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High-resolution THz gain measurements in optically pumped ammonia

Martin Mičica,[1,2,3,*] Sophie Eliet,[2] Mathias Vanwolleghem,[2,3] Roman Motiyenko,[4] Anastasia Pienkina,[4] Laurent Margulès,[4] Kamil Postava,[1,3] Jaromír Pištora,[1,3] and Jean-François Lampin[2]

[1] *Nanotechnology Centre, VŠB - Technical University of Ostrava, 17. Listopadu 15/2172, 708 33 Ostrava-Poruba, Czech Republic*
[2] *Institut d'Électronique de Microélectronique et de Nanotechnologie, UMR 8520 CNRS, Cité Scientifique, Avenue Henri Poincaré CS 60069, 59 652 Villeneuve d'Ascq Cedex, France*
[3] *IT4Innovations, VŠB -Technical University of Ostrava, 17. Listopadu 15/2172, 708 33 Ostrava-Poruba Czech Republic*
[4] *Université de Lille, CNRS, UMR 8523 - PhLAM - Physique des Lasers, Atomes et Molécules, F-59000 Lille, France*
[*] *martin.micica@vsb.cz*

Abstract: This study is aimed at the evaluation of THz gain properties in an optically pumped NH_3 gas. NH_3 molecules undergo rotational-vibrational excitation by mid-infrared (MIR) optical pumping provided by a MIR quantum cascade laser (QCL) which enables precise tuning to the NH_3 infrared transition around 10.3 µm. Pure inversion transitions, ($J = 3$, $K = 3$) at 1.073 THz and ($J = 4$, $K = 4$) at 1.083 THz were selected. The THz measurements were performed using a THz frequency multiplier chain. The results show line profiles with and without optical pumping at different NH_3 pressures, and with different MIR tuning. The highest gain at room temperature under the best conditions obtained during single pass on the (3,3) line was 10.1 dB×m^{-1} at 26 µbar with a pumping power of 40 mW. The (4,4) line showed lower gain of 6.4 dB×m^{-1} at 34 µbar with a pumping power of 62 mW. To our knowledge these THz gains are the highest measured in a continuous-wave MIR pumped gas.

OCIS codes: (300.6495) Spectroscopy, terahertz; (140.4130) Molecular gas lasers.

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1. Introduction

Terahertz photonics is one of the newer fields in electromagnetic research. The terahertz band lies between microwave and infrared frequencies thanks to which it possesses unique properties. It has promising applications in spectroscopy, nondestructive testing, wireless communications, imaging, detection of hazardous substances and much more [1, 2]. Despite some progress, THz photonics lacks a reliable, compact and powerful THz source. Thermal THz sources can be used in applications where incoherent higher THz frequencies are sufficient (Fourier Transform Infrared Spectroscopy), however, their power is very low and therefore they require cryogenic detectors. Optoelectronics [3] and electronic harmonic generation can be used to synthesize lower THz frequencies, but have a limited efficiency close to 1 THz and above. Another method uses nonlinear crystals (parametric effect, difference frequency generation) but generally needs very high power continuous or pulsed lasers. Direct THz generation - lasers, can be divided in two categories: optically pumped molecular lasers (OPMLs) and semiconductor lasers (terahertz quantum cascade lasers and p-Ge laser). Terahertz quantum cascade lasers (THz QCLs) are relatively new sources demonstrated in 2002 [4]. They can be small with reasonable output power and some tunability, however they suffer from decreasing power with temperature and still require cryogenic cooling [5]. THz OPMLs were demonstrated in 1970 and the original design can still be found in commercial THz lasers [6, 7]. They are powerful but they are bulky, not efficient and have limited tunability. Optical pumping in the mid infrared (MIR) region can be seen as a drawback if a CO_2 (or N_2O) discharge laser is used. The tunability of these kind of lasers is also limited due to the required coincidence of the line provided by the pumping laser with the MIR absorption line of the THz active molecular gas. This condition limits also drastically the choice of the molecule and of the transition. Thanks to the development of new MIR quantum cascade lasers (MIR QCLs) we may overcome some disadvantages of the CO_2 pump laser. MIR QCLs are small, have a high efficiency and can be precisely tuned which allows to excite lines of molecular gases that cannot be accessed by a conventional CO_2 pump. In addition, commercially available MIR QCLs are operated around room temperature.

