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Optimal repetition rates of excitation pulses in a Tm-vapour laser

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Abstract. The optimal excitation pulse repetition rates (PRRs) for a gas-discharge Tm-vapour laser with indirect population of upper laser levels are determined. It is shown that, under the same excitation conditions, the optimal PRRs increase with a decrease in the energy defect between the upper laser acceptor level and the nearest resonant donor level. The reasons for the limitation of the optimal PRRs in Tm-vapour laser are discussed. It is shown that the maximum average power of Tm-vapour laser radiation may exceed several times the Cu-vapour laser power under the same excitation conditions and in identical gas-discharge tubes.

Keywords: Tm-vapour laser, frequency–energy characteristics, collisional excitation transfer, relaxation of metastable states.

1. Introduction

Tm-vapour lasers belong to gas-discharge rare-earth metal (REM) vapour lasers.

REMs form a series of 13 elements (Ce–Yb) in the periodic table; the inner 4f shell in the atoms of this series becomes gradually completed with increasing atomic number, while the outer 6s shell is completely filled throughout the series. As a result, REM atoms have both unscreened [for example, $4f^{13}(^2F^0_{7/2})6s6p(^1P^0_1)$] and screened [for example, $4f^{12}(^3H_5)$ - $5d_{5/2}6s^2$] excited states. In turn, the screened excited states give rise to transitions that are characterised by zero (or small) line broadening in collisions of REM atoms in these states with heavy particles [1, 2]. Lasing was obtained in vapours of seven REMs: Eu, Sm, Tm, Yb [3, 4], Ho, Dy, and Er [5–7] (in both visible and near-IR ranges).

A specific feature of REM-vapour lasers is that the upper levels in most laser transitions (from more than 50 transitions on which lasing was obtained) have the same parity as the ground state and cannot be excited by an electron impact from the ground state. For this reason the upper laser levels are

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$$\mathbf{M}_{i}^{*} + \mathbf{M}_{0} \nleftrightarrow \mathbf{M}_{k}^{*} + \mathbf{M}_{0} \pm \Delta E, \tag{1}$$

$$\mathbf{M}_{i}^{*} + \mathbf{B} \nleftrightarrow \mathbf{M}_{k}^{*} + \mathbf{B} \pm \Delta E, \tag{2}$$

where M_i^* and M_k^* are the REM atoms in excited *i* and *k* states with similar energies, M_0 is the REM atom in the ground state, B is the inert gas atom, and ΔE is the energy defect between the *i* and *k* states. Processes (1) and (2) are efficient at $\Delta E < k_B T_g$, where k_B is the Boltzmann constant and T_g is the gas temperature. A schematic diagram of the formation of population inversion in the aforementioned lasers is shown in Fig. 1. If the resonant level R is screened, the collisional excitation transfer from this level to the upper laser level UL may occur only via reaction (2). At the same time, the excitation transfer from the unscreened resonant level to the upper laser level occurs mainly via reaction (1) [8].

The average lasing power P_{las} of pulsed lasers is determined by the expression

$$P_{\rm las} = E_{\rm c} V f = E_{\rm pul} f,\tag{3}$$

where E_{pul} is the lasing pulse energy, E_c is the specific laser energy extraction; V is the active-medium volume, and f is the excitation pulse repetition rate (PRR). The E_c and E_{pul} values and, therefore, P_{las} depend strongly on f. Under specified



Figure 1. Schematic diagram of population of the upper laser level UL via the collisional excitation transfer in reactions (1) and (2) from a close-lying resonant level R: LL is the lower laser level, λ_{gen} is the laser transition wavelength, and *E* is the energy defect between levels R and UL.

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excitation conditions, three characteristic rates for each laser transition can be selected in the operating range of f: a rate optimal for the pulse energy $[f_{opt}(E_{pul})]$, a rate optimal for the laser radiation power $[f_{opt}(P_{las})]$, and a limiting rate (f_{lim}) . The rates $f_{opt}(E_{pul})$ and $f_{opt}(P_{las})$ are determined by the onset of decrease in, respectively, the pulse energy E_{pul} and radiation power P_{las} with an increase in f, while the rate f_{lim} corresponds to lasing stop. No systematic studies of the frequency–energy characteristics of REM vapour lasers with indirect excitation of upper laser levels have been performed.

