

Single-molecule mid-infrared vibrational spectroscopy and detection in plasmonic nanogaps

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The detection of molecular vibrations in the mid-infrared (MIR) range ($\lambda=3\text{-}30\mu\text{m}$) at room temperature has a wide range of applications, including gas sensing, chemical reactivity, medical imaging, astronomical surveys, and quantum communication. However, current technologies are limited by thermal noise, and rely on energy-intensive cooled semiconductor detectors such as mercury cadmium telluride (MCT). In this article, we present our recent advancements using plasmonic nanogaps to overcome this challenge by upconverting low-energy MIR light into high-energy visible wavelengths ($\lambda=500\text{-}800\text{nm}$) that can be detected using silicon technologies capable of detecting single photons [1-3].

By strongly coupling surface plasmons between two metallic nanostructures spaced a few nanometers apart, light can be confined to extreme dimensions, allowing for coherent coupling to electronic and vibrational states of the molecules assembled in the nanogaps. The measured light-matter coupling strengths approach strong light-molecule coupling at room temperature [4,5], enabling Purcell-enhanced light emission [6] and optomechanics with bond vibrations [7].

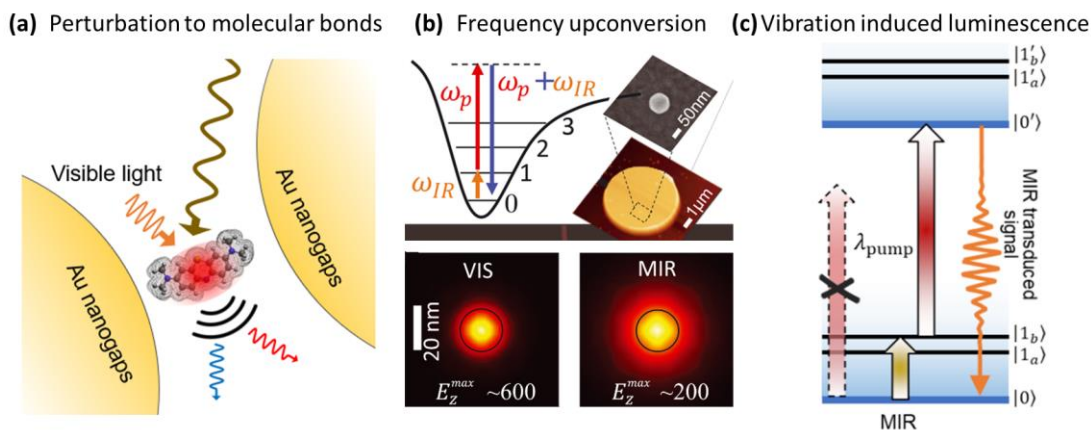


Fig. 1 Three different methods for detection and spectroscopy of molecular vibrations in the mid-infrared range at room temperature.

Recent advances in surface-enhanced Raman scattering (SERS) and enhanced light emission have opened up new possibilities for detecting molecular vibrations in the mid-infrared range at room temperature. We present three different methods for achieving this (Fig.1). The first method involves using nanoparticle-on-foil (NPOF) nanocavities that support both visible and MIR plasmonic hotspots, allowing for modulation of molecular SERS signals in the presence of MIR photons due to MIR absorption in the phonon resonance of the substrates [2,8]. In the second approach, phonon absorption is suppressed, and a 140% amplification of the SERS anti-Stokes emission is observed when an MIR pump is tuned to a molecular vibrational frequency [3,10]. The third approach involves assembling molecular emitters into a nanoscale cavity and continuously pumping them with optical energy, resulting in the transduction of MIR light absorbed by the molecular vibrations into visible luminescence [1,9].

These demonstration opens unique possibilities not just for molecular spectroscopy and sensing but has wide implications in quantum preparation of vibrational states for mode-selective chemistry and nano-optics.

References

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