A terahertz molecular laser optically pumped by a MIR QCL based on ammonia (NH_3) was demonstrated recently [8] and offers many new possibilities. It is essential to know the most important parameters [9] and the behavior of the lasing lines if one wants to optimize the design of a laser. In this paper a study of the THz gain properties of two selected optically-pumped NH_3 lasing lines near 1 THz at room temperature is presented. Compared to our previous results [10, 11] important information about the gain, the lineshape and the pressure dependence are obtained.

2. Theoretical background

Ammonia (NH_3) is a well known molecule that was used in some lasing experiments starting with the first demonstration of stimulated emission and the construction of the first maser [12]. High dipole moment (1.4 D), high rotational constant and fast relaxation rate makes it a good choice for THz OPMLs [8]. Absorption of NH_3 in the infrared spectral range around 960 cm^{-1} is associated with the ν_2 normal modes which correspond to a symmetrical deformation of N-H interatomic bonds, also called umbrella mode. The energy levels are also split due to the tunneling of nitrogen atom through the barrier created by the three hydrogen atoms [13]. These levels are split into the lower energy symmetric (s) and the higher energy antisymmetric (a) wavefunctions (Fig. 1). NH_3 is also a symmetric top molecule whose rotational state is expressed by the quantum numbers (J, K) where J is the total rotational angular momentum and K is the projection of the angular momentum to the symmetry axis. The energy differences between the a and s levels for the vibrational ground state $\nu_2 = 0$ lie in the microwave region. The first maser was demonstrated by inversion of population between a and s levels of the (3,3) line at a frequency close to 24 GHz [12]. In the first vibrational excited state $\nu_2 = 1$ the difference between a and s increases and the splitting becomes wider with a frequency around 1 THz. The selection rules for rotational inversion transitions are $\Delta\nu = \Delta K = 0$, $\Delta J = 0, \pm 1$, $a \leftarrow s$ and $s \leftarrow a$. The selection rules for parallel vibrational transitions like ν_2 are $\Delta\nu = \pm 1$, $\Delta K = 0$, $\Delta J = 0, \pm 1$ and $a \leftarrow s$, $s \leftarrow a$ [14].

In this article we study excited molecules optically pumped using Q-branch transitions $\Delta\nu_2 = 1$, $\Delta J = \Delta K = 0$, $a \leftarrow s$ and population inversion on pure inversion transitions $\Delta J = \Delta K = 0$, $s \leftarrow a$ (Fig. 1). The molecules relax by collisions with other molecules or with cavity walls. For the experiment we selected two lines: (1) pumping at 967.3463 cm^{-1} (saQ(3,3) transition) and probing around 1073049.6 MHz ($\nu_2 = 1$ asQ(3,3)), (2) pumping at 966.8147 cm^{-1} (saQ(4,4) transition) and probing around 1082592.4 MHz ($\nu_2 = 1$ asQ(4,4)) [15].

3. Experiment

The experimental setup is shown in Fig. 2. The THz probe beam was generated by a frequency multiplier chain (Virginia Diodes, Inc.) equipped with WR-0.65x3x3 [16] stages (9 times frequency increase). The input signal for the multiplier was synthesized by a high precision microwave generator (Agilent) with an amplitude modulation used for a lock-in detection. An InSb hot electron bolometer (QMC Instruments Ltd.) cooled to 4 K was employed for the detection of the THz radiation. The NH_3 gas flowed through a gas cell made from a 50 cm long copper tube with 10 mm inner diameter. The dimensions of the tube were chosen based on a compromise between gain, mode nonuniformity and waveguiding effect [17]. The gas cell was terminated by THz and MIR transparent high resistivity Si window inclined at the Brewster angle in order to avoid any reflection in the cell. The cell was tested using a helium leak detector with no leaks

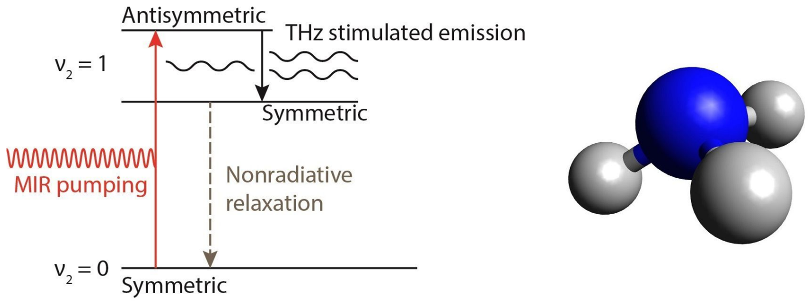

Fig. 1. The lasing process of NH_3 on an energy diagram (left). 3D view of the NH_3 molecule (right).

