Spectroscopic measurements of CH_3OH pumped by cw CO_2 laser

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1. Introduction

The development of the far-infrared (FIR) region was greatly hampered in the past by the lack of good sources with extensive spectral coverage. This kind of an obstacle is now rapidly disappearing due to the development of optically pumped FIR lasers. In general, pumping occurs through the absorption of vibrational energy supplied by a tuned pump laser, and laser action occurs by one or more pure rotational transitions in an excited vibrational state. The attractiveness of optical pumping is that it is a specific, intense, nondestructive and universal method [1]. The performance of FIR lasers is based on an overlap of available pump lines. Therefore, for an effective pumping the molecules excited by the infrared radiation must possess an absorption line the frequency of which closely coincides with that of the laser emission line.
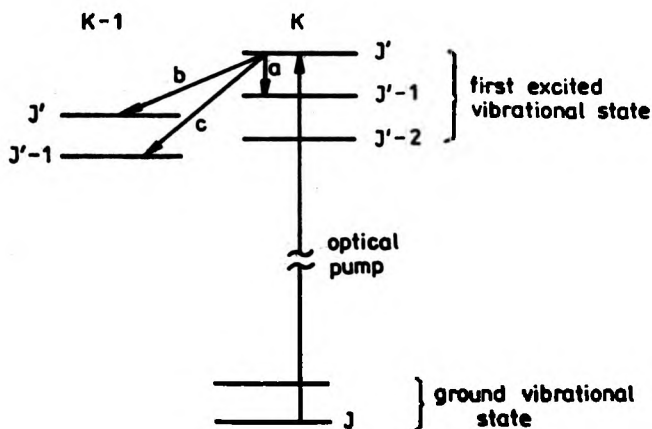

Fig. 1. Model of optical pumping in CH_3OH molecule. J, K – quantum numbers representing the total angular momentum and its projection on the CH_3 symmetry axis, respectively

A good choice for an optical pump is the CO_2 laser which has many lasing lines around 10.4 μm and 9.4 μm. The next problem is to select a polyatomic molecule having one of its fundamental vibrational states close to this spectral region.

Methanol (CO_3OH) was one of the first molecules to be used for FIR generation by optical pumping [2]. The CO_3OH molecule is a weakly asymmetric top with hindered internal rotation, and its success in optical pumping is due to the fact that the first excited C–O stretch state is centred at 1033.9 cm^{-1} [3], i.e., just in the middle of the CO_2 laser spectrum. In such a case there is a number of coincidences close enough to transfer the methanol molecules by resonant absorption from a level J in the ground state to the J' one in the excited vibrational state, as indicated in Fig. 1. This absorption may create population inversion between the pumped level and the levels below. The stimulated emission may occur on the transitions, denoted by a, b and c in Fig. 1.

This paper reports the measurements of the overlapping degree of CO_2 emission line and CH_3OH absorption line.

2. Experimental set-up

Figure 2 shows the set-up used in all measurements performed in this work The cw CO_2 laser used in the spectroscopic measurements was equipped with a two-section pyrex discharge tube 120 cm long and 1.4 cm in diameter. The water cooled discharge tube was closed with NaCl Brewster-angle windows
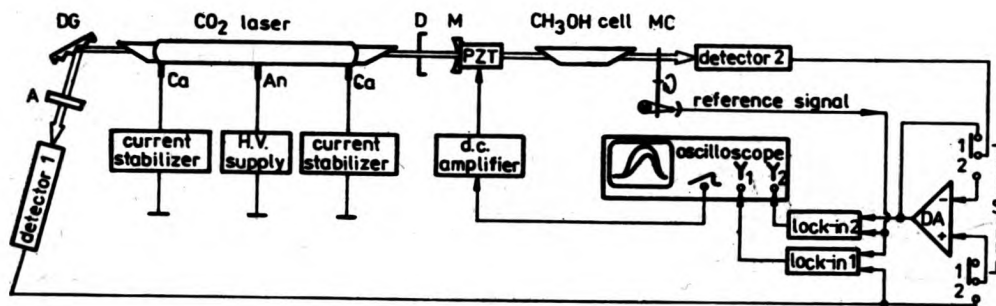

