Surface enhanced Raman spectroscopy via isolated plasmonic nanoantennas integrated on silicon nitride waveguides

Ali Raza^{1,2}, Javier Losada³, Stephane Clemmen^{1,2}, Roel Baets^{1,2}, Amadeu Griol³ and Alejandro Martínez³

¹ Photonics Research Group, INTEC Department, Ghent University-imec, Technologiepark-Zwijnaarde, 9052 Ghent - Belgium

² Center for Nano- and Biophotonics, Ghent University – Belgium

³ Nanophotonics Technology Center, Universitat Politècnica de València, Camino de Vera s/n, 46022, Valencia - Spain

e-mail: amartinez@ntc.upv.es

ABSTRACT

In this work, we demonstrate experimentally surface enhanced Raman spectroscopy (SERS) via bowtie metallic nanoantennas integrated in silicon nitride waveguides. Two different configurations are considered: nanoantenna on top of the waveguide and nanoantenna inside a gap created in the waveguide. Even though both configurations allow for observing the Raman peaks of 4-nitrothiophenol (NTP), the latter shows higher peaks as a result of a better coupling from the Raman active centres to the fundamental TE waveguide mode. Our results pave the way towards the realization of massive SERS detectors on silicon-compatible photonic chips.

Keywords: silicon photonics; SERS; nanoantennas; hybrid photonic circuits.

1. INTRODUCTION

Recently, there has been a growing interest on the integration of plasmonic nanostructures in silicon waveguides. The main reason is that this hybrid plasmonic-photonic approach [1] reunites the best of two worlds: on one side, the extreme optical properties of plasmonic nanostructures enabling optical functionalities (sensing, modulation, non-linear processing) at low power and in sub-micron foot-prints; on the other side, the possibility of massive fabrication of photonic integrated circuits (PICs) using silicon-compatible technologies.

One of the possible plasmonic structures to be integrated in silicon-based PICs is a metallic nanoantenna supporting a localized surface plasmon resonance (LSPR) either in the near-infrared [2] or in the visible [3]. Silicon and silicon nitride waveguides are chosen respectively for each wavelength regime. In the simples approach, plasmonic nanoantennas are built on top of the dielectric waveguides so that they are excited via coupling between the evanescent part of the waveguide mode (typically, the fundamental TE-like mode) and the nanoantenna LSPR. Although this approach has enabled different interesting experimental results, the weak coupling (~10%) between the waveguide mode and the LSPR makes typically necessary to include a set of nanoantennas for an efficient detection (for instance, in the demonstration of on-chip surface-enhanced Raman scattering (SERS) [4]). Stronger coupling (and as a result, larger amplitude contrast at the waveguide output) can be obtained if the nanoantenna is placed in a subwavelength gap created in the silicon waveguide, so that the nanoantenna is perfectly aligned with the waveguide optical axis [5]. In both cases, the nanoantenna – supporting strongly localized fields in its surroundings - would act as an efficient transducer between the Raman-active centres of a certain substance and the waveguide modes, thus ultimately enabling massive SERS detectors in a silicon-compatible PIC.

In this work, we demonstrate SERS via isolated bow-tie Au nanoantennas integrated in silicon nitride waveguides using the previous approaches and establish a comparison between them from numerical and experimental results.

2. NUMERICAL SIMULATIONS

Figure 1 shows a sketch of the two different configurations to be considered in this work: bow-tie Au nanoantenna on top of a silicon nitride waveguide (CONF1) and inside a subwavelength gap etched in a silicon nitride waveguide (CONF2). Notice that CONF2 requires first etching the waveguides (and the gap), then depositing SiO_2 so that the nanoantenna can be aligned with the waveguide axis and then building the nanoantenna inside the gap [5]. Full 3D numerical simulations using the software *CST Microwave Studio* were used to analyse the performance both devices. Two parameters have been considered in the simulations in order to know about the performance of the system: the β -factor, which is determined by the ratio of the Stokes power coupled into the fundamental TE-like mode of the waveguide; and the intensity enhancement, which is obtained by monitoring the local electric field enhancement in the middle of the nanoantenna gap when the waveguide is fed by the fundamental TE-like waveguide mode.

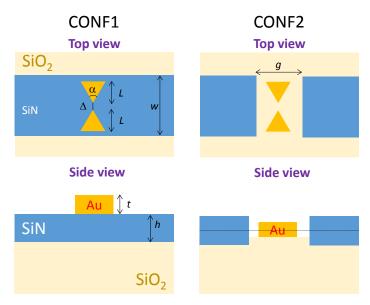


Figure 1. Sketches of the two devices under study in this work: nanoantenna on top of the waveguide (left-hand side) and nanoantenna in a gap created in the waveguide (right-hand side).

We started by simulating CONF1 using the nominal parameters of the system that was used in Ref. [4] to measure SERS in a chain of nanoantennas. These parameters are: α =60°, L=100 nm, Δ =50 nm, w=700 nm, h=220 nm, and t = 30 nm (plus 3 nm of Ti for adhesion). Figure 2 shows on the left the β -factor and intensity enhancement as a function of the wavelength for this structure. Notice that we have observed that the LSPR of the nanoantenna is redshifted with respect to the laser wavelength to be used in the experimental system (λ =785 nm) as well as with respect to the expected Stokes peaks. Then, we simulated CONF2 and optimized the parameters in order to maximize both the β -factor and intensity enhancement. The results are shown in Fig. 2 (right column) for an optimized device with α =64°, L=120 nm, Δ =20 nm, w=700 nm, h=220 nm, g= 250 nm and t = 30 nm (again plus 3 nm of Ti for adhesion). In Fig. 2 we have also included the results for the fabricated structures after extracting the dimensions from scanning electron microscope images (see Fig. 3). Notice that, in general, CONF2 can lead to one order of magnitude larger values of the β -factor and the intensity enhancement as a result of the improved interaction between the waveguide mode and the nanoantenna. However, we want to highlight here that CONF1 could be further optimized to improve its response.

