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Collisional lasing on a self-terminating transition $2\,^1P_1^o-2\,^1S_0$ in helium atom

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Abstract. **Lasing on a self-terminating transition $2\,^1P_1^o-2\,^1S_0$ (λ = 2.058 μm) in helium atom is studied for a single- and double-pulse operation regimes under electron beam pumping in pure helium and its mixtures with H_2, N_2, O_2, CO_2, H_2O, NH_3, and N_2O. In pure helium, the maximal pulse duration is ~50 ns, which agrees with the calculated value. Recovery of lasing in the second pulse is observed at a time delay between the pulses of longer than 1.25 μs. In adding CO_2, N_2O, NH_3, and H_2O, the relaxation rate for population of the metastable state $He(2\,^1S_0)$ increases, which makes the delay, needed for recovering lasing, shorter up to pulse merging in the case of H_2O. At the exciting pulse base-level duration of 1.2 μs, in mixtures of helium with NH_3 and H_2O, laser pulses with a duration of ~0.8 μs are observed, which testifies that collisional quasi-cw lasing occurs. Mechanisms of collisional lasing are discussed.**

Keywords: *collisional lasing, helium laser, constants of de-excitation.*

1. Introduction

Depopulation of a lower laser level through collisions with heavy particles is actively used for obtaining an inversion population in atoms, molecules, and their ions, which resulted in creation of high-power highly efficient lasers (lasers on CO_2 and CO molecules, iodine atoms, etc.). The idea of using collisional de-excitation of working states for producing a medium with negative absorption was first suggested [1] just prior to creation of a first laser. Later, Bennett [2] and Gould [3] defined the requirements to active media of the lasers (sometimes called 'collision lasers') based on fast relaxation in a system of groups of closely residing upper and lower levels with the energy separation in groups $\Delta E \leqslant k_B T_g$ (here, k_B is the Boltzmann constant, T_g is the active medium temperature).

As was mentioned in [4], successful implementation of Gould's suggestion on creating a collision laser is a laser on a CO_2-N_2-He mixture. However, attempts to create such lasers on active media with atomic or ion transitions by conventional excitation schemes [a gas discharge or electron beam (EB)] were generally not successful [5]. Under optical excitation [6] or by mixing with a preliminarily excited carrier of energy [7], population inversion is produced and lasing is obtained, for example, in rubidium and caesium atoms [6] or iodine atoms [7]. Although the relaxation and energy transfer in these atoms at $\Delta E \leqslant k_B T_g$ only occur in an ensemble of upper levels, most powerful and prospective lasers are created and are still being developed on their basis [8, 9]. One more successful tendency in creation of lasers with collisional de-excitation is search for and realisation of the systems with fast relaxation of lower working levels in a single collision act at $\Delta E \gg k_B T_g$. In particular, in a He–Zn–Eu mixture, the transition $Eu^+(z\,^7P_4-a\,^7D_5^o)$ at λ = 1.002 μm exhibits a maximal gas laser radiation energy per unit mass [10].

Among a great variety of collisional processes, which lead to fast relaxation of atomic state populations at $\Delta E \gg k_B T_g$ and result in lasing [11–23], only the penning effect in atoms and quasi-resonance recharge in ions are of sufficiently general character. Recently [24], it was reported about collisional lasing on a transition $He(2\,^1P_1^o-2\,^1S_0)$ at λ = 2.058 μm with a simultaneous employment of other universal quenching mechanisms, namely, collisions with molecules and slow electrons. An attractive feature of the laser is its possibility to produce lasing on the transition from a resonance state (RS) to a metastable state (MS), suggested in [25] and realised for atoms Ba [22, 26], Eu [10, 22], Ca and Sr [12, 13], Pb [22], and for ions Ca^+, Eu^+, and Sr^+ [21]. Here, we report results of a detailed study of collisional lasing in helium atom.

2. Experimental

In studying the lasing characteristics, we used the laser cavity and pump generator described elsewhere [24, 27]. Lasing on a self-terminating transition was obtained under pumping by a radial EB, which was generated by an open discharge (OD) [28] in a double pulse regime with the FWHM pulse duration of 25 ns in a 12-cm-long cavity 3.1 cm in diameter. The amplitude of the current in the EB reached 2.5 kA and the working voltage in the OD did not exceed 7 kV. The properties of collisional lasing were also studied under pumping by single pulses with the duration of up to 1.2 μs generated by an artificial line with a wave impedance of 4 Ω.

The MS relaxation rate for $He(2\,^1S_0)$ was measured by absorption in a test cavity with a length of 9 cm and diameter of 5 cm, which was also pumped by the radial EB with duration of ~15 ns generated in the OD. For probe radiation we used radiation of a laser on the self-terminating transition $He(2\,^1P_1^o-2\,^1S_0)$ at λ = 2.058 μm pumped by the radial EB. A near-axis domain was cut from the laser beam by a diaphragm 0.3-cm in diameter and the radiation was directed along an axis of the test cavity. The nonresonant losses in the laser cavity were βl = 0.34 (here, β is the reduced coefficient of nonreso-

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nant losses and l is the length of the laser active medium), and the pumping was chosen so that the gain at the Doppler profile centre be $\alpha = 0.69$ (α is the gain). The lasing profile calculated for this case in [29] is given by the expression $I_{las}(\nu) = \alpha(\nu) lI_s[1 - \sqrt{\beta/\alpha(\nu)}]^2$ {$I_s = A_{rm}h\nu_0/[2\sigma(\nu)]$ is the line centre saturation intensity at the frequency $\nu_0 = c/\lambda$, A_{rm} is the transition probability for He($2\,^1P_1^o - 2\,^1S_0$); h is Planck's constant, $\sigma(\nu)$ is the absorption cross section for Doppler broadening} and is presented in Fig. 1. In the same figure, the Doppler profile for the absorption line in the test cavity is shown for the transition He($2\,^1S_0 - 2\,^1P_1^o$) at room temperature.

