Ultrafast Dichroism via Photoinduced Symmetry Breaking in Plasmonic Metasurfaces

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Introduction
Motivations

Ultrafast control of light at the **nanoscale** is attracting increasing attention and inspiring novel paradigms for high speed all-optical **modulation**:

- **Metasurfaces** upon **ultrashort** illumination
- **Plasmonic** nanostructure optical **nonlinearities**

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All-optical broadband modulation with **full return to zero within 1 ps** remains an open challenge, e.g. for ultrafast **polarisation** switching

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- What is faster than hot carrier relaxation?
- How to exploit such effect for optical modulation?
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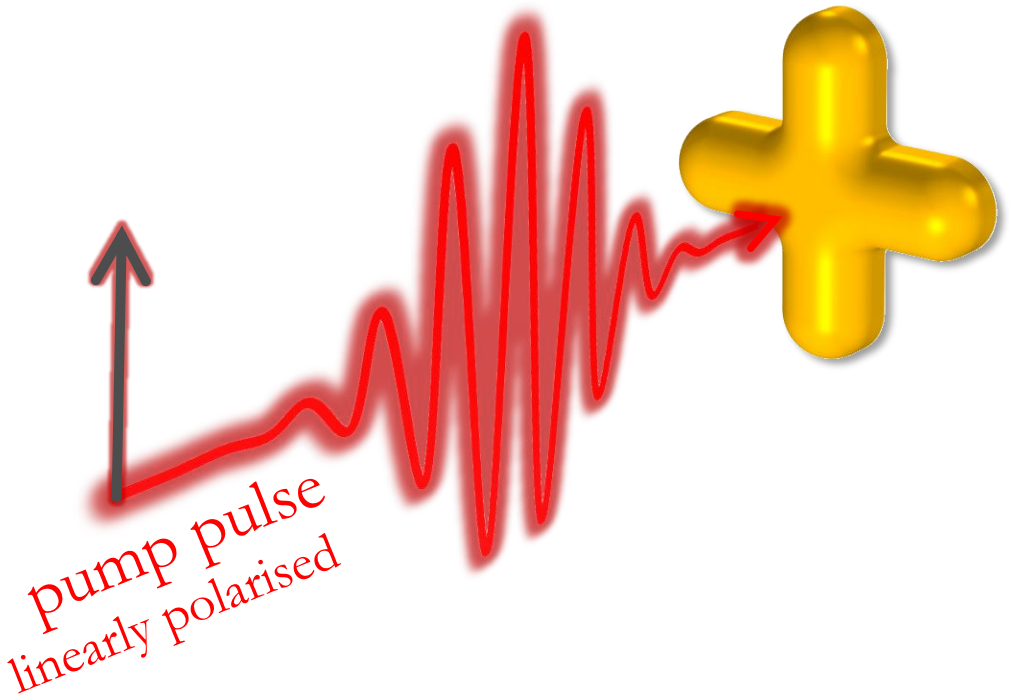
Spatial inhomogeneities
Hot carrier photogeneration



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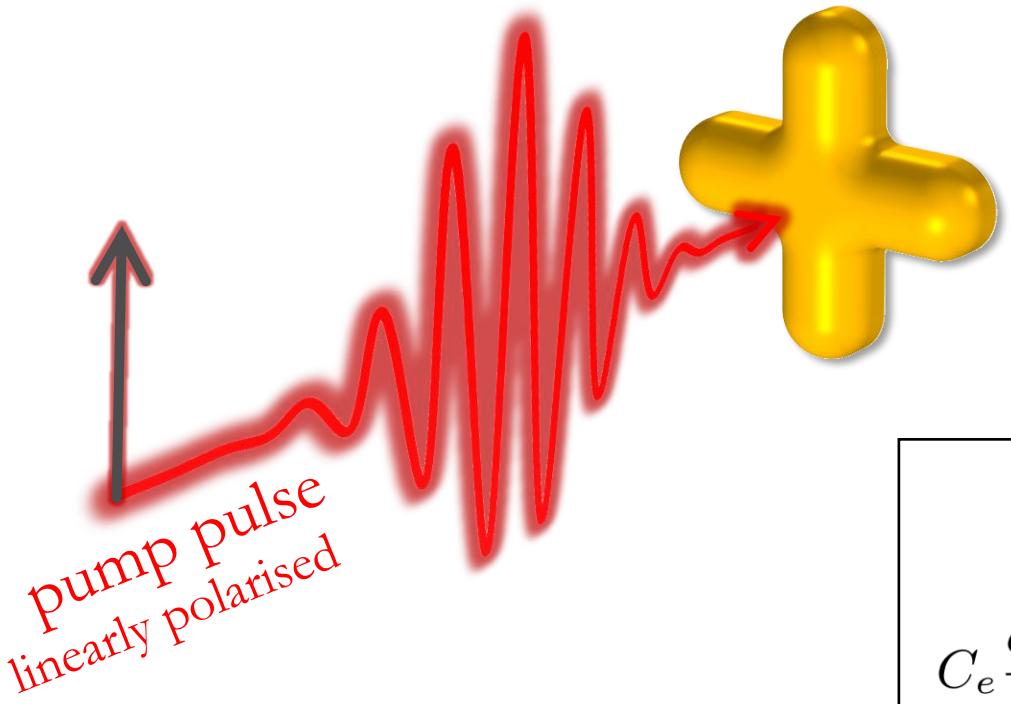

Our **I3TM:**

$$\frac{\partial N}{\partial t} = -aN - bN + P_{abs}(\vec{r}, t)$$

$$C_e \frac{\partial \Theta_e}{\partial t} = -\nabla\Big(- \kappa_e \nabla \Theta_e \Big) - G\Big(\Theta_e - \Theta_l\Big) + aN$$

$$C_l \frac{\partial \Theta_l}{\partial t} = \kappa_l \nabla^2 \Theta_l + G\Big(\Theta_e - \Theta_l\Big) + bN$$

[1-4]