The purpose of this study was to analyse the specific behavioural features of the rates $f_{opt}(E_{pul})$ and $f_{opt}(P_{las})$, depending on the energy defect ΔE between the resonant and upper laser levels and on the buffer gas (helium) pressure in Tm-vapour laser.

2. Experimental

A Tm-vapour laser [9] was chosen for experiments because there are many (more than 20) laser transitions with nonresonant upper laser levels and a variety of energy defect values ΔE_{R-UL} between the upper laser and close-lying resonant levels in thulium atoms. We studied four transitions, which provided lasing at a saturated thulium vapour pressure of 0.1 Torr. A schematic diagram of the transitions under study is presented in Fig. 2.

The energy defect values ΔE_{R-UL} spread from 27 to 461 cm⁻¹ (Table 1) [10, 11]. The upper level UL1 of the laser transition with the wavelength λ_1 was populated during reaction (1) from the two unscreened resonant levels R1 and R2. The upper levels UL2–UL4 of the other laser transitions with the wavelength $\lambda_2 - \lambda_4$ were populated via reaction (2) from the two screened resonant levels R3 and R4. Note that the transitions with λ_2 and λ_4 are competing because of their common lower laser level LL2.

The laser operated in the self-heating regime. The heatinsulating Al₂O₃ ceramic gas-discharge tube (GDT) had an inner diameter of 20 mm and an active-region length of 300 mm ($V = 94.25 \text{ cm}^3$). Small pieces of metallic thulium (with the purity of 99.83%) were placed on the inner surface of the GDT throughout its length. A working capacitance of 2.35 nF, charged to a voltage of 14 kV, was switched to the GDT via a TGI1-1000/25 hydrogen thyratron. The buffer gas (helium) pressure p_{He} was varied in the range of 1–3 Torr, and the PRR ranged from 1 to 30 kHz. To select individual laser lines, we used a tunable cavity with a diffraction grating having 300 grooves mm⁻¹ as a highly reflecting mirror and a glass plate as an output mirror. The experimental setup was described in more detail in [12]. The technique for measuring the energy characteristics of the laser was determined by its



Figure 2. Schematic diagram of the laser transitions in a thulium atom. The dashed arrows indicate population of resonant levels from the ground state via the discharge electron impact. Laser transitions are shown by solid lines. The fractional numbers near the levels are the values of total angular momenta J.

self-heating working regime. After reaching the optimal temperature conditions, the PRR was changed for a short time, and the lasing pulse amplitude and width were measured with an oscilloscope. The temperature of the gas-discharge channel hardly changed over this short time interval. The fairly high thickness of the heat-insulating GDT layer did not make it possible to control the average lasing power at high PRRs because of GDT overheating.

3. Results and discussion

Figure 3 shows the dependences of the experimentally measured lasing pulse energy E_{pul} on the PRR *f* for the chosen transitions in a thulium atom. These curves clearly indicate that, under the same excitation conditions and concentrations of thulium and helium atoms, the larger the energy defect ΔE_{R-UL} between the upper laser acceptor level and the nearest resonant donor level (from which this laser level is basically populated), the earlier E_{pul} begins to decrease with an increase in *f*.

For example, at a helium pressure $p_{\text{He}} = 3 \text{ Torr}$ (Fig. 3a) the decrease in the energy E_{pul} on the transition with λ_3 ($\Delta E_{\text{R-UL}} = 323.130 \text{ cm}^{-1}$) begins even at the rate $f_{\text{opt}}(E_{\text{pul}}) = 2 \text{ kHz}$,

Table 1. Spectroscopic parameters of the laser transitions in thulium atoms [10, 11].

