Frequency modulation spectroscopy with a terahertz quantum-cascade laser

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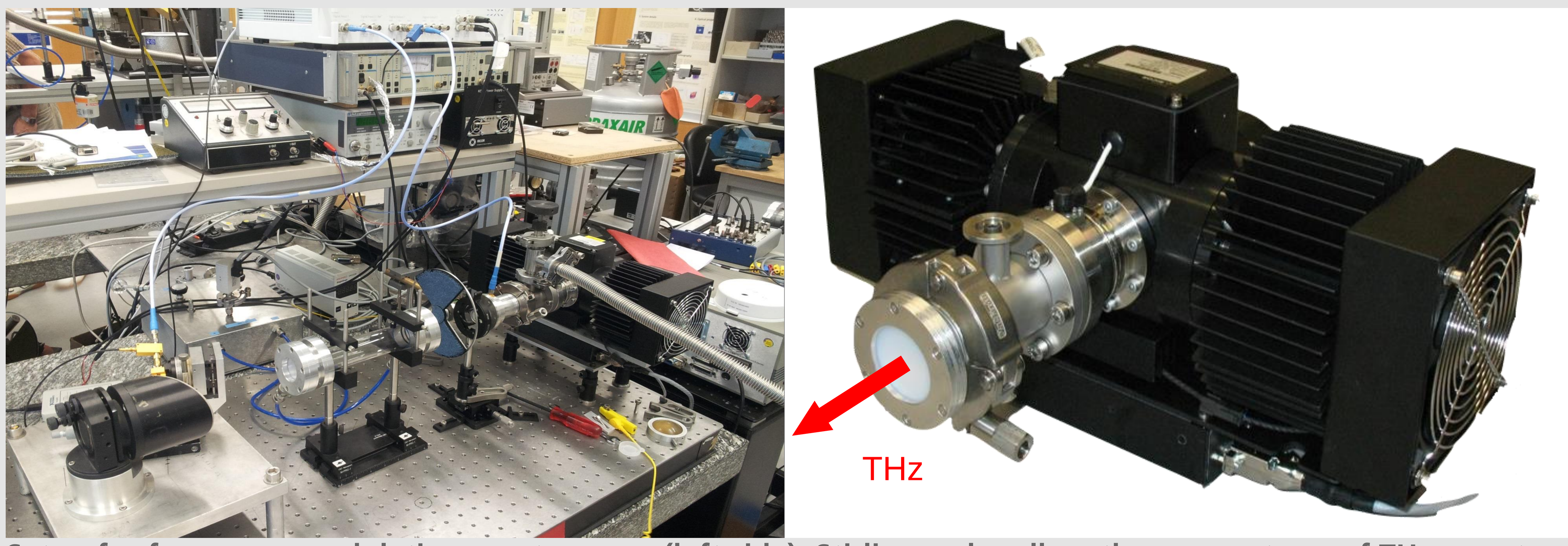


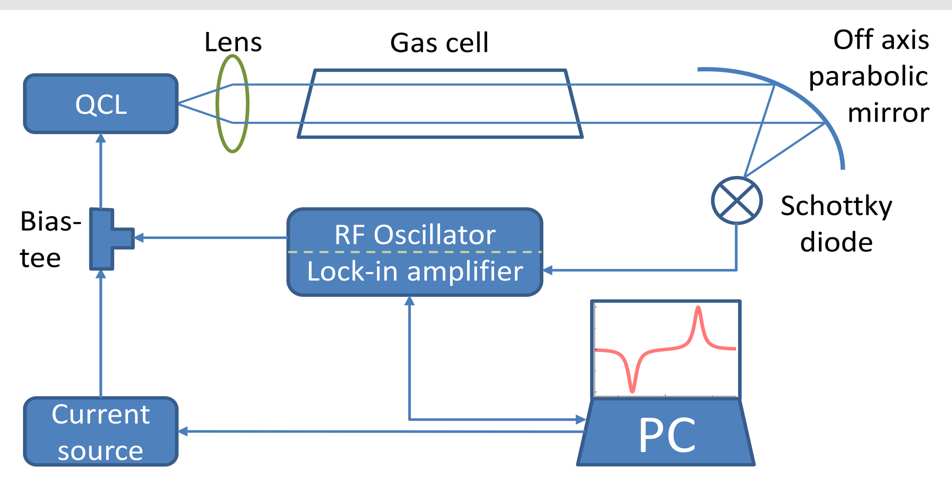
Setup for frequency modulation spectroscopy (left side). Stirlingcooler allow the compact use of THz-quantum cascade laser at cryogenic temperatures down to 35 K (right side).

