Driving and Imaging Achiral-to-Chiral Phase Transitions

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Chiral molecules owe their handedness to the spatial arrangements of their *nuclei*, which form structures which are not superimposable with their mirror images. However, chirality can also be related to *electronic* degrees of freedom. Indeed, as shown by Ordoñez and Smirnova, even the hydrogen atom can have chiral superpositions of electronic excited states [1].

Synthetic chiral light [2] enables ultrafast and highly efficient imaging of molecular chirality. Such light is *locally* chiral: the tip of the electric-field vector draws a chiral (3D) Lissajous figure in time, at each point in space. Interestingly, such tailored light can also be used to create chiral electronic states in atoms [3,4], which emit chiral photoelectron currents and exhibit photoelectron circular dichroism [3]. Here we show how to apply synthetic chiral light which is both locally and *globally* chiral [2] to create ultrafast chiral electronic currents in systems which are initially achiral, such as atoms, thus driving ultrafast achiral-to-chiral phase transitions.

We solved the time-dependent Schrödinger equation for the hydrogen atom exposed to an intense and ultrashort locally chiral field. The induced polarization, proportional to $\langle\psi(t)|\mathbf{r}|\psi(t)\rangle$, is depicted in Fig. 1. As shown in Fig. 1a, the laser drives ultrafast chiral electron motion at its fundamental frequencies (800 and 400nm), although nonlinear effects are also evident. Indeed, the high-frequency components create a chiral structure in time, see Fig. 1b, and give rise to chiral high harmonic generation (HHG). Chiral HHG is suppressed after the pulse is gone, but the chiral electron motion does not stop: the atom remains excited in a chiral coherent superposition of different energy and angular momentum stationary states, and thus the electron continues to undergo ultrafast chiral dynamics, see Fig. 1c.

The ultrafast chiral current shown in Fig. 1c gives rise to chiral free-induction decay radiation with elliptical polarization. As we will show in this conference, the imprinted handedness is recorded in the polarization of the emitted light. This work creates exciting opportunities for driving and monitoring achiral-to-chiral phase transitions in all-optical setups, as well as for driving chiral photo-chemical reactions using achiral reagents.

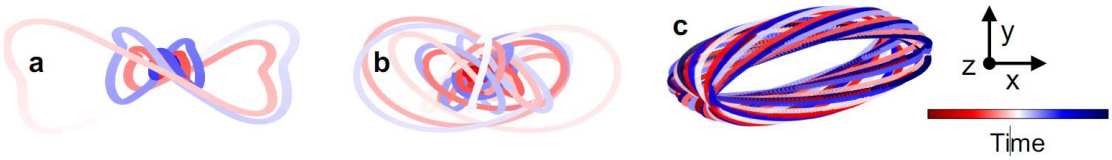

Figure 1: Ultrafast polarization induced in the hydrogen atom by our locally chiral field [2]. **a**, total polarization in the presence of the field. **b**, high-order components, resulting from filtering out the linear response in **a**. **c**, induced polarization after the pulse is gone. Laser parameters: wavelengths 800 and 400nm, intensity 10^{14} Wcm^{-2}, pulse duration 7fs (FWHM).

References

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