Wavelength	Resonant level R		Upper laser level UL		Lower laser level LL		$\Delta E_{\mathbf{R}}$ _111
	Energy $E_{\rm R}/{\rm cm}^{-1}$	Electronic configuration	Energy $E_{\rm UL}/{\rm cm}^{-1}$	Electronic configuration	Energy $E_{\rm LL}/{\rm cm}^{-1}$	Electronic configuration	/cm ⁻¹
$\lambda_1 = 1101.115 \text{ nm}$	25745.117 25656.019	$\begin{array}{c} 4f^{13}({}^{2}\mathrm{F}{}^{0}_{7/2})6s6p({}^{1}\mathrm{P}{}^{0}_{1})\\ 4f^{13}({}^{2}\mathrm{F}{}^{0}_{5/2})6s6p({}^{3}\mathrm{P}{}^{0}_{0}) \end{array}$	25536.116	$4f^{12}(^{3}\text{H}_{6})6s^{2}6p_{3/2}$	16456.913	$4f^{12}(^{3}\text{H}_{6})5d_{5/2}6s^{2}$	209.001 119.903
$\lambda_2 = 1310.06 \text{ nm}$	22929.717 22791.176	$\frac{4f^{12}({}^{3}\mathrm{H}_{5})5\mathrm{d}_{5/2}6\mathrm{s}^{2}}{4f^{12}({}^{3}\mathrm{H}_{5})5\mathrm{d}_{3/2}6\mathrm{s}^{2}}$	22902.127	$4f^{12}(^{3}\text{H}_{6})6s^{2}6p_{1/2}$	15271.002	$4f^{12}({}^{3}\mathrm{H}_{6})5\mathrm{d}_{3/2}6\mathrm{s}^{2}$	27.590 -110.951
$\lambda_3 = 1069.39 \text{ nm}$	22929.717 22791.176	$\frac{4f^{12}({}^{3}\mathrm{H}_{5})5\mathrm{d}_{5/2}6\mathrm{s}^{2}}{4f^{12}({}^{3}\mathrm{H}_{5})5\mathrm{d}_{3/2}6\mathrm{s}^{2}}$	22468.046	$4f^{12}(^{3}\text{H}_{6})6\text{s}^{2}6\text{p}_{1/2}$	13119.610	4f ¹² (³ H ₆)5d _{3/2} 6s ²	461.671 323.130
$\lambda_4 = 1338.01 \text{ nm}$	22929.717 22791.176	$\frac{4f^{12}({}^{3}\mathrm{H}_{5})5\mathrm{d}_{5/2}6\mathrm{s}^{2}}{4f^{12}({}^{3}\mathrm{H}_{5})5\mathrm{d}_{3/2}6\mathrm{s}^{2}}$	22742.777	$4f^{13}(^2\mathrm{F}^0_{7/2})5\mathrm{d}6\mathrm{s}(^3\mathrm{D})$	15271.002	$4f^{12}(^{3}\mathrm{H}_{6})5\mathrm{d}_{3/2}6\mathrm{s}^{2}$	186.940 48.399





Figure 3. Dependences of the lasing pulse energy E_{pul} at the transitions studied for the Tm-vapour laser on the PRR *f* at helium pressures $p_{He} =$ (a) 3, (b) 2, and (c) 1 Torr.

whereas on the transition with λ_1 ($\Delta E_{\text{R}-\text{UL}} = 119.903 \text{ cm}^{-1}$) it begins at the rate $f_{\text{opt}}(E_{\text{pul}}) = 6 \text{ kHz}$. On the transitions with λ_4 ($\Delta E_{\text{R}-\text{UL}} = 48.399 \text{ cm}^{-1}$) and λ_2 ($\Delta E_{\text{R}-\text{UL}} = 27.590 \text{ cm}^{-1}$), which have a common lower laser level LL2 (Fig. 2), no decrease in the energy E_{pul} was observed with an increase in f to 30 kHz, and their optimal rate $f_{\text{opt}}(E_{\text{pul}})$ exceeded the maximum PRR value that was used in the experiment. At $p_{\text{He}} = 2 \text{ Torr}$ (Fig. 3b) the decrease in E_{pul} became pronounced on the transitions with λ_2 , and lasing with λ_3 stopped. At $p_{\text{He}} = 1 \text{ Torr}$ (Fig. 3c) we observed only two strong lines with λ_1 and λ_2 , and the limiting rate $f_{\text{lim}} = 24 \text{ kHz}$ on the transition with λ_2 was even within the PRR range used in our experiment. Figure 4 shows the dependences of the average lasing power P_{las} on the PRR

Figure 4. Dependences of the average lasing power P_{las} on the PRR *f* at helium pressures $p_{\text{He}} = (a) 3$, (b) 2, and (c) 1 Torr. In view of the smallness of power values for the transitions with λ_1 and λ_3 , they are increased by factors of 5 and 20, respectively, in panel (a).

f. The powers P_{las} were calculated from formula (3), because, as was noted in Section 2, GDT overheating made impossible power measurements at high *f*. It can clearly be seen that the behaviour of the calculated optimal rates $f_{\text{opt}}(P_{\text{las}})$ qualitatively repeats that of the experimental rates $f_{\text{opt}}(E_{\text{pul}})$. Figure 5 presents the dependence of the optimal PRR $f_{\text{opt}}(E_{\text{pul}})$ on the helium pressure p_{He} for the transitions with λ_1 , λ_2 , and λ_4 .

