IR - MILLIMETER-WAVE DOUBLE RESONANCE FOR PRECISION MOLECULAR SPECTROSCOPY

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Metrology-grade spectroscopic measurements of molecules are of high importance for numerous applications such as testing fundamental physics¹, molecular physics², and astrophysics³. In this context, we tested the well-established double resonance technique^{4,5,6,7} using both infrared and millimeter wave transitions to provide high resolution and frequency metrology dataset. The hyperfine components of rotational transitions involving both the ground and the $v_2 = 1$ vibrational state of NH₃ as well as the $v_2 = 1$ vibrational state of D₂O have been investigated trough IR-THz Doppler-free and cross-over-free double resonance spectroscopy. Spectra were obtained using both a free running external cavity quantum cascade laser (QCL installed at SOLEIL synchrotron) as well as a sub-Hz QCL⁸ (installed at LPL Villetaneuse) frequency-stabilised on the REFIMEV signal⁹. The lasers have been used to optically pump the molecules inducing a velocity class selection. We will present the major interests of this approach for precision spectroscopy and the current limitations we meet.

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