Probing optical chirality in the near-field of plasmonic nanostructures

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Abstract: We directly probe the optical chirality in the near-field of plasmonic nanostructures coated with a dye-doped polymer film by measuring the photoluminescence enhancement and analysing it with respect to its circular polarisation state.

Chirality – or handedness – is a structural property of matter with an extensive history. However, Tang and Cohen recently introduced optical chirality as a measure of the chiral density of states in electro-magnetic fields [1].

Plasmonic nanostructures provide one way of achieving strong chiral fields because the phase difference between the electric and magnetic field component can be engineered *via* the geometry of the structures or, more precisely, *via* the relative arrangement of several nanostructures [2]. The strong near-fields of plasmonic nanostructures can generally enhance the emission of emitters in close proximity and this enhanced emission, in turn, follows the polarisation state of the plasmonic resonance associated with the near-field enhancement [3]. So far this has only been studied for linear polarisation; here we extend this basic concept to measuring circular polarisation states of the emission.

For our experiments we used arrays of silver nanorod pairs where the centre of one rod is shifted along the long axis with respect to the other rod; the handedness of the structure is determined by the direction of the lateral shift. The samples were fabricated by a standard electron-beam lithography process on a glass substrate and subsequently coated with a thin dye-doped polymer film (Styrly 9 incorporated into a polyvinyl alcohol matrix). Using these we measured the photoluminescence (PL) enhancement and analyse the emisison with respect to its circular polarisation states.

The PL enhancement factor f_{PL} on the chiral nanorod pairs shows a distinct asymmetry between left- and right-handed emission, leading to a negative differential enhancement factor Δf_{PL}. This effect is reversed when probed on an array of enantiomeric nanorod pairs where the PL enhancement is greater for the other handedness and consequently Δf_{PL} becomes positive (see Figure 1). In a reference measurement on symmetric nanorod pairs, which possess no optical chirality, we find no difference between the PL enhancement for left- and right-handed polarisation. Additionally, in the absence of nanoparticles the dye-doped polymer film does not exhibit any measurable difference between left- and right-handed emission. As a further control experiment we have introduced a 200-nm thick spacer layer between the nanostructures and the dye-doped film and repeated the experiments described above. In this case, when we probe the transmission of photoluminescence generated away from the plasmonic near-fields, we do not detect a differenc in the handedness of emission, regardless of the chirality of the nanoparticles.

In conclusion, we utilised the near-field coupling between plasmonic nanostructures and emitters to directly probe the optical chirality in the near-field of these structures by measuring the asymmetry in the PL enhancement factor with respect to the circular polarisation state of the enhanced emission.

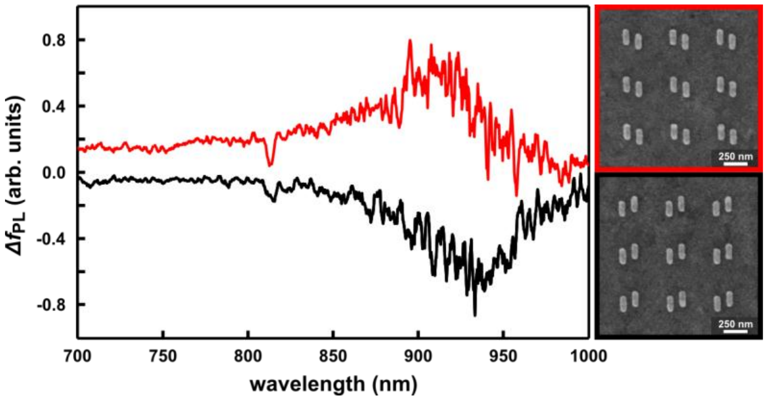

Figure 1. Differential photoluminescence enhancement factor Δf_{PL} for left- and right-handed emission, measured on two enantiomeric arrays of silver nanorod pairs coated with a dye-doped polymer-film.

References

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