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A submillimeter CH_3OH laser with a high-power optical pumping: new emission lines

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***Abstract*. Five new emission lines in the wavelength range of 70 – 140 μm are obtained from a CH_3OH submillimeter laser pumped by high-power periodic (500 Hz) quasi-stationary 70-μs pulses from a CO_2 laser. The wavelengths (with an error of 0.5 %), the threshold pump powers, and the optimal pressure are measured. It is shown that no saturation is present at pump powers of 1000 – 3000 W.**

Keywords: *submillimeter laser, high-power optical pumping, multifunctional CO_2 laser.*

One of the main trends in the studies of optical pumping of submillimeter (SMM) lasers has always been the search for new lasing lines in active media based on many well-known substances used in optically pumped SMM lasers [1]. The works devoted to solving this problem utilise mainly the following methods: the use of isotope-substituted modifications apart from the basic molecules [2, 3]; extending the frequency range of the pump radiation [2 – 5]; improving the design of the SMM cavity [6, 7] and increasing the pump power (a pulse mode is used) [8 – 10], which allows the observation of weaker lines or lasing lines in a certain spectral region.

Note that, in these studies, the active media are usually pumped optically either in a continuous mode at a power of 40 W or by pulses with a duration of ~ 100 μs and a maximum pulse power of 100 W.

The aim of this work was to study the effects occurring upon pumping molecules by high-power laser radiation. The main attention was drawn to the search of new lasing lines in the SMM wavelength range. Freon (CH_2F_2) and methanol (CH_3OH) were chosen as the active media. Extensive studies of SMM lasers based on these media have demonstrated their high efficiency. This is explained by a good coincidence of the absorption bands of these molecules with the frequency range of the CO_2 laser lines and also by the significant dipole moment of molecules and their rich vibrational – rotational absorption spectrum [11, 12]. A repetitively pulsed mode of pumping CH_2F_2 and CH_3OH molecules was chosen on the basis of earlier results [11 – 14], but in our case the pulse pump power was increased to several kilowatts.

In the experimental setup described in our previous paper [15], the design of SMM laser (*1*) was significantly modified (Fig. 1). The SMM laser used had a 1-m-long open cavity that consisted of two gold-plated metal mirrors 40 mm in diameter. The flat input mirror had a 4-mm-diameter coupling hole; the output mirror was spherical ($R = 2.36$ m) and had a 3-mm-diameter hole for extracting SMM radiation. A 0.4-mm thick silicon plate installed in the mirror's central part had a coating with a high reflectivity for the pump radiation. The loss introduced by this coating for radiation in the SMM range was insignificant. Thus, the pump-radiation loss was lowered and the exit crystal-quartz window of the SMM laser was protected. The laser also included system (*13*) for scanning the cavity length designed for automating the wavelength-calculation procedure.

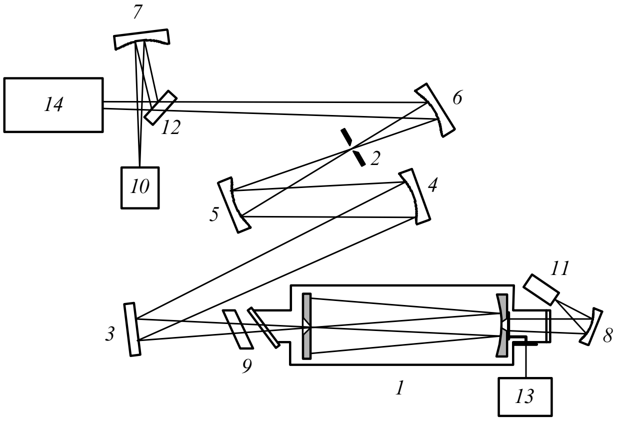

Figure 1. Scheme of the experimental setup: (*1*) submillimeter laser; (*2*) diaphragm; (*3–8*) mirrors; (*9*) attenuator; (*10*) IR detector; (*11*) SMM detector; (*12*) reflecting wedge; (*13*) system for scanning the cavity length; (*14*) CO_2 laser.

A multifunctional CO_2 laser [16] generating up to 70 lines in the 9.4 and 10.4-μm bands which could be tuned from the line centre within a range of 8 MHz, was used for pumping. The laser emitted ~ 70-μs pulses with a repetition rate of 500 Hz. The maximum time-averaged pump power was limited by the radiation resistance of the reflecting silicon plate at the centre of the output mirror and amounted to 120 W. The mean pulse power thus exceeded 3 kW.