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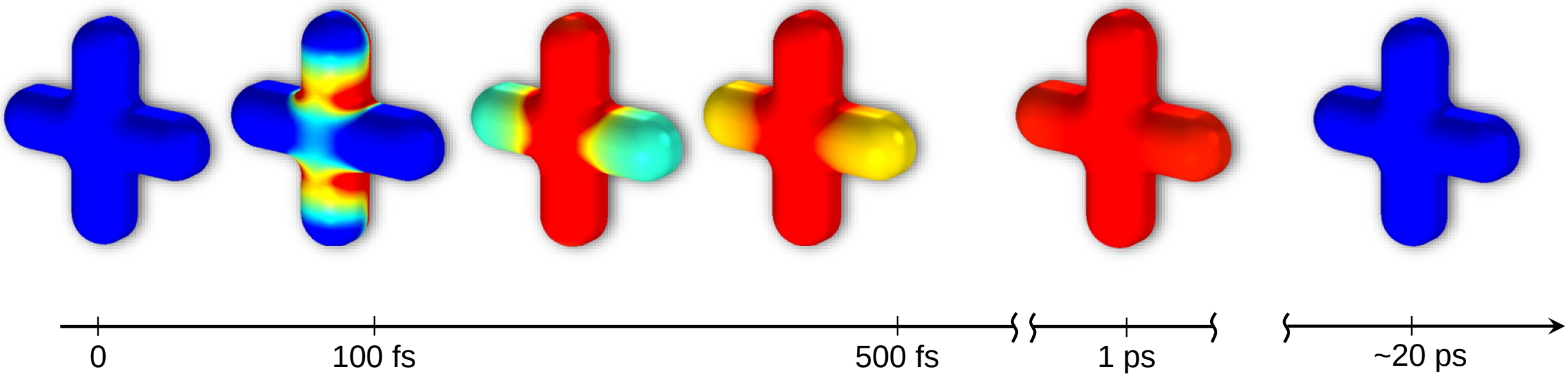

Photoexcitation creates a **spatially non-uniform** hot carrier distribution dictated by the **resonant mode** of the plasmonic structure and evolving in time as a **diffusion** process

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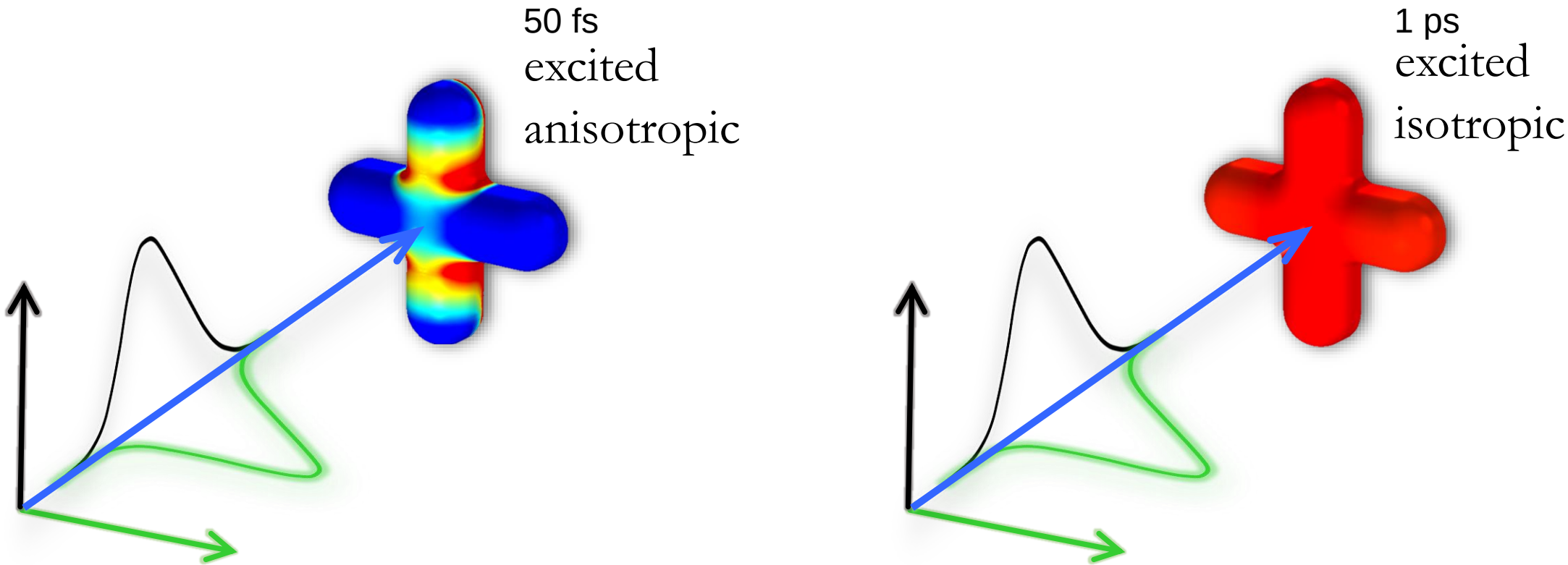
Transient symmetry breaking
Photoinduced sensitivity to polarisation



Although the symmetric geometry, **asymmetric** hot carrier spatial distribution opens a **transient** symmetry-breaking window: the structure exhibits **polarisation dependence**[5-7]