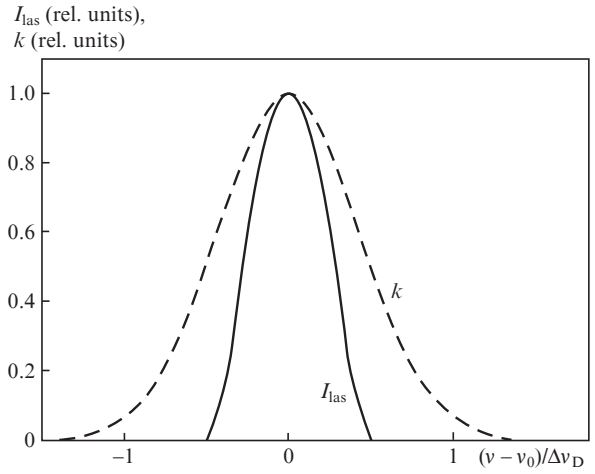

Figure 1. Intensity of laser radiation I_{las} and the absorption coefficient k for the Doppler-broadened line He($2\,^1S_0 - 2\,^1P_1^o$) vs. detuning from the line centre [$\lambda = 2.058$ μm, $\Delta\nu_D$ is the Doppler (FWHM) linewidth].

With a monochrome probe radiation frequency ν_0 corresponding to the centre of the absorption line, the absorption coefficient $k(\nu_0)$ is determined by the ratio of the energies w_{out} and w_{in} for the probe radiation at the cavity output and input, correspondingly, $w_{out}/w_{in} = \exp(-ks)$, where s is the absorption path length. Because the monochrome probe radiation at the wavelength $\lambda = 2.058$ μm is absorbed on the transition He($2\,^1S_0 - 2\,^1P_1^o$), then the absorption coefficient is $k = k(\nu_0) \propto n_m$, where n_m is the MS population for He($2\,^1S_0$). In view of the time evolution $n_m = n_m^0 \exp(-At)$, the rate of quenching A is determined by the expression

$$At = -\ln k + b = -\ln[-\ln(w_{out}/w_{in})] + c,$$

where b and c are constants. The constant of the process is found from the MS de-excitation rate: $k_i^m = A/n_i$, where n_i is the concentration of the ith reagent (electrons and various molecules).

In the case where the frequency of the probe radiation $\nu \neq \nu_0$, the absorption coefficient reduces and, consequently, w_{out}/w_{in} and the population n_m are determined with an inaccuracy $\Delta n_m/n_m = [k(\nu_0) - k(\nu)]/k(\nu_0)$. If the probe radiation is not monochromatic, then

$$w_{out}/w_{in} = \int I_{las}(\nu)\exp[-k(\nu)s]\,d\nu \Big/ \int I_{las}(\nu)\,d\nu = \exp(-k_{av}s)$$

(k_{av} is the resulting absorption coefficient) and the coefficient k_{av} obtained differs from $k(\nu_0)$. The inaccuracy of measuring the de-excitation constant $\Delta k_i^m/k_i^m$ due to the finite spectral width of the probe radiation was calculated for various values of the absorption coefficient $k(\nu_0)$ as follows

$$\frac{\Delta k_i^m}{k_i^m} = 1 - \frac{\ln(k_{av})}{\ln[k(\nu_0)]} = 1 - \frac{\ln[-\ln(w_{out}/w_{in})]}{\ln\{-\ln[\exp[-k(\nu_0)]]\}}.$$

Figure 2 shows the calculation results for the intensity distribution of the probe laser radiation over frequency presented in Fig. 1. One can see that the inaccuracy raises as $k(\nu_0) \propto n_m$ increases (the ratio w_{out}/w_{in} reduces). For example, in order to provide the accuracy of $\Delta k_i^m/k_i^m \approx 2\%$ in determining the de-excitation constant $k_i(\nu)$, the ratio w_{out}/w_{in} should be greater than 0.06.

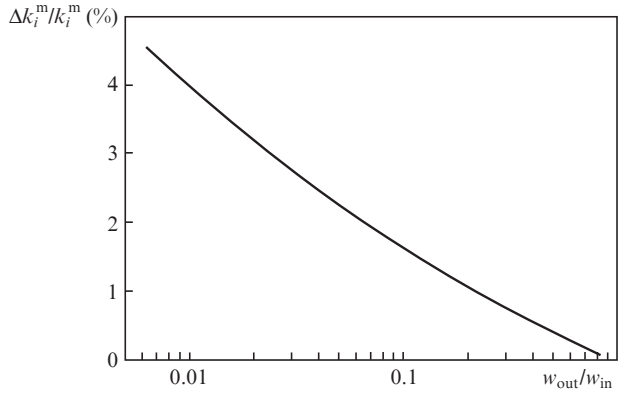

Figure 2. Inaccuracy $\Delta k_i^m/k_i^m$ of measuring the de-excitation constant vs. the energy ratio w_{out}/w_{in} of output radiation passed across the test cavity and input probe radiation.

The parameters of the laser pulse radiation were measured with an InSb sensor (PD24-03) with the time resolution of 4 ns and with a four-channel TDS-2024B oscilloscope with the bandwidth of 200 MHz. Spontaneous emission from working cavities was studied by using a monochromator and a FEU-106 photomultiplier with the time resolution of 3 ns.

