Raman spectroscopy using photonic waveguides

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We propose to use silicon nitride (Si_3N_4) based photonic waveguides in a lab-on-a-chip context for Raman spectroscopy. The evanescent tail of the guided modes can efficiently excite Raman active molecules located in the cladding of a waveguide. Similarly, a significant fraction of the total emitted Stokes power is evanescently coupled to the same mode. We calculate the spontaneous Raman scattering efficiency as a function of Si_3N_4 strip waveguide dimensions and show that under typical conditions, the efficiency is in the order of 10^{-8}. Preliminary experimental results are reported, as the first demonstration of waveguide assisted Raman spectroscopy (to the best of our knowledge).

Introduction

Sensors based on integrated photonics have evolved as a very promising route for lab-on-a-chip sensing applications [1]. One class of sensors is aimed at waveguide assisted spectroscopic techniques, such as absorption spectroscopy, fluorescence spectroscopy and Raman spectroscopy. These spectroscopic sensors are based on light-matter interaction of the evanescent field of the waveguide and allow a direct measurement of the specific spectrum of the molecules rather than other indirect methods using evanescent fields, such as refractive index sensing. Further, compared to free-space spectroscopy, the enhancement effects inherent to the waveguide, alongside with the long interaction length, lead to an increased light-matter interaction, resulting in a higher sensitivity as required by spectroscopic applications, especially in the context of Raman spectroscopy. In this paper, we summarize the theoretical and experimental results to realize Raman sensors based on Si_3N_4 photonic waveguides.

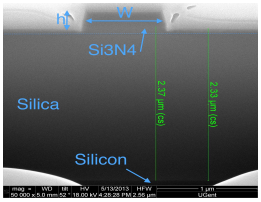

Fig. 1: SEM picture of the complete Si_3N_4 waveguide cross-section discussed in this paper including underlying SiO_2 and silicon substrate.

Recently, we investigated the scattering efficiency of a particle located in the vicinity of a channel waveguide for several geometries [2]. We theoretically determined the overall efficiency (η) of (Raman) scattering from a guided mode and subsequent emission coupled back to the fundamental mode (both TE and TM) of a rectangular channel waveguide by uniformly distributed particles in its surrounding. We define η as the ratio between the emitted power collected by the waveguide and the incident pump power in the mode. It was shown that η is highly dependent on the mode area that further depends on the waveguide index and geometry. The same methodology can be applied to more

popular strip waveguide geometry (Fig. 1) for silicon nitride (Si_3N_4) waveguides [5]. In this paper, we numerically evaluate η as a function of the waveguide width (w) for several thickness (h) values for Si_3N_4 (n=2) waveguides in water (n=1.33) cladding. The preliminary result of the on-going experimental investigation to verify the aforementioned theoretical results is also reported.

Theoretical investigations for collection of Raman signal for silicon nitride waveguides

Assuming that the Raman scattering mechanism can be modelled as the radiation of an oscillating dipole, and neglecting the Stokes shift, in the weak coupling regime, it can be shown that the efficiency of spontaneous Raman collection from molecules of scattering cross section σ and density ρ lying in the surrounding of a waveguide into a mode of a channel waveguide of length l is given by [2]:

$$\eta = \frac{\pi^2 \lambda_0^2}{(\varepsilon_0 \varepsilon)^2 n} (\kappa n_g^2 \Gamma^2 l) \sigma \rho \qquad (1)$$

Where, n_g is the group index of the mode, $n=\sqrt{\varepsilon}$ is the refractive index of the surrounding medium, and ε_0 is the permittivity of the vacuum, κ is a constant relating the polarizability and the cross-section of the molecule, Γ is a parameter dependent on the overlap of the mode area and the sensing area, given by:

$$\Gamma^2 = \int_{sens} ds \left(\frac{\varepsilon_0 \varepsilon |E(\vec{r}_0)|^2}{\iint \varepsilon_0 \varepsilon(\vec{r}) |E(\vec{r})|^2 d\vec{r}} \right)^2 \qquad (2)$$

