European Conference on Optical Communication (ECOC) 2022 ©
Optica Publishing Group 2022

Molecular Optomechanical Springs for Infrared Metasurface Detectors

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Abstract *We show strong coupling of a mid-infrared microscale resonator with ultra-localised plasmonic nanocavity modes. This enables new types of mixing between IR absorption and SERS, opening up new types of ultrasensitive molecular detection.*

Introduction

Of key importance in molecular spectroscopy is analysis of the infrared electromagnetic spectrum. Infrared spectral information accesses frequency bands for absorption of molecules in the atmosphere (greenhouse gases and ozone) or thermal emitted radiation (thermal imaging) as well as traces molecular fingerprints.

Nanometre-scale metallic cavities can confine photons into 'hotspots' of nanometre volume which boost the light-matter interaction. Using such nanocavities in spectroscopy for extreme light localisation now resolves movements of single atoms in molecule [1, 2]. One promising approach to detect infrared radiation is frequency upconversion via optomechanical coupling. Optomechanical interactions allow coherent conversion of signals between optical and mechanical domains. Nanocavities containing single molecular layers act as mechanical springs/oscillators, with infrared vibrational modes excited by a visible pump laser via their Raman polarizability (Figure 1a). Interaction of light and matter in these sub-nm mode volumes allows extreme optomechanical coupling and single MIR photon sensitivity [3]. Here we achieve frequency upconversion of 9-10 μm MIR incoming photons to visible photons via SERS at a pump wavelength of 785 nm.

Results

In this study, we combine bottom-up assembly of nanocavities with top-down photolithography to form the disk resonator (nanoparticle-on-resonator, NPoR). This is achieved by placing Au nanoparticles (60 nm diameter) on top of a 100 nm-thick disk microresonator (μ-resonator) of variable (here 6 μm) diameter (Figure 1b). The nanocavity gap can be controlled at sub-nm scales using a dielectric molecular spacer which is self-assembled on top of the Au disk.

Crucial for high upconversion efficiency is the optimal overlap of visible and MIR radiation. This is fulfilled through engineering doubly-resonant antennas, NPoRs, that focus long- and short-wavelengths into the same active region (Figure 1b). We demonstrate that effective optical coupling of a μm-scale disk and nm-scale molecular gap cavity lead to enhanced light localization (E^2/E_0^2>10^5). This leads to 200% higher integrated antiStokes SERS signal when the MIR pump beam is on, tuned with Raman and infrared molecular modes at wavelengths of 9-10 μm (Figure 1c). Further, the superposition of higher-order modes of the μ-resonator with the optical resonances of self-assembled nanocavities is found to control the near-field resulting in modulation of SERS intensities with nanoparticle location.

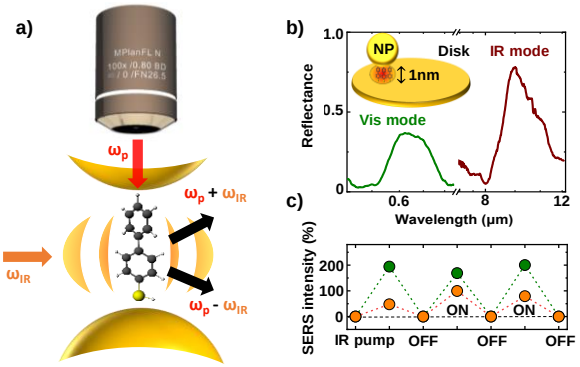

Fig. 1: Molecular nanocavity optomechanics for efficient infrared radiation detection via frequency upconversion. a) The physical mechanism: a mid-infrared (MIR) signal resonantly excites molecular vibration, which is parametrically coupled to light via optomechanical interaction in an ultrathin cavity. b) The doubly-resonant nanoparticle-on-resonator (NPoR) antenna has both visible and infrared resonances. c) The observed percentage change in antiStokes SERS integrated area on two different nanostructures (orange and green) obtained by switching the MIR beam ON/OFF at a wavelength of 10μm.

Conclusions

We demonstrate that effective optical coupling of a μm-scale disk resonator and nm-scale molecular gap cavity lead to enhanced light

localisation (>10^5). This provides new possibilities in single-molecule spectroscopies with vibrational peaks of molecular monolayers giving >200% higher SERS intensity (>10000% predicted under fully optimized conditions) and a 10-fold stronger electronic scattering compared to standard plasmonic constructs used before. This dual resonator approach can be extended to higher-Q optical μ-resonators that address specific vibrational modes to Stokes or antiStokes sides. Our results on efficient frequency upconversion of infrared radiation in visible regime via molecular optomechanics open new potential in plasmon-based low-cost infrared spectroscopic techniques.

Acknowledgements

A.X. acknowledges support from EMPA internal funding call (IRC 2021).

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