detected. The NH_3 gas was injected at constant flow from a cylinder through a needle valve, which was used to control the desired pressure, into the gas cell and pumped by a turbomolecular pump. A small flow of NH_3 during measurement prevents from a cell walls contamination and helps to stabilise the pressure. The temperature of NH_3 was approximately 20°C. MIR optical pumping was provided by a distributed feedback MIR quantum cascade laser (QCL) with a central wavelength around 10.3 µm (AdTech) and a power up to 100 mW. A polymer lens (Zeonex) was used to focus the THz beam and a 200 mm focal length ZnSe lens was used to focus the MIR beam into the gas cell. The THz beam was collected by gold parabolic mirrors and focused into the bolometer. A step attenuator (Lasnix) was used to reduce the power of the MIR beam when needed. A semi-insulating SiC wafer which reflects MIR and is transparent for the THz beam was used as a beam combiner. Polarization (p) and propagation directions of the THz and MIR beams entering the gas cell were the same. The spot size of the MIR beam at the entrance of the cell was approximately 2 mm diverging up to 10 mm at the end of the cell.

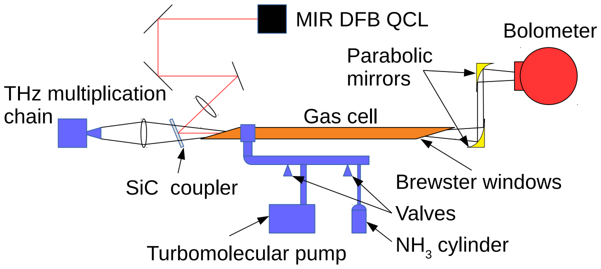

Fig. 2. Schematic of the experimental setup.

4. Results

First the THz spectra of the two lines were recorded without MIR pumping (QCL off). The NH_3 pressure was stabilized and the multiplication chain was swept with a frequency step of 108 kHz over a sufficient range around the line center. Figure 3 shows profiles of the NH_3 (3,3) (left) and (4,4) (right) lines at different pressures. The output power of the multiplication chain measured by a pyroelectric detector was 3.2 µW at 1.073 THz and 6.8 µW at 1.082 THz. The Gaussian shape of the absorption lines at such low pressure is caused dominantly by Doppler broadening [18]. The line center is slightly shifted from its theoretical value with increasing pressure by the collisional shift [19].

Figure 4 shows (3,3) (left) and (4,4) (right) lines with optical pumping by the MIR QCL at

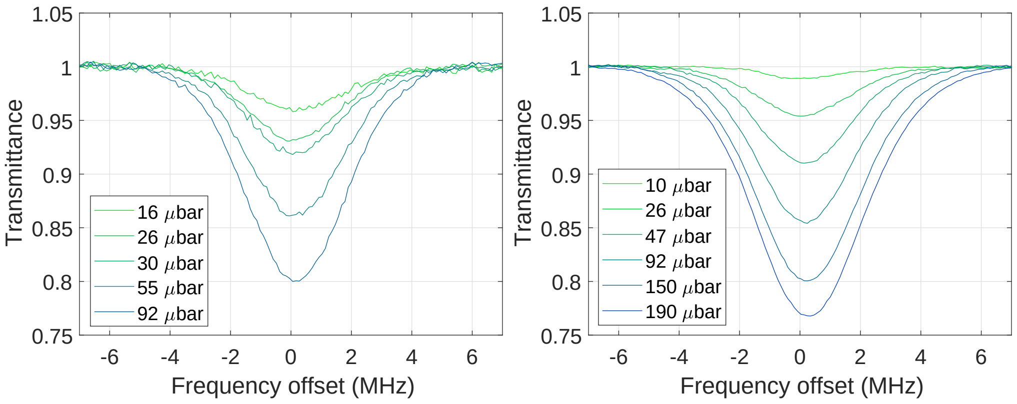

Fig. 3. THz absorptions of NH_3 (3,3) (left) and (4,4) (right) at different pressures, offset from the theoretical line center: 1073049.6 MHz for (3,3) and 1082592.4 MHz for (4,4).

a NH_3 optimal pressure of 26 μbar and 34 μbar respectively (for determination of the optimal pressure, see Fig. 5 right). The lasing linewidth of the MIR QCL is narrower than the Doppler broadening of the MIR transitions of NH_3.

