Silicon metasurfaces for enhanced fluorescence-based chiral sensing

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Chirality is omnipresent in biochemistry, where molecules with opposite handedness can have markedly different properties. Detecting molecular chirality is, however, plagued by low sensitivity, limiting the possible use cases to high concentrations and large volumes. We propose using silicon nanophotonic structures to create optical resonances of electric and magnetic dipole origin. The strongly enhanced sensitivity can enable the detection of the handedness of molecules in small volumes. We also demonstrate a new polarization-modulation microscope built to push the limits of chiral sensing using fluorescence.

Detecting the handedness of chiral molecules is critical in biomedicine, where the functionality of biomolecules such as proteins, amino acids, and carbohydrates can strongly differ between enantiomers. Circular dichroism (CD) is commonly used to distinguish enantiomers based on the differential absorption of right- and left-handed circular polarizations of light by chiral molecules. This transmission-based method requires measuring very small changes in absorption, which is inherently difficult at low concentrations due to the large background caused by the light source itself. As a result, the sensitivity is reduced, requiring a high density of molecules and long measurement times. Conventional methods are thus unsuitable for critical applications like detecting protein aggregates linked to various diseases.

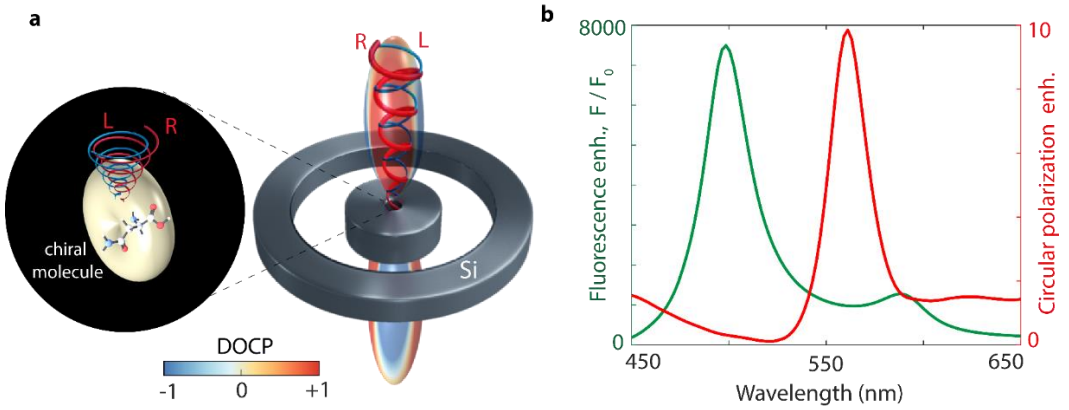

Figure 1 Silicon nanoantennas tailored to boost fluorescence-based chiral sensing.

Fluorescence-based chiral sensing bypasses some of the difficulties of CD by detecting light emitted at a different wavelength than the excitation source, allowing for the complete removal of the source background from the signal. In general, fluorescence-based chiral sensing can be split into two categories. Firstly, fluorescence detected circular dichroism (FDCD), where the differential absorption is measured as a difference in emitted fluorescence intensity. Secondly, circularly polarized luminescence (CPL), where molecules are excited by unpolarized light and

the circular polarization of the emitted fluorescence is measured. Nanophotonics offers two routes to enhance the sensitivity of chirality detection [1], [2], [3], [4]. Super-chiral fields, offering a higher chirality than circularly polarized plane waves, can exist within the near field of nanophotonic structures. Furthermore, nanophotonics can enhance the detected fluorescence intensity through excitation, Purcell, and collection enhancements. Our simulations using silicon nanostructures show a 10-fold polarization enhancement while simultaneously enhancing the fluorescence intensity 1000 times. This combination of enhancement is promising for probing molecular chirality at the single-molecule level, eliminating the impact of ensemble averaging.

We present our experimental progress, including fabrication methods for achieving such nanostructures using crystalline silicon for its high refractive index and lower losses than amorphous silicon. Furthermore, we introduce a custom-built microscope setup for measuring degrees of circular polarization below 1%. We use photoelastic modulation for precise polarization modulation and an avalanche diode for photon counting to enable lock-in detection with suppressed noise.

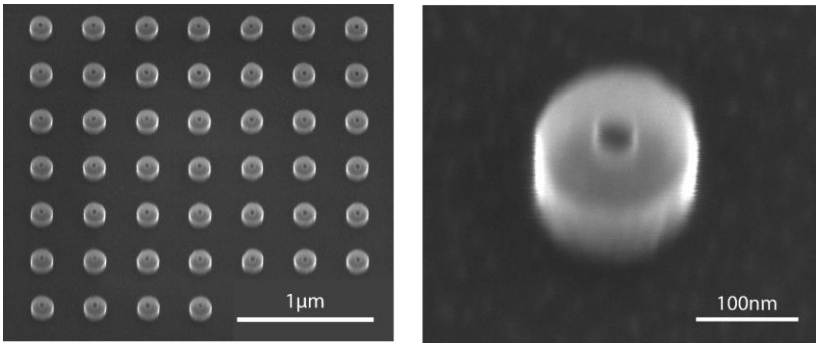

Figure 2 SEM images of fabricated nanophotonic structures for chiral sensing.

In conclusion, we combine advancements in silicon metasurfaces tailored for fluorescence-based chiral sensing and a novel polarization-resolved microscope to increase the sensitivity toward single-molecule chirality detection.

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