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Additional changes introduced into the optical pump system ensured a more uniform distribution of the pump radiation intensity over the SMM cavity and prevented undesirable CO_2-laser self-excitation effects associated with a back reflection of pump radiation from the output mirror of the SMM laser. The pump CO_2-laser radiation passed through system of spatial filtration (*6*), (*2*), (*5*) that transformed its beam into a near-Gaussian beam and multiply attenuated the pump radiation intensity propagating in the opposite direction in the optical path. The pump radiation was then focused to the coupling hole in the input mirror of the cavity by a system with a focal length $F = 1.2$ m.

The SMM laser radiation focused by spherical mirror (*8*) with $R = 30$ mm was detected by pyroelectric detector (*11*). The latter was preliminarily calibrated in the wavelength range of up to 200 μm, which allowed the absolute value of the output laser power to be determined. When searching for new lines, all the lines of the CO_2 laser were sequentially verified by pumping the active medium at three pressures (70, 200, and 350 mTorr).

The wavelength of the observed SMM radiation was determined using the technique of scanning the cavity length [17], which was smoothly changed within the range of 15 mm by the output mirror of the SMM laser. Upon each change in the wavelength by an integer number of half-waves, we observed the output-power maximum corresponding to a successive change of the longitudinal modes in the cavity. The readings of the SMM detector were recorded simultaneously with displacements of the output mirror using a recorder. The wavelength was calculated from the expression $\lambda = 2\Delta L/N$, where ΔL is the change in the cavity length corresponding to the change of N longitudinal modes. The error in determining the wavelength by this method was 0.5 %. Five new lasing lines in the short-wavelength SMM spectral region were obtained for CH_3OH molecules, while no new lines were observed for CH_2F_2 molecules.

In this work, CH_3OH with a purity of 99.9 % was used as the active medium, but we had no information on its isotope composition. Therefore, all of the lasing lines observed were verified by comparing with the data obtained earlier for all of the existing isotope modifications of the working molecule [18]. All the lasing lines corresponded to the $^{12}CH_3{}^{16}OH$ molecule.

Table 1 presents the parameters of the SMM lasing lines of CH_3OH molecules and the corresponding pump lines; the SMM radiation power, the optimal CH_3OH pressure, and the corresponding pump power are given. The polarisation of the SMM-laser output radiation was not recorded.

As the pump power increased, the lasing power increased linearly at all new lines. The lasing thresholds for all the lines are 600 – 1500 W (Table 1). It is probably the high threshold power that causes the generation of new lines. It is known [19] that, upon high-power optical pumping, the energy levels responsible for SMM lasing can be shifted in high-intensity fields (the field shift). As a result, the pump lines and the absorption band of the active medium may overlap, thus leading to lasing. Since the field shift in optically pumped SMM lasers has not been properly studied, it is planned in the future to perform such studies, which is necessary for determining the energy levels participating in the radiative transitions upon SMM lasing on lines with high thresholds.

Comparatively low lasing powers (0.12 – 1.45 mW) were obtained at all new lines. This is explained by the fact that a nonoptimised high-Q cavity was used in this study. Increasing the output power requires that optimal cavity parameters be selected for each lasing line.

Therefore, five new lines have been discovered for CH_3OH molecules in the setup with high-power optical pumping. The optimal pressure, the threshold power, and the wavelength (with an accuracy of 0.5 %) were measured for each lasing line. At pump powers of 1000 – 3000 W, the absence of saturation is typical of all the lines.

The results obtained point to the high efficiency of applying high-power optical pumping in SMM lasers, which can be used in a search for new lasing lines of both the well-known and insufficiently studied molecules. The data of more detailed studies can also be useful in molecular spectroscopy.

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Table 1. Lasing lines obtained for CH_3OH molecules.

CO_2-laser pump line	Pump power/W	SMM lasing wavelength/μm	SMM lasing power/mW	Optimal pressure /mTorr	Pump threshold power/W
9P(10)	900	77.3 ± 0.3	1.44	500	650
9R(30)	1440	137.4 ± 0.07	0.31	310	500
10P(30)	2760	118.3 ± 0.2	1.45	500	800
10R(8)	1800	97.4 ± 0.2	0.12	420	650
10R(14)	3300	80.1 ± 0.2	0.23	330	1600

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