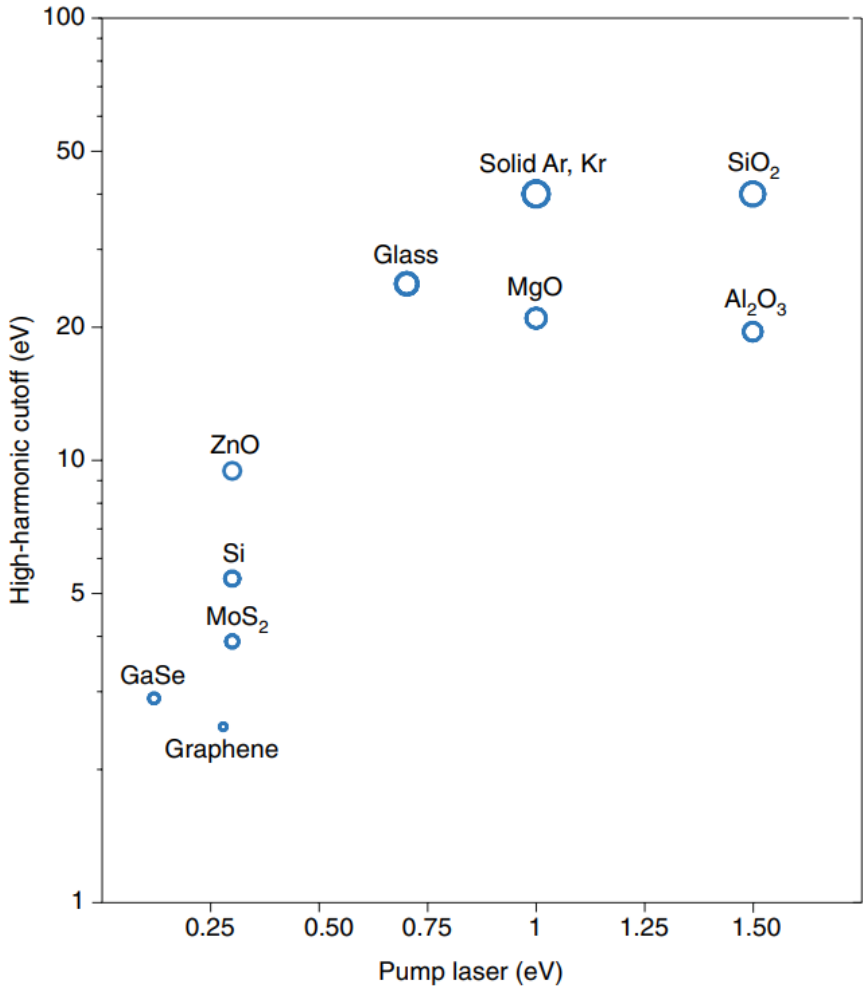
PetaHertz currents inside solids



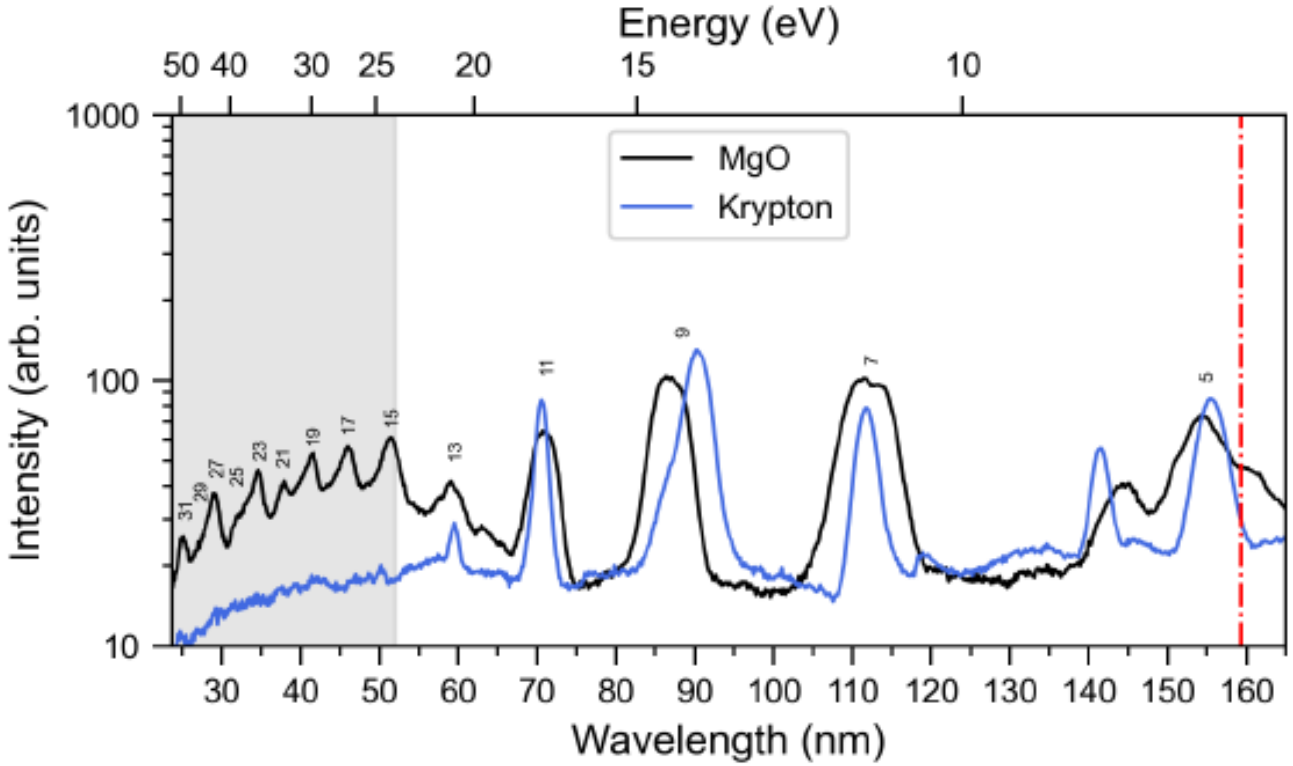
Three steps generate currents

Each current term is petaHertz rate

