Mid-Infrared Quantum Correlation Upconversion Spectroscopy

Yujie Cai[1], Yu Chen[1,*], E Wu[1,*]

[1]*State Key Laboratory of Precision Spectroscopy, East China Normal University, 200062, Shanghai, China*

The mid-infrared (MIR) spectral region contains rich molecular vibrational and rotational information and is important for the identification of complex substances in biology, materials science, and environmental monitoring. However, conventional MIR spectroscopy usually relies on MIR detectors with intrinsically high noise and on Fourier-transform systems with scanning structures. For applications such as weak-light detection, low-photodamage sample characterization, and stable measurements in complex environments, the reliable extraction of MIR spectral information at the single-photon level remains a key challenge.

Here, we develop a single-photon MIR spectroscopy scheme based on quantum correlations and frequency upconversion [1]-[2]. Broadband nondegenerate correlated photon pairs are generated through spontaneous parametric down-conversion (SPDC). One photon lies in the MIR band, covering the 2.76-3.94 μm spectral range, and serves as the probe for the sample. It is then synchronously upconverted to a wavelength range detectable by a silicon single-photon detector, while preserving its temporal and spectral correlations with the heralding photon. The conjugate heralding photon lies in the near-infrared telecom band. We further introduce a nonlocal wavelength-to-time mapping scheme by inserting a long dispersive fiber in the heralding arm, so that the wavelength information of the correlated photons is directly mapped onto the arrival-time difference of the photon pairs [3]. Based on coincidence measurements, true MIR signals can be distinguished from upconverted SPDC noise, upconverted thermal radiation, and detector dark counts. The scheme overcomes the limitations of conventional broadband MIR upconversion spectroscopy under weak-illumination conditions, with the MIR probe power as low as approximately 370 fW.

Combined with raster-scanning microscopy, we demonstrate MIR quantum hyperspectral microscopy for label-free tissue imaging. The hyperspectral data cube is formed from the coincidence spectra of all image pixels. Via nonlocal wavelength-to-time mapping, MIR spectra can be acquired without wavelength scanning, which increases the photon flux incident on the detector and improves the speed of hyperspectral data acquisition. In contrast to stained histopathology, the MIR band directly probes functional-group vibrations and therefore allows chemical-bond information to be identified across different regions of biological tissue sections [4]-[5]. Our MIR quantum correlation upconversion spectrum spans the protein N-H stretching band at 3030 nm and the lipid-related band at 3417 nm arising from the asymmetric CH_2 stretching vibration. The hyperspectral results indicate distinct molecular compositions in different spatial regions of the tissue and enable the identification of pathological changes.

MIR quantum hyperspectral microscopy is a label-free single-photon imaging method that provides intrinsic chemical information from functional-group vibrational spectra. It enables MIR spectroscopy under extremely low-illumination conditions with non-destructive and robust operation, and highlights the potential of quantum light sources for future biomedical applications.

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