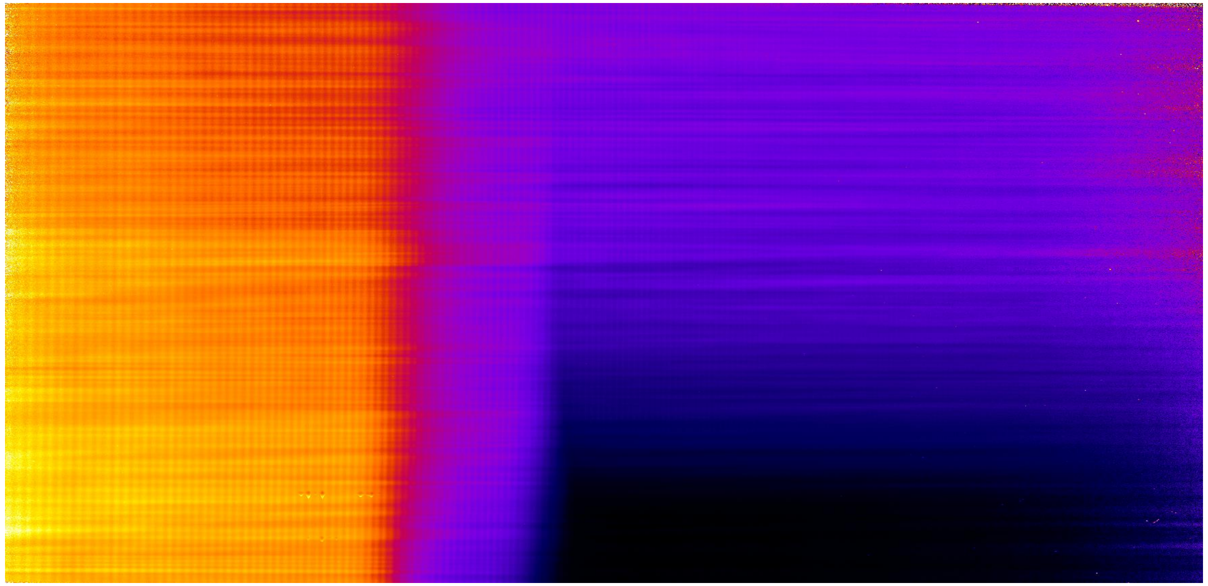
Laser driven accelerators for a new frontier in ultrafast physics

Brendan Dromey



Studying nascent proton driven radiation chemistry in H_2O in real time

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Acknowledgments

- Mark Coughlan
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- Rong Yang
- Martin Speicher
- Jorg Schreiber

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Overview

- Ultrafast sources of laser driven radiation
- Optical streaking technique
- Pulsed radiolysis in SiO_2: The role of dimensionality
- Pulsed radiolysis in H_2O: Solvated electron dynamics on ultrafast timescales

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Laser driven ion accelerators – a novel source for ultrafast physics

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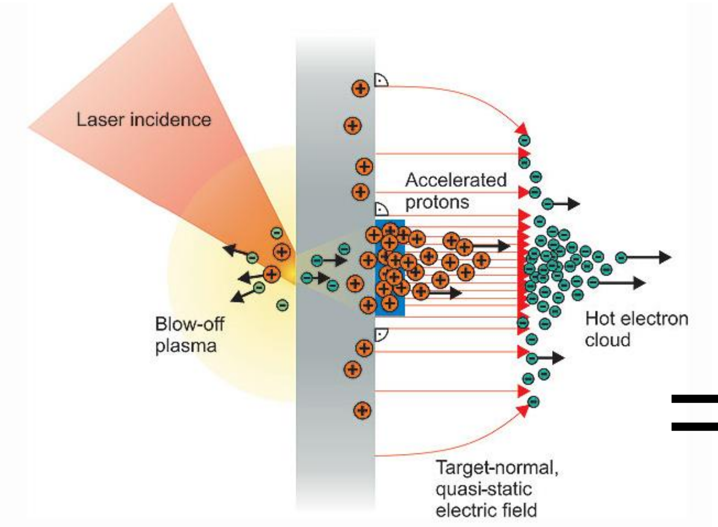
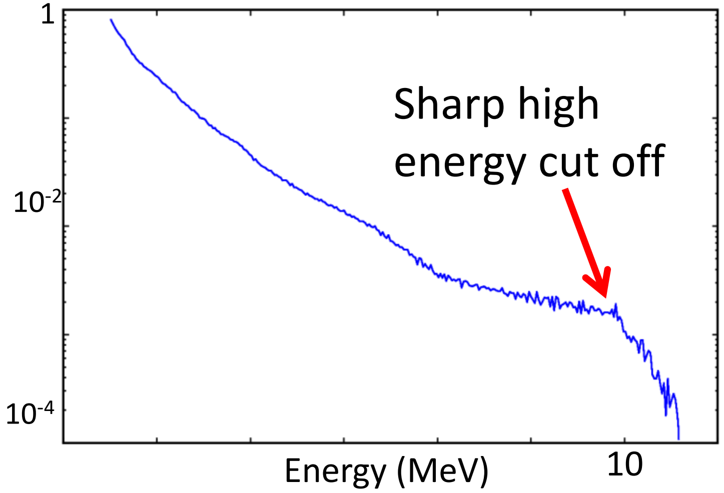
Traget Normal Sheath Acceleration, TNSA



H. Schwoerer, "Laser-plasma acceleration of quasi-monoenergetic protons from microstructured targets", Nature, **439**, 445-448, 2006

⇒

PIC simulation: Broadband energy spectrum

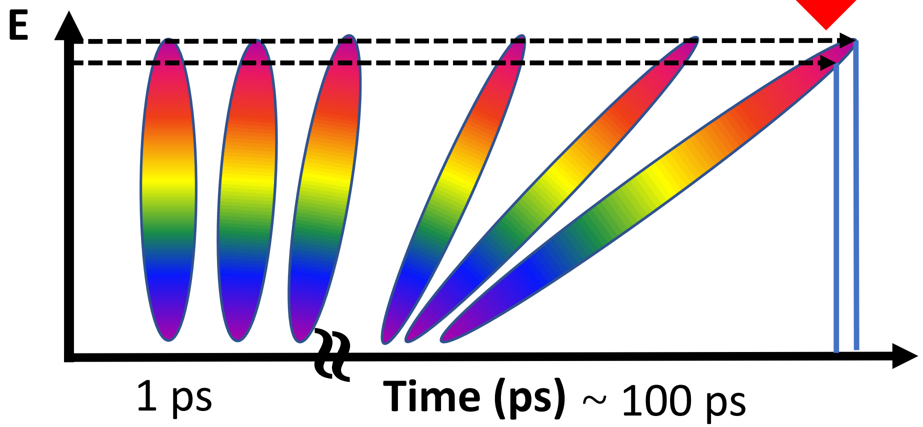


⇒ **Velocity dispersion**

⇓

Phase space rotation

Critically, ultrafast pulse duration is preserved in narrow energy bandwidths of the TNSA spectrum



By slowing and stopping lower energy components of the ion energy spectrum the ultrafast pulse duration can be recovered to permit ultrafast pulsed radiolysis

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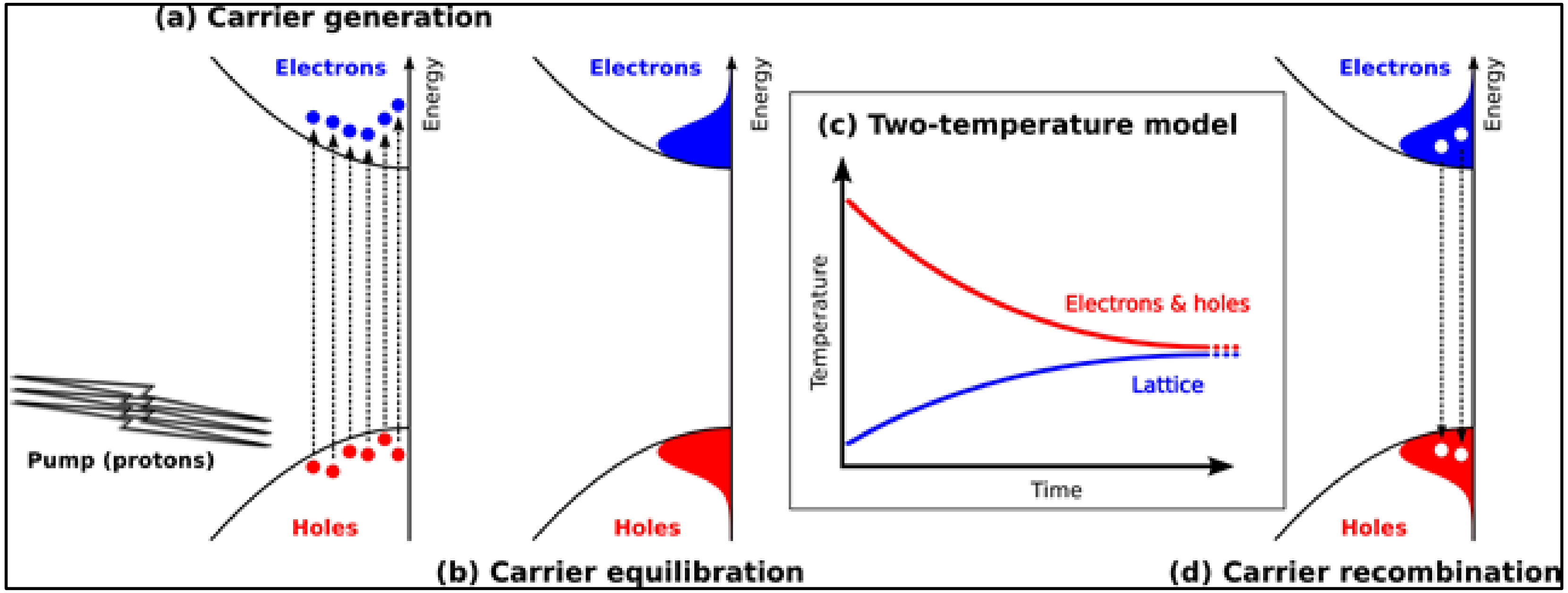
Transparent Dielectrics (or aqueous solutions)



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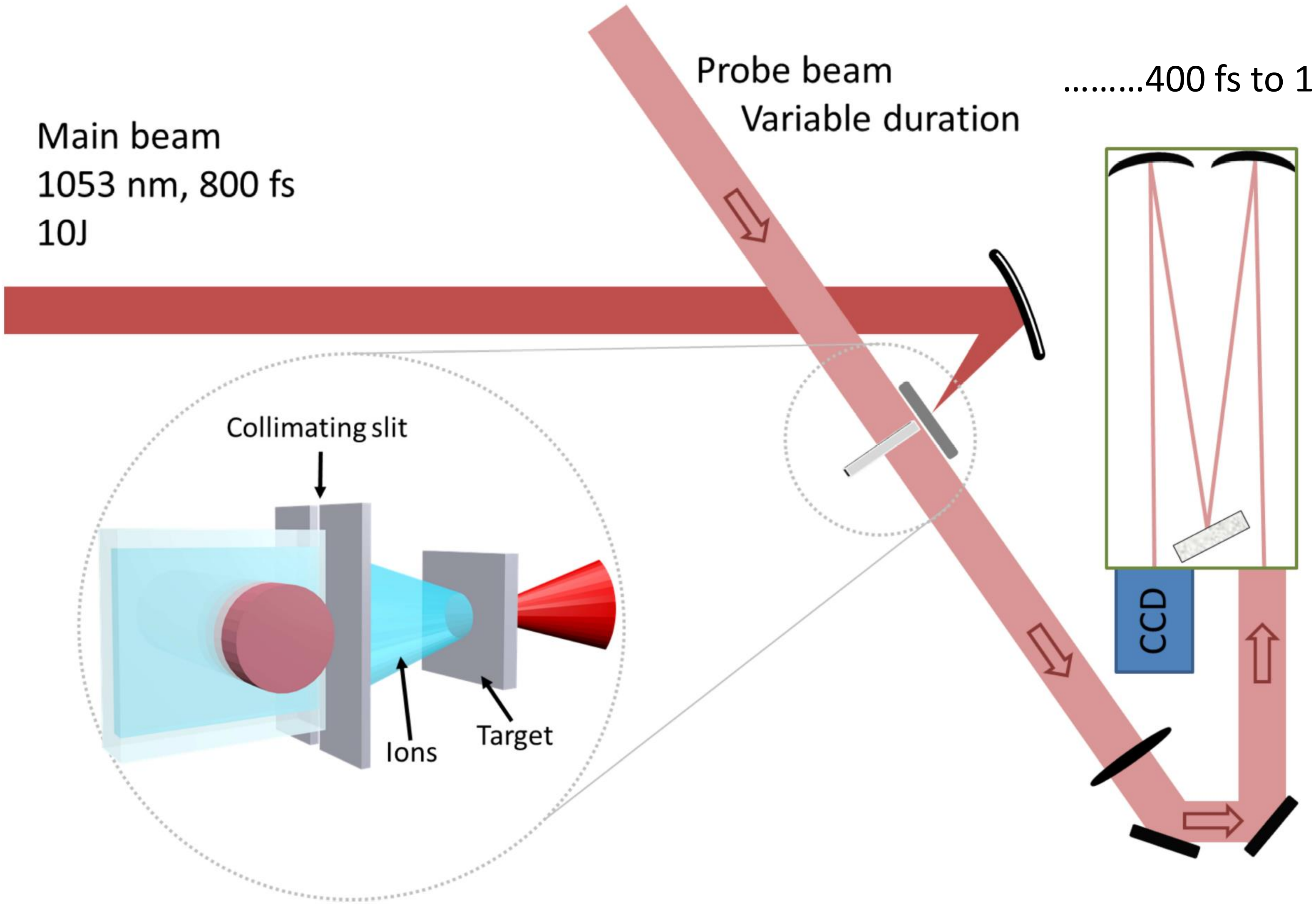
Observing ultrafast proton interactions in a single shot



Schematic
(not to scale)