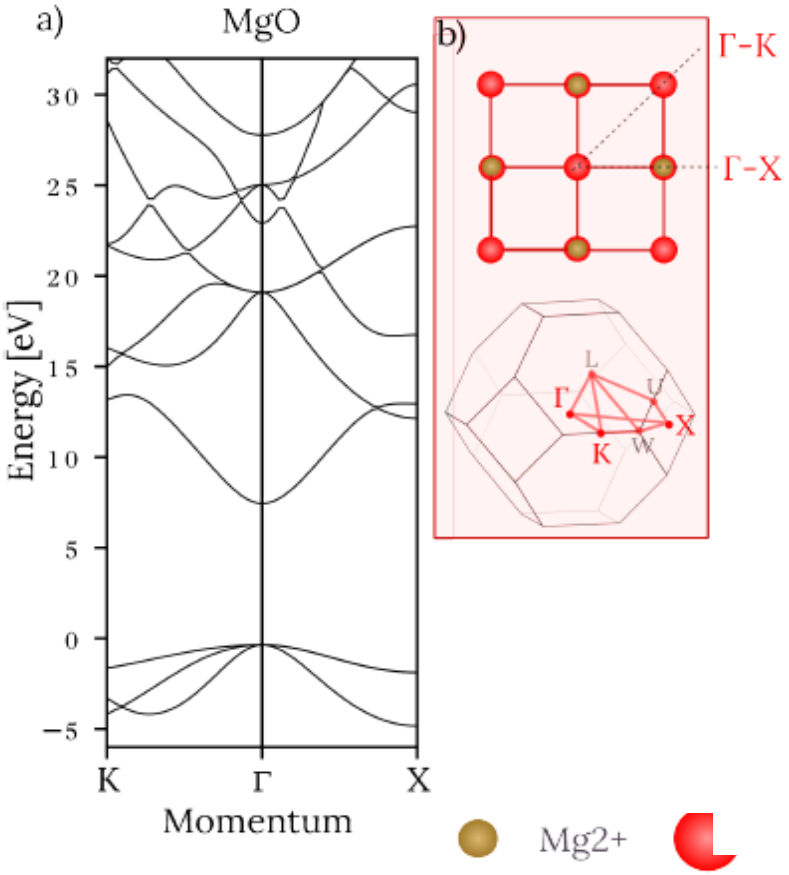
New source of EUV and attoseconds

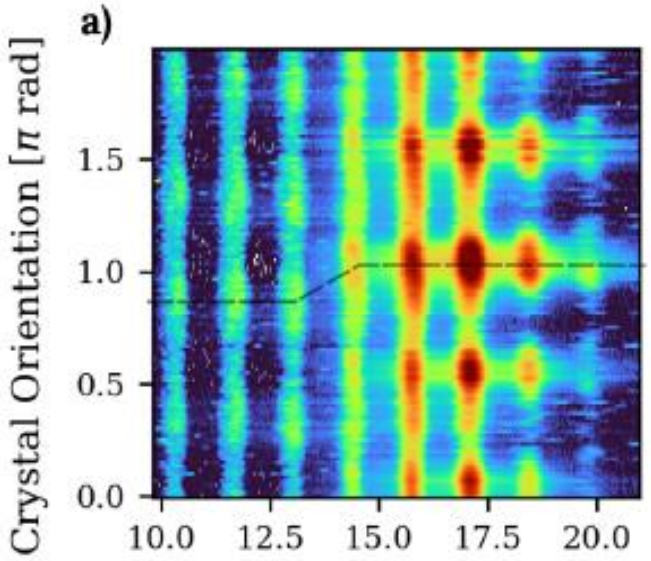


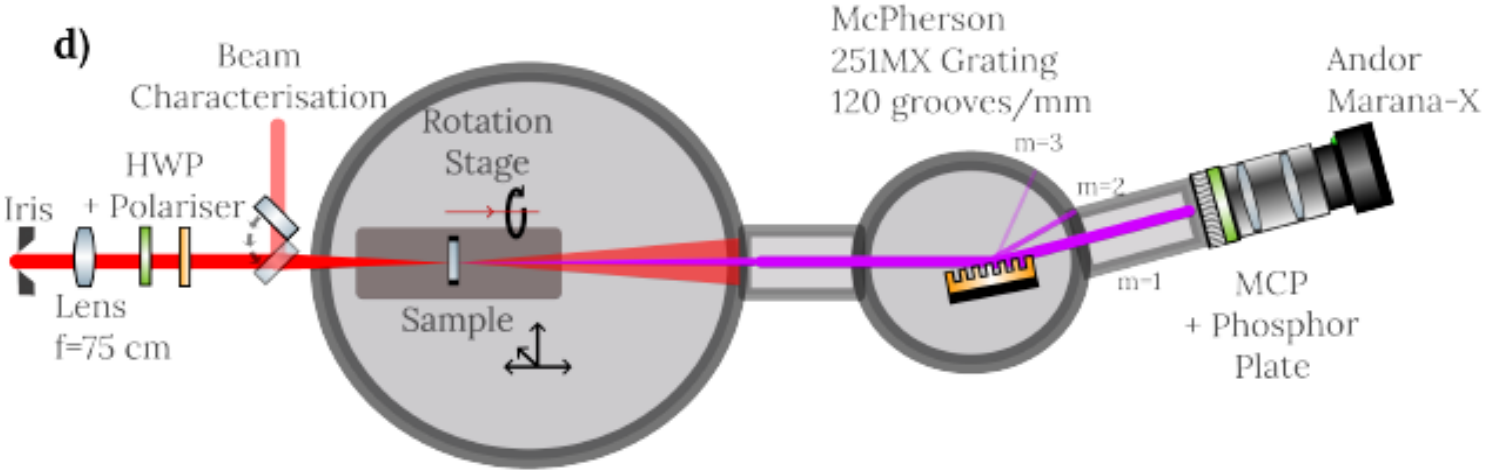
Sample	Thickness (μm)	Drive Wavelength (μm)	Pulse Duration (fs)	Cutoff Energy (eV)	Ref.
Solid Ar & Kr	5	1.33	50	40	[8]
Al_2O_3	430	0.80	12	31	[9]
MgO	200	1.70	11	25	[10]
MgO	100	1.30	50	30	[11]
SiO_2	0.120	0.80	~2	33	[2]
SiO_2	0.120	0.83	~2	34	[12]



