

## molecular spectroscopy

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# PhD Thesis Defense HANYU YE 'Novel Pulsed Optical Parametric Sources in the Mid-Infrared and the Application Towards High-Resolution Molecular Spectroscopy'

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Friday, May 3, 15:00. ICFO Auditorium

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Optical Parametric Oscillators

ICFO-The Institute of Photonic Sciences

The mid-infrared (mid-IR) spectral region coincides with fundamental rotational-vibrational transitions of a large number of molecules in the gas phase. The strong interaction between

mid-IR light sources and these gaseous molecules has enabled experimental investigations of energy-level structures of various molecules and also led to a series of spectroscopic applications in, for example, biomedicine, environmental monitoring, and combustion diagnostics. Nanosecond pulsed optical parametric sources tunable in the mid-IR are versatile tools offering the potential to access different molecular transitions. Their compact size and relatively high pulse energy allow convenient and sensitive detection in various types of spectroscopy.

Achievable narrow linewidths close to Fourier transform limit also make it possible to resolve Doppler-limited fine absorption lines and distinguish different species with high selectivity. As such, this thesis aims at the further development of pulsed optical parametric sources in the mid-IR region, employing different nonlinear materials and down-conversion configurations, as well as their application towards high-resolution molecular spectroscopy.

We have demonstrated optical parametric generation (OPG) in the newly invented nonlinear crystal, orientation-patterned gallium phosphide (OP-GaP). Pumped by a Q-switched Nd:YAG laser, the OPG source at 25 kHz repetition rate generates tunable radiation across 1721-1850 nm and 2504-2787 nm. Detailed characterization including temperature tuning, pump transmission, and OPG threshold validated the performance of OP-GaP as the next-generation quasi-phase-matched nonlinear material.

Following the development of the OPG, we further successfully demonstrated and characterized a singly-resonant optical parametric oscillator (OPO) based on OP-GaP. Driven by the same Nd:YAG laser at 50 kHz repetition rate, the OPO delivers mid-IR idler in the spectral range of 2.8-3.1  $\mu\text{m}$ . Absorption-induced thermal effects in the OP-GaP sample were revealed and studied using different pump power levels.

We also demonstrated a stable, pulsed degenerate OPO at 2.1  $\mu\text{m}$  based on MgO:PPLN pumped by the same Nd:YAG laser with variable repetition rates from 65 kHz to 90 kHz. The OPO, in a Littrow-grating-cavity configuration, can provide up to 2.7 W of degenerate output with good pulse-to-pulse and long-term power stability. The spectral narrowing effect of the grating-cavity was also examined and compared to a plane-mirror linear cavity. Finally, we developed a high-resolution difference-frequency spectrometer in the spectral range of 3308.5-3317.3 nm. The mid-IR source for the spectrometer was based on single-frequency mode-hop-free tunable, pulsed difference-frequency-generation (DFG) in MgO:PPLN. A par

of the methane spectrum in the Q-branch of  $\nu_3$ -band was resolved and compared to HITRA simulation in both atmospheric pressure and reduced pressure. This conceptual technique can be extended to broader mid-IR regions for detecting various other molecules or to higher energy level for nonlinear spectroscopy with high resolution.

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