Detection of Surface-enhanced Raman Signals from a Single Nanoplasmonic Antenna Integrated on a Single Mode Waveguide

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We present the first demonstration of on-chip Raman spectroscopy using a single nanoplasmonic antenna integrated on a single mode nanophotonic waveguide. To achieve this goal, shot noise associated with waveguide background is investigated.
OCIS codes: (130.3120) Integrated Optics Devices; (250.5403) Plasmonics; (170.5660) Raman Spectroscopy.

High index contrast nanophotonic waveguides allows an efficient excitation and collection of Raman scattered light via evanescent coupling to an analyte [1,2]. Such waveguide based Raman systems are ideal for dilute solutions and monolayers [3] but they don't provide sufficient field enhancement to probe tiny analyte volumes. On the other side, plasmonic antenna provide even greater field enhancement leading to what is known as surface enhanced Raman spectroscopy (SERS). SERS has been reported from single antenna [4]. The combination of those two technologies is expected to lead to on-chip SERS but so far the shot noise associated with waveguide material background has always required using multiple antenna to collect an observable Raman spectrum [5]. Indeed, the realization of single antenna based surface-enhanced Raman spectroscopy (SERS) requires an antenna structure with high field enhancement and background mitigation strategies. In our present demonstration, we measure an on-chip-SERS signal from an evanescently excited single bowtie antenna covered with a monolayer of 4-nitrothiophenol (NTP) as depicted in Fig.1 (a). To understand the limitations and possible further improvements, we also analyze the effect of waveguide length on signal-to-noise ratio.

Our waveguides and bowtie plasmonic antenna are fabricated via e-beam lithography [5] and the exact geometry is depicted in Fig.1. The monochromatic beam at 785 nm is coupled into the silicon nitride (SiN) waveguide through horizontal coupling via the input facet. At the output, the waveguide is terminated by an inverted taper to avoid spurious reflections. The waveguide loss without any antenna has been estimated at 4.2dB/cm using the cut back method at 785 nm pump wavelength.

For measuring a Raman spectrum that is solely attributed to the antenna and not to the evanescent field of the waveguide, we have coated NTP that selectively binds to gold. We acquire Raman spectra in a back-reflection configuration on a conventional Raman microscope (Witec Alpha 300R+, equipped with 100x/0.9 objective, 785 nm

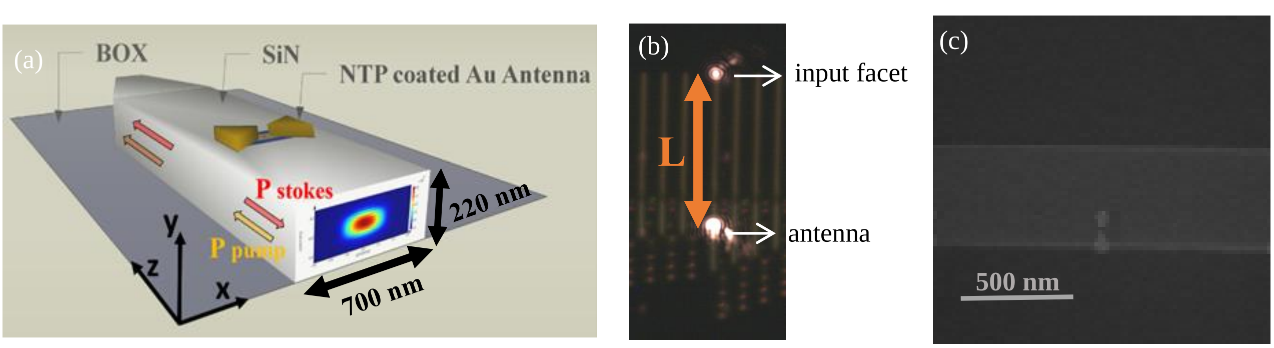

Fig. 1 (a) Schematic of bowtie antenna on SiN rib waveguide followed by an inverted taper. The profile of the fundamental TE mode at 785 nm is shown in the facet. The thickness, gap and longitudinal height of the antenna is 30, 40 and 110 nm respectively (b) An optical microscope top view of the waveguide and the bowtie antenna showing light scattering from antennas. L is the distance from input facet to antenna (c) SEM image of bowtie antenna on SiN waveguide.