3. Lasing characteristics in the double-pulse regime

The lasing parameters in the second pump pulse, i.e., the dependences of w_2/w_1 on the time delay between the pulses Δt (here, w_2 and w_1 are the energies of the second and first pulses, respectively), were measured both in pure He and in its mixtures with H_2, N_2, O_2, H_2O, NH_3, and N_2O. A characteristic example of lasing recovery in pure He in the second pulse is curve (*1*) in Fig. 3a. One can see that lasing arises at $\Delta t_1 = 1.25$ μs and completely recovered lasing is observed at $\Delta t_2 = 5$ μs. The rate of the recovery weakly depends on the pump pulse energy. Such a dependence is typical for electron-beam-pumped self-terminating lasers, and is determined by the rate of MS relaxation in the afterglow [22, 30, 31].

Molecular admixtures accelerate the process of lasing recovery in the second pulse. The recovery time substantially depends on the type of the admixture and, under some conditions, on a pressure of the latter p_{mol}. In the case of dimers (H_2, N_2, O_2), at increasing p_{mol} the parameters Δt_1 and Δt_2 first rapidly reduce and then remain constant up to the limiting pressure, at which lasing still can be observed (Fig. 3a). Most

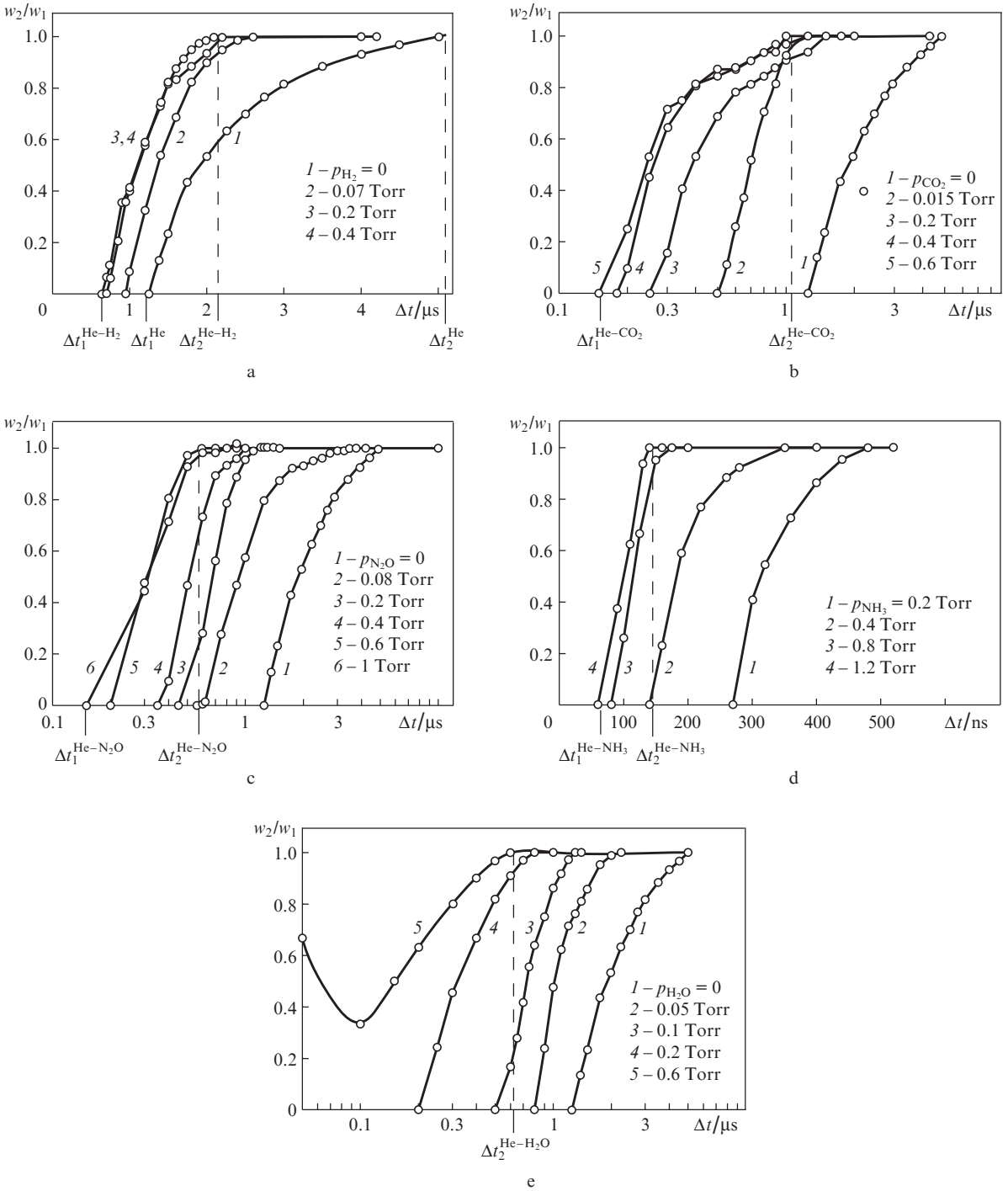

Figure 3. Ratios w_2/w_1 vs. the delay Δt between the pump pulses at p_{He} = 7 Torr under double pulse excitation in mixtures $He-H_2$ (a), $He-CO_2$ (b), $He-N_2O$ (c), $He-NH_3$ (d), and $He-H_2O$ (e).

rapidly lasing recovers in the mixture $He-H_2$, in which case Δt_1 = 0.64 μs and Δt_2 = 2.2 μs.

Lasing is more rapidly recovered in mixtures of He with triatomic molecules: a symmetric CO_2 molecule and weakly polarised N_2O molecule, which has a dipole moment d_{N_2O} = 0.16 D (1 D = 0.33×10^{-29} K m) (see Figs 3b, c). However, similarly to the case of dimers, the fall of Δt_1 and Δt_2 vanishes as the mixture pressure exceeds a certain value. For a N_2O molecule, the minimal values of Δt_1 and Δt_2 are, respectively, 0.15 and 0.6 μs, and for CO_2 molecule these are 0.15 and 1 μs.