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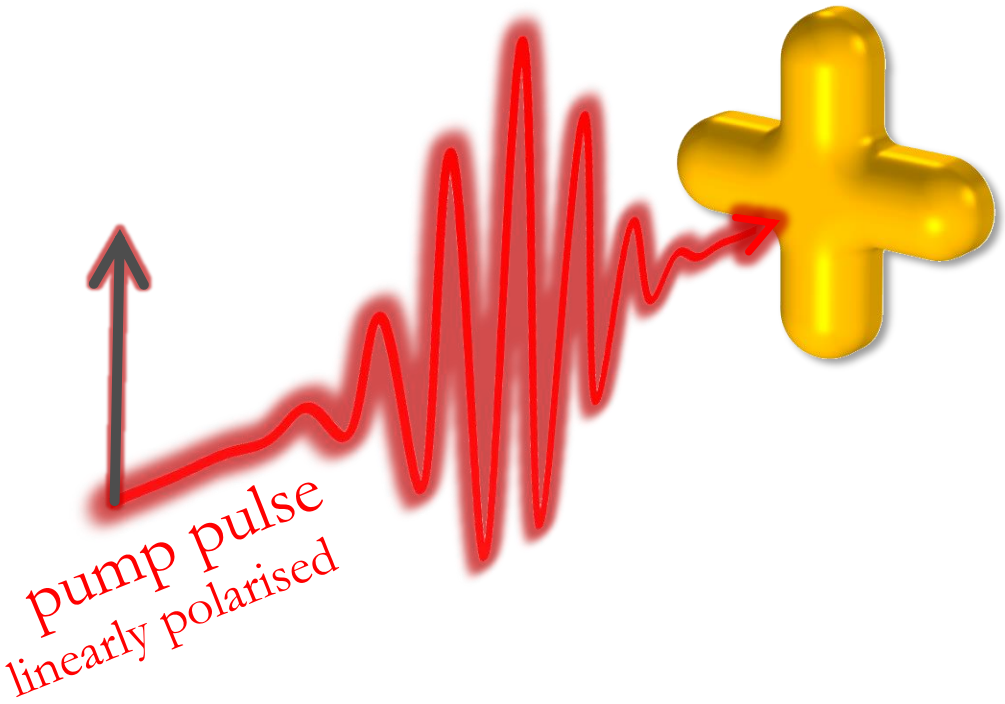
Broadband dichroism
Pump-probe differential transmittance



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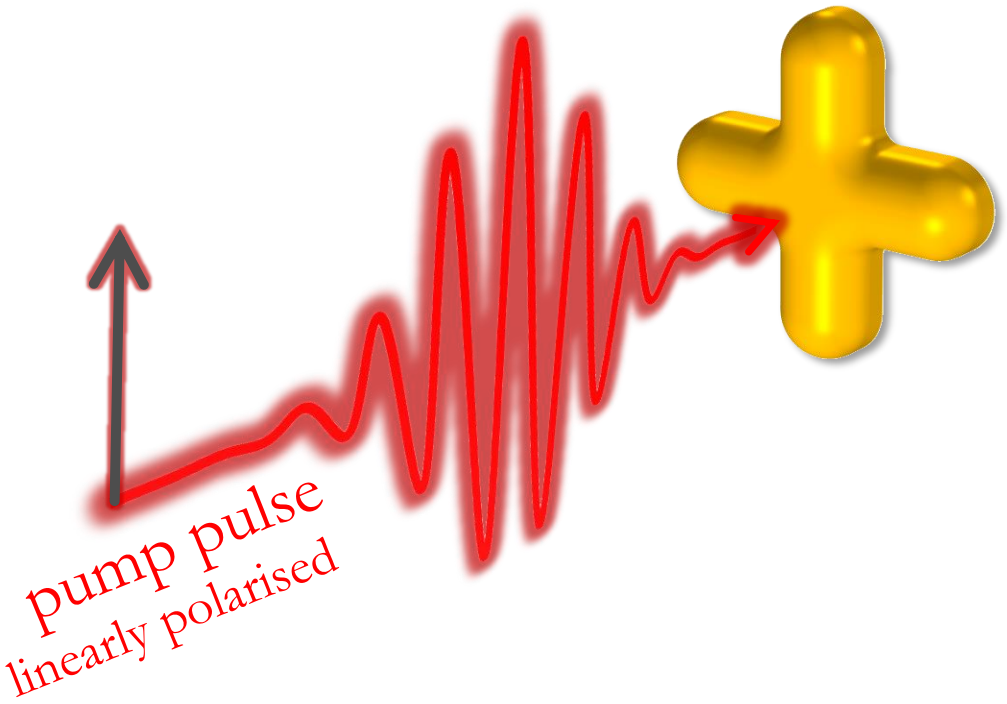
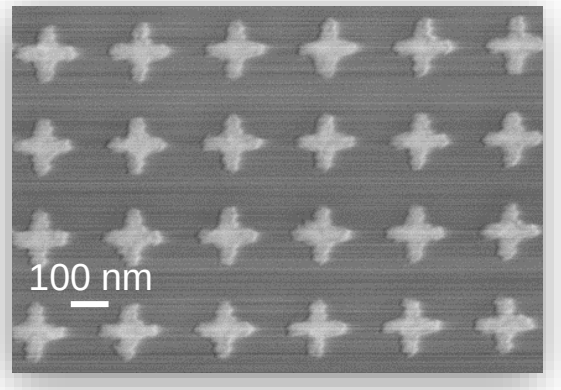
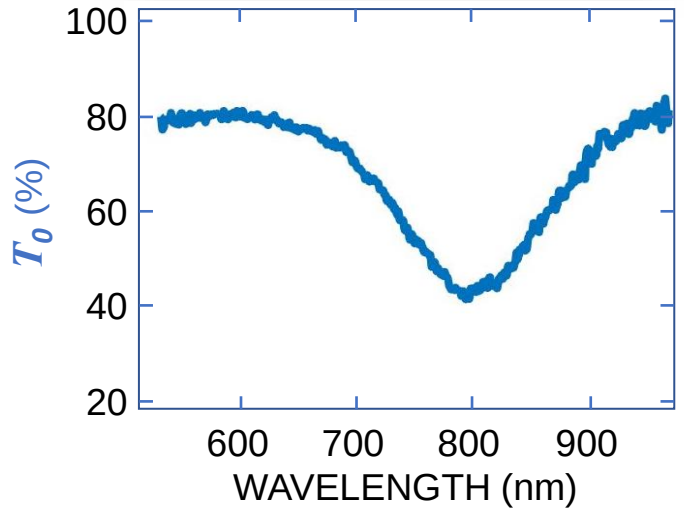
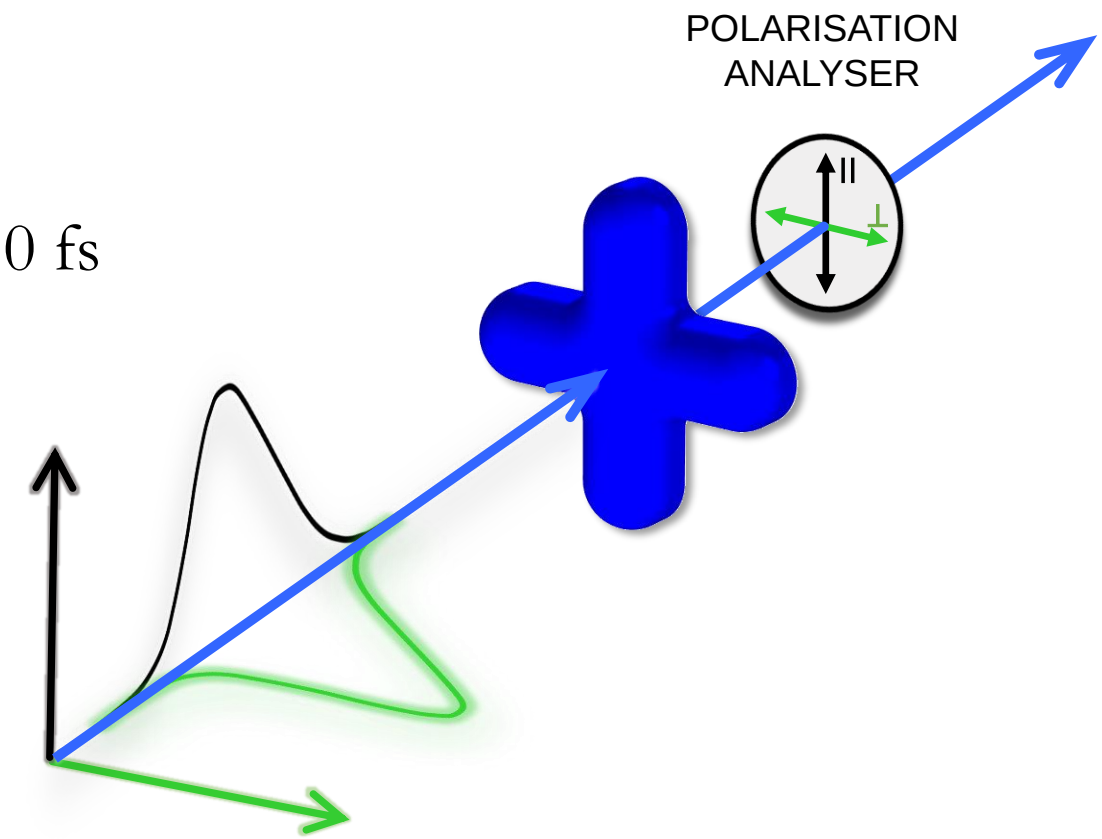


