Boosting Fluorescence-Based Chiral Sensing with Nanophotonics

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Abstract:
The handedness of chiral molecules can be detected in their circularly polarized fluorescence, which is typically very weak. Here, we propose dielectric nanophotonics to increase both the fluorescence intensity and polarization contrast.

1. Introduction

Chirality detection of enantiomers is crucial for numerous applications in medicine and biochemistry. Conventional chiral sensing tools such as circular dichroism (CD) exploit the dissymmetric absorption of large ensembles of chiral molecules for right- and left-circularly polarized light. Nanophotonics has recently offered a viable route to increase the chiral sensitivity through superchiral fields [1–3]. However, it remains impossible to lower the detection limits down to a single molecule using absorption-based methods.

In contrast, emission-based chiroptical methods such as circularly polarized luminescence or fluorescence-detected circular dichroism are a promising path for probing the molecular chirality at the single molecule level. Such analysis could uncover the conformational dynamics of individual biological molecules, usually masked by ensemble averaging. Circularly polarized luminescence (CPL) exploits the dissymmetry in circularly polarized emission for a chiral molecule. It is quantified by the degree of circular polarization (DOCP), which is expressed as:

$$\mathrm{DOCP} = \frac{I_R - I_L}{I_R + I_L} \tag{1}$$

where I_R and I_L are the intensities for right- and left-handed circularly polarized emission, respectively. For a small chiral molecule, DOCP values are typically below 0.001 (inset in Figure1 a). Tailored resonant nanostructures can enhance weak chiral contrast [4, 5]. However, must Studies thus far, are based on the electric field suppression at the location of the chiral molecule [6], which essentially degrades the fluorescence signal. This suppressed emission, has therefore been a key limitation towards achieving single-molecule sensitivity because of poor signal-to-noise ratio. Here, we introduce nanophotonic resonators to boost chiral sensing based on circularly polarized luminescence. Our fluorescence-enabled approach can enhance both the degree of circular polarization and the total fluorescence intensity (S). To increase these two quantities together, we propose a high-index dielectric system composed of a silicon holey disk surrounded by a ring (Figure 1a). We consider a chiral dipole with intrinsic DOCP = 0.01 placed at the center of the hole and calculate the fluorescence and the DOCP enhancements for the coupled system in the normal direction (Figure 1b).

Our approach enables simultaneous 1000- and 10-fold enhancements in fluorescence signal and polarization, respectively. For different nanostructures, higher enhancements of either fluorescence or polarization are possible, subject to trade-offs. Our proposed method offers outstanding performance for single-molecule chiroptical detection in a variety of applications.

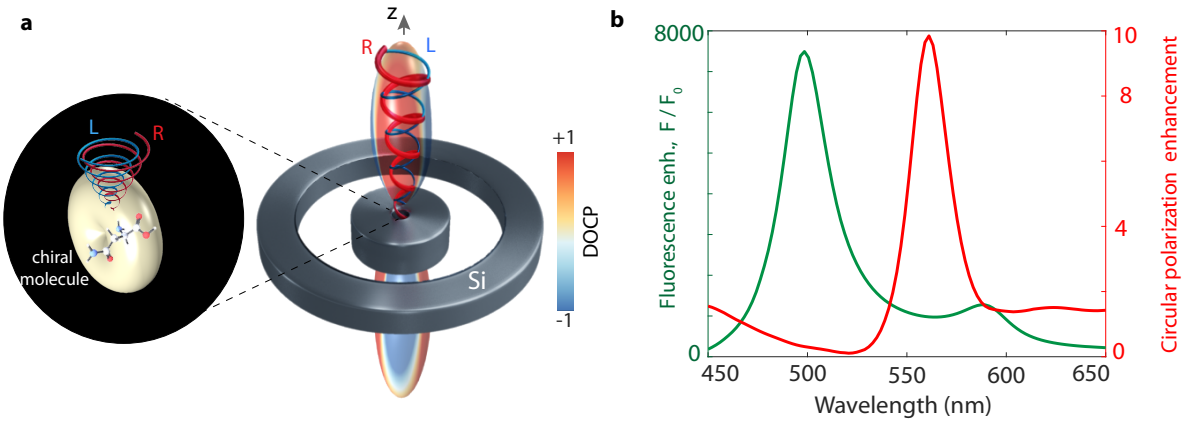

Fig. 1. A high-index dielectric nanostructure for boosting circularly polarized luminescence of small organic chiral molecules. (a) A fluorescent chiral molecule with low dissymmetry factor. A holey disk-ring system made of silicon can increase the dissymmetry factor and the fluorescence signal of the chiral molecule at the same time. The diameter and height of the holey disk are 160 nm and 90 nm, while the inner and outer radii of the ring are 320 nm and 420 nm, respectively. The heights of the ring and disk are the same. The diameter of the hole is 20 nm and the chiral molecule is placed at its center. (b) Enhancements in fluorescence and degree of circular polarization for a chiral emitter with intrinsic DOCP = 0.01 and intrinsic quantum yield η = 0.001. The system provides one order of magnitude amplification in DOCP at λ =560 nm and a 1000-fold enhancement of fluorescence at the same wavelength.

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