Shortest lasing recovery times in the second pulse are observed in mixtures with polar molecules NH_3 and H_2O possessing large dipole moments: d_{NH_3} = 1.46 D, d_{H_2O} = 1.84 D (see Figs 3d, e); in the mixture with H_2O, lasing does not vanish at $p_{H_2O} > 0.5$ Torr. For the second pulse, at $\Delta t < 100$ ns an increase in the ratio w_2/w_1 is observed, which is a consequence of temporal overlap of laser pulses. Note that this phenomenon – overlap of laser pulses as the similar pump pulses become closer – is observed for self-terminatig transition lasers for the first time.

4. Lasing parameters under long excitation pulses

Figure 4 presents the oscillograms of the pump pulses and laser radiation while exciting pure He by long pulses. In all the experimental conditions (p_{He} = 6–50 Torr, the EB current of 0.16–2.34 kA, discharge voltage of U = 2.4–6.2 kV), a longest pulse base-level duration was 50 ns with a statistical error of ±3 ns. A similar maximal duration of the laser pulse is obtained in helium mixtures with H_2, N_2, O_2, CO_2, and N_2O up to the admixture pressures yet providing lasing. Quite differently lasing occurs in helium mixtures with H_2O and NH_3. Adding admixtures H_2O (up to 0.6 Torr) and NH_3 (up to 1.3 Torr) at p_{He} = 6 Torr results in a greater generation energy (by a factor of 30 in the mixtures with H_2O) and pulse duration due to a stretched trailing edge of the pulse [see Fig. 5, oscillograms (*1–4*)].

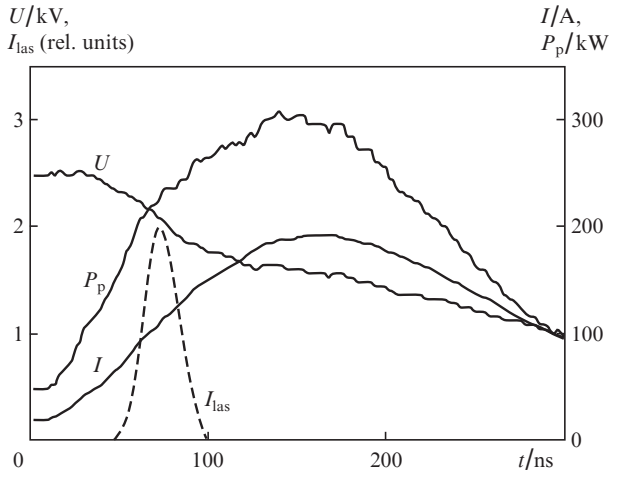

Figure 4. Oscillograms of voltage U, current I, power P_p of the discharge, and laser radiation pulse (I_{las}) at p_{He} = 12 Torr.

In exciting by single pulses, the shape of the laser pulses depends not only on the pressure of a molecular additive p_{mol}, but also on the pump power P_p (see Fig. 6). The maximal feasible pump power, in turn, depends on the OD duration and is limited by the instability of EB generation, which arises if the pump power overrides a certain value. For example, in the He–H_2O mixture at pressures of p_{He} = 6 Torr and p_{H_2O} = 1.3 Torr, the highest pump power is 480 kW at the FWHM pulse duration of τ_p = 280 ns, 150 kW at τ_p = 500 ns and 72 kW at τ_p = 680 ns.

At higher pressures p_{H_2O} and low pump powers, the laser pulse has no leading peak and appears in a long delay relative to the onset of the current pulse. In mixtures of He with H_2O and NH_3, lasing occurs at a considerably lower pump power as compared to pure helium. For example, in the He–H_2O mixture at the pressures p_{He} = 6 Torr and p_{H_2O} = 1 Torr, lasing starts at the voltage U = 1.08 kV corresponding to the maximum current I = 100 A, whereas in pure helium the threshold values for U and I needed for lasing to occur are 2.4 kV and 180 A, respectively. The range of optimal helium pressures in adding an admixture shifts to lower values of p_{He}. Lasing at 2.058 μm is observed in the He–H_2O mixture at p_{He} = 0.7 Torr and p_{H_2O} = 0.7 Torr, whereas in pure helium a minimal working pressure is p_{He} = 6 Torr. The range of pressures (p_{He} > 9 Torr) in which lasing may occur in mixtures of helium with molecular admixtures is limited by the arising instability of EB generation.

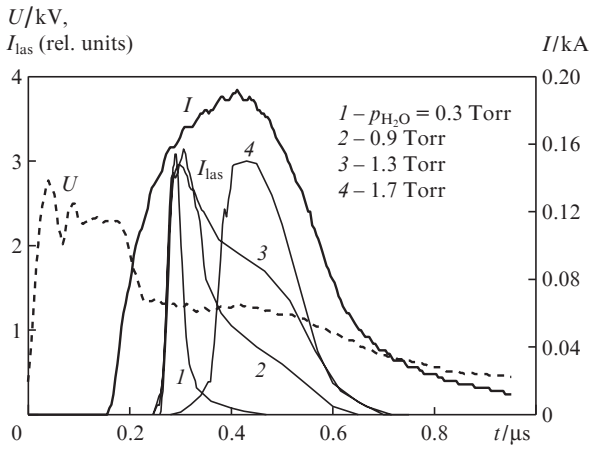

Figure 5. Oscillograms of laser radiation pulses (I_{las}) at various water vapour pressures, p_{He} = 6 Torr and the same discharge voltage U and current I, which are also shown.