Probe ion induced
a) opacity (free electrons in conduction)
b) Photabsoption bands
c) Polaristation effects

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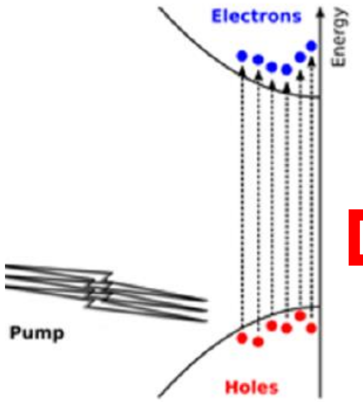
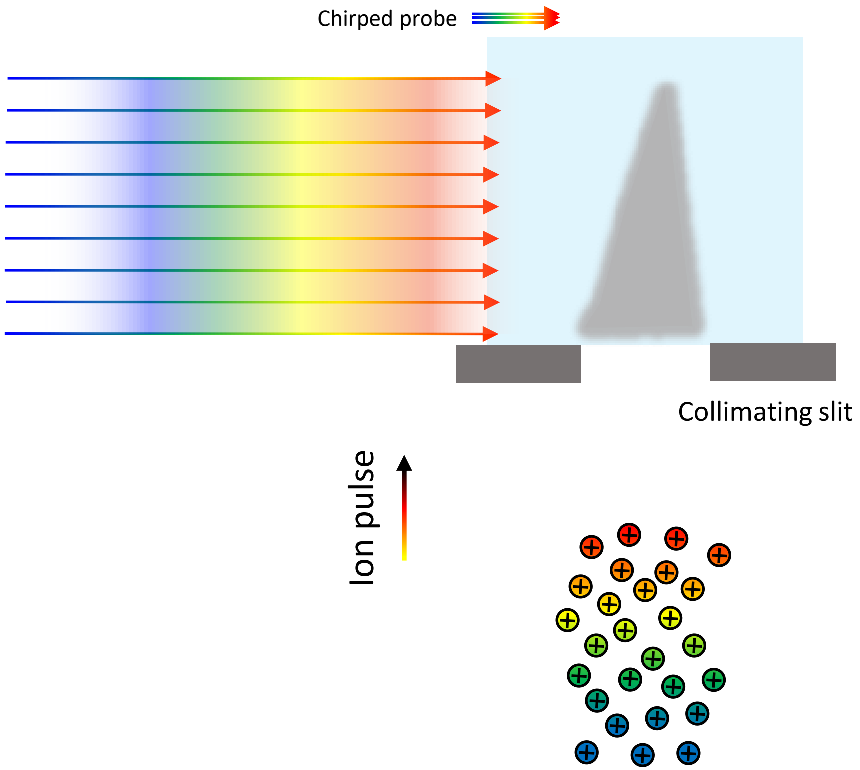
……..Ultrafast optical streaking for proton matter interactions

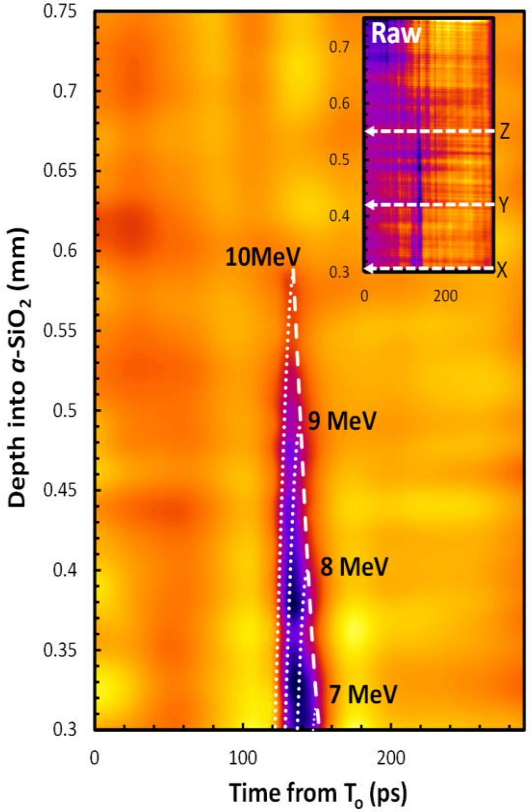
Excitation/ionisation to allow free-free absorption of probe photons ⇒ Chirped pulse optical streaking ⇒ Example of normalised transient opacity data (SiO_2)



Sample can be
Trasparent Dielectric
H_2O

Target:
H_2O, biological samples





Dromey et al. "Picosecond metrology of laser-driven proton bursts", Nat. Comms, 7, 10642 (2016)

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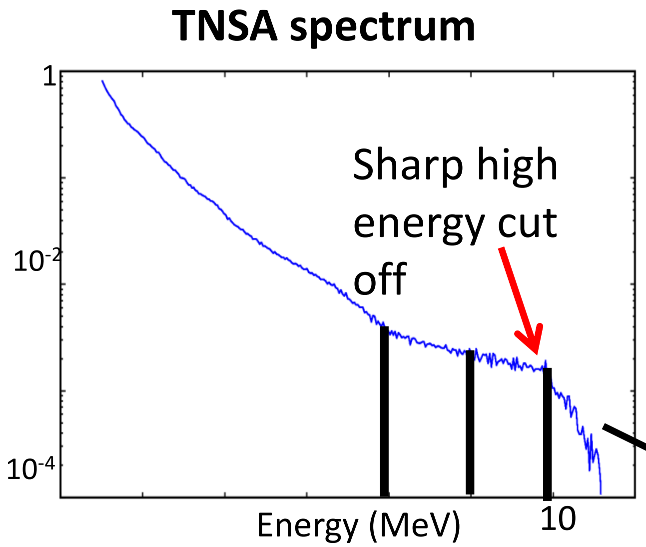
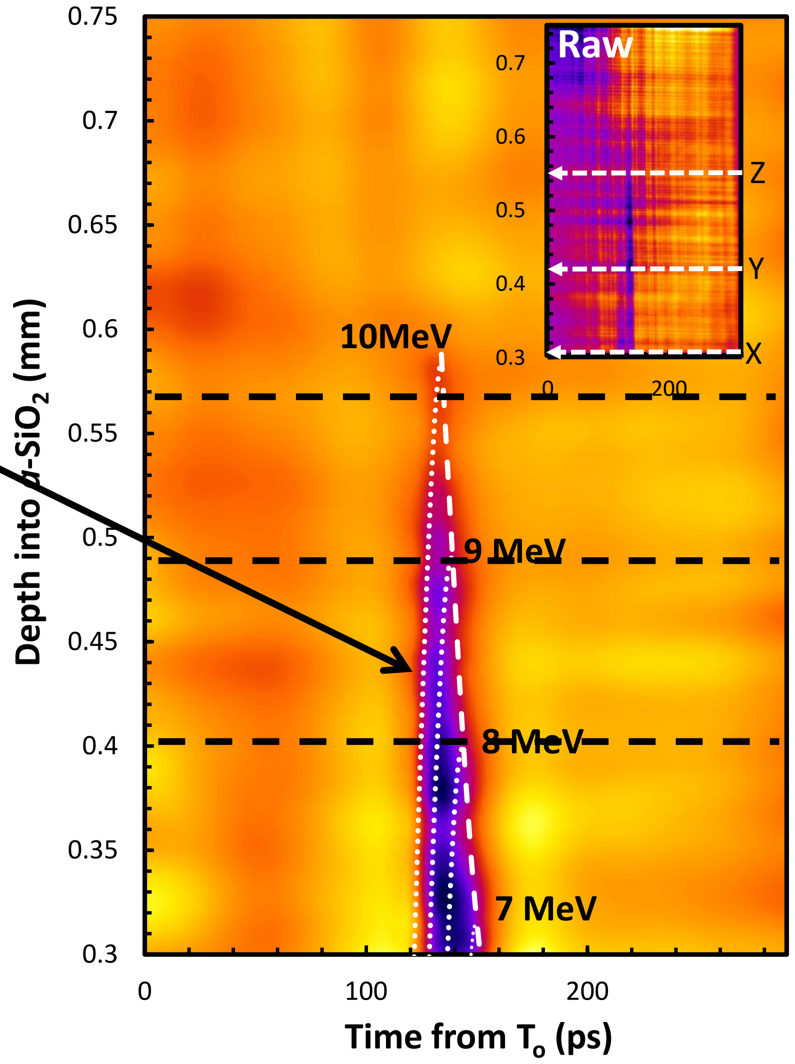
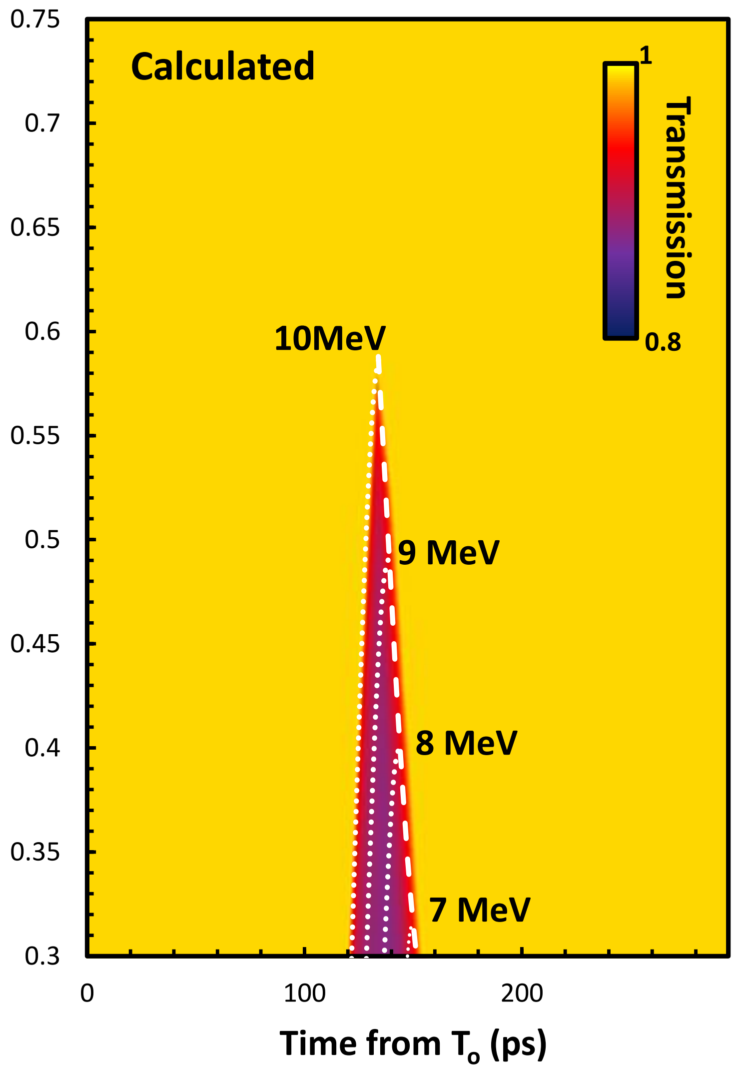
Results: Optical streak of opacity in SiO_2

Dromey et al. "Picosecond metrology of laser-driven proton bursts", Nat. Comms, 2016







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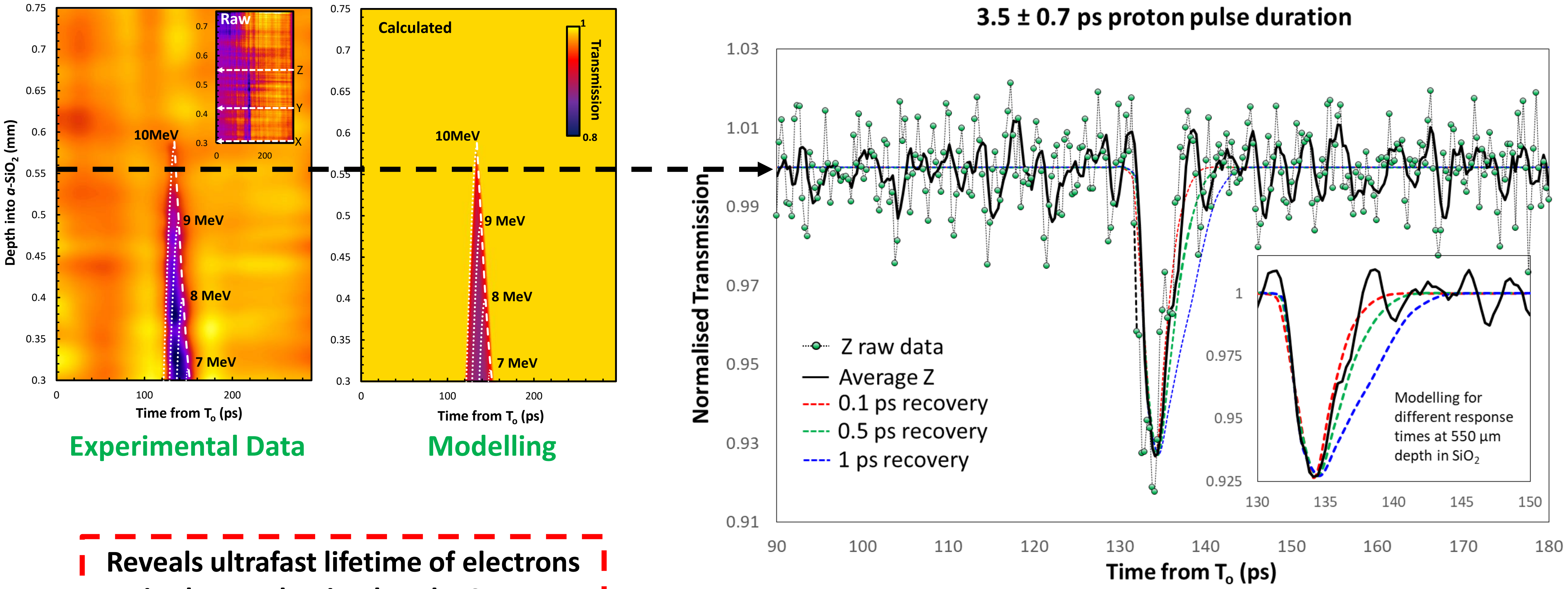
Revealing ultrafast dynamics