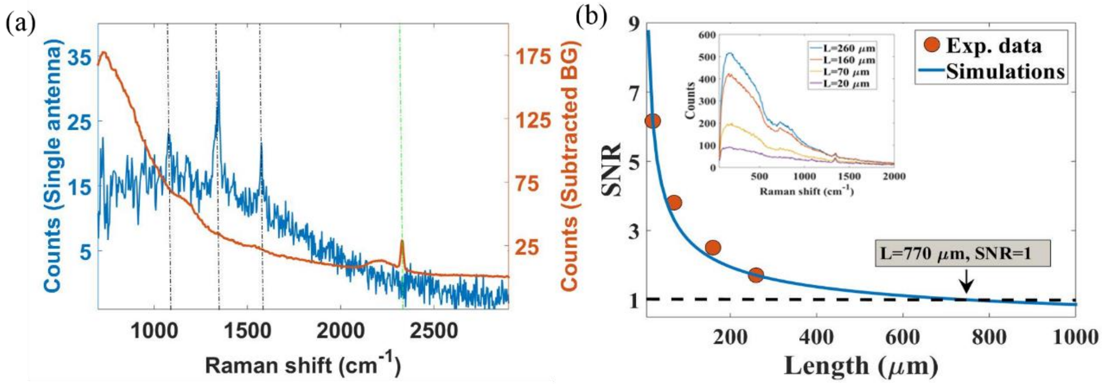

Fig. 2 (a) The Raman spectra measured from a waveguide functionalized with a single N=1 antenna (blue). The background spectrum is shown on the right axis. (b) The measured and fitted Signal-to-noise ratio for a single antenna on waveguides of length L. The small inset shows the Raman spectra for waveguides of L = 20, 70, 160 and 260 µm.

laser and -70 C cooled Andor IDUS 401 CCD camera) using a power of 0.5 mW and a 30 x 1 s integration time. The polarization is aligned to the TE-mode of the waveguide. Fig 2(a) shows the Raman spectrum of NTP measured from a waveguide with L=20 µm coupled to a single antenna. The background that is subtracted from this NTP-spectrum is measured on a long waveguide without antenna and rescaled to match the length of the waveguide used in combination to the antenna. Better background subtraction is possible and currently under investigation. The lines corresponding to NTP (black dotted line) are clearly identified on the antenna spectrum but are absent on the background spectrum that instead shows a peak at 2330 cm^{-1} (green dotted line) due to the Raman response of SiN.

In order to analyze the signal-to-noise-ratio, an analytical formula is derived that incorporates the backscattered signal generated by an antenna and background associated with SiN. If we assume a) same waveguide loss α at pump and Stokes wavelength, b) no back reflection of pump from antenna and c) no back reflection of Stokes and Pump from the output waveguide facet then the SNR for the backscattered signal is given as

$$\mathrm{SNR}=\frac{\eta_{NTP}\sigma_{NTP}\sigma_{NTP}\times\exp(-2\alpha L)}{\sqrt{(\eta_{NTP}\sigma_{NTP}\rho_{NTP}\exp(-2\alpha L)+((\eta_{SiN}\sigma_{SiN}\rho_{SiN})\times(1-\exp(-2\alpha L))\times(2\alpha)^{-1}))}}$$

where η,σ and ρ are the single antenna conversion efficiency, Raman cross section and molecular density respectively. η quantifies the enhanced Raman signal from an antenna coupled to a waveguide mode [5] and computed using Lumerical FDTD simulation software. The exponential decay term in analyte signal corresponds to decay of forward propagating pump before excitation and backscattered signal after excitation. Likewise, in SiN background, the exponential decay is the result of integrated backscattered signal over the whole length. As shown in Fig 2(b), this formula fits well to the experimental data measured from waveguides of different length. For a given pump power and integration time, SNR will drop below 1 when the background associated shot noise becomes too high. As a result, there is a maximum waveguide length for any specific Stokes wavelength for which the SERS signal can just overcome the inherent background of the waveguide material. Above this length, the Raman signal will become undetectable. Given this length, is on the orders of hundreds of microns, on-chip filtering is certainly a possible way to mitigate this problem. Further improvement of the field enhancement of plasmonic antenna [6] would make that maximal length even greater.

In summary, we have, for the first time, demonstrated surface-enhanced Raman scattering from a single antenna integrated to a single mode SiN waveguide. Furthermore, we analyze the background associated with the SiN waveguide and conclude that the maximum waveguide length over which the SNR becomes insufficient is compatible with on-chip spectral filtering technologies such as ring cavities. This work therefore paves the way towards fully chip-integrated SERS sensor.

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

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