Motivation

Many physical phenomena have characteristic energies, which correspond to terahertz (THz) frequencies. For example, high-resolution spectroscopy allows for the investigation of the structure and the energy levels of molecules and atoms. THz quantum-cascade lasers (QCLs) are promising radiation sources for such a type of spectroscopy, because they are frequency tunable, and they exhibit mW output powers as well as a narrow line width. So far, absorption spectroscopy with QCLs employed modulation of the QCL frequency on the order of kHz and phase-sensitive detection (1).

We describe a spectrometer based on a QCL using frequency modulation (FM) spectroscopy with frequencies up to 50 MHz. The condition for FM spectroscopy is achieved when the modulation frequency is large compared to a characteristic width of the spectral feature of interest and only one sideband probes the spectral feature. Potentially, the method is very sensitive, because the modulation frequency is well above the frequencies of the most important noise sources (2).

Setup



The QCL used emits at 3.1 The and has a 1.16 mm long Fabry-Pérot cavity and a single plasmon waveguide (3). The QCL is mounted in a compact air cooled Stirling cooler (Ricor model K535). The emission spectrum of the QCL contains several modes with a spacing of about 26 GHz as measured with a Fourier transform spectrometer (Bruker model Vertex 80V). The output power of the QCL was 1 mW at a current 550 mA and a temperature of 45 K. The beam was collimated with a lens made from polymethylpentene (TPX©) and guide through an absorption cell. An off-axis parabolic mirror was used to focus the radiation onto the Schottky diode. The absorption cell was 31 cm long and has 1 mm thick windows made from HDPE. The pressure inside the cell was measured with a capacitive manometer. Fine tuning of the QCL frequency was achieved by tuning the driving current or the heat sink temperature. A maximum tuning range of approximately 3 GHz was obtained for each of the laser modes.

The driving current of the QCL was superimposed with a small sinusoidal current with a modulation frequency ω_m up to 50 MHz using a bias-tee. The modulation current was generated by a Zurich Instruments Lock-in amplifier model HF2LI (LIA). We used modulation amplitudes up to 4 mA corresponding to a frequency modulation of up to 32 MHz, which is nearly equal to the FWHM of the CH_3OH absorption line at a pressure of 1 hPa. The signal from the Schottky diode was detected with the same LIA.