Reveals ultrafast lifetime of electrons in the conduction band < 0.5 ps

Dromey et al. "Picosecond metrology of laser-driven proton bursts", Nat. Comms, 7, 10642 (2016)

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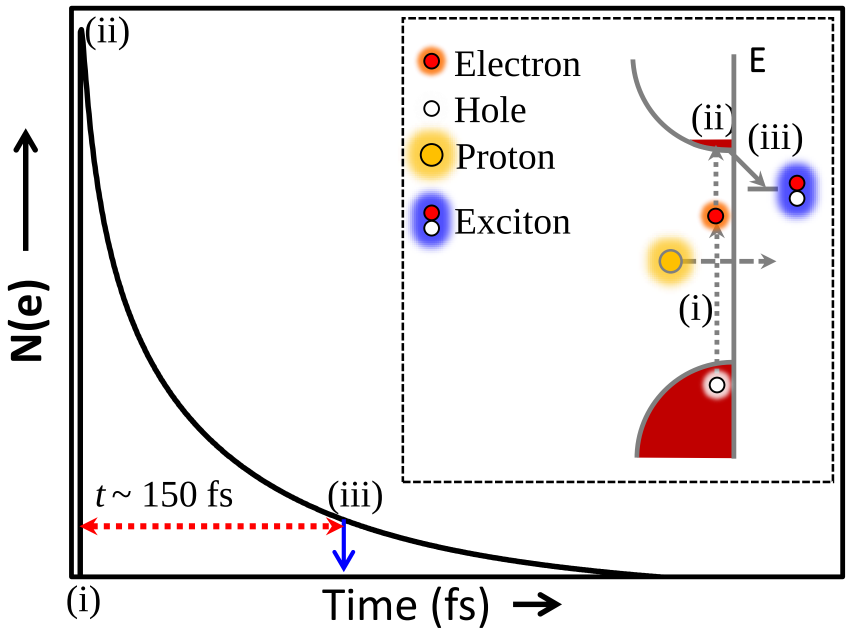
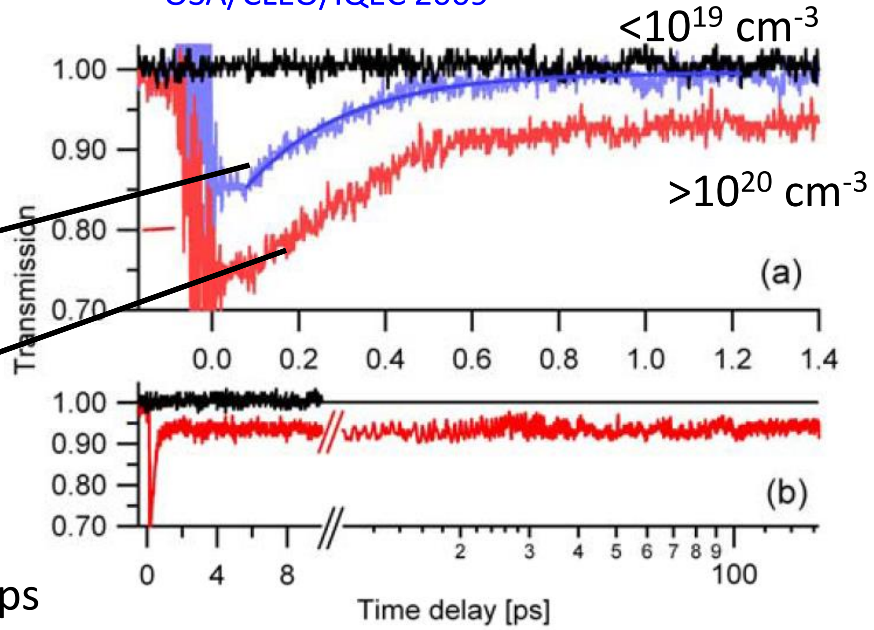
Temporal response of SiO_2 to ionisation – Exciton formation



Exciton (correlated electron hole pair) formation provides a rapid decay channel
⇒ electron is no longer 'free'

Audebert et al., Space-Time Observation of an Electron Gas in SiO_2, PRL 73, 1990 (1994)

D. Grojo "Time-Evolution of Carriers after Multiphoton Ionization of Bulk Dielectrics" " IThI3, 2009 OSA/CLEO/IQEC 2009



"Two speed decay"

Transition from free electron gas to electron hole plasma

$<10^{19}$ cm^{-3} Rapid decay

$>10^{20}$ cm^{-3} Slow decay

Conduction band electron component exists for >100 ps

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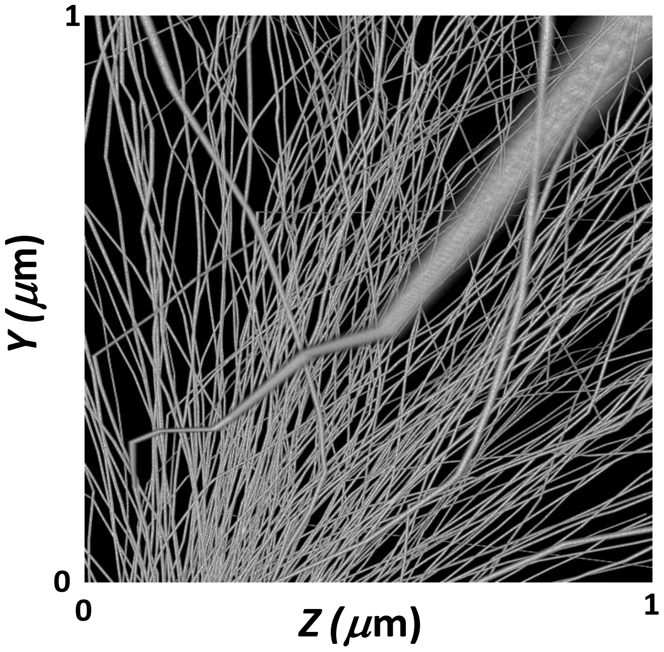
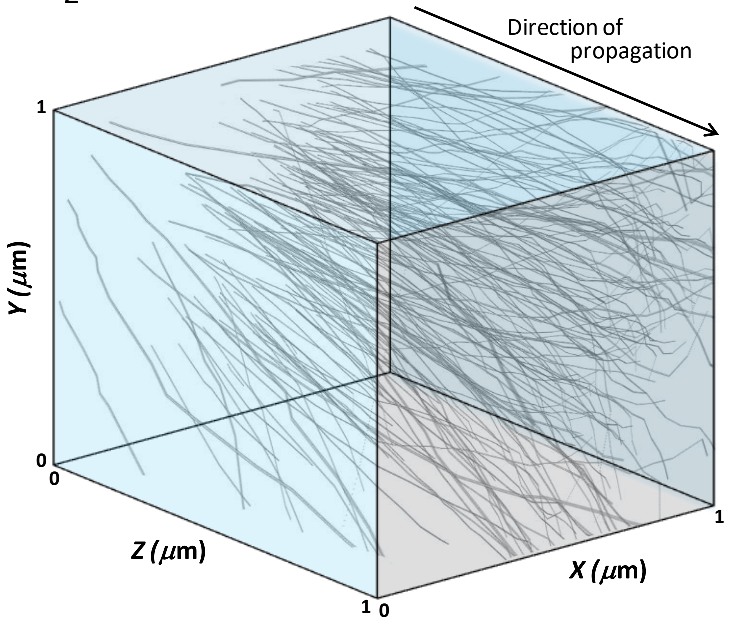
Nanostructured dose distribution of protons in SiO_2



Results suggests near solid density ionisation along each track >10^{21} cm^{-3}

Nanoscale tracks of ion damage from a combination of TRIM calculations (trajectories) and FLUKA simulations (track size) for protons in SiO_2



Flux: 50-100 μm^{-2} – same as the experimental conditions

Electron – Hole plasma conditions $\Rightarrow$ **not suitable for exciton formation**

Why are we seeing an ultrafast response in experiments?

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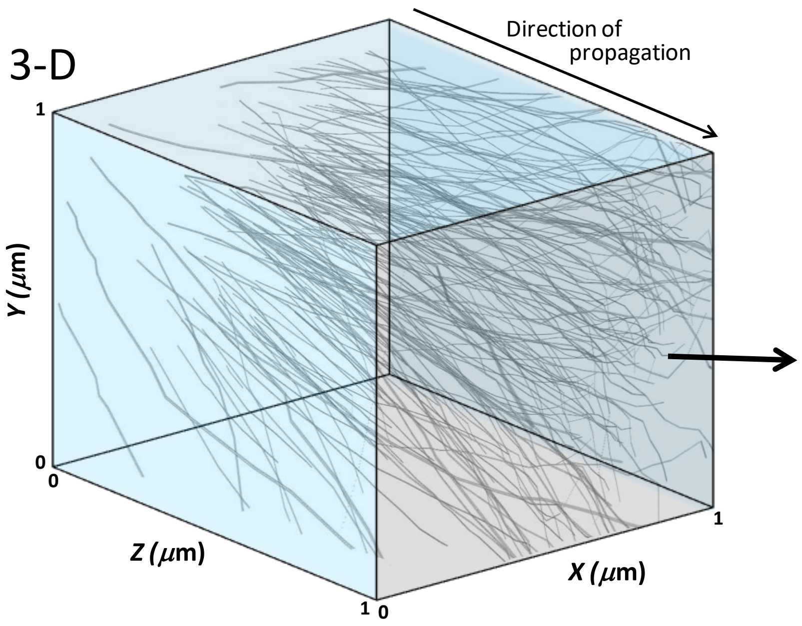
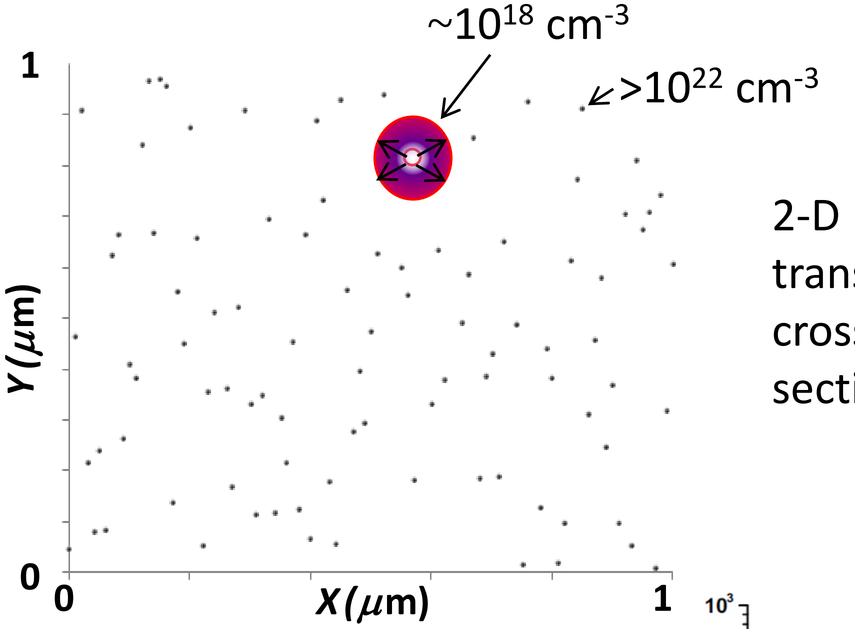
Hypothesis – rapid evolution of localised free density

This only the instantaneous picture…..

Nanometre scale energy density gradients

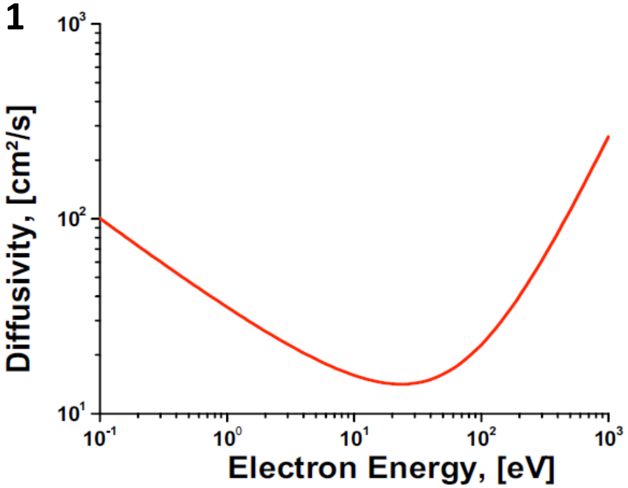




2-D transverse cross section

Such evolution would be consistent with predictions from Monte Carlo simulations for electron diffusivity