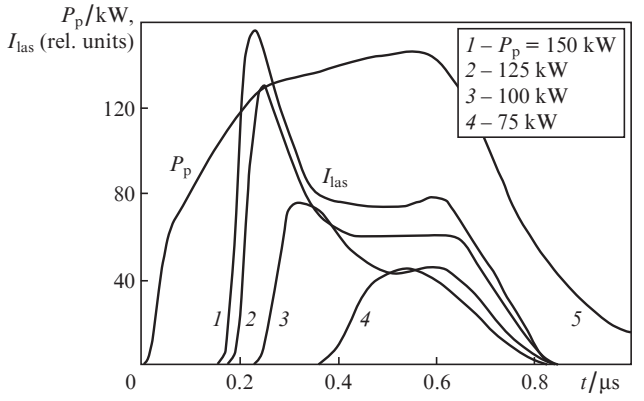

Figure 6. Oscillograms of the laser radiation pulses (I_{las}) at various pump powers P_p (*1–4*) and the oscillogram of the discharge power P_p (*5*) at p_{He} = 6 Torr and p_{H_2O} = 0.9 Torr.

5. Measurements of the relaxation rate and de-excitation constants for the He($2\,^1S_0$) state in mixtures with molecular additives

Clear understanding of the mechanism of arising quasi-cw lasing necessitates information about de-excitation constants for the metastable state He($2\,^1S_0$). Unfortunately, certain conditions should be met in performing the corresponding measurements for obtaining reliable data. To the best of our knowledge, there is only one work in which the constants of quenching atoms He($2\,^1S_0$) by complex molecules were measured [32], though indirectly and without specifying accuracy. The main difficulty is that the rate of quenching k_e^m of the He($2\,^1S_0$) state by electrons to the He($2\,^3S_1$) state is extremely high and amounts to 4×10^{-7} cm^3 s^{-1} [33]. Actually yet at a small electron concentration n_e needed for obtaining reason-

able quantity of atoms $He(2\,^1S_0)$, the populations of the states $He(2\,^1S_0)$ and $He(2\,^3S_1)$ relax into the ground state $He(1\,^1S_0)$ as a unit with the constant $k_e(2\,^3S_1) = 4.2\times10^{-9}$ cm^3 s^{-1} [34, 35]. In addition, atoms $He(2\,^1S_0)$ may participate in fast quenching when colliding with each other, with atoms $He(2\,^3S_1)$ and $He(1\,^1S_0)$ [33, 35].

Thus, the EB parameters and He pressure in the test cavity were chosen in such a way that the quenching rate of the state $He(2\,^1S_0)$ by electrons $k_e^m n_e$ would not exceed 10^5 s^{-1}, and the total quenching rate in all other processes $\sum_i k_i^m n_i$ including diffusion on walls would not exceed 10^4 s^{-1}. In particular, in the experiments with the test cavity diameter of 5 cm, the measurements were taken at a pressure $p_{He} = 8-15$ Torr, EB current below 5 A, FWHM pulse duration of 15 ns, and voltage amplitude of ~4 kV. By calculating the concentration n_e according to [27], we estimated an upper limit of this parameter 2×10^{11} cm^{-3}. Measurements at such a value of n_e guarantee negligible subpopulation of the MS in recombination processes, which was also verified by the intensity of recombination radiation in singlet and triplet systems of helium levels. Note that even at the pressure of additive $p_{mol} \sim 10^{-2}$ Torr, the intensity of recombination radiation falls by a factor of greater than an order and its duration substantially reduces. This is explained by recharge of helium ions on additive molecules with their following recombination. At $p_{mol} \sim 10^{-1}$ Torr, the recombination radiation of helium actually ceases.

Figure 7 shows an example recovery curve for a medium transparency in the test cavity after the excitation pulse stops. The rate constant k_e^m for electron de-excitation of the $He(2\,^1S_0)$ state calculated from this data was $(3.8\pm0.4)\times10^{-7}$ cm^3 s^{-1}, which within an acceptable accuracy coincides with a reference data ($k_e^m = 4\times10^{-7}$ cm^3 s^{-1} [33]). In the same figure, the ratio w_{out}/w_{in} is shown versus the time delay Δt for the mixture $He-H_2O$. Similar dependences were obtained for the mixtures of helium with NH_3, CO_2, and N_2O, which stronger affect the rate of lasing recovery in the second pulse. The relaxation rates of the MS $He(2\,^1S_0)$ found from these results are shown in Fig. 8 versus the admixture pressure, and the quenching constants are presented in Table 1, which also includes (without specifying accuracy of measurements) data from [32] obtained, as was mentioned, by indirect methods.

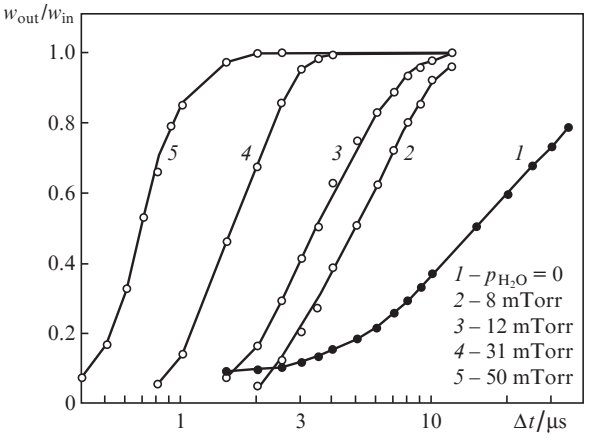

Figure 7. Ratio of energies w_{out}/w_{in} of the probe radiation at the output and input of the test medium vs. a time delay Δt between the pulses in the mixture $He-H_2O$ at various pressures of water vapours and $p_{He} = 8$ Torr.

