Frequency modulation spectroscopy with a terahertz quantum-cascade laser

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Many physical phenomena have characteristic energies in the terahertz (THz) frequency range. 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. We describe a spectrometer based on a QCL using frequency modulation (FM) spectroscopy with frequencies up to 50 MHz. This type of spectroscopy is an alternative to wavelength modulation techniques in order to investigate the dispersion of molecular transitions. 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. The QCL used in these experiments has a single-plasmon waveguide and a Fabry-Pérot cavity with both facets uncoated and is optimized for low electrical pump power. It operates on several modes centered at 3.1 THz. Frequency tuning was achieved by varying the driving current of the QCL. The beam of the QCL is collimated with a TPX lens and guided through a 30 cm long absorption cell, which is filled with CH_3OH at a pressure of 100 Pa, and focused onto a Schottky diode. FM is achieved by superimposing an AC current of about 1−2 mA with a frequency up to 50 MHz onto the QCL driving current. The AC amplitude results in a modulation of the QCL emission frequency with a magnitude corresponding to the full width at half maximum of an absorption line of CH_3OH, which is approximately 16 MHz at a pressure of 100 Pa. The absorption is measured as a function of the laser driving current using a lock-in amplifier with a bandwidth of 50 MHz.