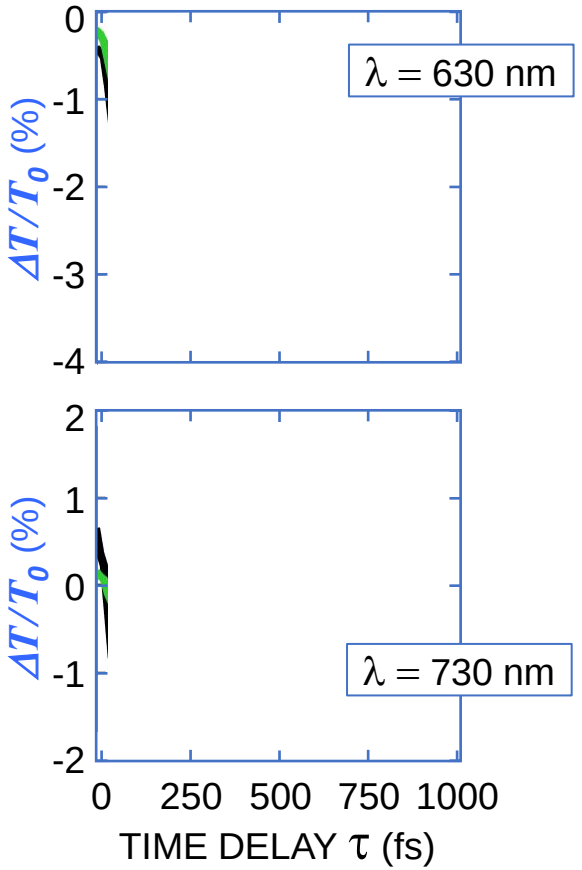




Broadband dichroism
Pump-probe differential transmittance

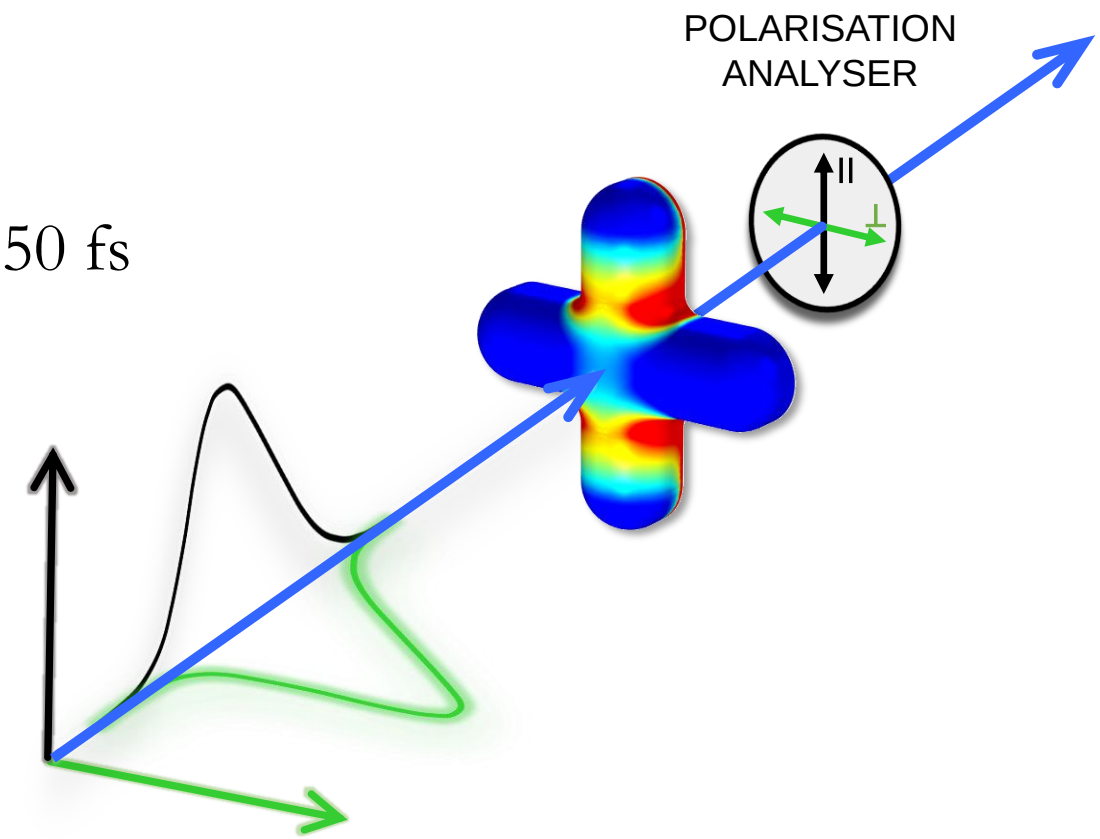
probe at $\tau = 0$ fs

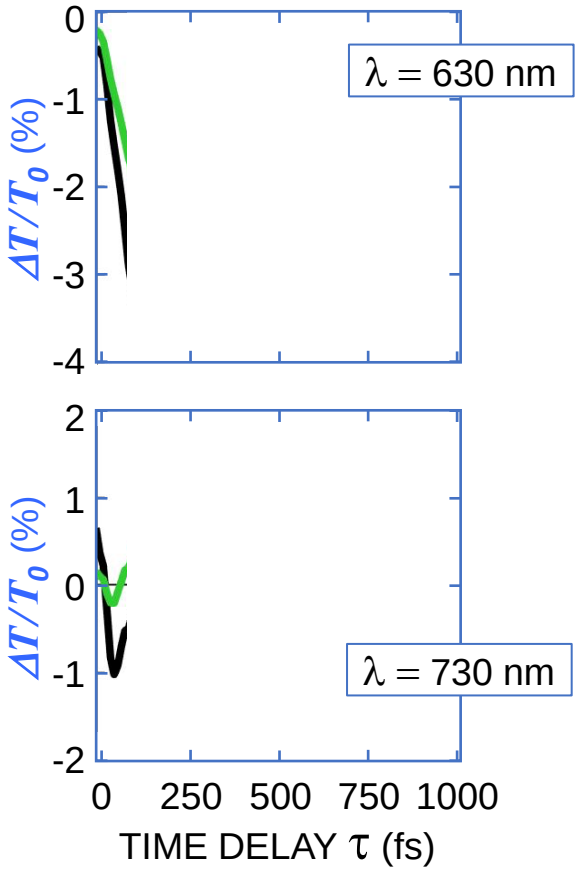




Broadband dichroism
Pump-probe differential transmittance

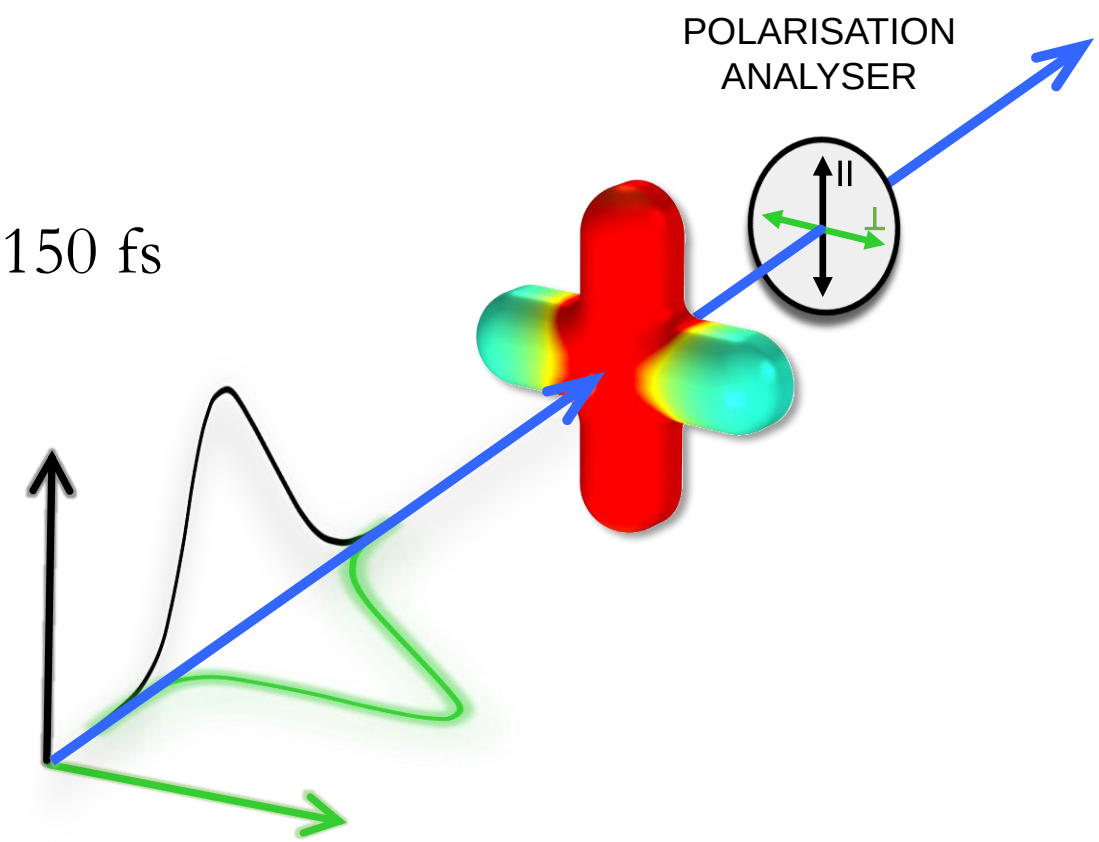
probe at $\tau = 50$ fs

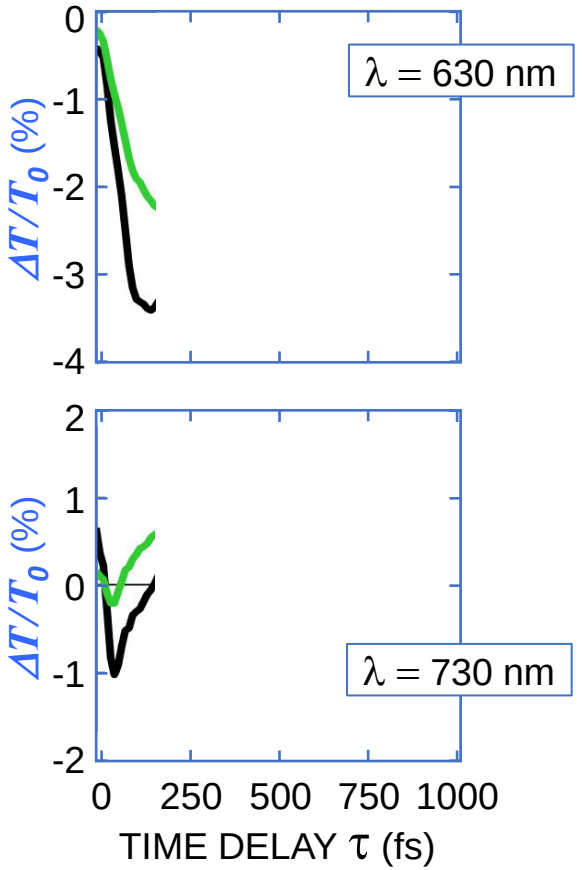




