Resonant enhancement mechanisms in lab-on-chip Raman spectroscopy on a silicon nitride waveguide platform

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Abstract: Lab-on-chip approaches, based on CMOS-compatible silicon nitride photonic waveguides, may have a large potential for Raman spectroscopy. The role of various resonant mechanisms to enhance the Raman signal is discussed.

1. Introduction

Raman spectroscopy gains importance as a label-free and foolproof detection method for a variety of chemical substances and biomolecules. The major barrier to the widespread use of Raman spectroscopy is the fact that the spontaneous Raman scattering process is extremely weak, leading to the use of pump lasers with relatively high power or the use of long detection integration times, especially for low analyte concentrations. These factors limit the application range. While in most Raman spectroscopy systems the light-matter interaction is organized by means of a confocal microscope or a fiber probe, here we discuss the potential of a lab-on-chip approach based on silicon nitride (Si_3N_4) photonic waveguides. The basic concept is analogous to the use of hollow fibers – with the analyte in the core – for Raman spectroscopy [1], but now the analyte forms the cladding of a nanophotonic waveguide structure on a chip, with relatively high refractive index contrast. In this way a strong interaction is created between the mode(s) in the waveguide structure and the molecules of the analyte, thereby leading to a large molecular cross-section for the scattering process. Furthermore the waveguide structure on the chip may be functionalized with extra enhancement mechanisms such as nanoplasmonic antennas or photonic cavities. All these elements can lead to dramatic improvements of the Raman scattering efficiency, thereby opening the route towards Raman spectroscopy with mW-level pumps and short sub-second integration times even for analyte concentrations below 1000 ppm. The fact that the nanophotonic chips can be produced by CMOS-compatible process steps, paves the way for compact, low-cost systems with high performance. The chip can then be a use-once disposable chip.

The key figure of merit in Raman spectroscopy is the minimum detectable analyte concentration for a given pump power and integration time. In certain applications the volume of probed analyte needs to be as small as possible, e.g. in high resolution imaging applications or in intracellular sensing. In this case the relevant limit of detection is the minimum detectable number of analyte molecules for a given pump power and integration time. In the limit this can lead to single molecule detection.

2. Waveguide enhancement

Waveguide modes are transverse electromagnetic resonances and provide confinement of light over long distance. For a given guided mode power the electric field strength is high and therefore the interaction with molecules is strong. In a configuration whereby the pump beam is the fundamental mode of a single mode waveguide and the Stokes light is collected into this same mode, the collected power scales with waveguide length (waveguide losses notwithstanding). Furthermore the Stokes light is collected in a single electromagnetic mode, which provides the ideal situation to maximize the spectral resolution and/or minimize the footprint of the spectrometer.

The efficiency of collection of the omnidirectional Raman scattering into a waveguide mode depends heavily on the index contrast of the waveguide. In [2-3] it was shown that this dependence is quadratic with the index contrast. While silicon waveguides provide a very high index contrast and are available through standard technology platforms in CMOS-fabs, their opacity at wavelengths below 1 micrometer renders them less ideal for Raman spectroscopy, especially of biological media. We have therefore developed a Si_3N_4 waveguide platform in a CMOS fab [4]. Through the use of this platform we have demonstrated Raman spectroscopy of

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Isopropylalcohol covering the Si_3N_4 waveguide with a collection efficiency far higher than in a Raman microscope [5]. An as of yet unresolved issue is to deal with the background in the Raman spectrum that can directly affect the limit of detection of the device because of the shot noise associated with the background. The origin of this background – somehow related to the waveguide – is not fully clear yet and depends on the technological parameters of the waveguide fabrication. Future work will focus on its reduction through a variety of strategies.

3. Plasmonic enhancement

Nanoplasmonic structures have been known to enhance the Raman efficiency by many orders of magnitude and over a wide wavelength range through Surface Enhanced Raman Scattering (SERS). Nanoplasmonic structures can be integrated on top of Si_3N_4 waveguides in a variety of ways: through the adsorption of gold nanoparticles, through the use of nanosphere lithography or through the nanopatterning of gold nano-antennas such as bow-tie antennas. For the latter case, in [6] we have demonstrated through simulation that the efficiency of Raman scattering can be enhanced by ten orders of magnitude. Obviously this enhancement takes place over only extremely small volumes near the tips of the bow-tie antenna. An interesting side-effect of the large field gradient near the tip is that particles (with a size of the order of 100nm) can be trapped. This can have a stabilizing effect on the SERS-spectra of molecules in such particles, when the trapping time becomes larger than the required integration time.

We have also demonstrated the use of ultra-small Si_3N_4 chips (with dimensions of a few micrometers) as intracellular Raman probes [7]. These chips were made SERS-efficient through the adsorption of gold nanoparticles on their surface and incorporated into fibroblast cells through electroporation.