An analysis of the experimental data (Figs 3, 5) also suggests that the limitation of PRR in Tm-vapour laser is mainly related to the relaxation rate of the lower laser (metastable) levels of the corresponding transitions during the interpulse period. In this case, the depopulation [of at least screened metastable states (Table 1)] occurs in collisions with helium



Figure 5. Dependences of the optimal PRR $f_{opt}(E_{pul})$ on helium pressure p_{He} for the transitions with λ_1, λ_2 , and λ_4 .

atoms. Indeed, on the one hand, the rate $f_{opt}(E_{pul})$ for the laser transition with λ_1 is little sensitive to variations in p_{He} , because the population of the upper level UL1 of this transition is determined by reaction (1), and the thulium vapour density in the experiment did not change. However, on the other hand, an increase in $p_{\rm He}$ leads to an increase in the rate at which lasing may occur (Fig. 3). Thus, the higher the He concentration, the more efficiently the lower laser level LL1 is depopulated during the time between pulses and, therefore, the higher the limiting PRR f_{lim} . The upper levels UL2 and UL4 of the laser transitions with λ_2 and λ_4 are populated in reaction (2); therefore, an increase in the helium concentration increases both the limiting PRR f_{lim} and the optimal PRR $f_{\text{opt}}(E_{\text{pul}})$ for these transitions. Nevertheless, one cannot exclude other possible mechanisms for limiting PRR. For example, in lself-terminating manganese- and copper-vapour lasers the main process that limits PRR is the recombination during the time between pulses [13].

The character of the dependence $f_{opt}(E_{pul})$ on p_{He} for the competing transitions with λ_2 and λ_4 (Fig. 5) suggests that their common screened level LL2 does not relax directly to the ground state: the relaxation occurs via population of their upper laser levels in reverse direction; i.e., in collisions with helium atoms, metal atoms pass from the lower laser level to the upper level and then relax via reaction (2) to the nearest resonant level, which optically decays to the ground state during the time between pulses. In this case, the population rate dN_R/dt of the resonant level R in reaction (2) is determined by the well-known equation [14]

$$\frac{\mathrm{d}N_{\mathrm{R}}}{\mathrm{d}t} = N_{\mathrm{UL}} \langle \sigma_{\mathrm{UL-R}} \bar{v}_{\mathrm{He}} \rangle N_{\mathrm{He}}.$$
(4)

Here, σ_{UL-R} is the excitation transfer cross section from the upper laser level UL to the close-lying resonant level R, \bar{v}_{He} is the average thermal speed of helium atoms, and N_{He} is the helium concentration. The concentration of thulium atoms N_{UL} at the level UL is found by integrating (over the interpulse period 1/f) the equation for the population rate of this level:

$$\frac{\mathrm{d}N_{\mathrm{UL}}}{\mathrm{d}t} = N_{\mathrm{LL}} \langle \sigma_{\mathrm{LL}-\mathrm{UL}} \bar{v}_{\mathrm{He}} \rangle N_{\mathrm{He}},\tag{5}$$

where $\sigma_{\text{LL}-\text{UL}}$ is the excitation transfer cross section from the lower laser level LL to the upper level UL and N_{LL} is the concentration of thulium atoms at the level LL (Fig. 1). The depopulation of the level LL2 via this channel explains the general regularities in the behaviour of the rate characteristics of the lasing on the transitions with λ_2 and λ_4 (Fig. 5). The small discrepancy in the $f_{\text{opt}}(E_{\text{pul}})$ values for these transitions is due to the difference in the energy defects $\Delta E_{\text{R}-\text{UL}}$. Nevertheless, this suggestion must be additionally checked.