Osmani et al. *e-J. Surf. Sci. Nanotech. Vol. 8 (2010) 278-282*



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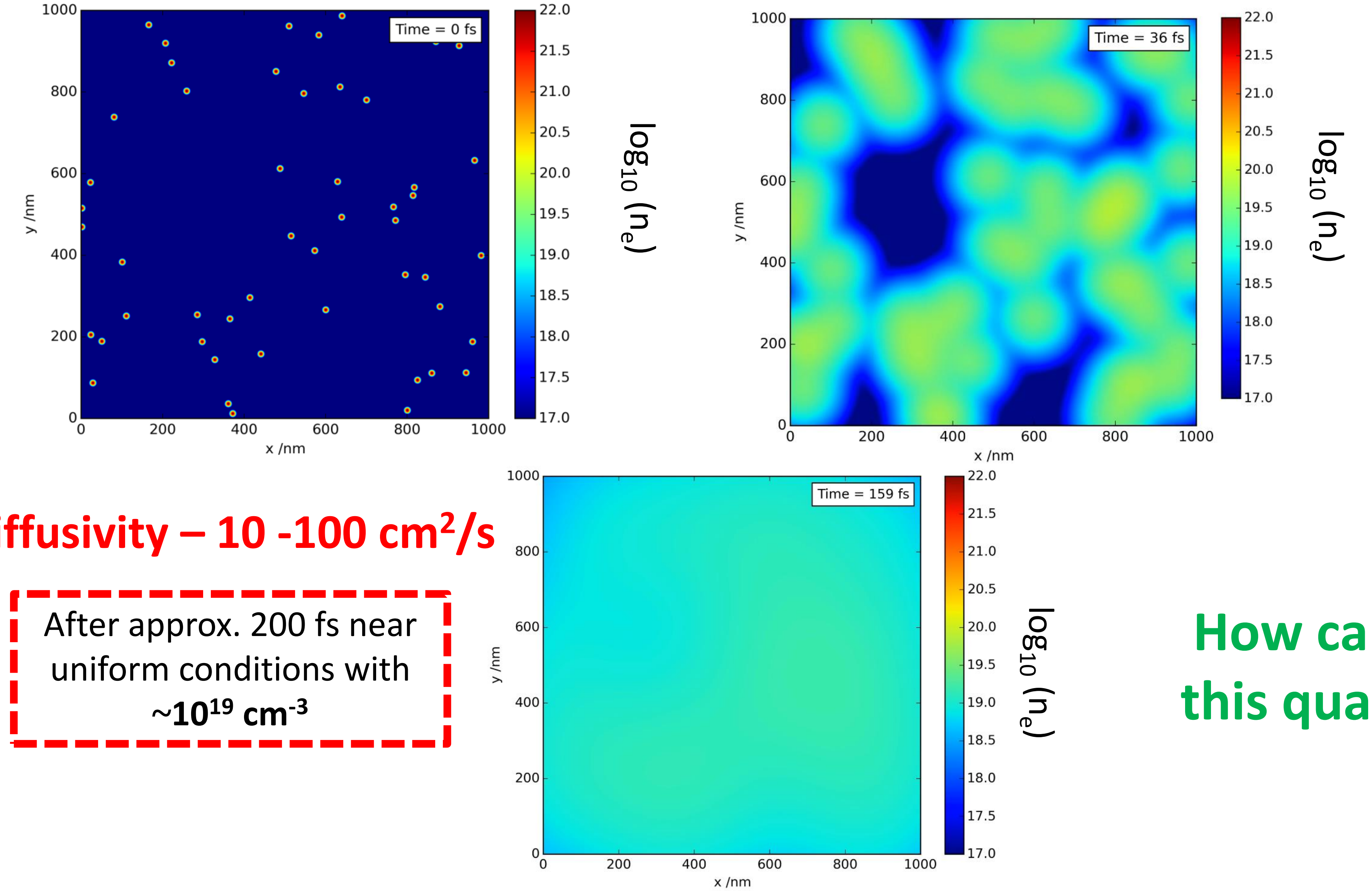
Implied rapid evolution of density (simplified)



Diffusivity – 10 -100 cm²/s

After approx. 200 fs near uniform conditions with **~10¹⁹ cm⁻³**

How can we study this quantitatively?

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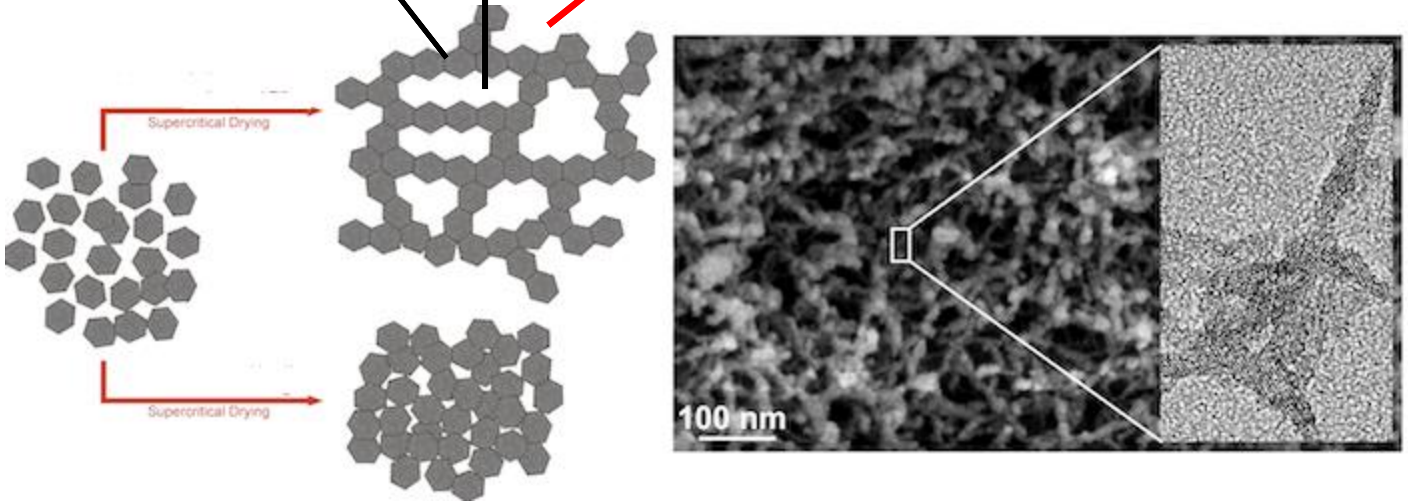
SiO_2 Aerogel – reduced dimensionality

20 - 30 nm voids

2-4 nm structures

Tortuous nanostructured matrix of SiO_2

2-4 nm particles of SiO_2



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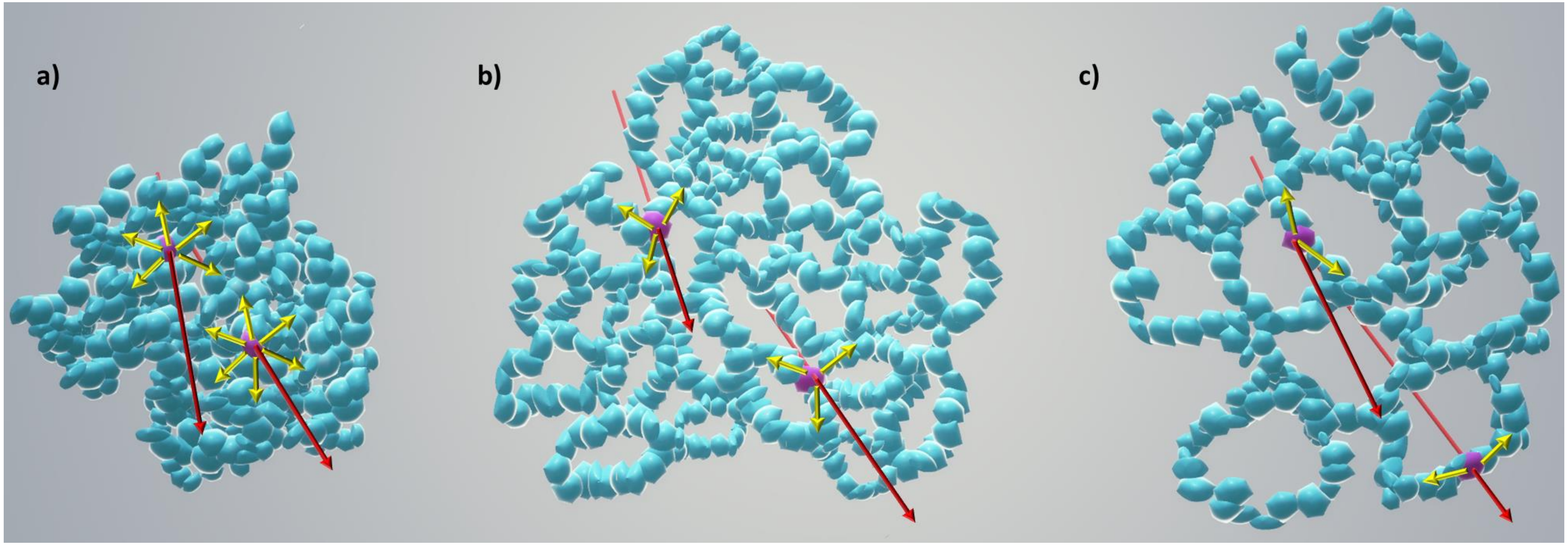
SiO_2 Aerogel – reducing dimensionality

(schematic, for illustrative purposes only)

2.6 gcm^{-3} .26 gcm^{-3} 0.8 gcm^{-3}



3D → Approx. 1D

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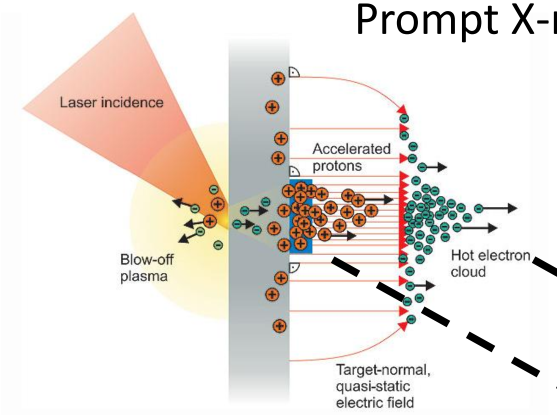
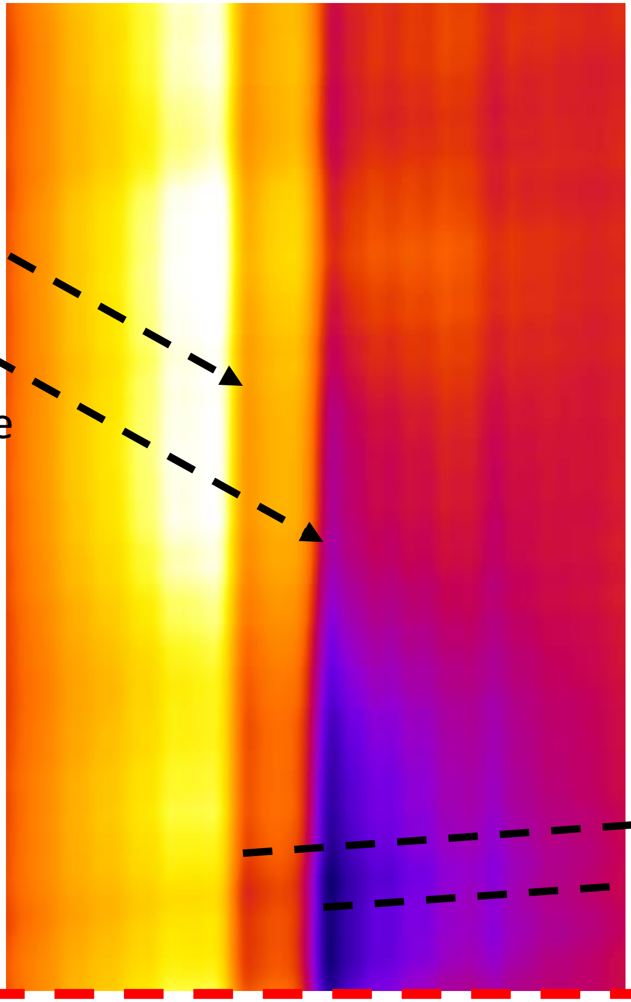
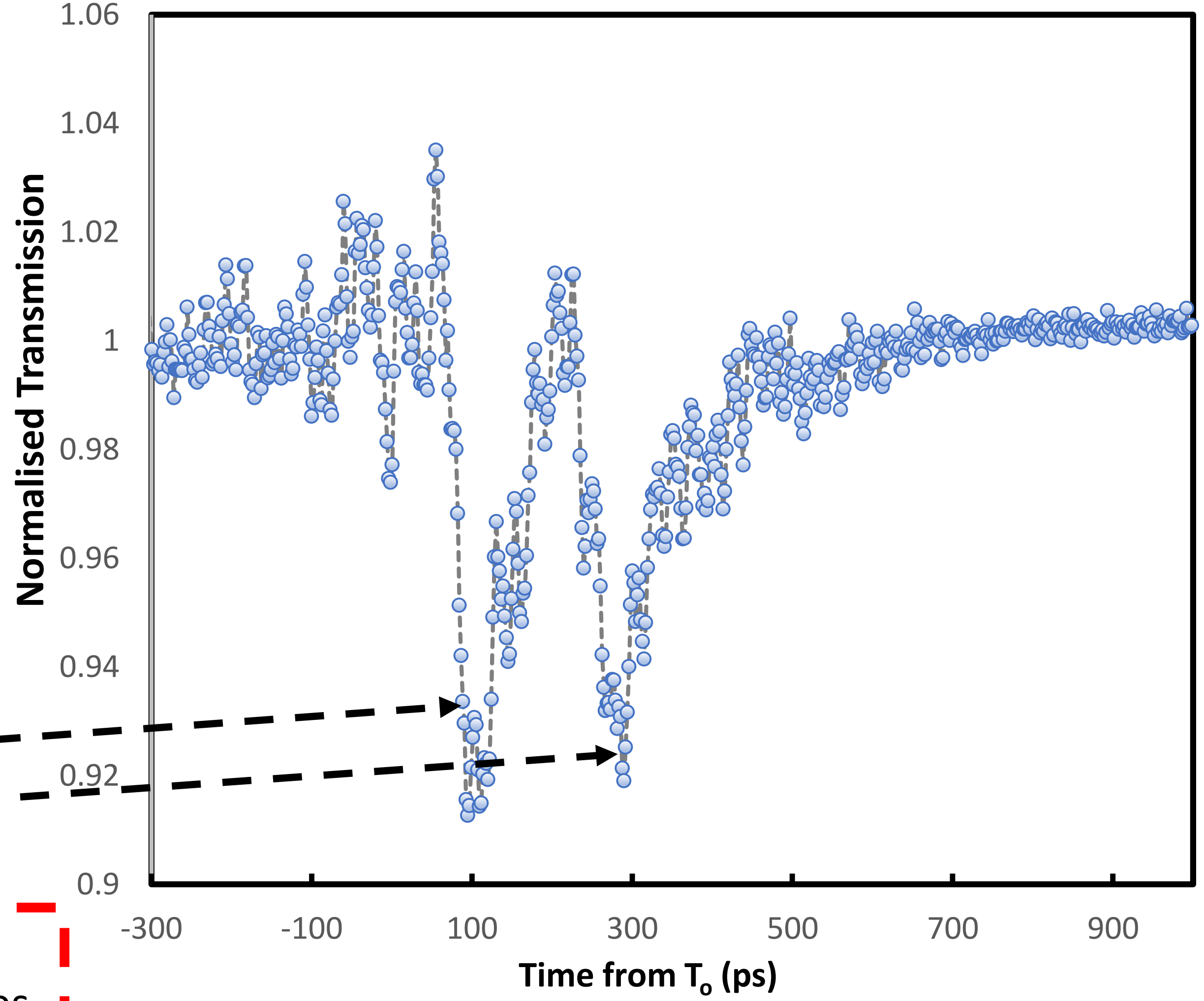
Results: Optical streak of opacity in ~ 0.26 gcm^{-3} Aerogel