Fig. 2. Experimental set-up. A–attenuator, DG–diffraction grating, D–diaphragm, M–output mirror, MC–mechanical chopper, S–change-over swich, DA–differential amplifier. All measurements were carried out with a synchroneous detection method

The gas mixture used consisted of $CO_2 : N_2 : He = 1 : 1 : 4$ under an optimal total pressure of 13 Torr. The discharge tube was operated with non-flowing gas at a stabilized discharge current of about 10 mA for each section. The laser cavity was equipped with a plane reflecting diffraction grating DG (150 lines per mm), used to select laser lines in 10.4 µm and 9.4 µm bands for both P and R branches. On the opposite side of the grating, the cavity was terminated by the concave (R = 10 m) totally reflecting gold-coated mirror M. The laser beam was coupled out through a 3.5 mm hole in the centre of the mirror

[4]. The position of the output mirror could be adjusted by means of a piezo-electric transducer PZT [5] supplied with a saw-tooth signal from an oscilloscope. The optical resonator length was 187 cm which gives an 80 MHz frequence distance between two adjacent axial modes. The laser operated in TEM_{00} transverse mode. Higher order transverse modes were compressed by means of a diaphragm D. The absorption cell used in the experiment was 60 cm in length and 2.5 cm in diameter. Both ends of the cell were terminated with NaCl Brewster-angle windows. The absorption cell was pumped out to 0.5×10^{-6} Torr pressure and next filled with CH_3OH vapour. The profile of the laser gain curve was monitored with detector 1 from the grating side. The CO_2 emission line deformed by CH_3OH absorption line was monitored with detector 2. Both profiles could be simultaneously observed by means of a two-channel oscilloscope (the change-over switch S in position 1). Non-cooled CdHgTe detector was used in the experiment [6]. All measurements were performed with a synchronous detection method in order to avoid thermal drifts of the detectors.

The presented experimental set-up gave also the possibility of a simultaneous observation of the CO_2 emission line and the CH_3OH absorption line (the change-over switch S in position 2). The absorption line could be obtained by subtracting the deformed profile of CO_2 emission line (detector 2) from the pure CO_2 emission line (detector 1) using a differential amplifier DA. This method enabled the measurement of the pump offset between the centres of CH_3OH absorption and the respective CO_2 emission lines. The voltage-supplied piezo-electric transducer was used for scaling of the resonator frequency.

3. Results

As a first step, coincidences between CO_2 pump lines and CH_3OH absorption lines were identified for 10.4 μm and 9.4 μm CO_2 laser bands. For every pre-selected CO_2 emission line the relative absorption of laser beam in CH_3OH was measured with detector 2 (see Fig. 2) when the cell was empty or filled with absorbing gas under 800 mTorr pressure. The measurements were carried out for the laser frequency tuned to the centre of gain curve using piezoelectric transducer PZT. Figure 3 shows the CO_2 laser emission spectrum and the relative absorption of laser radiation in CH_3OH as an absorber.

Then the investigations were performed on P34, P36 and P37 CO_2 emission lines of 9.4 μm band, i.e., on the mostly absorbed lines. Figure 4 shows these lines deformed because of the absorption in CH_3OH under 800 mTorr pressure. The evident deformation of pumping line was observed under pressure higher than 100 mTorr. The degree of deformation increased with the increasing pressure of the absorber. The experiment was carried out within the range of pressures applied in CH_3OH FIR laser cavity, i.e., 50–1000 mTorr [7, 8].

