Nanofabricated Plasmonic Resonators on Optically Active Materials for Hydrogen Photocatalysis

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Plasmonic resonators are metallic nanostructures that can highly concentrate optical energy into an ultra-small volume that has dimensions far below the wavelength.[1] When coupled with optically active materials, e.g. two-dimensional (2D) semiconductors and quantum dots (QDs), plasmonic resonators can be used in numerous applications like photovoltaics, photocatalysis, and emission enhancement.[2] This is because the resonators can provide extreme optical concentration and, in some cases, inject additional hot electrons into the materials. This then leads to enhanced generation of photocarriers and elevating performance in relevant applications.

Here we report the coupling of large-area plasmonic resonators with optically-active semiconductors, and then we discuss their potential applications in photocatalytic hydrogen conversion. Specifically, we have fabricated cm^2-scale silver nano-gratings using optical interference lithography (Fig.1**a** and **b**), which are then integrated with Cadmium Sulphide/ Cadmium Selenide (CdS/CdSe) core-shell quantum dots (QDs) (Fig.1**c**). The angle-resolved reflection spectra (Fig.2**b**) show that the plasmon resonances are highly dispersive, red-shifting in the visible range with increasing incident angles. The photoluminescence (PL) measurements (Fig.2**c**) reveal that when the plasmon resonances couple with the QDs, the PL from QDs are not only significantly (~ 100 times) enhanced, but also but also spectrally and directionally reshaped.

We will also discuss how the coupling between plasmonic resonators and 2D semiconductors, such as WS_2 monolayers and MoS_2/WS_2 hetro-bilayers, can benefit photocatalytic hydrogen production.

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[2] Ng, C., Wesemann, L., Panchenko, E., Song, J., Davis, T. J., Roberts, A., & Gomez, D. E, Plasmonic near-complete optical absorption and its applications. *Advanced Optical Materials* **7**, 1801660, 2019.

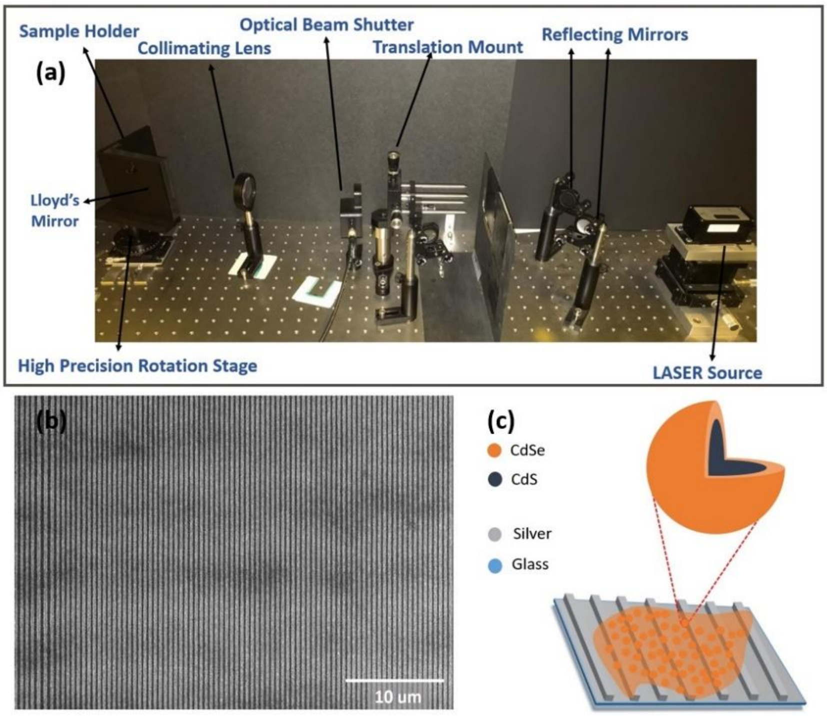

Figure 1**: **(a) The set-up for optical interference lithography ***(b)*** A scanning electron microscope (SEM) image of silver nano-gratings ***(c)*** A schematic of the silver nano-gratings coated with CdS/CdSe quantum dots (QDs).

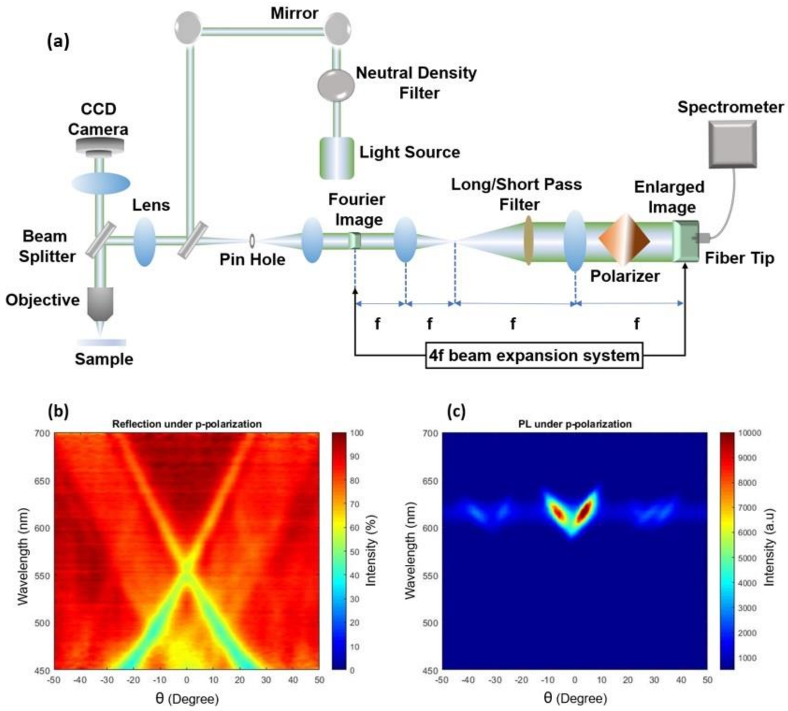

Figure 2**: **(a) A schematic for the set-up of angle-resolved optical spectra measurements ***(b)*** Reflection spectra under p-polarized illumination, i.e. the orientation of electric fields perpendicular to the plane of incidence. ***(c)*** Photoluminescence (PL) spectra with p-polarization.