Prompt X-rays/Fast electron pulse – gives very accurate t_o



TNSA ion pulse





Yields direct information about the absolute arrival time of various species

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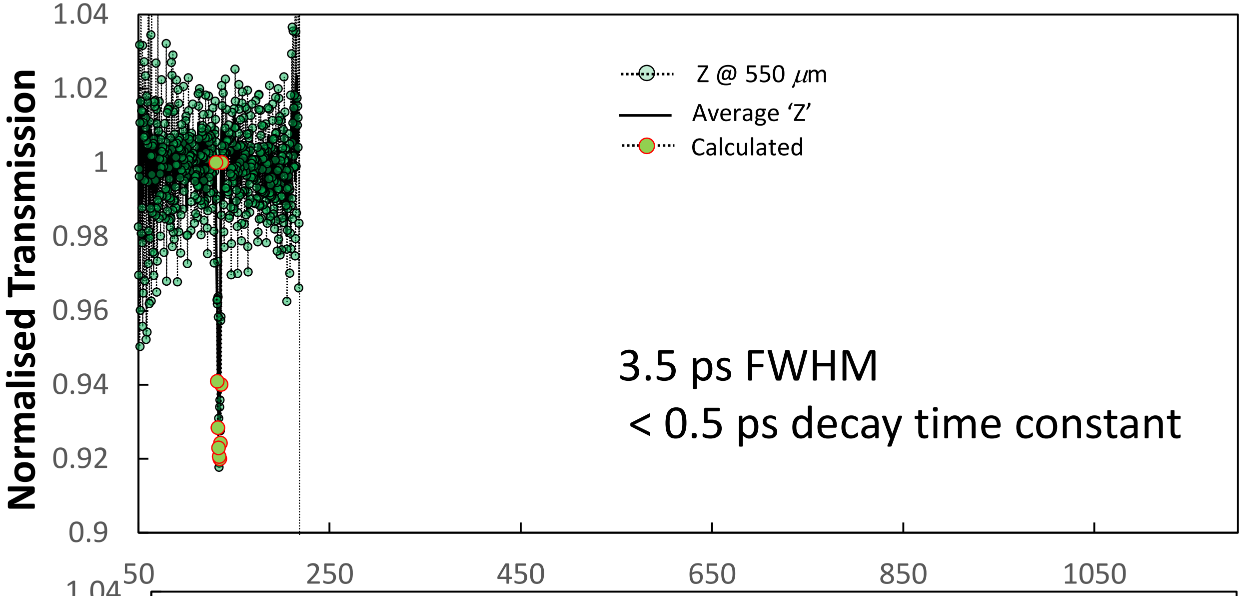
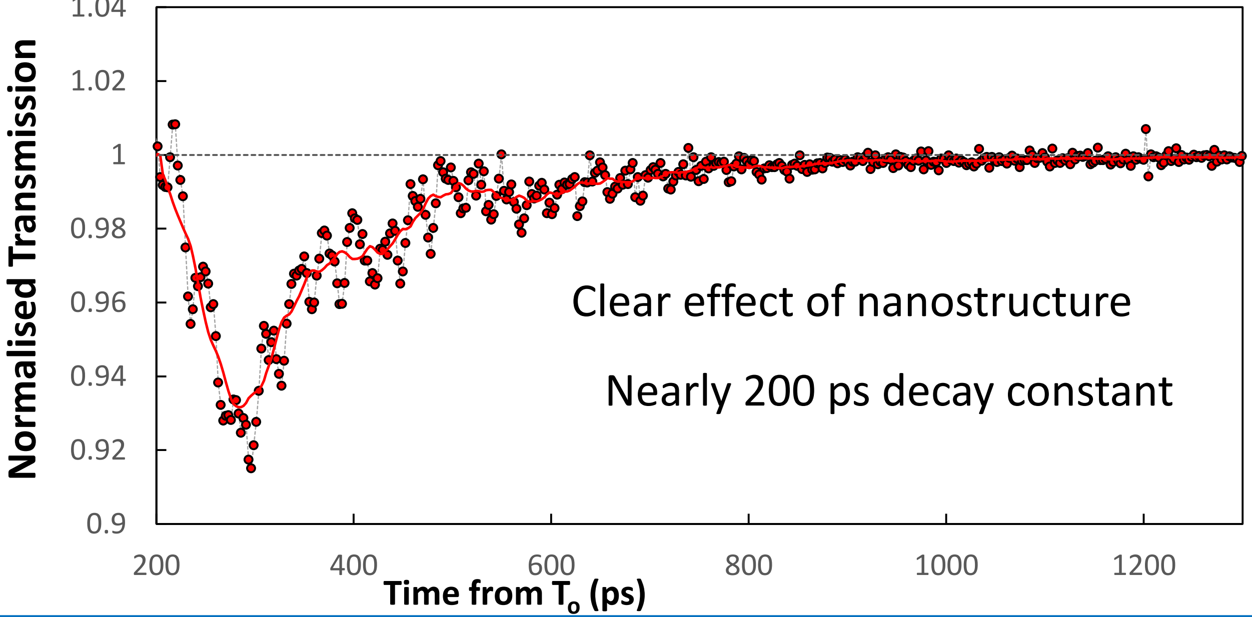
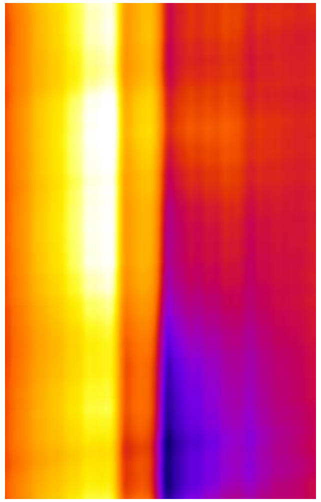
Direct comparison with solid density SiO_2



Streaks on the same timescales





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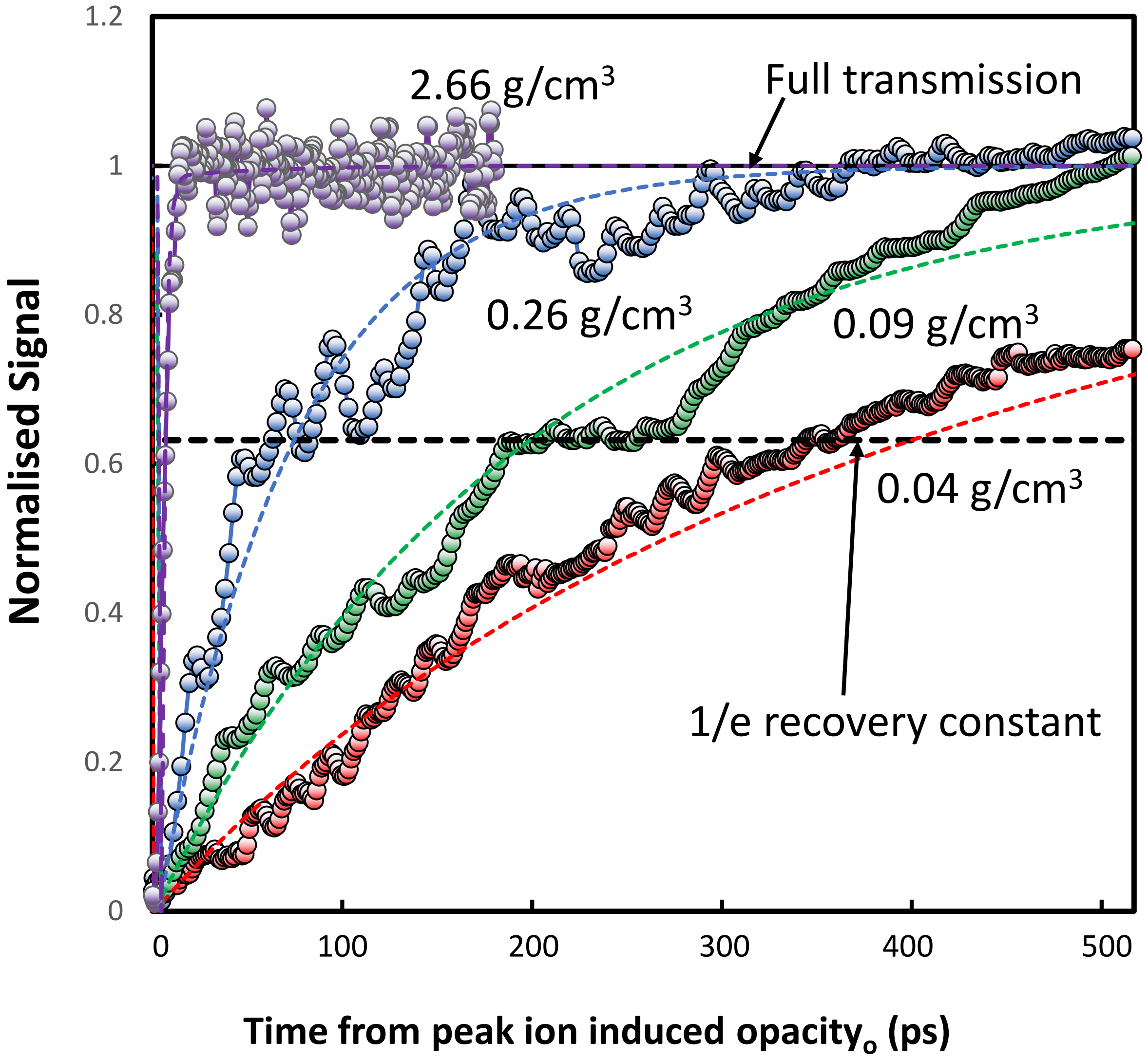
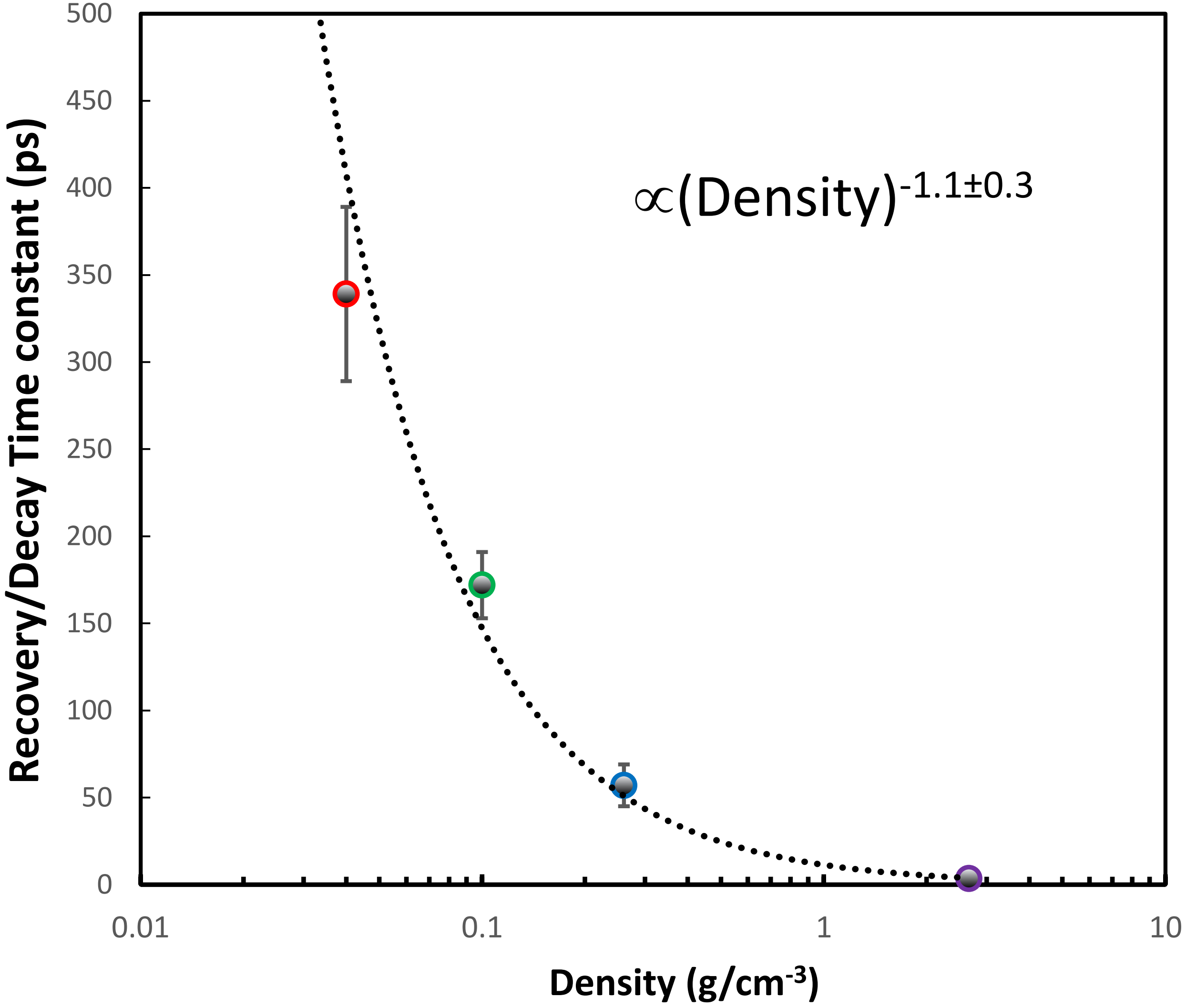
Scaling with average density/dimensionality