Broadband dichroism
Pump-probe differential transmittance

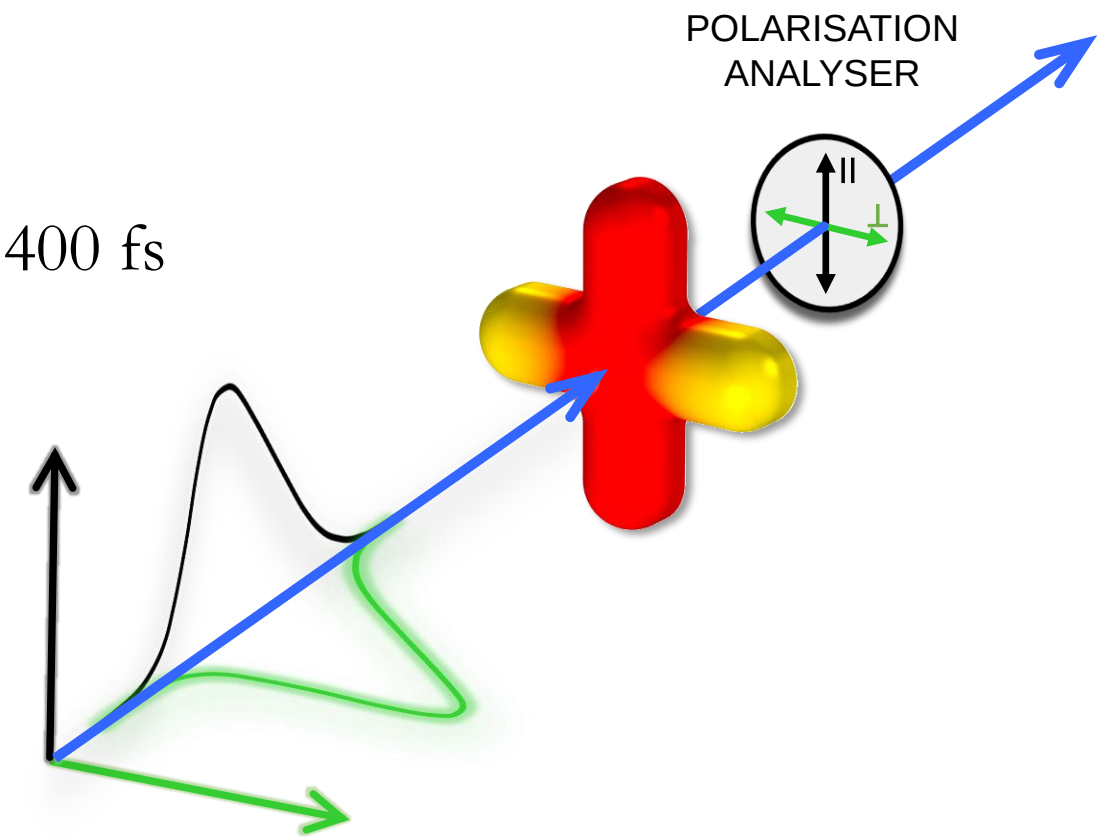
probe at $\tau = 150$ fs

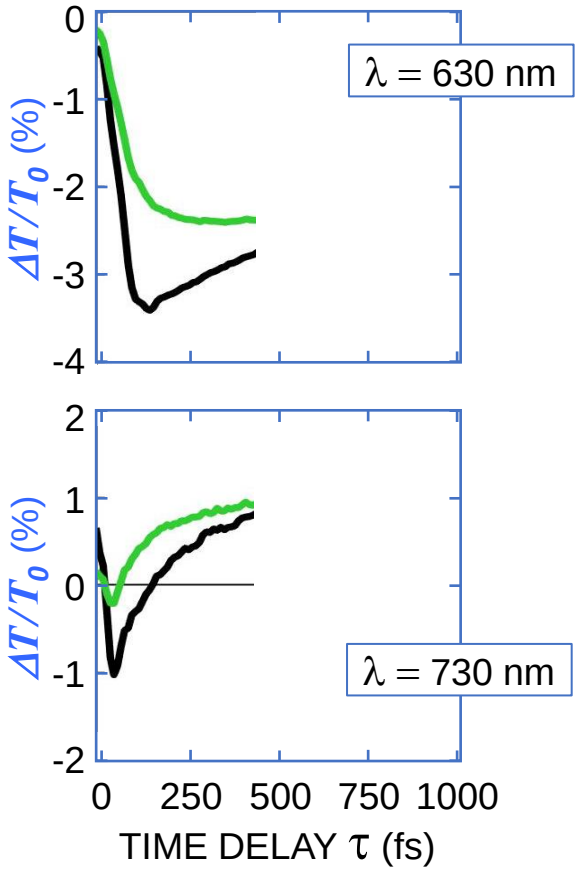




Broadband dichroism
Pump-probe differential transmittance

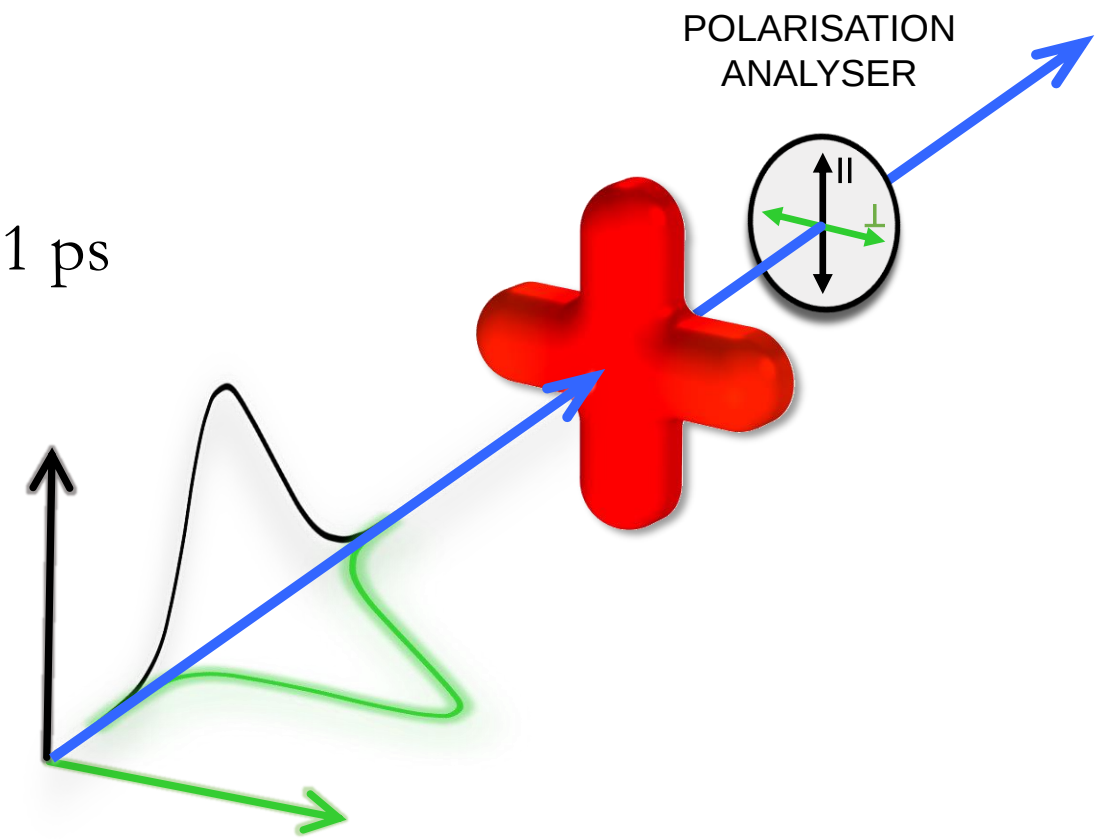
probe at $\tau = 400$ fs

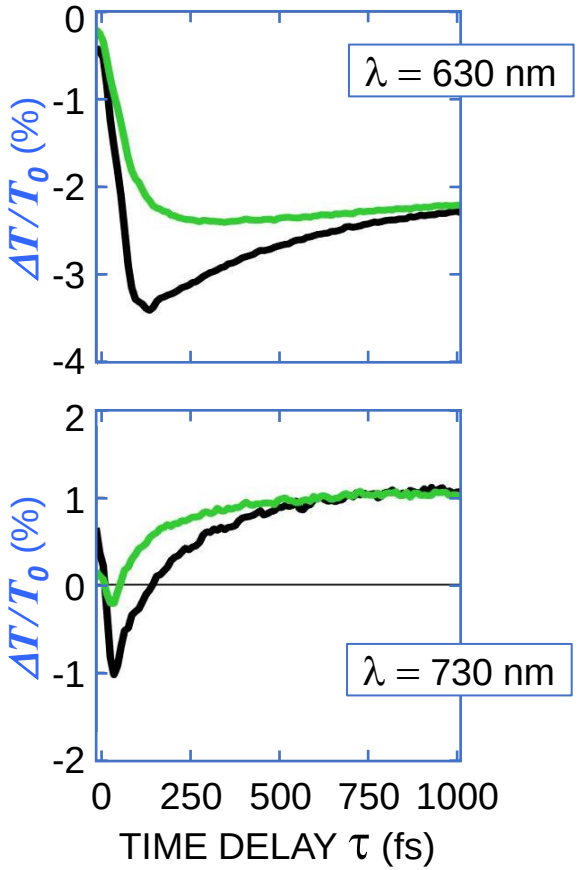




Broadband dichroism
Pump-probe differential transmittance

