Enantiosensitive Steering of Free-Induction Decay

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Synopsis: Chiral recognition has recently become an emerging frontier in ultrafast physics, with vivid progress achieved in multiphoton and strong-field regimes. Here we introduce a new phenomenon, enantiosensitive free-induction decay, steered by a tricolour chiral (TRICC) field at a gentle intensity, structured in space and time. We demonstrate theoretically that an excited chiral molecule accumulates an enantiosensitive phase due to perturbative interactions with the TRICC field, resulting in a spatial phase gradient steering the free-induction decay in opposite directions for opposite enantiomers.

Chiral recognition is an vital task in chemistry, whose origin dates back to the birth of the discipline with the discovery of the optical activity of biomolecules in solution. However, for dilute media and in gas phase, this effect is severely challenging to implement, since it relies on rather weak optical magnetic interactions. This creates a strong demand for an optical chiral discrimination method which relies purely on dipole-interaction physics.

In this work we propose an experiment allowing chiral recognition on an ultrafast timescale using non-destructive weak fields based on pure electric dipole interactions. Our scheme builds on recent advances on chiral synthetic light [1] to induce a controllable enantiosensitive quantum phase in the medium, which is then translated into easily measurable macroscopic observables. Specifically, we adapt the ability to steer bright and coherent free-induction decay (FID) radiation via its quantum phase manipulation in atomic gases [2] to chirally-sensitive drivers interacting with chiral media, thereby introducing an enantiosensitive Stark shift which gives rise to FID labelling of enantiomers (FIDLE).

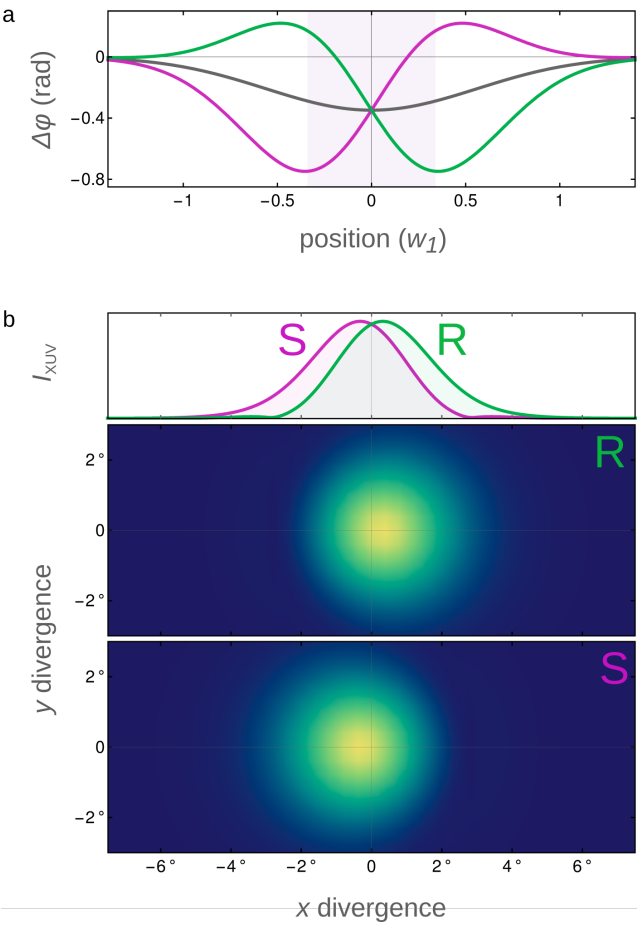

Figure 1: TRICC FIDLE simulations of (a) the enantiosensitive quantum phase and (b) far-field FID beam for the methyloxirane molecule

We model a chiral molecule promoted from its ground state into a FID-active excited state and which re-emits this photon energy by decaying back to the ground state. The direction of this emission is defined by the quantum phase of the FID-active state. The simplest way to introduce the chiral Stark shift is to consider the interaction of the FID-active state with two other excited states induced by a tricolour chiral (TRICC) field with noncollinear polarisations forming a chiral triplet. We create this chiral triplet macroscopically using tightly-focused Gaussian beams to provide a longitudinal polarisation component, resulting in a chiral time evolution of the electric field at every point. We show a clearly visible enantiosensitive steering of the FID emission for a real molecule [3].

References

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