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Summary SiO_2

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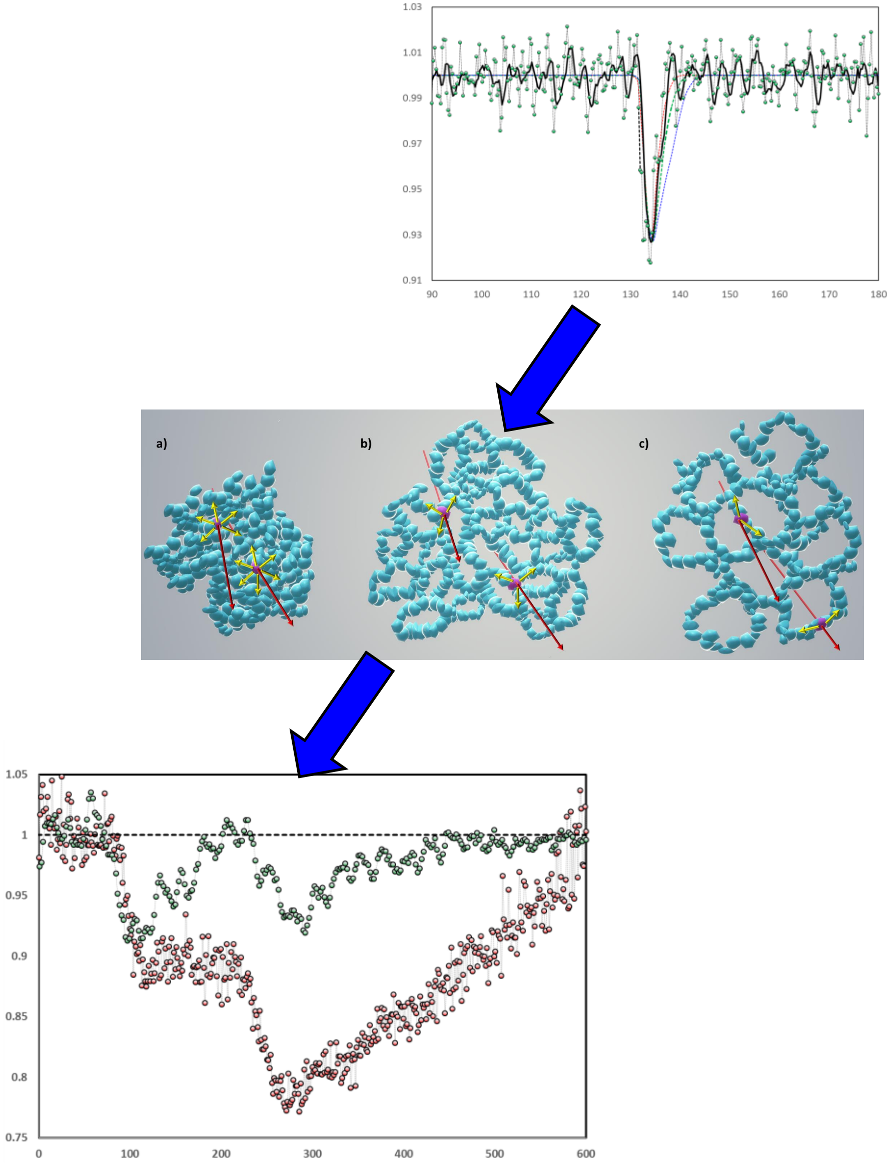
- Possible to directly measure proton damage in materials (here transparent dielectrics) on ultrafast timescales
- In bulk SiO_2 this allows direct measurement of the proton pulse duration.
- Nonlinear variation in recovery times for changing the dimensionality of the interaction for nanostructured SiO_2
- **Clear conclusion**: Reduced dimensionality inhibits the exciton pathway
- **Next step**: Absolutely verify scaling with average density/dimensionality and underlying physics



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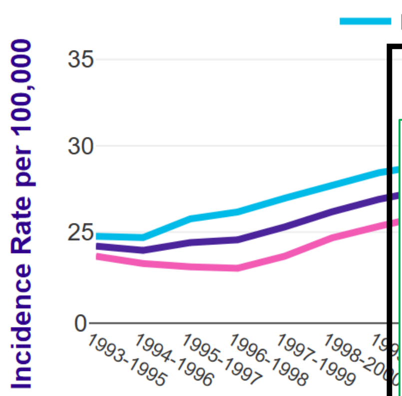
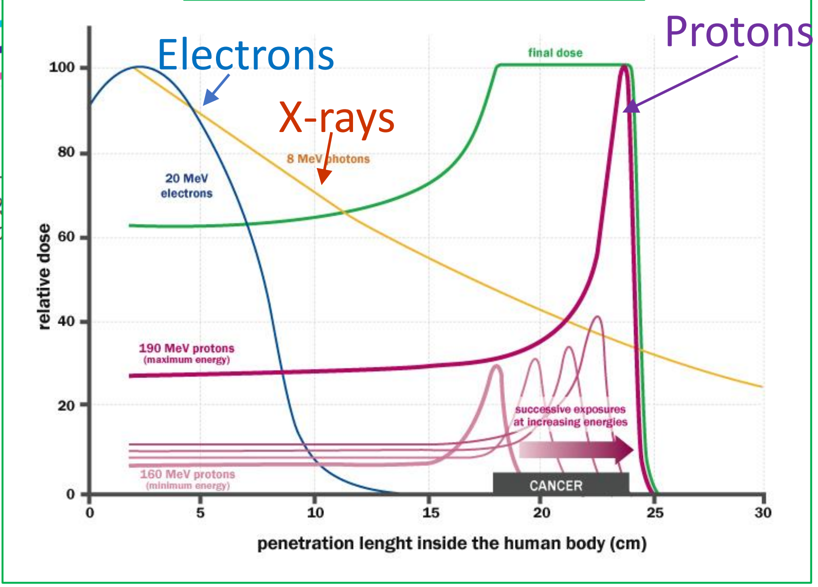
Addressing the rising incidence of cancer

Increasing cancer rates, in particular amongst young people (33% since 1993)

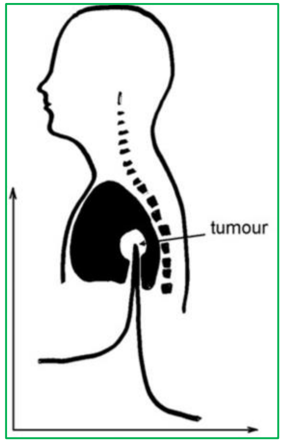


http://www.

Dose vs Depth curves



Ions - Characteristic Bragg peak



Rivista trimestrale dell'Istituto Nazionale di Fisica Nucleare
http://www.roma1.infn.it/rog/astone/didattica/asimmetrie6.pdf

1) Dose escalation
and/or
2) Highly targeted dose for radiosensitive treatment sites

This is driving major facility development and proliferation

https://www.klinikum.uni-heidelberg.de/

ULTIMATE GOAL - Improve long term prospects for young patients by reducing damage near the tumor site

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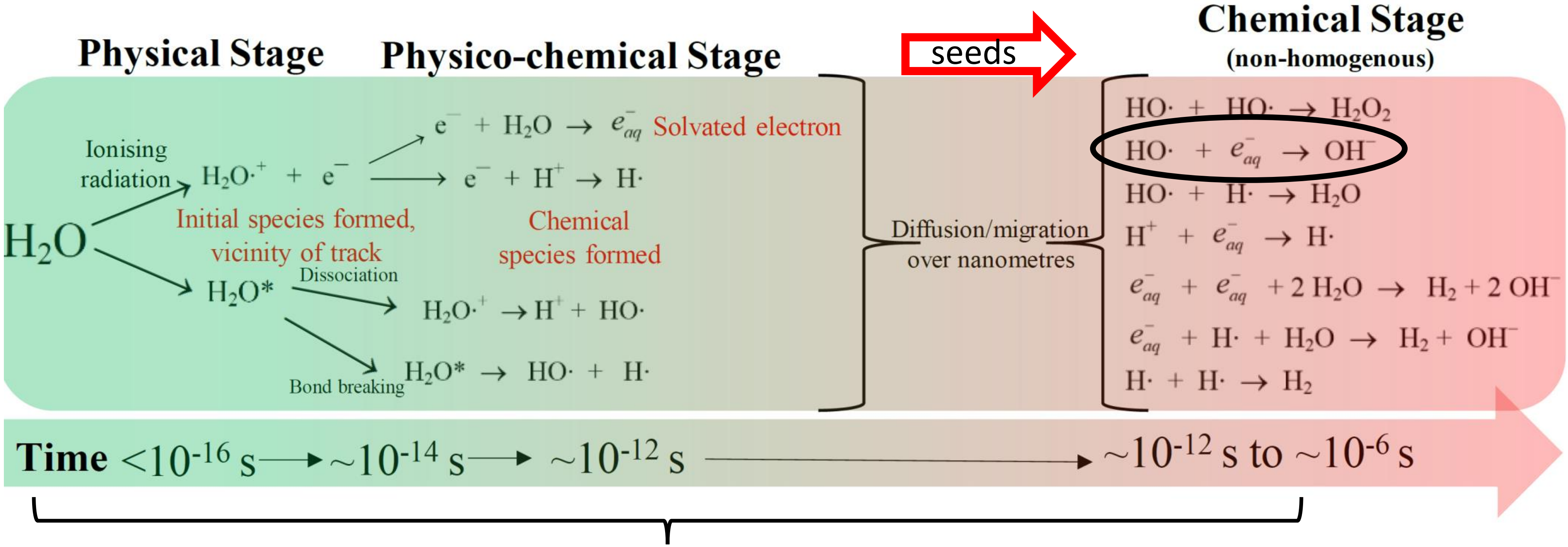
Ultrafast ion interactions in H_2O – nascent chemistry



e^{-}_{aq} is a powerful reducing agent for the cytotoxic HO·

This timeframe has been <u>inaccessible</u> to experimental observation to date

This prevents the benchmarking of *ab initio*, or "bottom up", numerical models

This in turn prevents unlocking the predictive power of these models

Baldacchino, G., "Pulse radiolysis in water with heavy-ion beams. A short review" Radiat. Phys. Chem. **77**, 1218– 1223 (2008)

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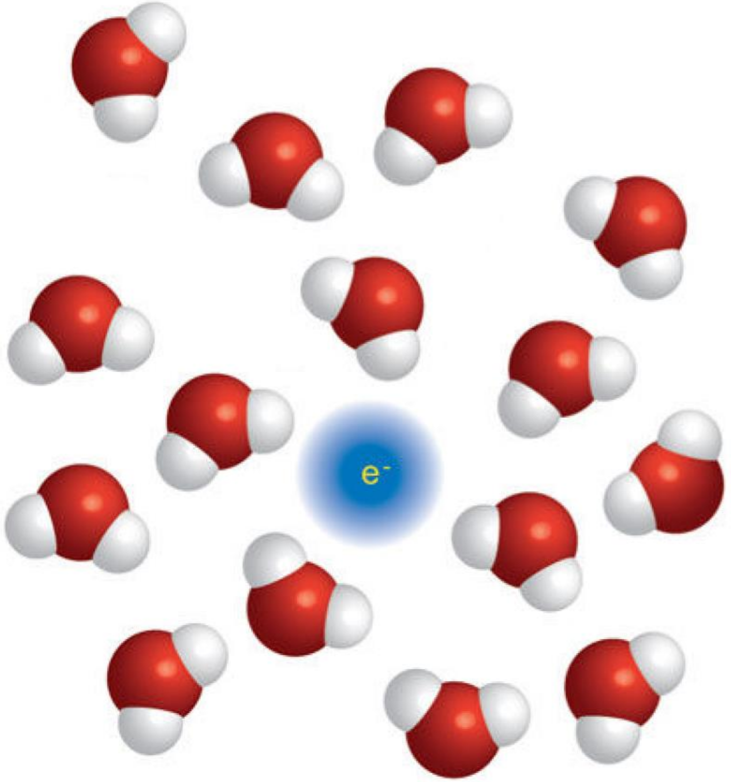
Solvated Electron

Protons ionise water molecules to produce a radical ion and free electron

$$H_2O \xrightarrow{\text{radiation}} H_2O^+ + e^-$$

Through dipolar interactions the electron is captured by the water, becoming solvated

$$e^- + H_2O \longrightarrow e_{aq}$$



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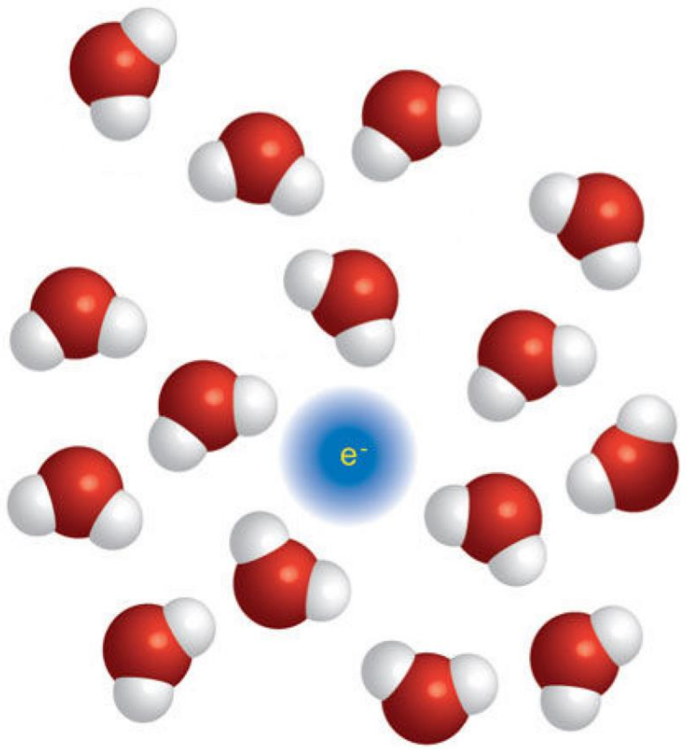
Studying the solvation process using ions

