Transparent Conductive Oxide Nanocrystals as Promising Materials for Magnetoplasmonics

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Active modulation of the plasmonic resonance in nanostructures enables the control of light propagation at the nanoscale, enabling a significant improvement of plasmon-based sensors and optical modulators. Compared to other external tools employed to this purpose, magnetic field presents several advantages: it is fast, easy to implement in devices, and its action on free charge carriers is fully reversible as it does not damage or modify the plasmonic material. However, achieving large magnetic modulation of the plasmonic resonance without broadening the optical response represents a great challenge in material choice for magnetoplasmonics, hampering the application in devices. Indeed, noble metals nanocrystals (NCs) have sharp optical resonances, but weak magneto-optical signal, proportional to the cyclotron frequency (ω_C); on the other hand, nickel ferromagnetic nanodisks or hybrid bimetallic nanostructures have large magnetic modulation, but suffer from the high optical losses of the magnetic metal, thus broadening the plasmonic resonance.

To overcome such limitations, we propose a paradigm shift in material choice by employing transparent conductive oxides (TCO) NCs, which are able to support a plasmonic resonance in the infrared, due to the free carrier density (10^{18}-10^{21} cm^{-3}) introduced through aliovalent doping.

In this work, we prepared TCO NCs by chemical synthesis, demonstrating up to 40-fold enhancement in the magnetoplasmonic response compared to Au NCs, detected through Magnetic Circular Dichroism (MCD). Such enhancement is ascribed to the lower effective mass (m*) of free carriers in TCOs with respect to most metals, which in turn boosts ω_C combined with a reduced plasmonic line width.

Employing colloidal dispersions of TCO NCs in a proof of concept field-modulated refractometric sensing experiment we achieved a superior refractive index sensitivity with respect to metal-based magnetoplasmonic systems reported in the literature. Our approach challenges the current state of the art of plasmonic refractometric sensing, with the advantage of not requiring curve fitting but simply tracking a change in magneto-optical signal at fixed wavelength.

Considering that non-magnetic TCOs have been used in this work, a further enhancement of the performance is potentially achievable by introducing magnetic dopants in plasmonic TCO NCs, exploiting carrier-mediated magnetic coupling between the introduced magnetic cations. We are currently investigating this approach in tin-doped In_2O_3 and Al-doped ZnO NCs, employing different magnetic cations as co-dopants.

Our results open up a new route toward high performance magnetoplasmonic materials and devices, discarding current approaches based on metallic nanostructures.