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Summary

High resolution molecular spectroscopy is a powerful tool to test fundamental physics, to measure fundamental constants or even to look for physics beyond QED. An essential ingredient for such experiments is a narrow-linewidth, frequency-stabilized light source, tunable and powerful enough to implement a Doppler-free technique and detect the molecular transition with a good signal-to-noise ratio. Moreover, dealing with molecular transitions, a key spectral region is the Mid-IR, where the strongest vibrational bands of simple molecules can be found. Indeed, the lack of suitable sources and key optical components has represented until now a severe limit for this kind of experiments in the 1–10 μm spectral region. Common spectroscopic methods working above 5 micron wavelength suffer from at least one of these problems: they are feasible only in a narrow spectral range; the laser power available for spectroscopy is limited; the frequency accuracy is poor. To overcome these limitations, we implemented a setup for high-resolution spectroscopy that can be applied in the whole 1–10 μm range. The setup combines the accuracy of a metrological Mid-IR source and the spectral resolution achievable with molecular beams. Our source is based on a quantum cascade laser (QCL), which provides tens of mW optical power, and on a coherent source relying on difference-frequency (DF) generation in a nonlinear crystal. As I will discuss, the latter is linked to the Cs frequency standard and provides high accuracy, which is transferred to the QCL by a phase-lock loop. We measured a vibrational transition in a highly-excited metastable state of CO around 6 μm with 11 digits of precision [35]. The main contribution to the molecular linewidth (900 kHz) was given by residual Doppler broadening.

The core of this thesis is dedicated to the spectral characterization of this source and to the improvement of the resolution of the spectrometer down to the limit given by transit time broadening. These two goals are strictly connected, as we will see. In order to overcome the limit to spectral resolution given by residual Doppler broadening, I modified the experimental setup to reduce the molecules velocity spread. Along z , the molecular beam propagation axis, it depends upon the dimensions and relative distance of the two pulsed UV lasers used to prepare the molecules in the excited metastable state (excitation laser) and to ionize them for detection after interaction with the QCL (ionization laser). I focused as much

as possible these two lasers and made the molecules and the QCL almost collinear, with a small angle of 3.1 degrees, so that the relevant velocity component for the Doppler broadening was the one along the molecular beam axis. This allowed a better selection of the velocity of the molecules and allowed to reduce the Doppler broadening by one order of magnitude, down to 80 kHz.

At the same time, I worked on the improvement of QCL stability. Working on the reduction of the molecular linewidth, I realized that the loop used to phase-lock the QCL to the DF radiation wasn't working at its best, leading to a not optimal laser spectral profile. Substantial improvements in the QCL phase-lock effectiveness led to an enhancement of the QCL power channelled into the DF mode from about 40% up to 70%, a value in line with the best results in literature. Under these new conditions, we expect that the QCL laser replicates the narrow-linewidth feature of its reference DF radiation.

As a final result, we were able to measure molecular transitions with a full width at half maximum of 177 ± 10 kHz, thus improving the spectral resolution of our spectroscopic apparatus by almost one order of magnitude. We are now almost limited by transit-time broadening.

The high spectral resolution of these measurements allowed also to provide a good estimation of the laser spectral linewidth. Indeed, considering the expected contributions to the line broadening coming from residual Doppler broadening, transit time and inhomogeneity of the magnetic field, an upper limit to the QCL linewidth in optimal locking conditions of few tens of kHz can be set. This estimation is much more precise than that achievable with more standard methods for lasers in our spectral region. As I will discuss in the text, alternative methods can be used to measure the laser linewidth or, better, the laser frequency noise spectral density. Unfortunately, these methods are often not easily implementable in the Mid-IR due to technical or physical limitations, making measurement of frequency noise spectrum of stable lasers a big challenge.

As an example, I measured the source frequency noise power spectral density using the mode of a calcium fluoride whispering gallery mode microresonator which acts as an optical frequency discriminator to convert frequency fluctuations into measurable amplitude fluctuations. Laser spectral linewidth has been calculated via numerical integration. This method proved to be effective in recording the frequency noise spectral density of our source in free-running conditions, but it demonstrated to be unsuited to properly measure the noise of the source in phase-locking conditions because of high dark noise contributions, as I will discuss in the text. Therefore, using this method, I could only measure an upper limit of 700 kHz for the laser linewidth.