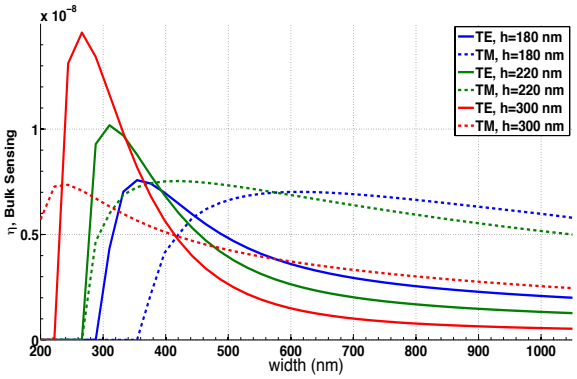

Fig. 2: Efficiency of Raman signal collection for the fundamental TE and TM mode of Si_3N_4 strip waveguides [5] as a function of waveguide widths for several heights (h) of the strip.

We use the COMSOL finite element eigenmode solver to determine the field of the waveguide modes for several waveguide geometries and solve Eq. (1). For simplicity, we neglect material dispersion, pump depletion, and assume $n_g \approx n_{eff}$ as this will have negligible impact. As an example, we take $\sigma=3\times10^{-30}$ $m^2Sr^{-1}molecule^{-1}$, as a typical cross-section of the dyes, ρ=1 mole/liter and calculate η for a 1 cm long waveguide. It is emphasized that the parameter values we have chosen do not affect the generality of our results because η scales in a simple way with these parameters as per Eq. (1).

Fig. 2 shows the calculated η as a function of the waveguide width (*w*) and height (*h*) for Si_3N_4 waveguides for bulk sensing for fundamental TE and TM polarized mode. The maximal efficiency, η_{max} is approximately $1.5x10^{-8}$, implying that under ideal conditions, for 1 mW of pump power in the waveguide, 15 pW of scattered signal can be collected by the guided mode. It is to be noted that the equivalent efficiency in a free-space confocal microscope system using diffraction-limited beams would be in the order of 10^{-10}. The efficiency is maximal near the cutoff widths and decreases sharply before becoming almost constant for wider widths.

Experimental observation of Raman peaks of Rhodamine 6G dye

The experimental setup is illustrated in Fig. 3. The setup is designed to measure the spontaneous Raman signal in a back-reflection configuration to reduce the collection of direct pump light by the spectrometer. A tuneable Ti-Sapphire CW laser (λ=720-975 nm) was used as the pump source. For the experiments, a wavelength of 785 nm is coupled to the waveguide by end-fire coupling using an aspheric lens of effective focal length 2.75 mm (NA=0.64). In order to avoid collecting the pump light, it passes via a dichroic beam splitter reflecting 785 nm and transmitting wavelengths longer than 790 nm. The counter-propagating Raman signal (Stokes) collected by the waveguide is then collimated via the same lens towards the dichroic filter. The collimated light is filtered using an edge filter, with the edge wavelength at 790 nm so as to block any stray pump light leaking into the spectrometer. The Raman signal is then focused to a multimode optical fibre of 50-microns core diameter using an aspheric lens of 11 mm effective focal length (NA=0.2) and measured using a commercial spectrometer (Avantes SensLine).

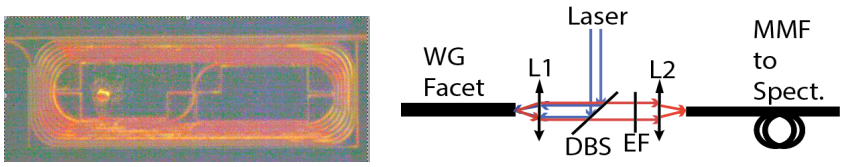

Fig. 3: (Right) The schematic of experimental setup. The pump laser at 785 nm is focused at the waveguide (WG) facet using an aspheric lens (L1). The counter propagating spontaneous Raman signal from the waveguide is coupled to the multimode fibre (MMF) using another aspheric lens (L2) via a dichroic beam splitter (DBS) and long pass edge filter (EF). (Left) The spiral used for measurement tinted with R6G dye used as Raman active molecules.

