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Ultrafast, all-optical and highly enantiosensitive response of uniaxially oriented chiral molecules to achiral tailored light

Chirality is a fundamental concept that pertains to the property of molecules that cannot be superimposed onto their mirror images, existing in two non-superimposable forms called enantiomers. Like our left and right hands, opposite enantiomers have identical properties unless they are in the presence of another chiral entity, such as another chiral molecule or field. Since many biologically active molecules are chiral, chirality plays a pivotal role in pharmaceuticals, where the effectiveness of a chiral drug often depends on its handedness.

Circularly polarized light has long served as a standard tool for chiral recognition. However, it is an inefficient chiral reagent [1]: the pitch of the helix that makes it spatially chiral is typically orders of magnitude larger than the molecule. The enantiosensitive response of the molecules, arising beyond the electric-dipole approximation, is tiny, thus requiring sensitive detection setups.

The electric-dipole "revolution"[1] is transforming the landscape of molecular chiral discrimination with a new generation of chiroptical methods offering orders-of-magnitude greater enantiosensitivity. Different members of this revolution rely on analysing different enantiosensitive observables, but share one common ingredient [1]: the enantiosensitive response of the molecules is driven solely by the electronic response of the

molecules to the local polarisation of the driving field –magnetic interactions are not required.

Here we show how uniaxial alignment of chiral molecules enables the generation of strongly enantiosensitive all-optical signals via purely electric-dipole interactions without the need of using chiral light. Our state-of-the-art numerical results show that the low-order and high-order nonlinear response of a prototypical chiral molecule to a cross-polarised ω,2ω field becomes strongly enantiosensitive upon uniaxial molecular alignment.

In contrast to other approaches, our proposal requires molecular alignment upon a molecular axis, not orientation, which can be realised using current optical technology. This creates exciting opportunities and potential applications for all-optical chiral imaging and manipulation, as well as for imaging and manipulating ultrafast chiral dynamics at their natural timescales.

[1] Ayuso, D., Ordonez, A.F. & Smirnova, O. Phys. Chem. Chem. Phys. 24, 26962-26991 (2022)

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