Structuring Light's Chirality to Induce Enantio-Sensitive Light Bending

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Structured light, which exhibits nontrivial intensity, phase, and polarization patterns in space, has key applications ranging from imaging and 3D micro-manipulation to classical and quantum communication [1]. However, to date, its application to molecular chirality [2] has been limited by the weakness of magnetic interactions. Here we show how to lift this limitation by structuring light's local chirality – a new type of chirality effective within the electric-dipole approximation and in the non-linear regime [3]. We introduce and realize an enantio-sensitive interferometer for efficient chiral recognition without magnetic interactions, which can be seen as a chiral version of Young's double slit experiment. We show that if the distribution of light's handedness breaks left-right symmetry, the interference of chiral and achiral parts of the molecular response leads to unidirectional bending of the emitted light in opposite directions in media of opposite handedness. Our work introduces the concepts of polarization of chirality and chirality-polarized light, exposes the immense potential of sculpting light's local chirality to control the molecular response, and offers novel opportunities for efficient chiral discrimination, optical molecular fingerprinting, and imaging on ultrafast time scales using intense light [4].

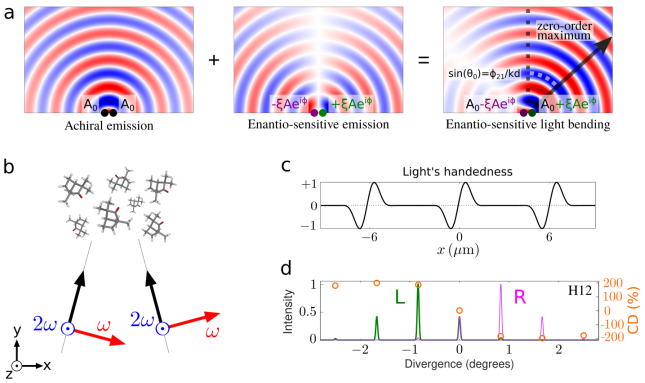

Figure 1: The response $P = P^{(\text{achiral})} + P^{(\text{chiral})}$ of a chiral molecule of handedness ξ can be decomposed into a non-enantio-sensitive part $P^{(\text{achiral})} = A_0$ and an enantio-sensitive part $P^{(\text{chiral})} = \xi e^{i\phi}$. A pair of molecules with their achiral response A_0 in phase (left panel), and their enantio-sensitive response $\pm\xi A e^{i\phi}$ out of phase (central panel), interfere to yield a shifted interference pattern (right panel), whose shift is enantio-sensitive. **b** We realize this *chiral Young's double slit* by considering high harmonic generation in a chiral medium driven by two non-collinear beams carrying linearly polarized ω and 2ω fields orthogonal to each other. **c** By controlling the two-color relative phase in both beams we achieve light with a *structured local handedness* $h(x)$ that resembles the charge density in a polarized dielectric. That is, we achieve light with *polarization of chirality*

References

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