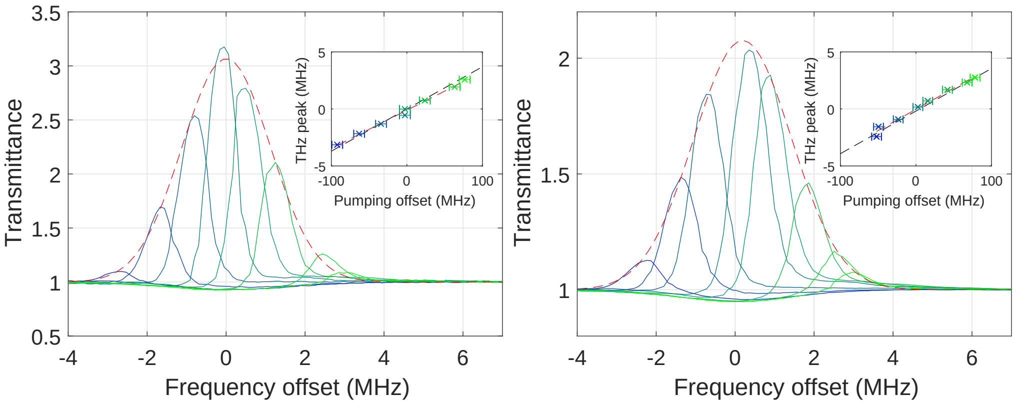

Fig. 4. Transmittance profiles of the (3,3) (left) and (4,4) (right) NH_3 lines versus THz probe frequency (offset by the line center frequency) at optimal pressures 26 μbar and 34 μbar (see Fig. 5 right), with pump power 40 mW and 64 mW respectively, at eight pump frequency settings. Inserts show the THz peak position (offset by the line center frequency) as a function of the MIR pumping frequency (offset by the line center frequency). Red dashed lines show estimated lineshapes of fully excited transitions.

The QCL pumps only a category of molecules with a particular velocity. The position of the THz gain peak then depends on the MIR pumping frequency and as a result the THz gain bandwidth is narrower than the Doppler linewidth. The maximum gain is obtained at the center of the THz lines when the QCL is tuned precisely to the center of the MIR transitions. The tuning curves (inserts in Fig. 4) of the THz peak gain frequency vs. MIR frequency have a linear trend with a slope equal to the ratio of the THz and the MIR transitions (27 for (3,3) and 26.8 for (4,4)). The power of the MIR beam measured by a thermopile at the exit of the evacuated cell was close to 40 mW for the (3,3) line and 62 mW for the (4,4) line with an estimated error of 5 %. The absorption of the pump power at 26 μbar in the 50 cm long cell was 53% for the (3,3) line and 26% for the (4,4) at 34 μbar. The linewidth of the MIR QCL is generally broadened by small current [20] and temperature fluctuations. Here, it is estimated to be 35 MHz and can be further improved by various methods [21].

Figure 5 (left) shows the transmittance as a function of the MIR pumping power. The transparency when the stimulated emission is equal to the absorption is about 1 mW for both lines. The gain dependence on the pump power is linear up to approximately 13 mW where probably depletion of the ground states starts, probably due to the slow vibrational relaxation of $\nu_2 = 1(s)$ states after stimulated emission [22]. The slope of the linear part of the gain for the (3,3) line is 0.08 mW^{-1} and 0.05 mW^{-1} for the (4,4) line. The effectiveness may be increased by adding different gas to help with the relaxation to the ground state [23].