Another distinctive feature of the results obtained is the significant increase in the energy E_{pul} (or E_c) on the transition with λ_2 with an increase in f. With a change in f from 2 to 30 kHz the pulse energy on this transition increases by a factor of 4 (Fig. 3a). The experimentally measured specific energy extraction E_c at f = 2 kHz was found to be 5 μ J cm⁻³, whereas at f = 30 kHz it increased to 20 µJ cm⁻³. Previously this effect has not been observed in self-terminating lasers. As an example Fig. 6 shows the behaviour of the energy extraction E_c with an increase in f [15], which is typical of Cu-vapour lasers. The energy potential of Tm-vapour laser with indirect population of upper laser levels was estimated by comparing the average powers of Tm- and Cu-vapour lasers under the same conditions and identical GDTs. Figure 7 shows the dependences of P_{las} on f for green and IR lines of the Cu and Tm-vapour lasers, respectively. The powers $P_{\text{las}}(f)$ for the Cu-vapour laser were calculated from formula (3) at known values of specific energy extraction E_c (Fig. 6) [15], and the powers $P_{\text{las}}(f)$ for Tm-vapour laser were estimated from the data obtained (Fig. 3a). It can clearly be seen that the average Tm-vapour laser power exceeds the average Cu-vapour laser power even at $f \approx 5$ kHz. At f = 30 kHz the average power $P_{\text{las}}(\lambda_2) \approx 60 \text{ W}$, and this value exceeds significantly the power of the Cu-vapour laser with a similar GDT $[P_{las}(\lambda_{Cu}) \approx 10]$ W]. Thus, Tm-vapour lasers can be promising coherent radiation sources in the IR spectral region, provided that the problem of heat removal from GDT is solved.



Figure 6. Dependence of the specific energy extraction of laser radiation, E_c , on the PRR *f* for a Cu-vapour laser ($\lambda_{Cu} = 510.6$ nm) [15].

The behaviour of the average power $P_{\text{las}}(f)$ in Cu-vapour laser is determined by the character of change in the pulse energy E_{pul} (or E_{c}) with a change in the PRR f (Fig. 6). The power of a four-level Tm-vapour laser greatly exceeds that of a three-level Cu-vapour laser because of the higher conversion efficiency of the excitation energy of resonant and upper laser levels into radiation in the Tm-vapour laser. In [16] it



Figure 7. Dependences of the calculated average lasing power P_{las} on the PRR *f* for a Cu-vapour laser ($\lambda_{\text{Cu}} = 510.6 \text{ nm}$) [15] and a Tm-vapour laser ($\lambda_2 = 1310.06 \text{ nm}$).

was shown that only a small fraction (a few percent) of the energy of the upper laser level is converted into radiation in the Cu-vapour laser; the reason is the dissipation of spontaneous radiation at the GDT wall. Such a strong depopulation of the resonant level is due to its short lifetime (~ 7 ns) (disregarding radiation trapping, which does not occur because the distance to the GDT wall is small). For the Tm-vapour laser the lifetime of the resonant level, from which the upper laser level is populated, is ~ 260 ns [17], a value exceeding the current pulse duration. Therefore, the resonant level is a reliable reservoir of excitation energy during lasing pulse formation.

4. Conclusions

(i) The main reason for limiting PRR in Tm-vapour laser is the relaxation rate of the lower laser (metastable) levels of the corresponding transitions during the time between pulses. Screened metastable levels are depopulated in collisions with helium (buffer gas) atoms. An increase in the helium concentration increases both the optimal PRRs $f_{opt}(E_{pul})$ and the $f_{opt}(P_{las})$ values for the laser transitions and the limiting PRRs f_{opt} . The relaxation (of at least the common lower laser level of the transitions with λ_2 and λ_4) occurs not directly to the ground state but via reverse population of their upper laser levels. Note that these conclusions are preliminary and must be experimentally verified.

(ii) In a Tm-vapour laser with indirect population of the upper laser levels the optimal excitation $f_{opt}(E_{pul})$ and $f_{opt}(P_{las})$ are determined by the energy defect ΔE_{R-UL} between the upper laser acceptor level UL and the nearest resonant donor level R. Under the same excitation conditions the optimal rates increase with a decrease in the defect ΔE_{R-UL} .

(iii) A comparison of the energy characteristics of Cu and Tm-vapour lasers (for $\lambda_{Cu} = 510.6$ nm and $\lambda_2 = 1310.06$ nm) shows that the calculated maximum average Tm-vapour laser power ($P_{las}(\lambda_2) \approx 60$ W) exceeds several times the average Cu-vapour laser power ($P_{las}(\lambda_{Cu}) \approx 10$ W) under similar excitation conditions and with identical GDTs used. Although the self-heating regime is implemented at lower *f* than the rates used in the experiment (30 kHz), the power $P_{las}(\lambda_2)$ exceeds $P_{las}(\lambda_{Cu})$ even at $f \approx 5$ kHz. Hence, Tm-vapour lasers can be considered as promising sources of coherent IR radiation.

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