Studies of molecular properties using the Self-Heterodyne effect using a chirped Quantum Cascade Laser

A.M. Juarez, G. Bustos, O. Magaña
Instituto de Ciencias Físicas, Universidad Nacional Autónoma de México, PO Box 48-3, 62251 Cuernavaca, Mor., Mexico

E Mail: amjuarez@icf.unam.mx

The external-cavity quantum-cascade laser (EC-QCL) has become an important radiation source in the mid-IR (3µ to 20 µm) portion of the electromagnetic spectrum. QCLs have been successfully implemented into a variety of spectral studies for different applications, such as atmospheric and environmental monitoring [1], detection of trace explosives [2], and human breath analysis [3]. QCLs generate tunable laser light by means of a periodic series of nanometer sized layers which leads to one-dimensional multiple quantum well confinement. In the pulsed mode, a high electron current is injected into the active medium and, therefore, inducing joule heating. This in turn modifies the refraction index of the QCL medium leading to a fast wavelength tunning induced by the current pulse thus allowing "chirped" operation of the laser. In our work, we use an Uber Tuner, pulsed, broadly tunable EC-QCL from Daylight Solutions with a emission range in the mid-IR tunable from 6.9 to 8.7 µm. The pulse width can be selected in a range from 40 ns down to 500 ns with a variable repetition frequency of 100Hz down to 100 KHz. The QCL delivers a maximum pulse power up to 350mW close to the middle of the tunning range. A Herriot cavity is coupled to the laser and is placed in between the QCL and the MCT. In order to capture the time dependency of the QCL laser for a given frequency we used a SRS 2000 boxcar gated integrator with picosecond resolution. With this arrangement, we have achieved measuring Self Heterodyne Excitation of the H_2O molecule. This phenomenon occurs when a pulse with a fast frequency sweep is made to interact with a resonant system in the gas phase. A simple picture of the self heterodyne process can be obtained if one considers that the original pulse, travelling through the cavity, adds up linearly with the delayed pulse as produced by the molecular sample leading to interference patterns in the total radiation detected at the end of the cavity, if filled up with the sample gas[4]. A more complete analysis of this process requieres the analysis of a closed two level system interacting with a swept field, in terms of the optical Bloch equations. In the present work we report pressure dependent self heterodyne signals as produced in the Herriot cell by the resonant absorption of water vapor in the cavity corresponding to a rotation-rotation transition J-J´ in the vibrational ground state of the H2O. Beyond this specific use of Quantum Cascade Lasers, I will also discuss potential applications of Quantum Cascade Lasers in state selective autodetachment of negative ions, studies of the competition of detachment-dissociation channels in negative molecular ions and the potential use of intracavity Mid Infrarred Spectroscopy as a potential diagnostic tool in ion storage rings.

References

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