EUROPEAN CONFERENCE ON INTEGRATED OPTICS

On-chip Enhanced Raman spectroscopy using metal slot waveguide

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Surface enhanced Raman scattering (SERs) is a technique that facilitates Raman spectroscopy of challenging samples such as thin layers, dilute solutions, or nanoparticles in a low concentration. SERS relies on localized plasmonic resonances that arise at ultra-small metallic gaps, tips or edges from a rough metallic surface, or an engineered nanostructure (antenna). Those plasmons lead to orders of magnitude of field enhancement but in a volume limited to a radius of 10-50 nm. However, plasmonic enhancement doesn't require a resonance. Propagating surface plasmon polaritons are electronic excitations that propagate in the metal over relatively long distances (several microns) while still enabling a large field enhancement. Here, we use it to increase the Raman scattering in a hybrid photonic-plasmonic structure whose centrepiece is a metallic slot waveguide.

The structure is depicted in figure 1a. It behaves as a perfect slot waveguide (fig. 1b) as the field is strictly confined to the void of the structure. Dielectric slot waveguides have been recently used for Raman spectroscopy as they increase the interaction length [1,2] with an analyte (liquid or monolayer). However, these dielectric waveguides have a moderate refractive index so that the confinement in the void of the structure is imperfect (see figure 1b) which results in lesser Raman scattering from the analyte and stronger photoluminescence from the dielectric core, hence limiting the signal to noise ratio. Having a perfect slot waveguide is therefore very appealing. A metallic slot waveguide relies on the actual excitation of a plasmon-polariton, thus limiting the interaction length due the exponential decay over a propagation distance of only a few microns. Furthermore, the mode profile in the metallic slot is significantly smaller than that of a regular dielectric (SiN) waveguide used to manipulate/analyze light on a photonic chip so that a dedicated tapering section is required to couple efficiently light between such a waveguide and the metallic slot-waveguide (fig. 1a). The taper efficiency and the field enhancement in the metallic slot-waveguide are strongly dependant on the void width. For optimization, a figure of merit (FOM) for this photonic-plasmonic device is formulated [3] that depicts the back-scattered Raman power P_s normalized with total pump power P_0 at input facet can be estimated:

$$FOM = \frac{Ps}{Po} = \frac{(\eta_{NTP}\sigma_{NTP}\rho_{NTP}) \times (\gamma)^2 \times (\Gamma)^2 \times \exp(-2\alpha_{SiN}L_{SiN}) \times (1 - \exp(-2\alpha_g L_g))}{2\alpha_g}$$

where η_{NTP} is the Raman conversion efficiency, the Raman crossection of NTP σ_{NTP}=0.358× 10^{-30} cm^2/Sr, its surface density ρ_{NTP} = 5.56× 10^{18} molecules/m^2, the coupling efficiency at the SiN waveguide facet $\gamma = 5dB$ and Γ is the photonics-plasmonic mode transition efficiency. We have optimized Γ and the FOM using FTDT LUMERICAL software as a function of the gap in the void. The results of this simulation (fig. 1b) show that the FOM varies over 2 orders of magnitude for width between 10 and 80 nm.

The silicon nitride waveguide and the gold slot are made using 2 subsequent e-beam lithography steps. The actual width of the void in the metallic slot is 31 nm, its length is 6 µm and the taper length is 1µm. To perform a proper characterization of the metallic slot-waveguide, we have coated it with a single layer of 4-nitrothiophenol (NTP) that selectively binds to gold but not to silicon nitride. A confocal Raman microscope is used to excite the TE mode of the silicon nitride rib waveguide from its facet. The guided light then excites the NTP-coated metallic slot-waveguide thus generating a Stokes field both in forward and backward directions. The microscope collects the backward scattered light resulting in a Raman spectrum displayed in fig. 2b showing clearly the expected NTP Raman resonances highlighted by dotted lines. An additional spectrum is measured from a SiN waveguide that is not coupled to any metallic slot-waveguide. The comparison shows that NTP spectrum is linked to the presence of the metallic slot-waveguide while the broad background and sharp peak at 2330 cm^{-1} are due to the photoluminescence of the 2.5 mm long SiN waveguide.

This first demonstration of Raman spectroscopy using a metallic slot-waveguide is a promising approach for enhancing Raman signal from analyte present in limited quantity or concentration such as a monolayer. A first measurement indicates negligible background associated to the plasmonic structure and further measurements are expected to quantify more accurately the enhancement in term of signal to noise ratio as compared to previously reported on-chip Raman sensors.

The authors acknowledge FWO and ERC-InSpectra for their financial support.

References

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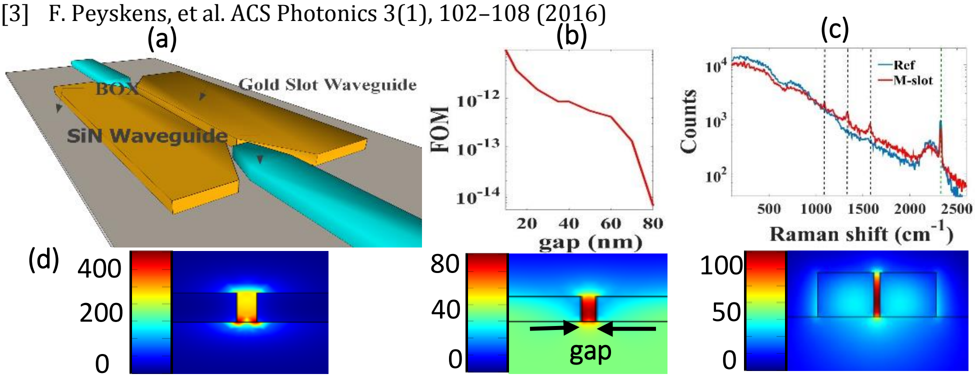

Fig. 1(a) Schematic of photonic-plasmonic structure (b) Optimized figure of merit (FOM) vs gap of metal slot waveguide. (c) Raman spectra measured from NTP-coated metallic-slot waveguide (red) and from bare SiN waveguide (blue) on same chip (laser power of 10mW and acquisition time of 1s) (d) Mode profile (Normalized E-field) of a metallic slot waveguide (left), a SiN slot waveguide of identical structure (center) and optimal dimension (right).