Probing the bandstructure out of equilibrium: MgO







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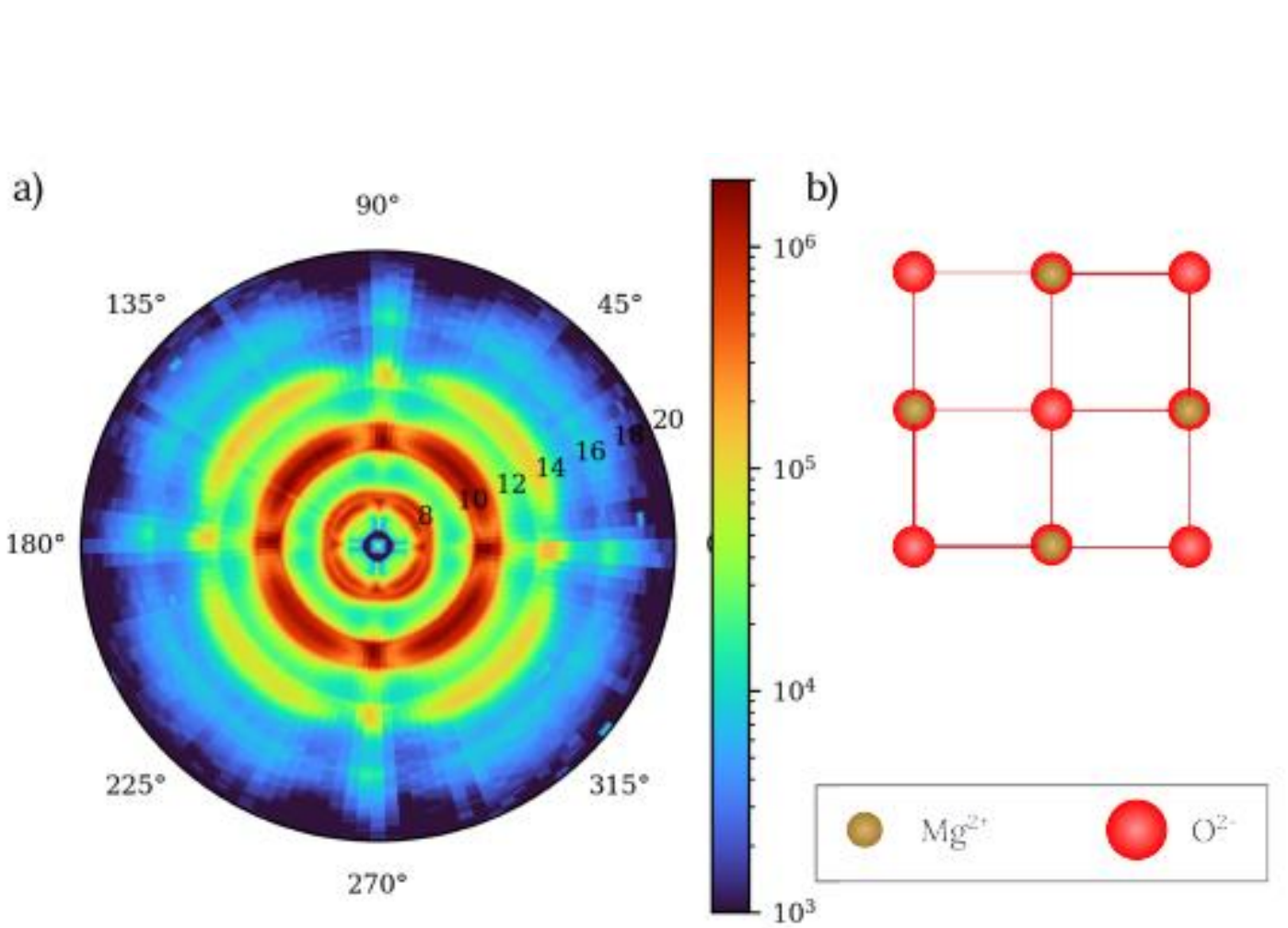