By subtracting the profiles shown in Fig. 4 using a differential amplifier
(see Fig. 2), the pure absorption profiles of CH_3OH spectral lines were obtained.
The example for 9P34 CO_2 emission line is shown in Fig. 5. Simultaneous re-
cording of the two profiles enabled us to measure the pump offset from the
frequency of the CO_2 line centre. The measured pump offsets were approxi-
mately $+30$ MHz, -27 MHz, and $+12$ MHz for 9P34, 9P36 and 9P38 emis-
sion lines, respectively. These values are in agreement with those reported
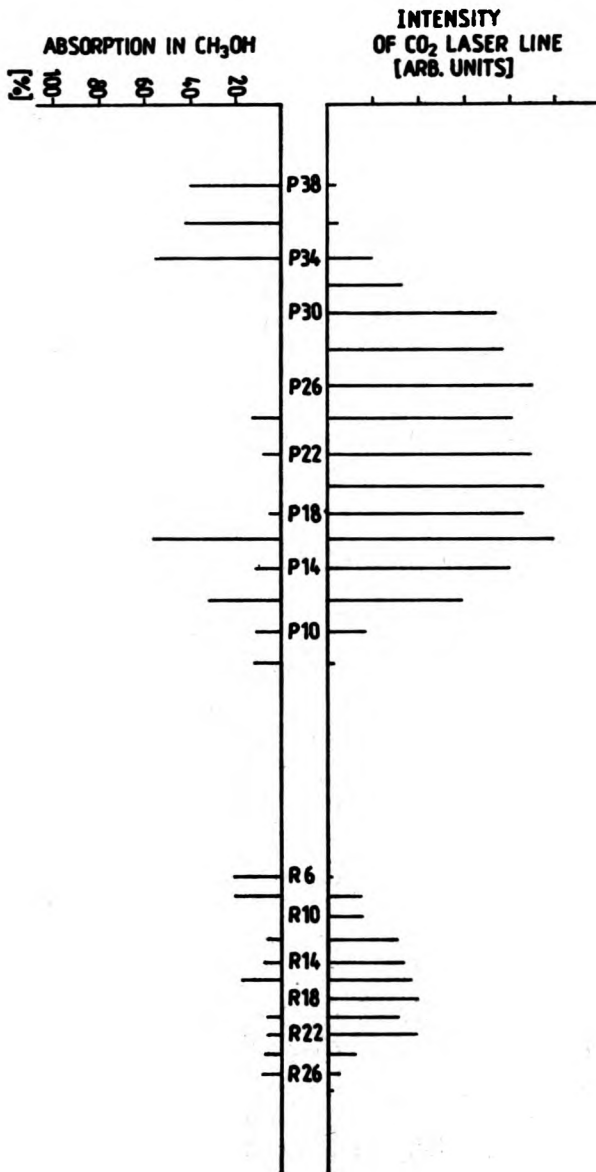in the papers of Inguscio et al. [9, 10].