We take a silicon nitride waveguide (w= 500 nm, h= 220 nm) on top of a 2.4 μm silica cladding on a silicon substrate (Fig. 1). A spiral waveguide (1 cm in length) is used as the sensing region for the experiment (Fig. 3). The laser power is set to 150 mW, but it was sub-optimally coupled (coupling loss: ~15 dB) to the waveguide so as to avoid pump hitting the substrate and under-cladding that would otherwise generate a background that saturates the detector. A drop of 0.41 mM Rhodamine 6G (R4127, Sigma Aldrich) dissolved in ethanol was dried on top of the waveguide spiral (Fig. 3). During this procedure, special attention was taken not to stain the facet with the solution that would make coupling of pump and signal difficult. The measured spectrum (60 s integration time) before and after the application of the dye is shown in Fig. 4. A noticeable feature of both spectra is that they have the strongest peak at 520 cm^{-1} corresponding to the silicon substrate and a broad fluorescence background due to the undercladding or the substrate of the waveguide. The small peaks due to the dye are

clearly visible, but are embedded in a broad background. In order to extract the spectrum, we take the spectrum in the range 1000-1700 cm^{-1} and apply the I-polyfit algorithm [3] to remove the background and compare the result (Fig. 4) with the Raman spectra of the dye provided by the manufacturer [4]. An excellent match between the spectra can clearly be seen after removing the fluorescence background.

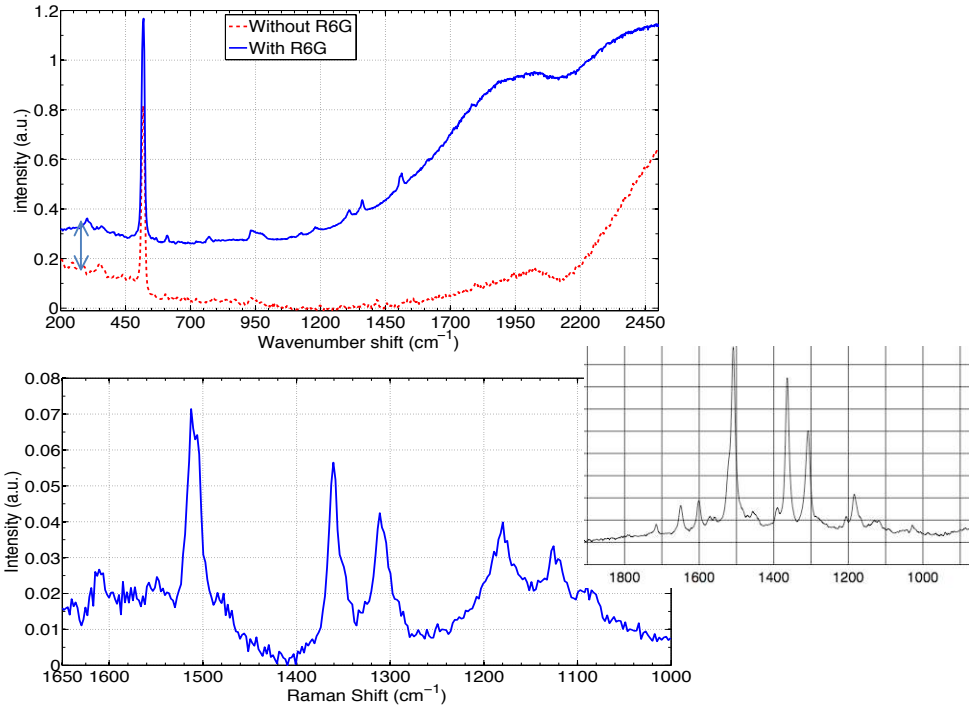

Fig. 4: (Top) Spectra measured before (red-dashed line) and after (blue-solid line) application of the dye. The spectrum with dye is shifted up by 0.2 units, for clear visualization. The peak at 520 cm^{-1} corresponds to Raman emission of silicon. The broad fluorescence from the under cladding forms the background for the emission from the dye, which are visible as small peaks. (Bottom) The spectra of dye extracted using I-polyfit algorithm [3]. In the inset, the section of spectrum provided by the manufacturer is also presented for comparison.

Conclusion

The theory predicts Raman signals of 10 pW per cm of waveguide per molar concentration of dyes for 1 mW pump. Initial experimental results suggest that photonic waveguides provide a very promising route for miniature Raman spectroscopy. More experiments are needed to confirm the numerical prediction of the theory.

The authors acknowledge ERC advanced grant InSpectra for the funding and imec, Leuven for processing of the waveguide.

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