Contribution ID: **38** Type: **Poster**

Experimental Realisation of Synthetic Chiral Light

Traditional chiroptical methods rely on the chirality of circularly polarised light (CPL) to determine molecular handedness. However, due to the large disparity between the pitch of the light's helix and the tiny size of the molecules, the interaction of CPL with chiral molecules is weakly enantiosensitive.[1] Synthetic chiral light has recently been proposed as an efficient alternative to CPL.[2] This new type of chiral light is locally chiral, i.e. the electric-field vector traces a 3D chiral Lissajous figure in time, that drives ultrafast chiral electron currents on the molecular scale. The nonlinear response of chiral molecules to such tailored fields is orders-of-magnitude more enantiosensitive than with CPL.[1, 2]

Here we present our experimental achievement: we have created locally chiral light in our laboratory. Our setup uses two ultrafast (50 fs) non-collinear laser beams of wavelength 800 nm linearly polarised in the plane of the optical table. In one of the two beams, an additional orthogonally polarised 400 nm component is generated from a type I BBO. The two beams are then focused and overlapped spatially and temporally.

To characterise this locally chiral field, we recorded the nonlinear response of a BBO crystal. Each non-linear process emits in specific directions due to the non-collinear geometry and conservation of non-linear momentum. The presence of a non-collinear second harmonic (400 nm) in between the two beams and two third harmonic generation (THG) signals either side confirms spatial and temporal overlap of the two 800 nm components. Likewise, the non-collinear sum-frequency generation signal (SFG) (266 nm) appears in the same position as one of the THG signals and confirms the overlap of the 400 nm and 800 nm components in opposite beams. This validates the simultaneous overlap of all three components and unambiguously demonstrates the generation of a locally chiral field.[2, 3]

The chirality of the field can be controlled by the sub-cycle phase of the 800 and 400 nm components of the field. This can be seen in the overall intensity of the 266 nm signal due to an interference effect between the SFG and THG signal from BBO. By scanning the phase in sub-fs increments we show a clear, reproducible interference effect which demonstrates the fine control we have over our locally chiral field. [2, 3]

We aim to further demonstrate the unique interactions of our locally chiral field with chiral biomolecules in their natural environment, the liquid phase. Not only does the experimental realisation of locally chiral fields create new methods for imaging chirality, but ultimately paves the way for highly efficient control, manipulation, and separation of chiral molecules.

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Session Classification: Poster Session