4. Cavity enhancement

Photonic microcavities – ring or disk resonators, DBR cavities, photonic crystal cavities - provide another means to enhance the electric field and therefore the Raman efficiency. At the same time they reduce the volume of analyte that is being sampled. The use of photonic microcavities will therefore definitely improve the molecule number detection limit while the improvement is less obvious for the concentration detection limit, unless Purcell enhancement is significant.. Various strategies can be envisioned. In a first case the cavity volume is very small (wavelength scale) such that the resonant enhancement is broadband across the entire Stokes spectrum, as in the nanoplasmonic case. In a second strategy the cavity is large such that the pump coincides with one cavity resonance and the Stokes spectrum is sampled with the free spectral range of the cavity. In a third strategy the cavity is only resonant for the pump and not for the Stokes spectrum. The best strategy will depend on the application envisaged.

5. Conclusion

Raman-on-chip holds the promise of opening up a myriad of applications in bio-chemical analysis, both for fundamental research on intracellular processes and for medical applications. The technology is still embryonic and presents many fundamental and technological challenges.

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On behalf of the IEEE Photonics Society and the Program Committee, it is my great pleasure as Program Chair to welcome you to LaJolla, California for the 27th IEEE Photonics Conference (IPC) – the IEEE Photonics Society's annual showcase event. In keeping with the tradition of offering a unique mixture of ground-breaking reports, in-depth educational offerings, and important attendee opportunities for contribution and discussion, the Program Committee has put together a strong and broad technical program for the entire photonics community to collaborate, educate and carry us into the future. We thank you for coming and hope you are able to take advantage of the multiple program enhancements we have added this year.

IPC is the one photonics conference where you can be exposed to the "must-know" technology and research through our **Plenary Talks** and over one hundred and twenty **Invited Talks**, receive in-depth educational opportunities through the five **Tutorials** and learn about the hottest new topics through the three **Special Symposia**, post deadline papers and the ground-breaking contributed talks. In addition to these mainstays of IPC, we have some exciting enhancements this year, including some new events for our **Sunday Program**, the official launch of **Women in Photonics** with a Tuesday night reception, a new opportunity for students with the **Student Poster Competition** on Wednesday and the introduction of a **Closing Ceremony** as the final stand-alone session on Thursday featuring the post deadline session and student awards to finish the week. Our Sunday program will include a Silicon Photonics Fabrication Short Course, a workshop featuring a "meet the editors" and "hot papers" of our own IEEE Photonics Journal and IEEE Photonics Technology Letters, and finish with a panel session on metamaterials allowing the community to discuss the challenges and opportunities ahead for this exciting field. We hope these enhancements along with our traditional program events will provide you with your best IPC experience yet.

IPC will have two Plenary sessions this year, on Monday and Tuesday afternoons, each featuring two distinguished speakers providing prospective and insight on major current topics in photonics. On Monday, Professor John Bowers, of the University of California-Santa Barbara, will be speaking on the topic of "Hybrid Silicon Photonic Integrated Circuits" and Professor Toshio Yanagida, of Osaka University, on "Single Molecule Imaging and Nanometry: Fluctuation and Function of Life". This session will be followed by the Awards Ceremony and finish the evening with the Welcome Reception. The second Plenary session, on Tuesday afternoon, will feature Sir Peter Night FRS, of Imperial College London, presenting "Exploiting Quantum Coherence for New Technologies: Timing, Navigation and Sensors" followed by Professor Kerry Vahala, of the California Institute of Technology, giving his talk entitled, "Cold-Mechanics to Stable-Microwaves: the Future of Optical Microcavity Research". This session will be followed by the Women in Photonics Reception, open to all and presided over by the current Photonics Society President, Dr. Dalma Novak.

The main technical program, put together by the volunteer photonics experts of our technical subcommittees, consists of sessions organized with Invited and Contributed talks covering research across all fifteen of our committee topic areas and three special symposia providing topic overviews and opportunities for discussion on Optoelectronic Devices for Solar Energy Harvesting, High Power Diode Lasers and Systems, and Optomechanics. In addition, I encourage everyone attending IPC to take advantage of the highly educational and inspiring series of Tutorials. This year we have five Tutorial speakers, each given their own hour and a half session to provide the most in-depth educational experience possible and charged with covering their respective topics with sufficient detail to catch you up or launch you into a new technical area. This year's tutorials feature experts from industry, government and academia covering a wide variety of topics including data-aided signal processing, optical nanoantennas, plasmonics, time and frequency metrology and quantum well photonics. Attending any or all of these events should be high on any attendee's list and is sure to stimulate new directions and ideas for many areas of research and development.

The organization of any conference is the result of many dedicated people and IPC is no exception. On behalf of the conference committee, I would like to express my most sincere thanks to all the volunteers who have contributed to the success of this year's meeting in planning, engaging speakers, reviewing papers, organizing sessions and the inevitable re-planning of many of the activities featured in the program. I would also like to thank the Photonics Society Conference Activities Staff for their hard work, excellent organizational skills and advice as well as their unending patience.

We hope everyone has a rewarding conference, enjoys the San Diego area, and is inspired to come along for the ride again next year in Reston, Virginia!

Sincerely,

Thomas Clark

Program Chair, 2014 IEEE Photonics Conference

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