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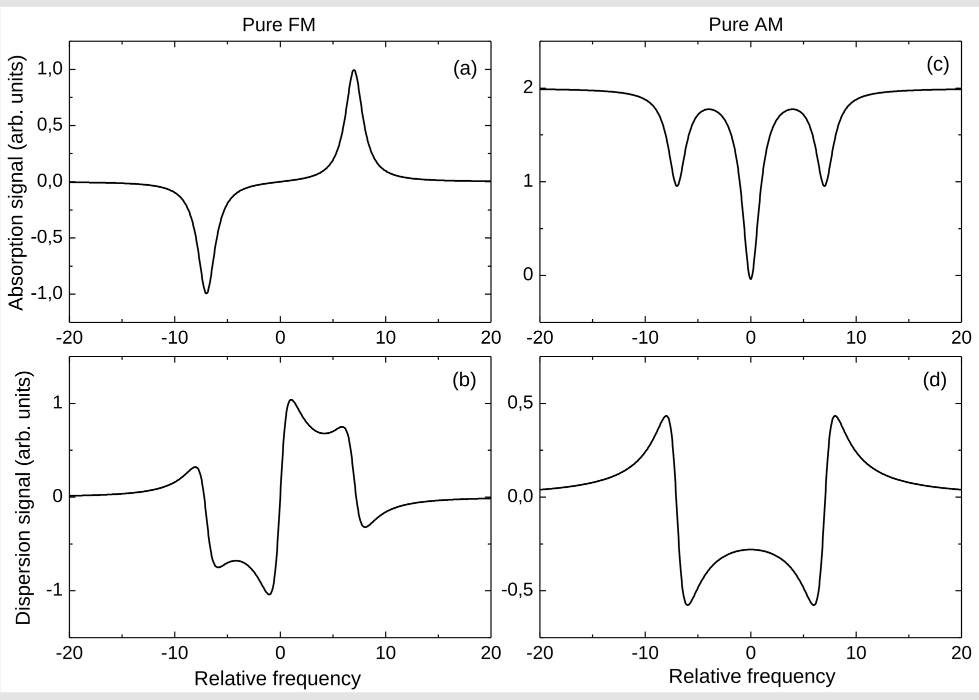
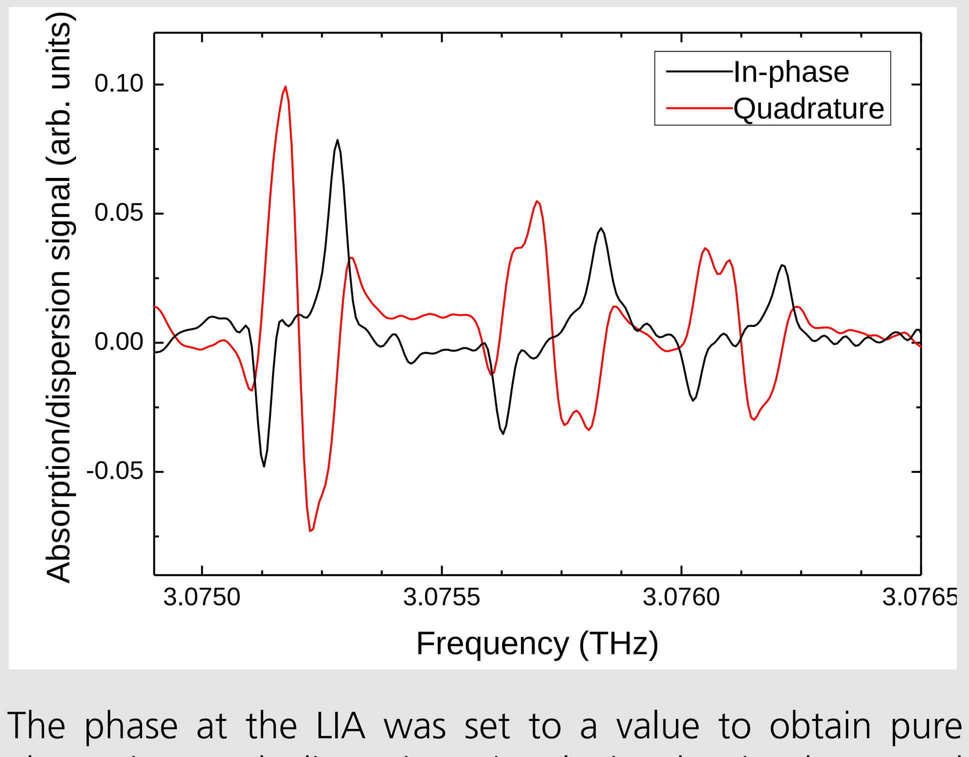
Theoretical background

The current modulation of the QCL causes not only FM, but also amplitude modulation (AM) of the laser emission. Fig. 2 shows the calculated in-phase and quadrature signals obtained at the output of the LIA for pure FM and pure AM. They were calculated for a modulation frequency of 50 MHz and an absorption profile, which is described by a Lorentz function with a full width at half maximum (FWHM) of 14 MHz (graph below, (a) In-phase and (b) quadrature for pure FM. (c) In-phase and (d) quadrature signal for pure AM).