Fig. 3. CO_2 laser emission spec-
trum and the relative absorption
of laser radiation in CH_3OH.
Absorption has been investiga-
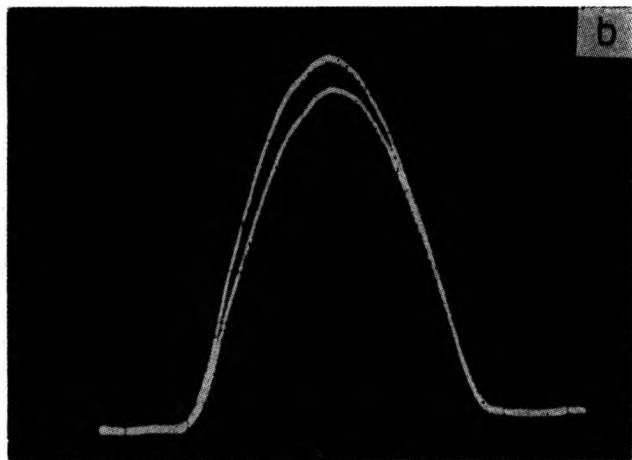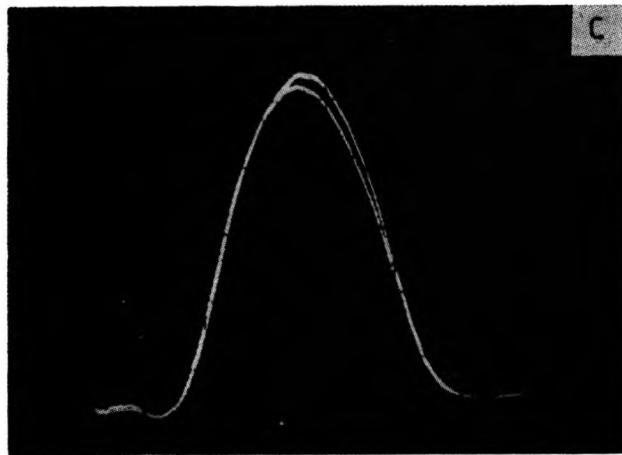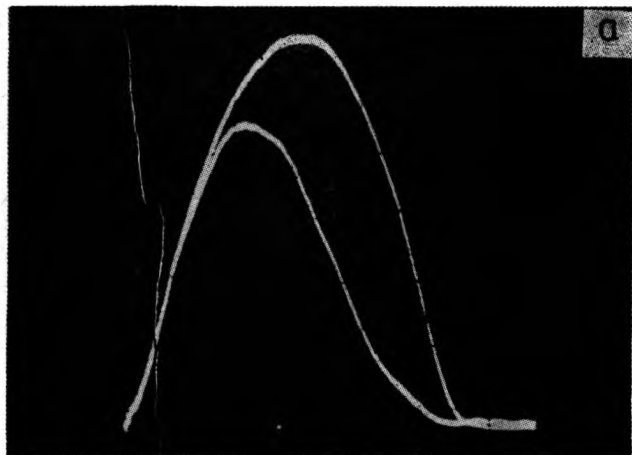ted for both P and R branches
of 9.4 μm band

Fig. 4. CO_2 laser emission lines (upper curves) and the same lines deformed because of the absorption in CH_3OH under 80 mTorr pressure (lower curves) for 9P34 (a), 9P36 (b), and 9P38 (c) lines
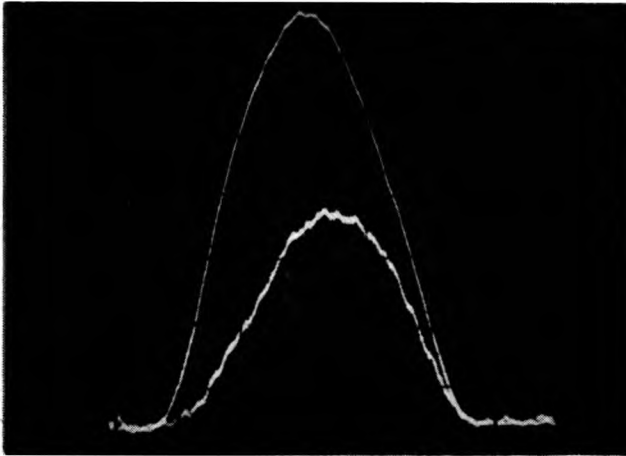
Fig. 5. 9P34 CO_2 emission line (upper curve) and CH_3OH absorption line (lower curve)

4. Conclusions

Summing up, we have demonstrated that the absorption of CO_2 laser beam in CH_3OH occurs for many emission lines of 9.4 μm band. The laser operating on these lines can be used for the optical pumping of CH_3OH FIR laser. As pointed out, usually the centre of CO_2 emission lines is shifted with respect to the centre of CH_3OH absorption lines. In general, it does not prevent the optical pumping of a FIR laser [11]. However, for the cw operation, the exact detuning of CO_2 laser frequency to the centre of CH_3OH absorption line is necessary in order to improve the pumping efficiency [12].

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