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THz gain measurements in optically pumped ammonia

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Abstract—We present experimental results about terahertz gain measurements in ammonia gas optically pumped by a quantum cascade laser. The gain was measured with the help of a vector network analyzer (VNA) and reached 17 dB/m at 1.07 THz.

I. INTRODUCTION

HE generation and detection of THz radiation remains a L challenge despite the maturity of THz technologies and their applications in radio-astronomy, biomedicine, communications, and the defense/aerospace industries [1]. Few practical sources of THz radiation exist even after extensive research in the area for many years. As classical optically pumped far-infrared (IR) lasers (OPFIR) are bulky and nonportable, and difference frequency photomixers produce broad linewidths and low output power, vacuum electronic devices, and multiplier chain sources struggle to reach frequencies above 1 THz. Despite recent advances, THz quantum cascade lasers, can operate only in pulsed mode using thermoelectric coolers and require cryogenic cooling for continuous wave (CW) operation. New possibilities are offered by exploiting the newly demonstrated THz molecular laser optically pumped by a mid-infrared (MIR) quantum cascade laser (OCL) based on ammonia (NH₃) [2,3]. NH₃ has several advantages for optical pumping: strong static dipolar moment, small molecular mass, high vapor pressure and strong absorption near 10 µm. By identifying the MIR/THz transitions pairs we have recently shown that optical pumping of NH₃ will potentially generate a huge quantity of new THz frequencies [4].

II. METHOD AND EXPERIMENTAL RESULTS

In order to design a laser, experimental values of the gain for each transition are needed even if the gain factor can be estimated using databases [4]. Gain values were previously measured for two transitions [5] and showed that the gains were the highest measured in a continuous wave pumped gas. Here we report new gain measurements for several pure-inversion transitions of NH₃. The experimental setup shown in Figure 1 is based on a 750–1100 GHz vector network analyzer (VNA). The two WR1 rectangular waveguide heads are fitted with horn antennas. A 46-cm long copper tube is used as a gas cell for NH₃. To avoid any parasitic reflection, a THz and MIR transparent high resistivity silicon window tilted at the Brewster angle is placed at both ends of the cell. The THz beam from the VNA is superposed to the MIR beam of a QCL tunable from 10.0 to 10.6 µm. The gain value is obtained by calculating the ratio between the THz transmission measured with and without the gas inside the cell.



Fig. 1.. Schematic of the gain measurement experimental setup

Figure 2 shows a typical experimental result for the strongest transition: the pure inversion transition with J=3 and K=3. The VNA allows to measure almost simultaneously the transmission coefficient in both direction: from port 1 to port 2 (S_{21} parameter) and from port 2 to port 1 (S_{12} parameter). A maximum gain of about 8 dB is measured for a MIR pump power of 23 mW and a pressure of 16 µbar. It corresponds to a lineic gain of 17 dB/m which is, to our knowledge, the highest value measured in a CW pumped gas in the terahertz range. The gain peak is measured at the same frequency for both direction of propagation showing that the QCL tuned was tuned very close to the center of the MIR absorption line.



Fig. 2. THz gain measured for the (J=3, K=3) pure inversion transition of NH_3 pumped at 967.346 cm⁻¹ (the spectrum is offset by 1073049.9 MHz). Pressure: 16 µbar.



Fig. 3. THz gain measured for the (J=3, K=1) pure inversion transition of NH_3 pumped at 967.449 cm⁻¹ (the spectrum is offset by 1014084.8 MHz). Pressure: 20 µbar.

The linewidth of the QCL is lower than the Doppler linewidth of the NH₃ transitions, then molecules with a particular class of radial velocity are pumped. When the QCL is detuned from the center of the line, the gain peak is shifted in two opposite directions for S_{21} and S_{12} due to the fact that the sign of the Doppler effect is reversed.

Figure 3 shows the weakest line we have measured (0.09 dB at a pressure of 20 μ bar, and this for J=3, K=1 transition) corresponding to a lineic gain of 0.2 dB/m. Measurement of the gain of other transitions will be shown at the conference.



Fig. 4. Measured gains (in m^{-1}) as a function of the calculated molecular gain factor G_m for six pairs of pump/probe transitions. The red line is the fit and the black dots are the measurements.

Figure 4 shows a summary of the measurement of the gain of six pure inversion transitions of NH₃ for the same pump power density P_p and pressure (20 µbar) versus the molecular gain factor G_m defined in [4] and calculated using the HITRAN database. The red line shows a fit considering a relaxation rate Γ of 2.5x10⁻⁵ s⁻¹. This value is compatible with the molecule-molecule and molecule-wall collision rate.

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