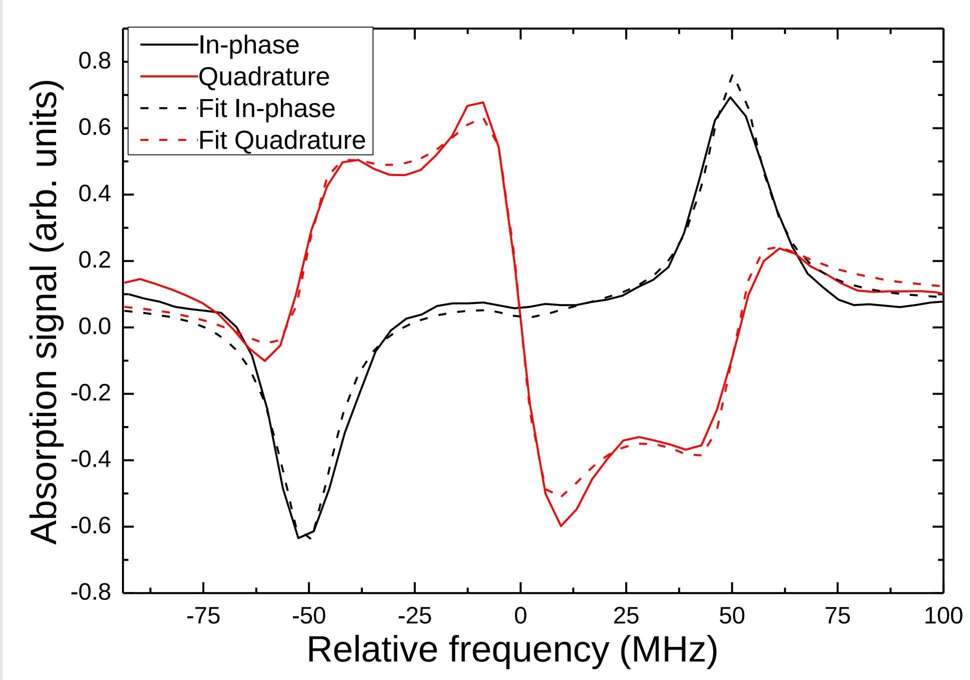


Experimental results

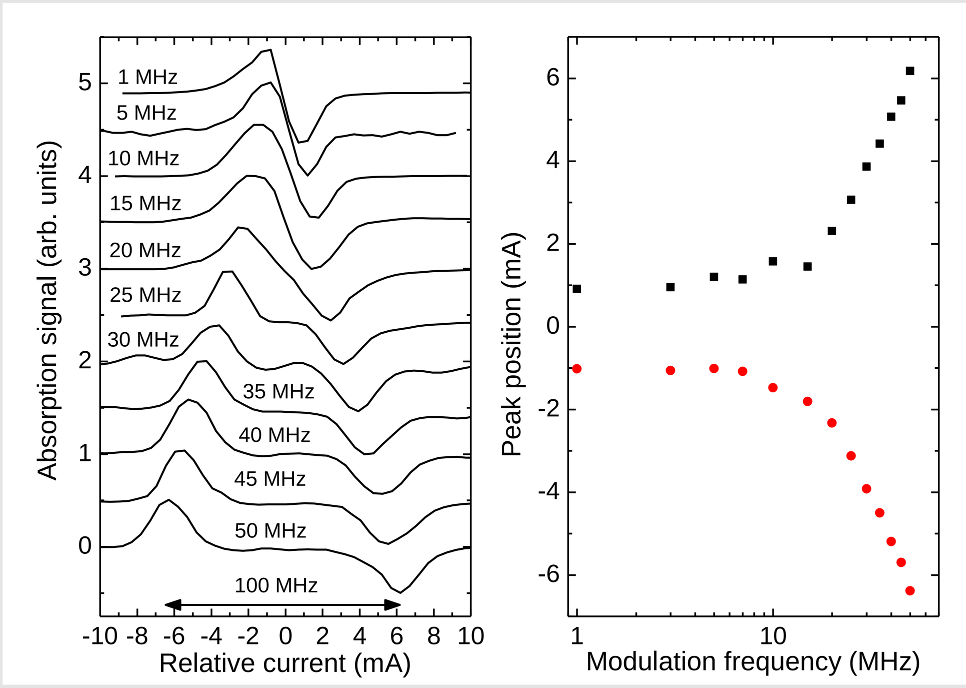
An example of the spectrum measured with FM spectroscopy at a modulation frequency of 50 MHz, with the corresponding in-phase and quadrature signals is shown in the graph below. A 1.5 GHz wide section with three absorption lines is visible. The spectrum was measured by tuning the QCL current in steps of 0.5 mA, which corresponds to approximately 6 MHz. Frequency calibration was done by comparison with a CH_3OH spectrum.