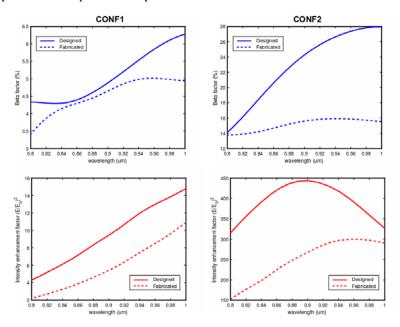


Figure 2. Numerical results obtained with CST Microwave Studio: β-factor (top) and intensity enhancement (bottom) for CONF1 (left) and CONF2 (right). Solid (dashed) lines show the results for the nominal (fabricated) parameters of the devices.

3. EXPERIMENTAL RESULTS

Samples with devices for both CONF and CONF2 were fabricated and teste in the lab. Details about the fabrication methods can be found in Ref.[4] (For CONF1 devices) and in Ref. [5] (for CONF2 devices). Both chips were coated with gold binding layer of 4-nitrothiophenol (NTP) using full night immersion in 1.55 g/L solution of NTP in ethanol. We recorded the Raman spectra in a back reflection configuration using a confocal Raman microscope (WITEC Alpha 300 R+, equipped with 40×0.6 NA objective, 785 nm wavelength and -70° C cooled ANDOR IDUS CCD camera). Both spectra shown in Fig 3 are recorded using 1mW power and 1 Sec integration time. Raman peaks are detected in both approaches, which shows the potential of the hybrid approach for on-chip SERS. Moreover, the Raman peaks – especially the one at $1,340 \text{ cm}^{-1}$ - can be more clearly distinguished in CONF2, in good agreement with the numerical simulations. We believe that larger signal-to-noise ratio in the Raman spectrum can be achieved by further design of the systems.

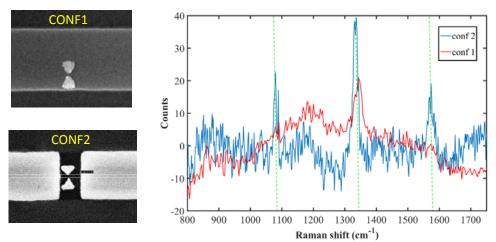


Figure 3. Background subtracted NTP Raman spectrum measured using CONF 1 (red) and CONF2 (blue) Green dotted lines corresponds to the position of three Raman modes of NTP. Scanning electron microscope images of the fabricated samples for both configurations are shown on the left.

4. CONCLUSIONS

We have demonstrated experimentally on-chip SERS in the 700–1000 nm region using isolated bow-tie nanoantennas integrated either on top or inside a gap of a silicon nitride waveguide. Remarkably, the active area of the sensing region is much less than 1 μ m², which should prospects for miniaturization well beyond what can be achieved using dielectric photonics. This finding shows that integration of plasmonic elements into silicon-based waveguides can pave the way towards hybrid PICs to be used in multiple applications in the classical and quantum domains.

ACKNOWLEDGEMENTS

A. M. acknowledges support from the Spanish Ministry of Economy and Competiveness (MINECO) under grants TEC2014-51902-C2-1-R and TEC2014-61906-EXP. The authors also acknowledge FWO and ERC-InSpectra for their financial support. We also acknowledge Dr. Frederic Peyskens for his useful discussion.

REFERENCES

- [1] F. J. Rodriguez-Fortuño, A. Espinosa-Soria, A. Martínez: Exploiting metamaterials, plasmonics and nanoantennas concepts in silicon Photonics, *J. Opt.* vol. 18, 123001, 2016.
- [2] I. Alepuz-Benache, C. García-Meca, F. J. Rodríguez-Fortuño, R. Ortuño, M. Lorente-Crespo, A. Griol, A. Martínez: Strong magnetic resonance of coupled aluminum nanodisks on top of a silicon waveguide, *Proc. SPIE* 8424, 84242J, 2012.
- [3] F. Peyskens, A. Z. Subramanian, P. Neutens, A. Dhakal, P. Van Dorpe, N. Le Thomas, R. Baets: Bright and dark plasmon resonances of nanoplasmonic antennas evanescently coupled with a silicon nitride waveguide, *Opt. Express* vol. 23, no. 3, pp. 3088–3101, 2015.
- [4] F. Peyskens, A. Dhakal, P. Van Dorpe, N. Le Thomas, R. Baets: Surface enhanced Raman spectroscopy using a single mode nanophotonic-plasmonic platform, *ACS Photon.* vol. 3, no. 1, pp. 102–108, 2016.
- [5] A. Espinosa-Soria, A. Griol, A. Martínez, Experimental measurement of plasmonic nanostructures embedded in silicon waveguide gaps, *Opt. Express* vol. 24, no. 9592-9601, 2016.