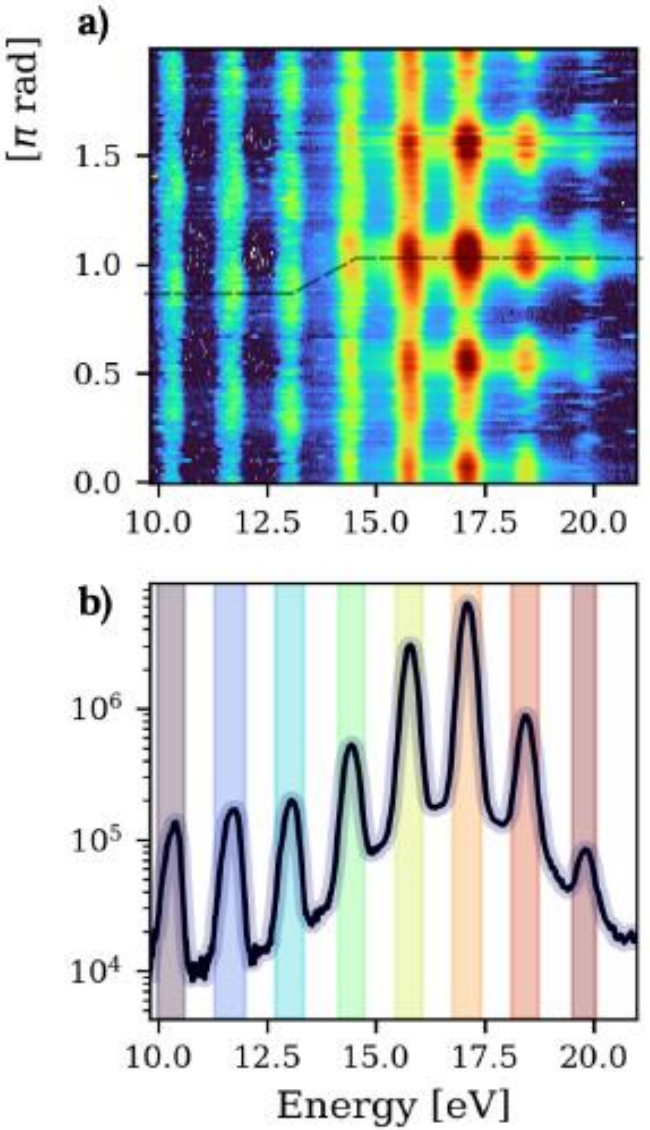

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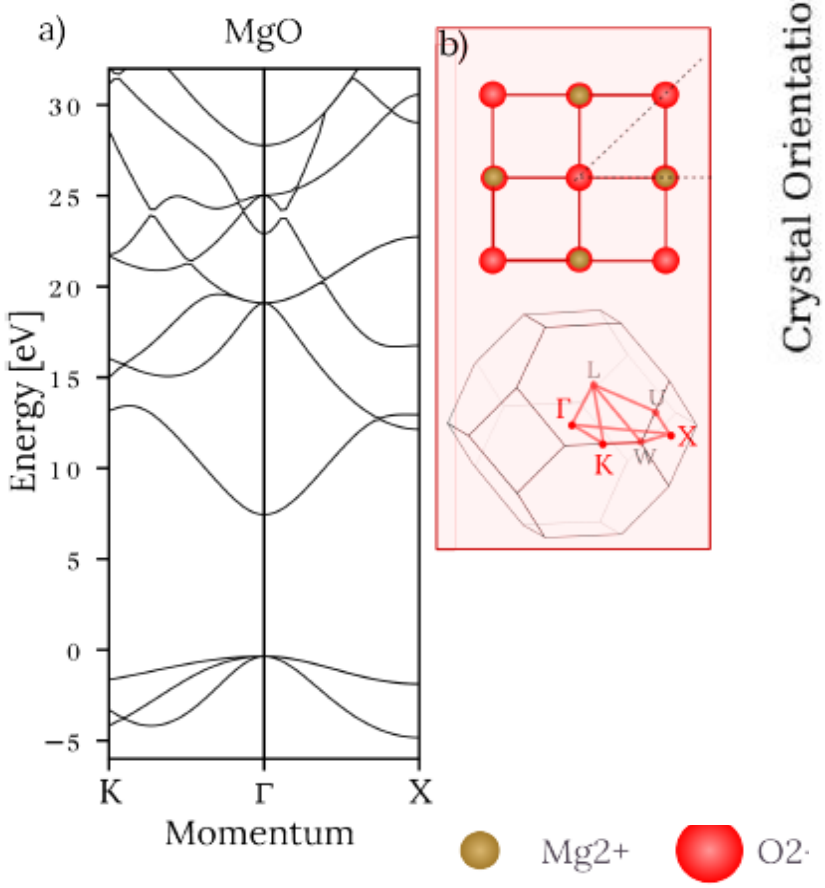