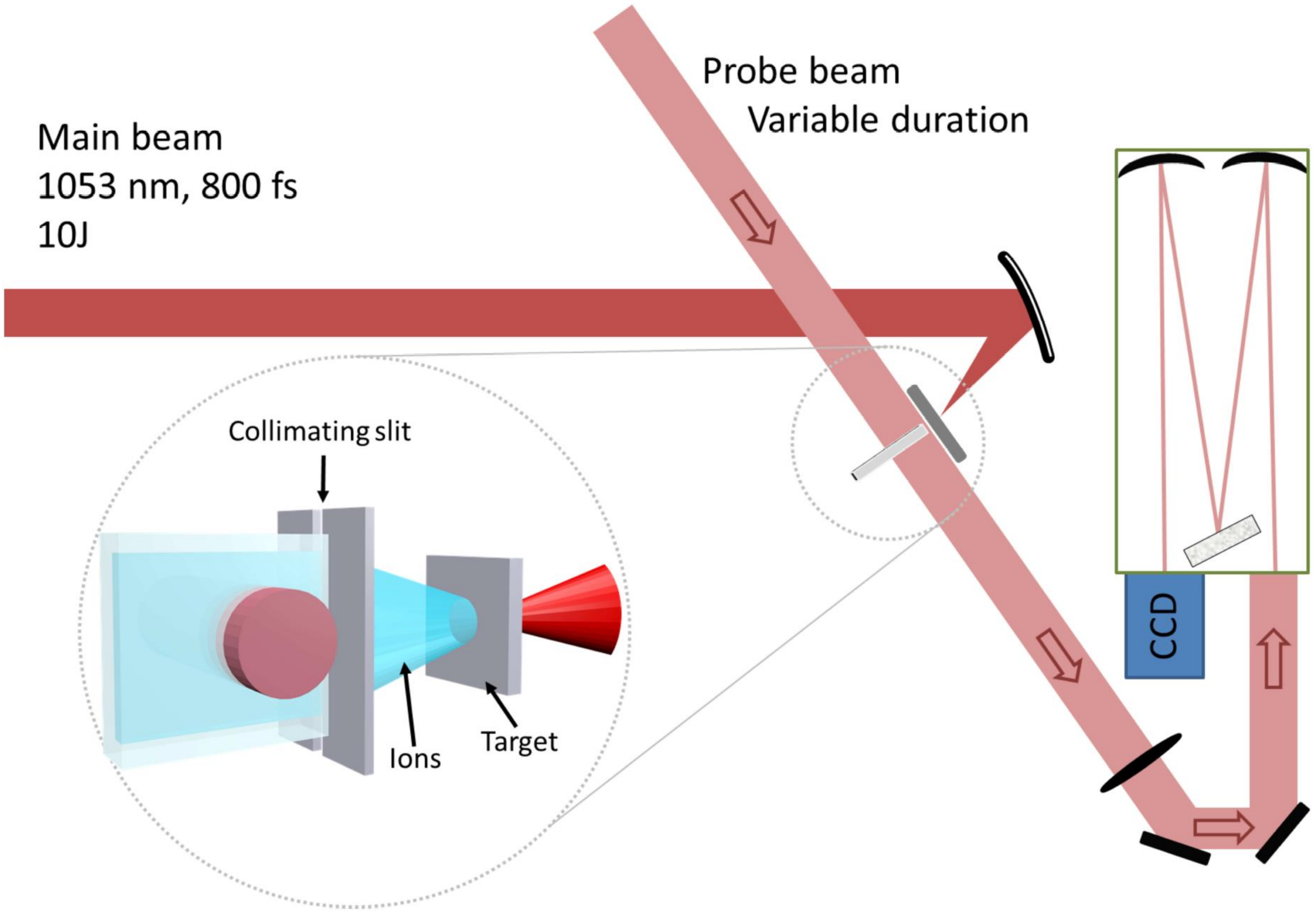
Pulsed ion-radiolysis so far limited by proton pulse duration and probe synchronisation.

Solvated electron extensively studied due to it's high absorptivity and broad absorption spectrum.

- Chemical scavengers added to determine yields of radiolytic products.
- For high temporal resolution – large uncertainty due to concentration of scavenger required.



Observing ultrafast proton interactions in a single shot



Schematic
(not to scale)

Probe ion induced
a) opacity (free electrons in conduction)
b) Photabsoption bands
c) Polaristation effects

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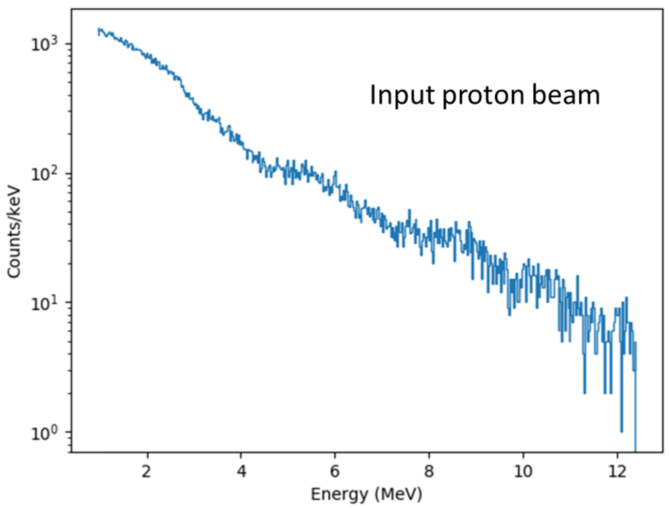
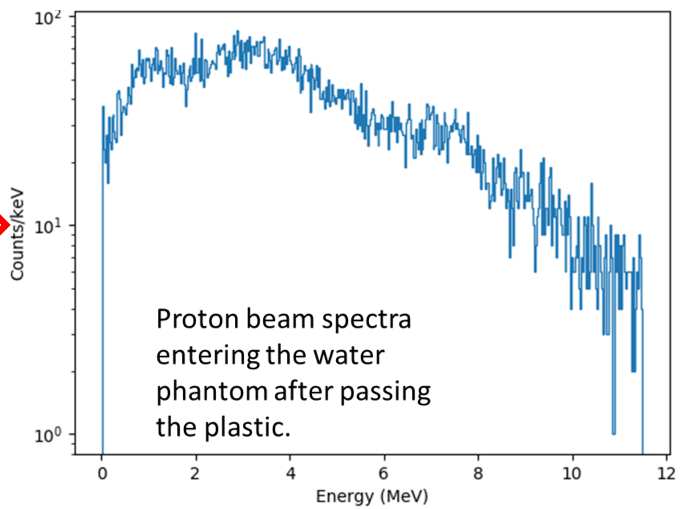
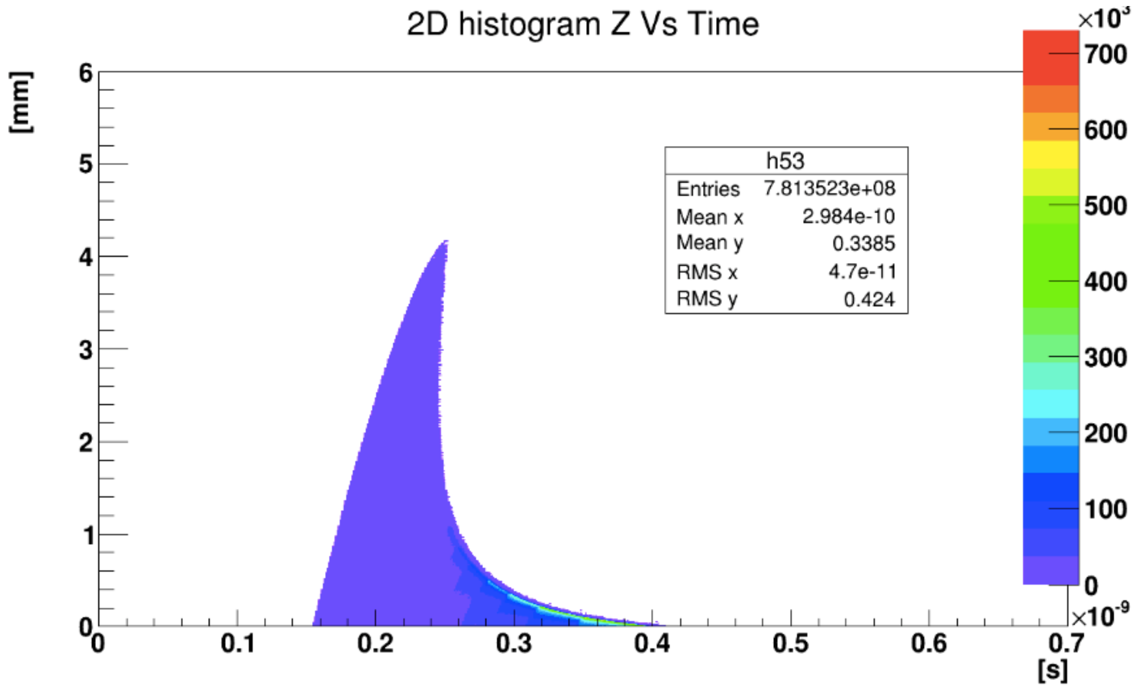
Full modelling of interaction – Geant 4







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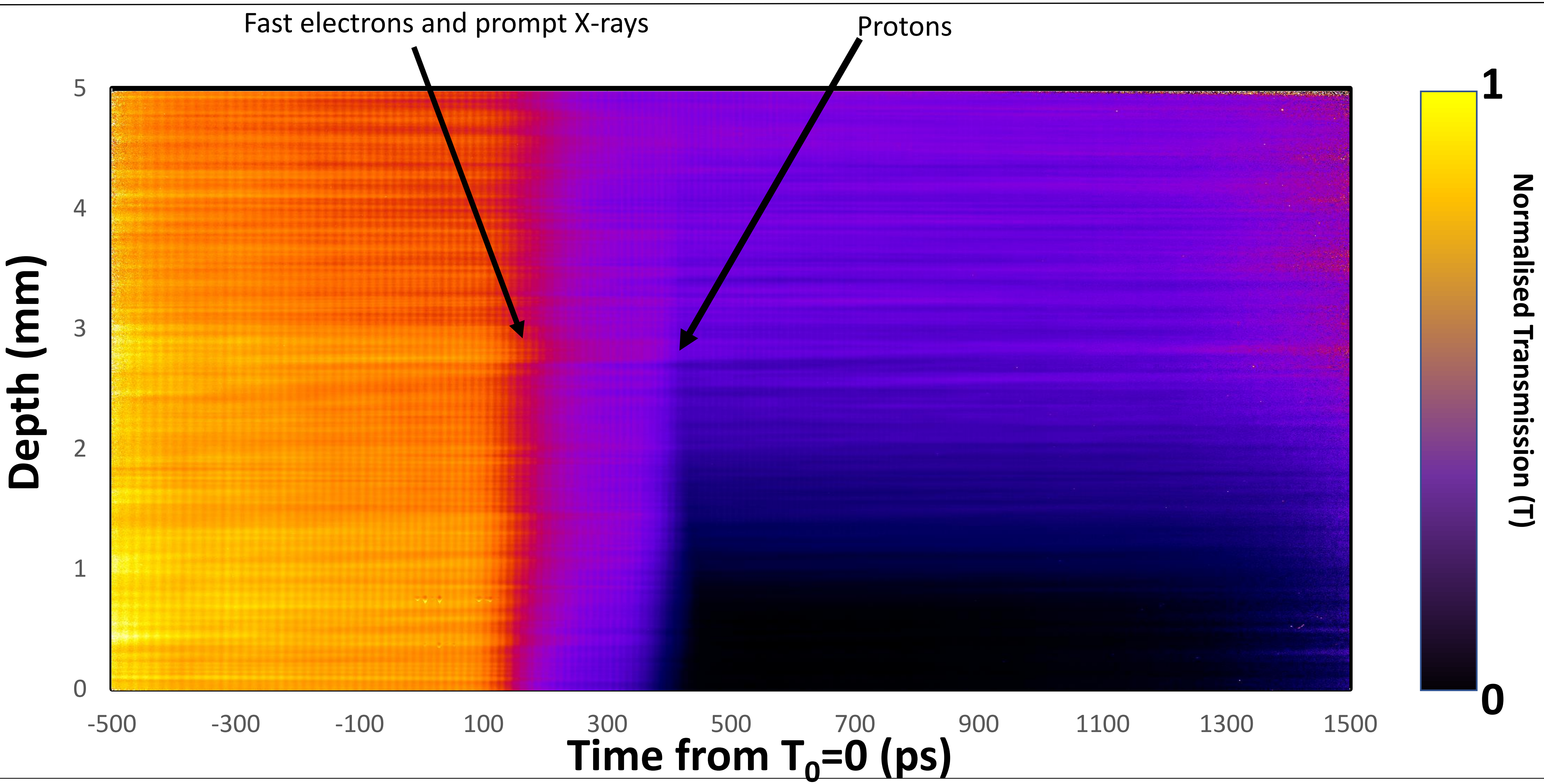
Experimental results optical streak of solvated electron dynamics