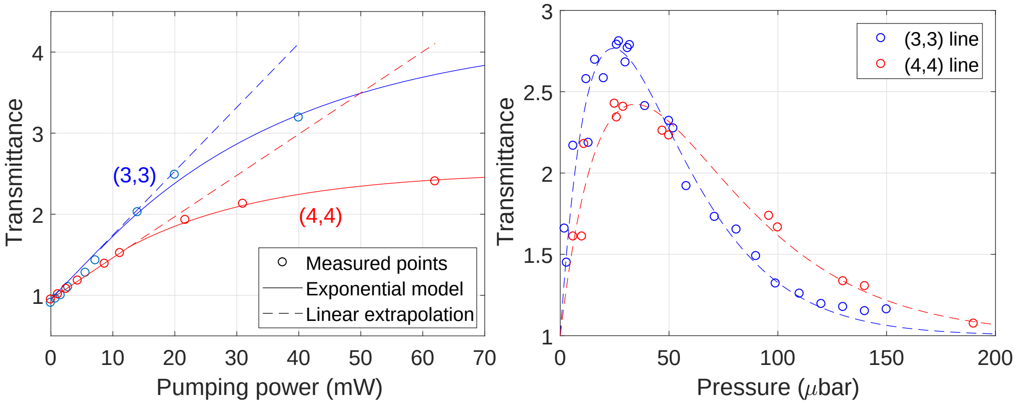

Fig. 5. Transmittance as a function of the MIR pumping power at the optimal pressure 26 µbar and 34 µbar (left) and the pressure with the constant pumping power 40 mW and 64 mW (right) for lines (3,3) and (4,4). Dashed and continuous lines are just guides for the eye.

The optimal NH_3 pressure is an important parameter that heavily affects gas laser performance [24]. Figure 5 (right) shows the THz gain as a function of the pressure. The gain increases linearly with the pressure due to the higher number of molecules in the volume up to a certain point where collisions between molecules and/or cavity walls cause increased nonradiative relaxation [22] and exponentially decrease of population inversion. The optimal pressure is between 20 – 30 µbar and 25 – 45 µbar for (3,3) and (4,4) line respectively.

The final NH_3 gain and pump absorption per unit length were calculated from the Beer-Lambert law for the single pass through the active medium [25]: $I(d) = I_0 \times \exp(\alpha d)$, where I_0 and $I(d)$ are the intensities at the input and at the output distance d (cell length), and α represents the absorption coefficient for negative values and gain coefficient for positive values. The maximum gain during the experiment for the (3,3) line at the best conditions (26 µbar, 40 mW pump power, 50 cm long path) was 3.2 which equals 2.3 m^{-1} or 10.1 $\mathrm{dB}\times\mathrm{m}^{-1}$. The maximum gain for the (4,4) line at the best conditions (34 µbar, 62 mW pump power, 50 cm long path) was 2.1 which equals 1.4 m^{-1} or 6.4 $\mathrm{dB}\times\mathrm{m}^{-1}$. A signal to noise ratio of the gain measurement was estimated to be higher than 20 dB. These values are to our knowledge the highest measured in a continuous wave MIR pumped gas in the THz range [9, 26, 27].

5. Conclusion

The THz optically pumped molecular lasers (THz OPMLs) are widely applied as sources of coherent THz radiation. Progress in tunable MIR quantum cascade lasers (QCLs) allows the replacement of normally used CO_2 laser MIR pump, which gives us the opportunity to excite transitions that were not accessible due to the required coincidence with CO_2 lines. The precise tuning of the MIR QCL allows a selective pumping and gives access to a number of new THz lasing lines in various molecular gases. NH_3 based THz OPML pumped by MIR QCL was successfully demonstrated [8] and opened up new possibilities in the field of THz lasers. In this article we presented measurements of important parameters of optically pumped NH_3. During optical pumping molecules undergo roto-vibrational inversion excitation to the vibrational level $v_2 = 1$ followed by stimulated emission on pure inversion transitions and nonradiative relaxation to the ground vibrational level. Two lines were selected at 1.073 and 1.083 THz. Measurements were performed at different pumping power, frequency and pressure values. Measured gains up to 10.1 $\text{dB}\times\text{m}^{-1}$ and up to 6.4 $\text{dB}\times\text{m}^{-1}$ were obtained during one-way copropagation for the (3,3) and (4,4) lines respectively. We attribute the high gain measured to the strong inversion transition chosen and to the fact that the QCL allows perfect resonant pumping. It is possible to achieve even higher gain by improving conditions, e. g. matching the THz probing beam with a preferred waveguiding mode of the cell [10, 17]. Obtained parameters will help a better understanding of the lasing gas behavior and may serve as an information for improving performance of the existing NH_3 THz OPML.

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Disclosures

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