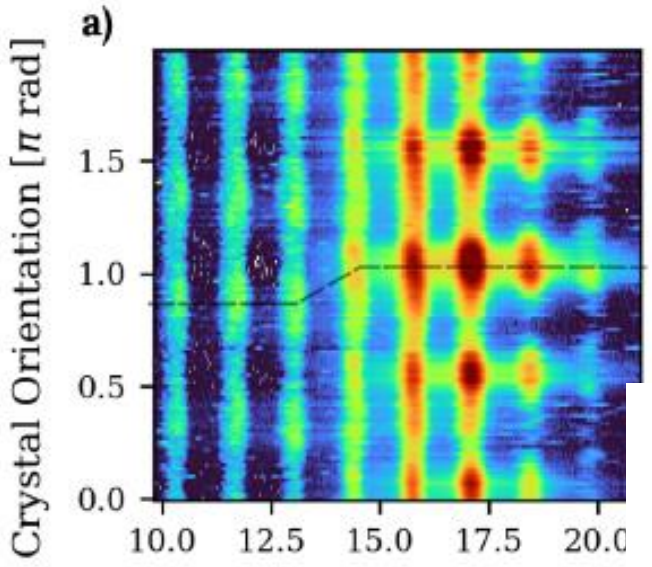
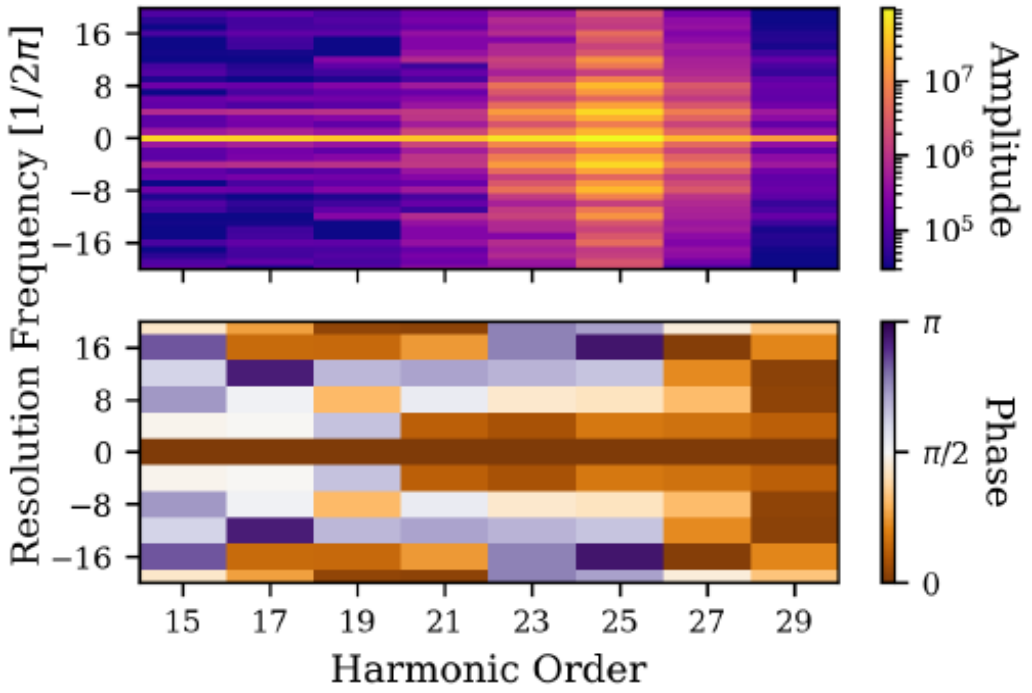
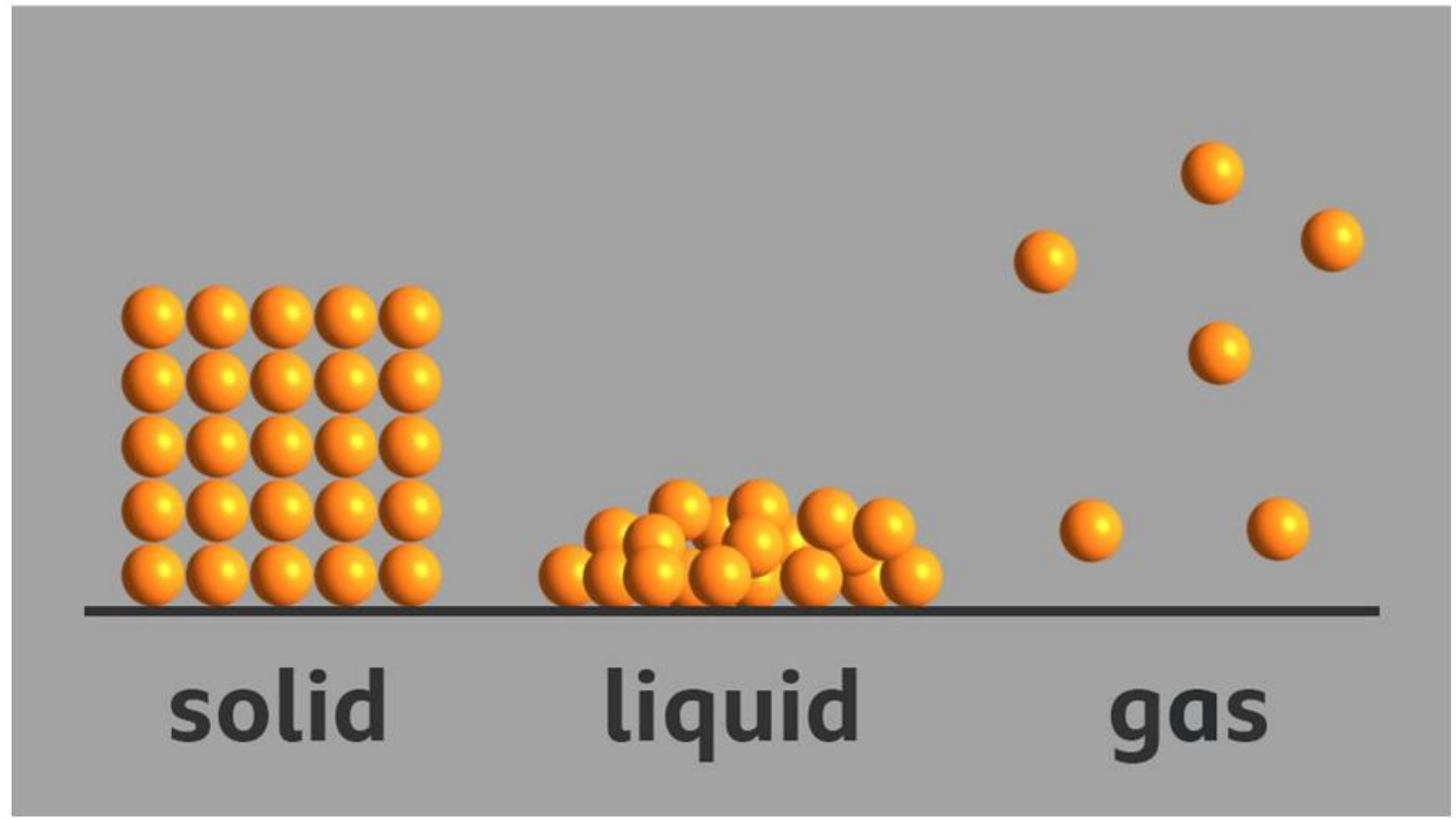