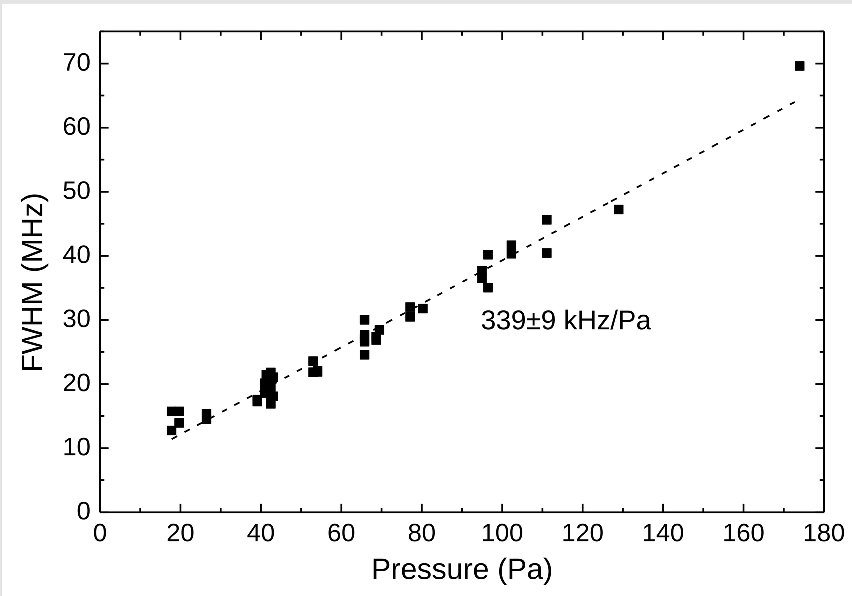
The phase at the LIA was set to a value to obtain pure absorption and dispersion signals in the in-phase and quadrature channels, respectively, of the LIA.



A single absorption line of CH_3OH measured at a pressure of 1hPa with the corresponding fit of the theory. The current amplitude was 1.6 mA, which translates into 15 MHz frequency modulation. The phase of the LIA was adjusted in such way that the absorption and the dispersion contributions to the FM signal appear purely in the in-phase and quadrature signals, respectively. The features for the in-phase measurement are separated by 100 MHz, which corresponds to $2\omega_m$.



The line width of the molecular transition limits the FM spectroscopy. The spectral line shapes were measured for different modulation frequencies ranging from 1 MHz up to 50 MHz. Almost no change of the peak positions occurs in the lower frequency range up to 10 MHz. Once the modulation frequency is significantly larger than the FWHM of the line, the separation of the two features approaches $2\omega_m$.



FM spectroscopy is also capable for the determination of the pressure broadening. The line width of CH_3OH was measured as a function in the pressure range from 20 to 180 Pa with a modulation frequency of 50 MHz. The pressure broadening was measured with an average value of 339±9 kHz/Pa. The Doppler width was determined with 5.6±0.6 MHz, compared to an expected value of 6.7 MHz.

Conclusion

This is the first demonstration of a THz absorption spectrometer based on a QCL and frequency modulation. It enables high-resolution molecular spectroscopy with simultaneous determination of absorption and dispersion.

Acknowledgments

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[3] M. Wienold et al.,"Low-voltage terahertz quantum-cascade lasers based on LO-phonon-assisted interminiband transitions," Electron. Lett. (45), 1030 -1031 (2009).

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