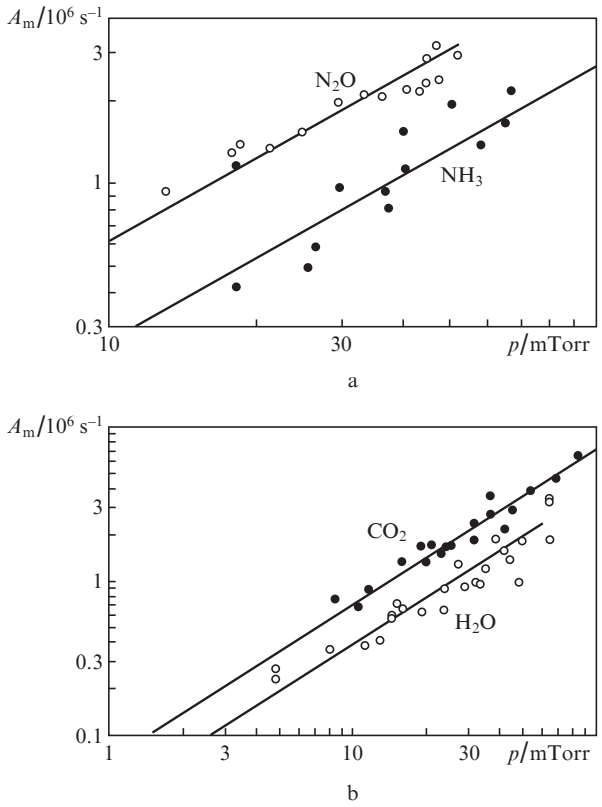

Figure 8. Rate of de-excitation of the state $He(2\,^1S_0)$ vs. the pressure of molecular additives NH_3 and N_2O (a), CO_2 and H_2O (b). Dots are experimental data, lines correspond to the calculated de-excitation constants from Table 1.

Table 1. Rate constants for $He(2\,^1S_0)$-state de-excitation by collisions with molecules.

Additive	$k_i^m/10^{-9}$ cm^3 s^{-1}	
	Present work	[32]
H_2O	1.2±0.3	–
NH_3	0.8±0.2	1.33
N_2O	1.9±0.2	0.92
CO_2	2.2±0.4	1.1

6. Discussion

6.1. Laser pulse duration in self-terminating regime

A longest duration observed in the present work τ_{max} under a linear pump power increasing with time (Fig. 4) was 50 ns. This is substantially shorter than the duration $\tau_{max} = 2/A_{rm} \approx 1$ μs $\{A_{rm} = 1.974\times10^6$ s^{-1} is the transition probability for $He(2\,^1P_1^o - 2\,^1S_0)$ [36]$\}$, determined in these conditions from the model [37]. Mechanisms that make the duration of the laser pulse shorter than the duration obtained in [37] are considered in [26]. It was shown [26], that τ_{max} substantially reduces if the decay of a resonance state into the ground state is taken into account along with the subpopulation of lower metastable states by electrons. Helium as an active medium is specific in that the states $He(k\,^1P_1^o)$ with $k > 2$ are also efficiently populated by electrons [38, 39]. Since these states rapidly decay to the MS $He(2\,^1S_0)$ (see Fig. 9, Table 2) the equation for the MS population should take into account, in addition to direct

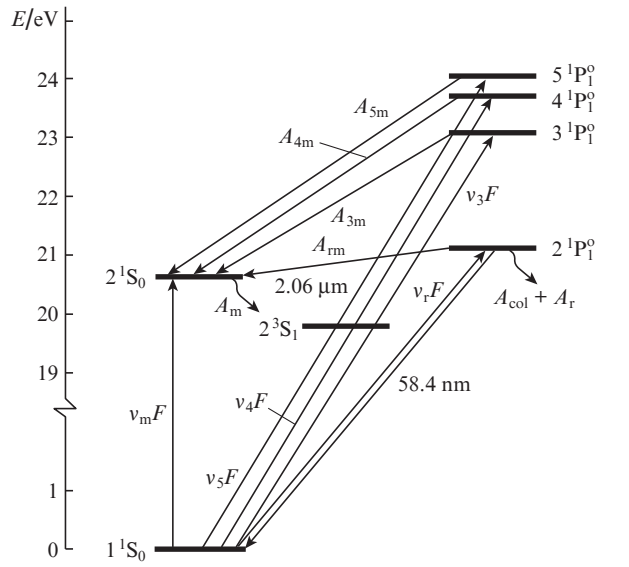

Figure 9. Atomic levels of helium and the processes under consideration.

Table 2. Relative frequencies ν_l (l = ion, m, r, k) for He atom ionisation and excitation from the ground state and probabilities A_{km} for transitions from levels $k\,^1P_1^o$ to level $2\,^1S_0$.

l	State	ν_l [39]	$A_{km}/10^6$ s^{-1} [36]
ion	$He^+(^2S_{1/2})$	1	–
m	$He(2\,^1S_0)$	0.055	–
r	$He(2\,^1P_1^o)$	0.35	1.99
$k=3$	$He(3\,^1P_1^o)$	0.11	13.4
$k=4$	$He(4\,^1P_1^o)$	0.044	6.93
$k=5$	$He(5\,^1P_1^o)$	0.022	3.92

excitation of the MS by electrons of the EB, the processes of a $He(2\,^1S_0)$ state population due to spontaneous emission from levels $He(k\,^1P_1^o)$. Finally, the system of equations describing the population of resonance and metastable states on the leading edge of the pump pulse (i.e., under the condition $n_0 \gg n_r \sim n_m \sim n_k$) takes the form:

$$\begin{aligned}
&\dot{n}_{ion} = n_0 \nu_{ion} F,\\
&\dot{n}_k = n_0 \nu_k F - n_k A_{km},\\
&\dot{n}_r = n_0 \nu_r F - n_r A_\Sigma,\\
&\dot{n}_m = n_0 \nu_m F + n_r A_{rm} + \sum_{k=3}^{5} n_k A_{km} - n_m A_m,
\end{aligned} \qquad (1)$$

where n_{ion} is the population of ions; n_0, n_r, and n_m are the populations of the ground, resonance, and metastable states, respectively; n_k (k = 3, 4, 5) are the populations of states $He(k\,^1P_1^o)$; A_{km} is the transition probability for $He(k\,^1P_1^o - 2\,^1S_0)$ [36]; $A_\Sigma = A_{km} + A_r + A_{col}$; $A_r = 0.238 A_0 \sqrt{\lambda_0/r} = 0.67\times10^6$ s^{-1} is the probability for the resonance radiation to be emitted from the tube centre [26, 40] (the tube radius is r = 2.5 cm); λ_0 = 58.4 nm; $A_0 = 1.8\times10^9$ s^{-1} is the transition probability for $He(2\,^1P_1^o - 1\,^1S_0)$ [36]; A_{col} and A_m are the probabilities of radiationless transitions from the RS and MS; F is the pump rate in numbers of interaction acts per single He atom; $\nu_{m,k,r}$ are the excitation frequencies for the corresponding helium levels from the ground state referred to the ionisation frequency ν_{ion} of helium in the ground state in the case of the EB with the energy of several keV (taken from [39]), see Table 2. The initial electron concentration $n_e(t=0)$ was taken zero.

In pure helium we have $A_{col} = A_m = 0$. Under the condition that lasing on the self-terminating transition vanishes at the instant when $\dot{n}_r/g_r = \dot{n}_m/g_m$ (g_r and g_m are the statistical weights for the RS and MS, respectively) [26], a solution of system (1) yields $\tau_{max}^{(1)} = 50$ ns at $F = F_0 t$, which coincides with the experimental result (see Fig. 4). For a rectangular pump pulse ($F = F_0$), we obtain $\tau_{max}^{(2)} = 24$ ns. Since in the mixtures of helium with H_2O and NH_3 the laser pulse duration is $\tau \gg \tau_{max}^{(1,2)}$ we may conclude that 'self-terminating' lasing transfers to 'collisional' lasing in these cases.

6.2. Mechanisms of lasing recovery in the second pulse

As was mentioned in Section 3, recovery of lasing on self-terminating transitions under electron-beam pumping is determined by the rate of MS relaxation. A most rapid de-excitation process is usually de-excitation through collisions with electrons [22]. Constant k_e^m for the state $He(2\,^1S_0)$ is extremely large and exceeds those for all known metastable states that are lower working levels in self-terminating transition lasers [22]. However, the recovery of lasing in the second pulse in pure helium occurs noticeably slower than, for example, in a lead vapour laser (λ = 722.9 nm) [30], for which Δt_1 = 0.45 μs and Δt_2 = 1.6 μs. Specificity of a He laser is a small energy difference for the states $He(2\,^1S_0)$ and $He(2\,^3S_1)$: $\Delta E \sim 0.66$ eV. The state $He(2\,^3S_1)$ is relatively slowly de-excitated by electrons $\{k_e(2\,^3S_1) = 4.2\times10^{-9}$ cm^3 s^{-1} [35]$\}$, which favours accumulation of atoms $He(2\,^3S_1)$ during the pulse. The rate of cooling of plasma electrons in elastic collisions with helium atoms $A_{eHe} = \langle\sigma_e v_e\rangle n_{He} m_e/m_{He}$ (σ_e is the cross section of plasma electrons with helium atoms, v_e and m_e are the velocity and mass of electrons, m_{He} is the mass of helium atom) is not high (1.24×10^5 s^{-1} Torr^{-1} for T_e = 1 eV). Hence, the electron effective temperature in near afterglow exceeds the value of 0.1 eV, which is characteristic for a stationary temperature T_e in a helium plasma beam [41]. The result is that the MS $He(2\,^1S_0)$ relaxes in near afterglow at a rate equal to the rate of electron cooling, which stipulates a comparatively slow recovery of lasing in the second pulse whereas the rate of lasing recovery is independent of n_e.

Introduction of molecular additives accelerates all the three processes affecting relaxation of the state $He(2\,^1S_0)$. First, due to a large cross section of elastic collisions between electrons and molecules [33, 42] and excitation of rotational and vibration molecule states [43], the cooling of electrons goes faster. Second, an additional channel arises for efficient de-excitation of the level $He(2\,^3S_1)$ by molecules [33], which also depopulates the level $He(2\,^1S_0)$. Third, the state $He(2\,^1S_0)$ quickly de-excitates directly in collisions with molecules (Table 1). However, the rates of the processes, especially of the first one, substantially differ for various kinds of gases. Excitation of vibration molecular states at $T_e > 0.5$ eV considerably increases the rate of electron cooling. Nevertheless, accumulation of molecules on vibration levels, in contrast, slows down the rate of electron cooling at low T_e because electrons acquire energy in colliding with molecules. In this case, in the critical interval of T_e for He lasers, the rate of electron cooling is determined by the rate of vibration state relaxation in mutual collisions between molecules (VT-relaxation), which is of crucial importance for lasing recovery in the second pulse. The rate of VT-relaxation raises in the series dimers – weakly polarised triatomic molecules – polar molecules [44–47], which helps bring the laser pulses closer up to their complete merge in mixtures of helium with H_2O.

6.3. A mechanism of quasi-stationary collisional lasing

As it follows from the above consideration, in the conditions of the present work, fast relaxation of the MS is implemented in two principally different processes: collisions with plasma electrons and heavy particles. In Fig. 10, we present the dependences of τ_{max}, found by solving system (1), on n_e and on the rate of the MS relaxation under collisions with molecules for various excitation conditions: by rectangular pulses and pulses in which the pump power linearly increases in time.