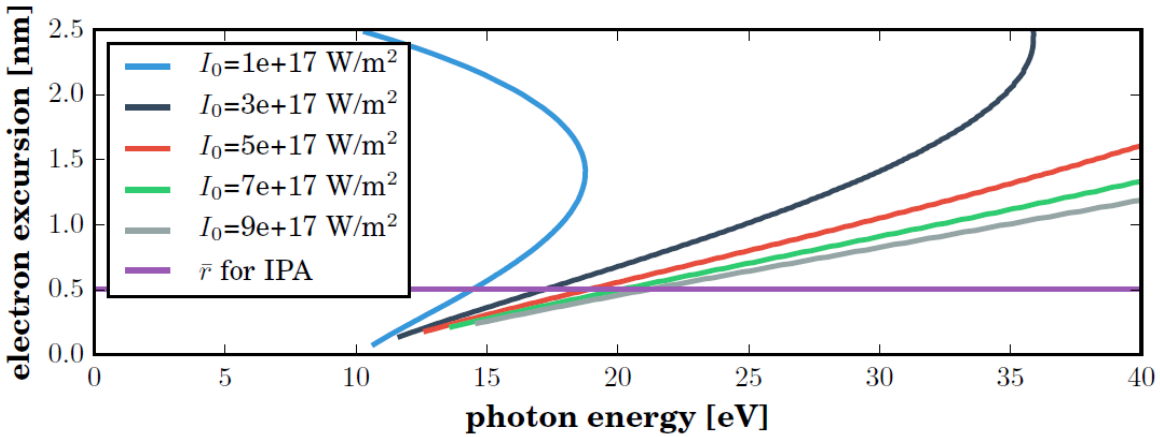


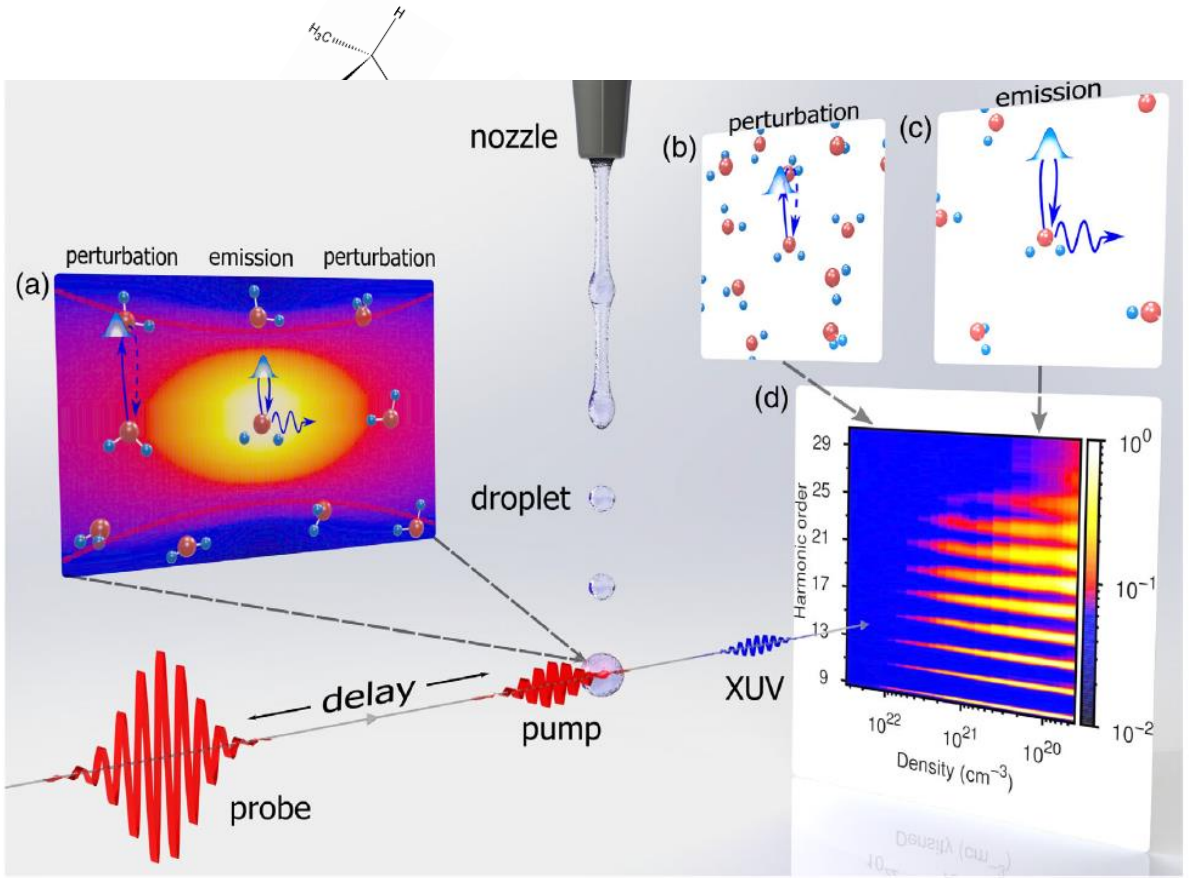
Can attosecond pulses be generated in different phases of matter?