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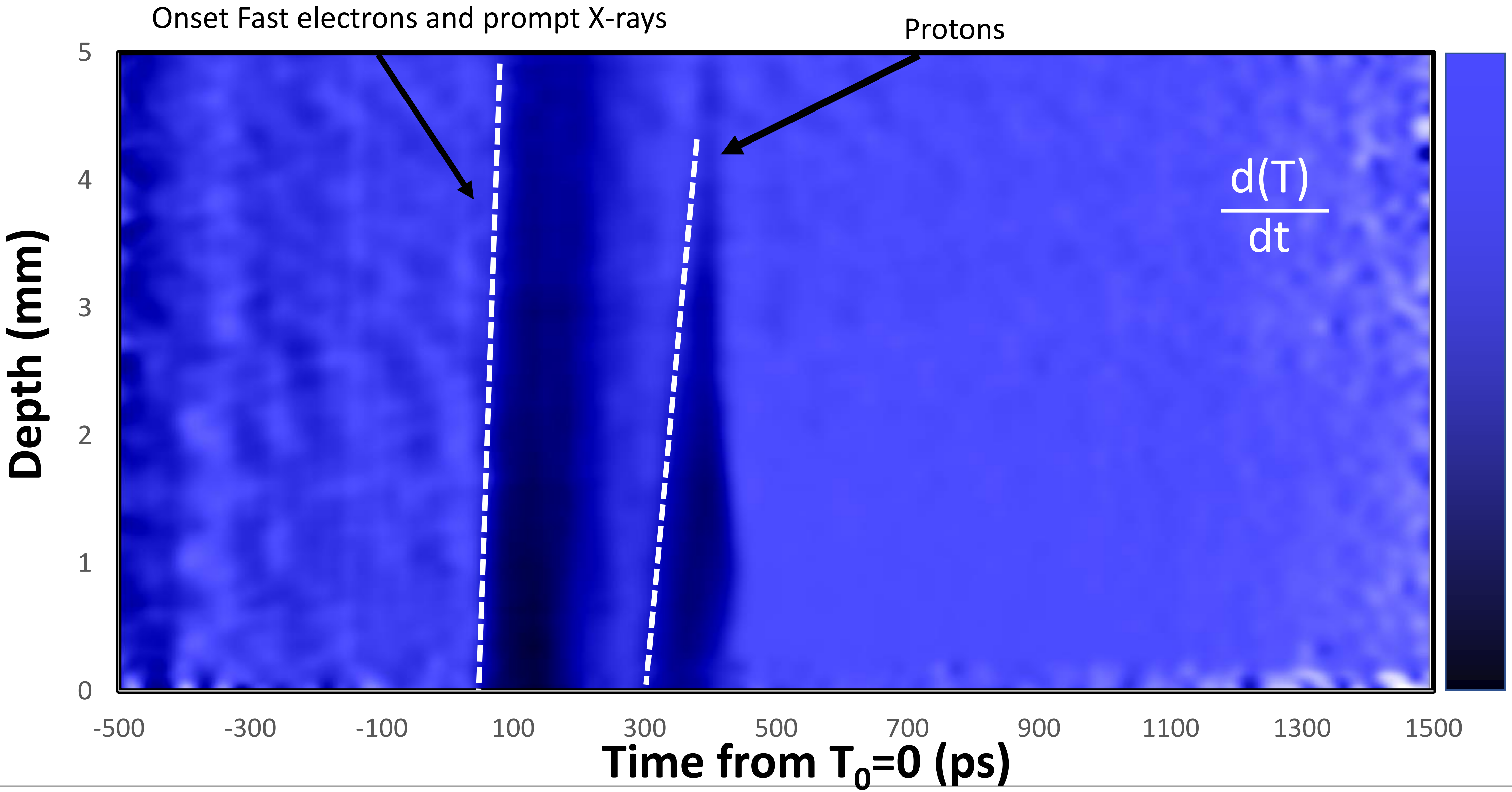

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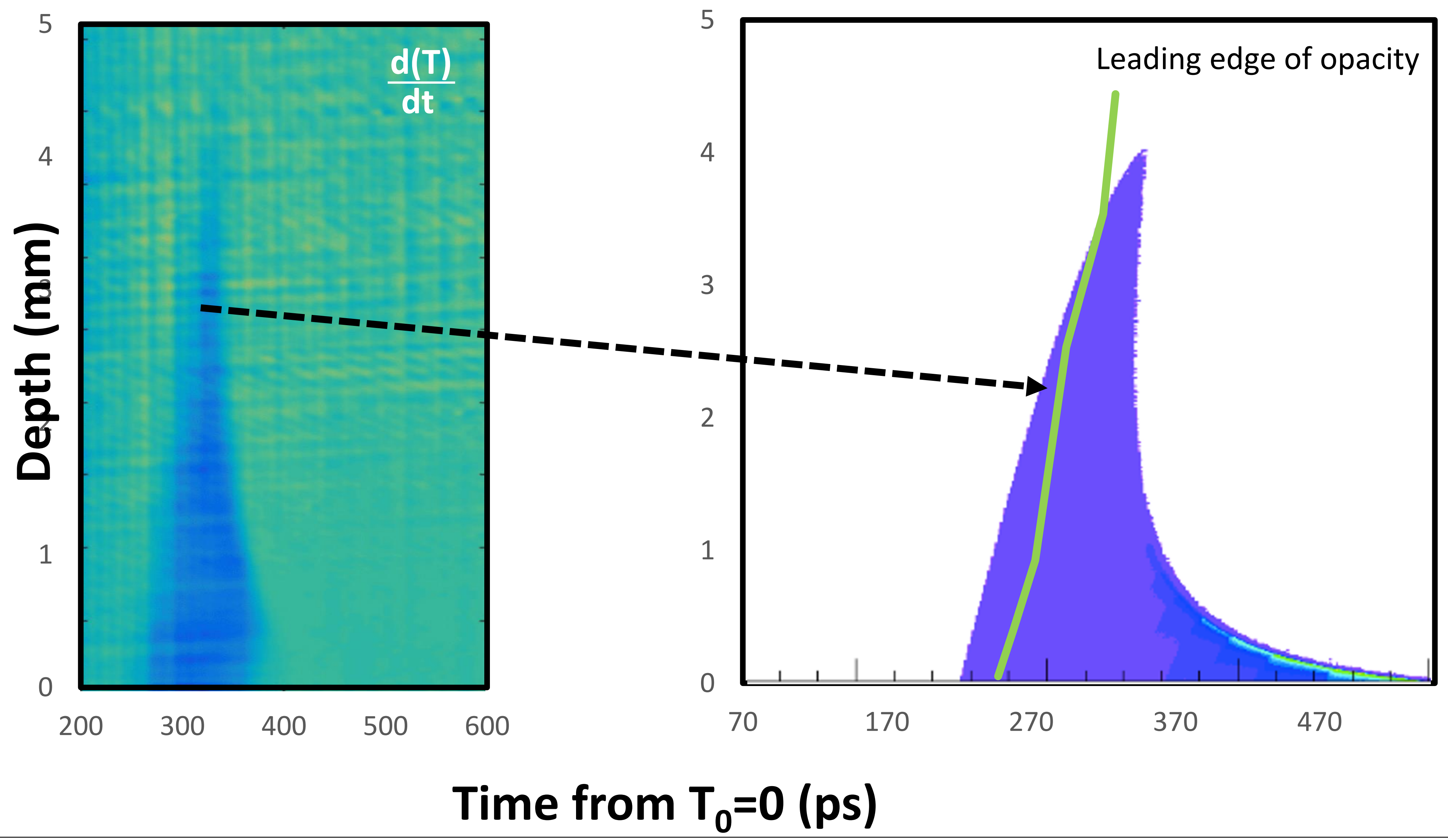
Mismatch between experiment and theory



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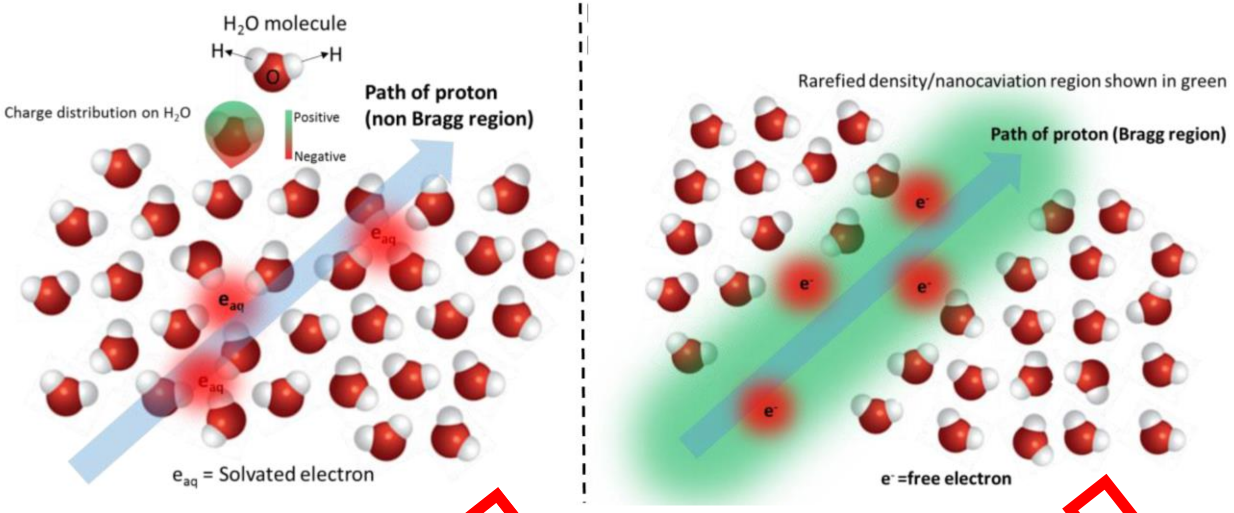
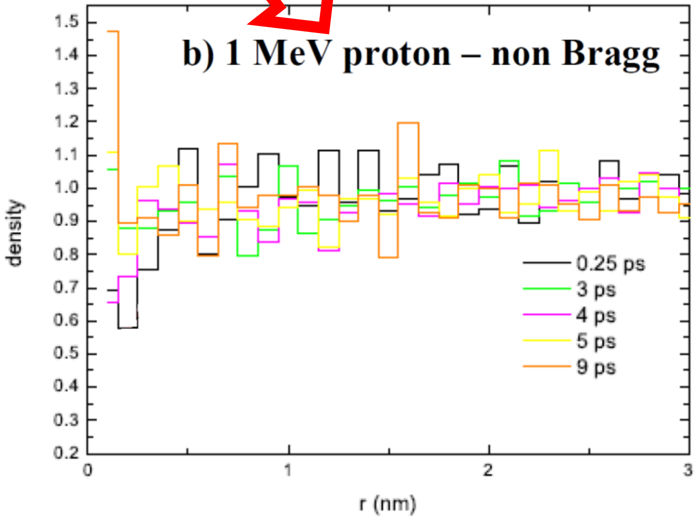
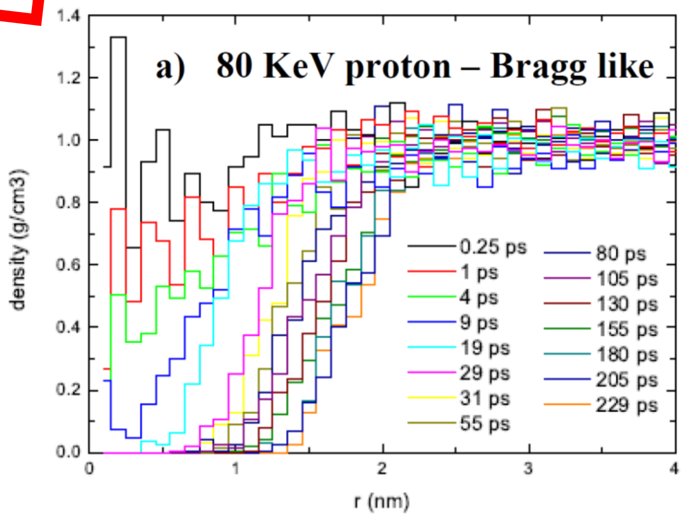
Proton induced dynamics in water – nanocavitation?

Outside Bragg peak region | **Bragg peak region**



Molecular dynamics (MD) simulations indicate a marked difference in H_2O response for increasing linear energy transfer (LET)





Simulation input by P. deVera and F. Currell, Univ. of Manchester

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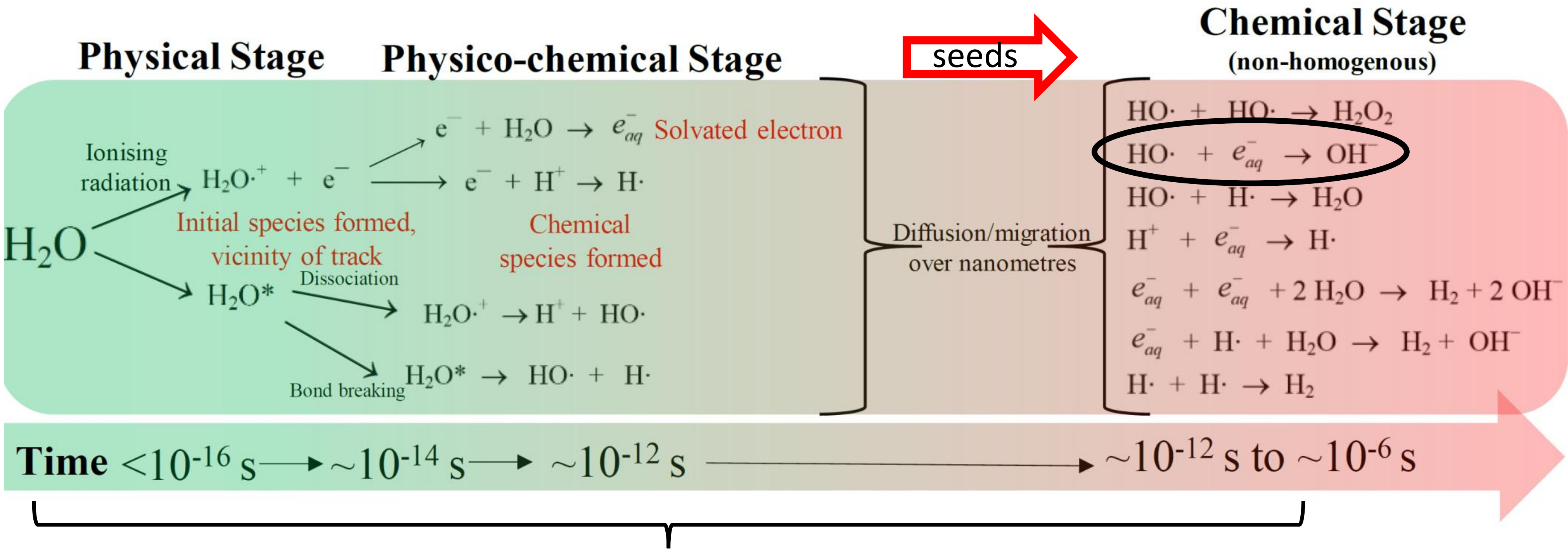
Ultrafast implications for nascent water chemistry?



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