probe at $\tau = 1$ ps





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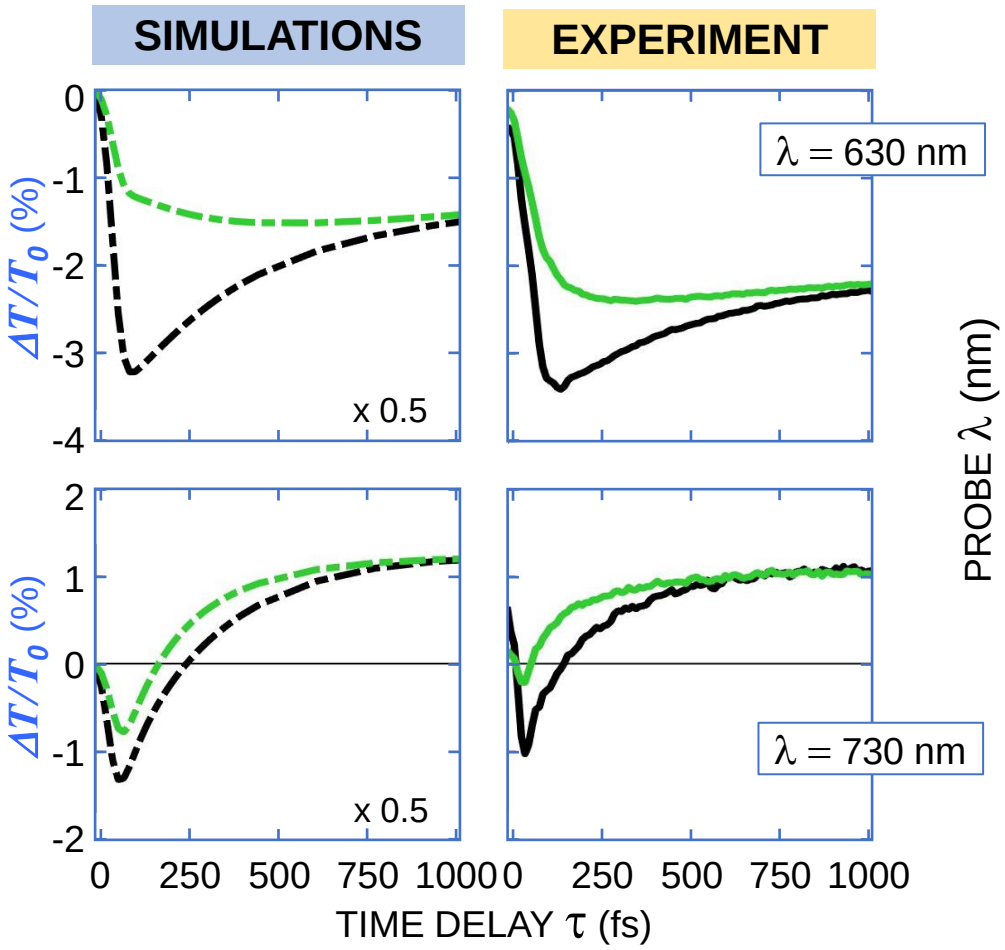
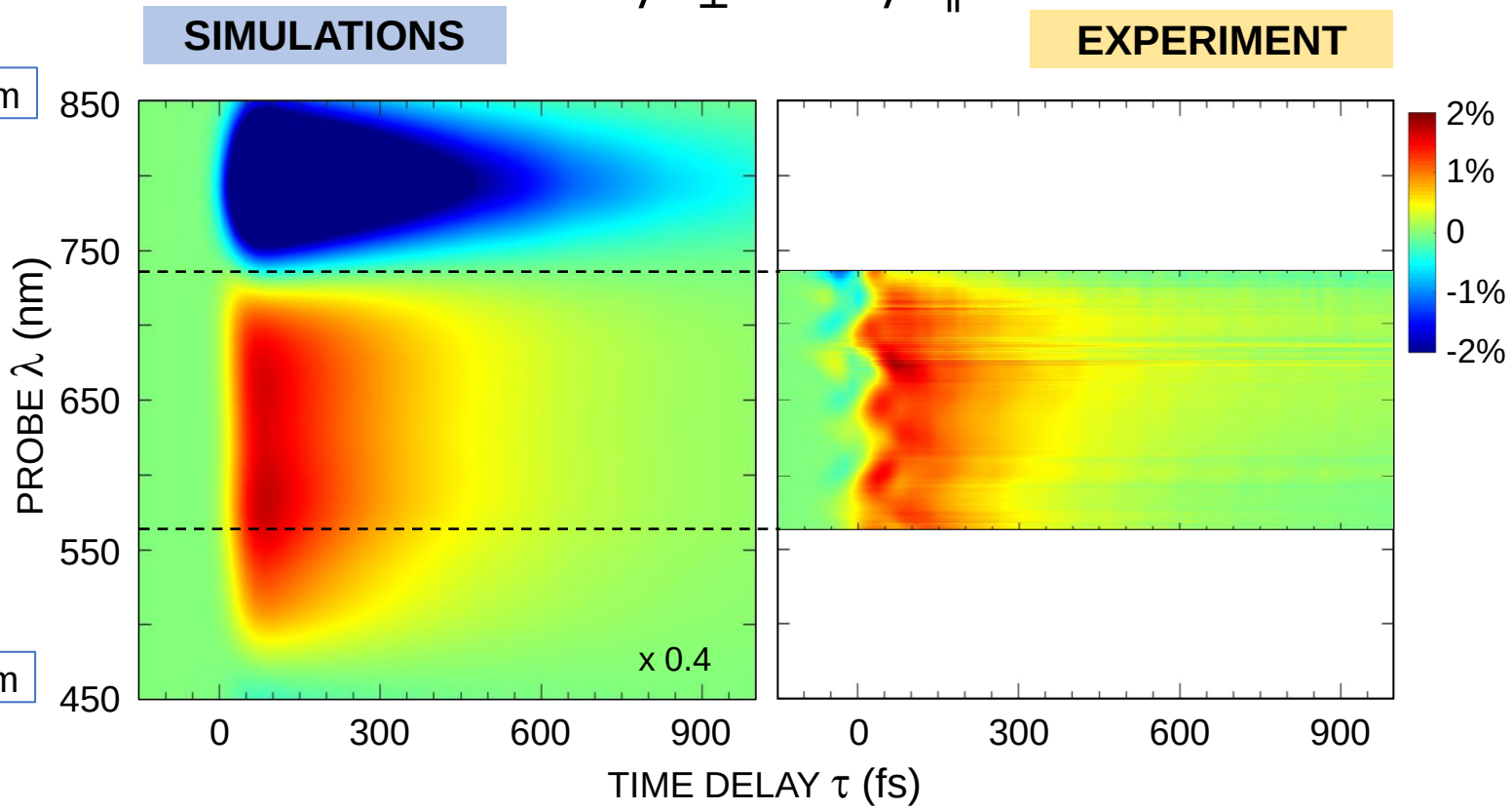


$\Delta T/T_{\perp} - \Delta T/T_{\parallel}$



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  - *Broadband transient dichroism with fully reversible recovery within 1 ps*

Conclusions and Perspectives

- The inhomogeneous distribution of photoexcited carriers is demonstrated to induce a **spatio-temporal** perturbation **breaking** the optical **symmetry** of the metasurface
- Photoinduced **anisotropy**, resulting in broadband dichroism, collapses **within 1 ps**, when excitation returns to symmetric state

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- Our results can disclose unprecedented routes for **ultrafast all-optical** light **control**

- Ultrafast dichroic devices for **high-speed** modulation of light **polarisation**
- Optimised platforms for hot-carrier-driven **photocatalysis**

Acknowledgments and References

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Thanks for your attention

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