Attoscience in the liquid phase

- Intermolecular/inter-atomic distances are much less than the electron excursion length



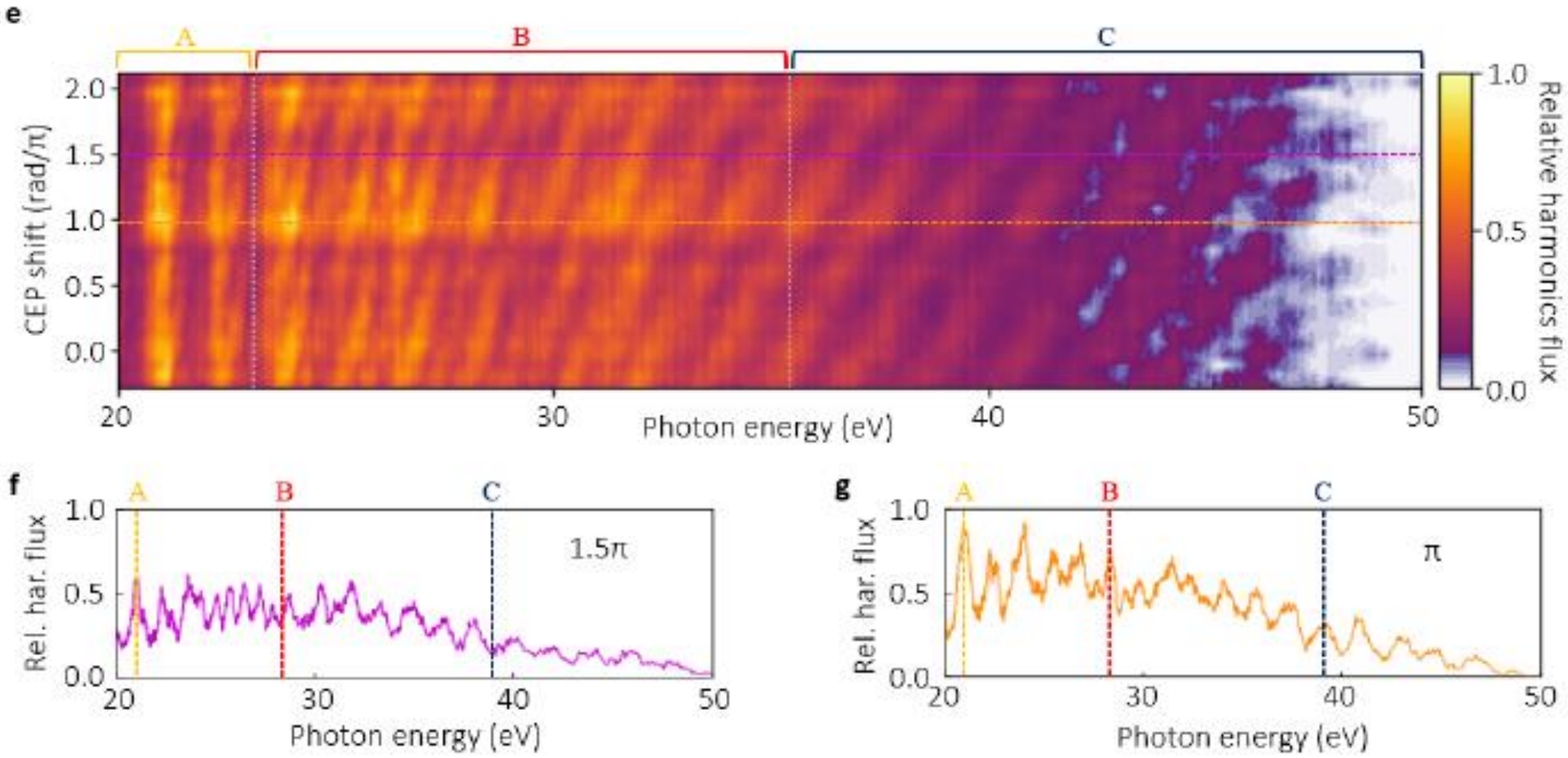


PHYSICAL REVIEW A **87**, 063811 (2013)

High-order-harmonic generation from dense water microdroplets

Heiko G. Kurz,[1,2,*] Daniel S. Steingrube,[1,2] Detlev Ristau,[2,3] Manfred Lein,[2,4] Uwe Morgner,[1,2,3] and Milutin Kovačev[1]

CEP dependent HHG in liquid Isopropanol



Simulations of HHG in liquid Isopropanol

$$A = \sigma' = k\sigma_{emp.}.$$

- Introduce macroscopic parameter ‘k’
- This accounts for screening, correlation potential and exchange
- Low energy ultrafast scattering captured