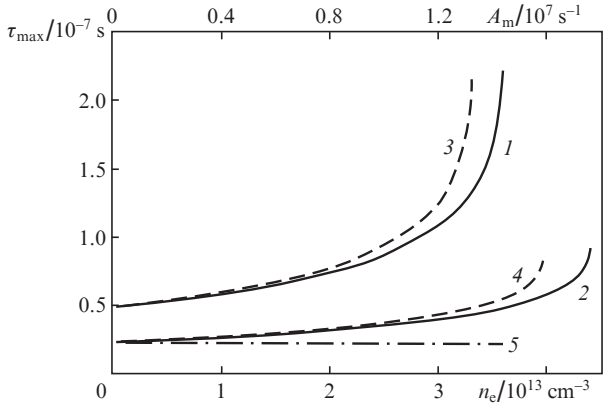

Figure 10. Maximal duration of a laser pulse τ_{max} vs. the concentration of plasma electrons n_e taking into account electron de-excitation of laser levels (*1*, *2*) and vs. the rate A_m of $He(2\,^1S_0)$-state de-excitation by molecules without RS quenching (*3*, *4*) and at $A_{col} = A_m$ (*5*). Curves (*1*) and (*3*) are obtained for a linear time dependence of the pump rate ($F = F_0 t$), curves (*2*), (*4*), (*5*) correspond to a rectangular pump pulse ($F = F_0$).

Curves (*1–4*) show that, at certain values of n_e and A_m, 'self-terminating' lasing should transfer to quasi-continuous one. However, in the experiments with pure helium, under no conditions up to $n_e \sim 10^{14}$ cm^{-3} the laser pulses were elongated. This may result from the two processes: de-excitation of the RS by plasma electrons and slow cooling of plasma electrons, which stipulates their high temperature T_e and, consequently, lack of inversion. The constant of RS quenching by electrons calculated by data from [48] ($k_e^r = 2.3\times10^{-8}$ cm^3 s^{-1}) and the measured time of lasing recovery in the second pulse (it recovers in approximately 5 μs, until the recombination processes not yet substantially reduce the concentration of electrons accumulated during the previous pulse) give evidence that the former process is of negligible importance. Hence, the second process that is a main obstacle to fast lasing recovery in the second pulse is also responsible for locking the development of quasi-continuous lasing.

On the other hand, under pumping by long rectangular pulses, lasing in mixtures of helium with H_2O and NH_3 develops with a noticeable delay Δt relative to the pump pulse leading edge (see Fig. 6). Lack of lasing for the time Δt may be explained by that the rate of relaxation of the RS in atom collisions with molecules conventionally is no smaller than that of MS relaxation (see, e.g., [33, 49]). In this case, when $A_{col} \sim A_m$, an inversion population cannot be reached [26]. As an example, in Fig. 10, curve (*5*) presents a result of solving system (*2*) under the condition $A_{col} \sim A_m$. This is only the 'onset' (in time Δt) of another process selective with respect to helium atoms, namely, electron quenching, which leads to lasing. To the instant of developed lasing, n_e according to solution of system (1) is as high as $\sim2\times10^{13}$ cm^{-3}, which corresponds to the rate of helium MS de-excitation $k_e^m n_e = 0.8\times10^7$ s^{-1}. At $k_{H_2O}^m = 1.2\times10^{-9}$ cm^3 s^{-1} and the concentration $n_{H_2O} = 10^{16}$ cm^{-3} ($p_{H_2O} = 0.3$ Torr) the probability $A_m = k_{H_2O}^m n_{H_2O}$ is $\sim1.2\times10^7$ s^{-1}, which is comparable to the rate of MS de-excitation by electrons. Thus, a quasi-stationary inversion in He lasers occurs under a combined action of the two universal mechanisms of lower working state de-excitation: by plasma electrons and by molecular gases. The influence of plasma electrons is responsible for the higher rate of de-excitation of the metastable state $He(2\,^1S_0)$ by molecules CO_2 and N_2O than that by molecules NH_3 and H_2O; however, no quasi-continuous lasing is observed with the former molecules, and the second-pulse generation develops with a delay due to a comparatively high temperature of the second relaxant – electrons.

7. Conclusions

In the present work, lasing on the transition $He(2\,^1P_1^o - 2\,^1S_0)$ at $\lambda = 2.058$ μm is studied in pure helium and in its mixtures with H_2, N_2, O_2, CO_2, H_2O, NH_3, and N_2O under electron-beam excitation by both single (with a duration of 25–1200 ns) and double pulses. In pure helium, typical 'self-terminating' lasing was observed with a maximal pulse duration of 50 ns. A recovery of lasing in the second pulse occurs with the second pulse time delay of above 1.25 μs. Introduction of additives CO_2, N_2O, NH_3, and H_2O increases the relaxation rate of MS $He(2\,^1S_0)$ population and shortens the time of lasing recovery in the second pulse up to overlapping of the pulses in mixtures $He-H_2O$. With the exciting pulse duration of ~1.2 μs in mixtures of helium with NH_3 and H_2O, lasing with the duration of ~0.8 μs was obtained, which indicates that a collision quasi-continuous operation regime is realised. Such a regime occurs under a combined action of the two universal de-excitation mechanisms for the lower working state of an atom: by plasma electrons and molecular gases. Hence, the assumption that in [24] the transfer from self-terminating to collisional lasing was realised by the new method for the first time, has been theoretically and experimentally confirmed. This opens a possibility for obtaining collisional lasing in other active media and, consequently, in other spectral ranges including the visible range.

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