 

***DISCUSSION***

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# Critical prepulse densities of electrons and metastable states in copper vapour lasers

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*Abstract*. **Two mechanisms of population inversion quenching were considered based on a detailed model of the active medium of a copper vapour laser. One of them is due to a high prepulse density of metastable states and the other is due to a high prepulse electron density. For a given pumping system, lasing was shown to terminate when the initial densities of metastable states or electrons exceeded some critical values. The results of calculations of the critical initial electron density are consistent with our simple estimates proposed earlier.**

***Keywords***: *copper vapour laser, pulsed mode, kinetic model*

## 1. Introduction

The fundamental question of what limits the pulse repetition rate in copper vapour lasers (CVLs) is still debated by scientists. In the works by G.G.Petrash et al., the main limitation is assigned to a high prepulse density of the lower metastable atomic working level, while in the works by P.A.Bokhan et al., to a high prepulse electron density. The history of the problem was discussed in detail in review [1]. It was shown [2] that there exists a critical electron density $N_{\rm e\,cr}$ in a CVL which limits the pulse repetition rate. This limitation appears because for $N_{\rm e} > N_{\rm e\,cr}$ the electron temperature does not reach the critical value $T_{\rm e\,cr} \approx 1.5 - 2$ eV, which is required for the production of population inversion, during the pulse that heats the plasma. The value of $N_{\rm e\,cr}$ was estimated in terms of the plasma parameters and the experimentally determined peak density of the pumping current in paper [2], where simple calculations confirming this estimate were also performed. Calculations carried out in the framework of more sophisticated models [3 – 5] confirm the results [2].

However, the existence of the critical electron density is questioned in a recent paper [6] entirely devoted to the critical analysis of Ref. [2]. In this connection there is good reason to revert to this problem and demonstrate the existence of $N_{\rm e\,cr}$ in the framework of a rather detailed kinetic model of the active medium.

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## 2. Kinetic model

We constructed a comprehensive model of the active medium which describes the time variation of the volume-averaged populations of levels of atomic copper and neon, the density of copper and neon ions, the electron temperature, and the intensity of laser radiation at the green and yellow lines of copper. This model was constructed in the traditional way (see, for example, Refs [7 – 9]).

### 2.1 Kinetics of excited states

The description of excited-state kinetics of copper atoms is based on the data on transition rates [10]. The model contains the population-density balance equations for nine states of atomic copper: $\mathrm{Cu}(4\,^2S_{1/2})$, $\mathrm{Cu}(4s^2\,^2D_{5/2})$, $\mathrm{Cu}(4s^2\,^2D_{3/2})$, $\mathrm{Cu}(4\,^2P_{1/2})$, $\mathrm{Cu}(4\,^2P_{3/2})$, $\mathrm{Cu}(5\,^2S_{1/2})$; for two states combining respectively three states $[\mathrm{Cu}(^4P^{\circ}, {}^4D^{\circ}, {}^4F^{\circ})]$ of $\mathrm{Cu}^*$ and four $[\mathrm{Cu}(5\,^2P_{3/2}, 5\,^2P_{1/2}, 4\,^2D_{5/2}, 4\,^2D_{3/2})]$ closely spaced levels of $\mathrm{Cu}^{**}$; and also for the ground state of $\mathrm{Cu}^+$ ions. For neon, account was taken of the ground (Ne) and first excited ($\mathrm{Ne}^*$) atomic state, and also of the ground state of $\mathrm{Ne}^+$ ions.

In Ref. [10], a bi-Maxwellian distribution function was employed to obtain the excitation rates for resonance and metastable levels. This means that a distribution function discontinuous at 16.6 eV (the energy of the first excited state of Ne) was used whose shape below the discontinuity energy and above it corresponded to the Maxwellian one, but the temperatures corresponding to this shape were calculated separately. The magnitude of the discontinuity is determined by the distribution-function normalisation. We used in our model the conventional Maxwellian energy distribution function. Test calculations showed that both approaches give almost coincident excitation rates for copper levels in the parameter range of interest.

The rates of reactions involving neon corresponded to the rates which we previously used to model XeCl [13], ArF [14], and XeF [15] lasers (see also review [16]) and KrCl lamps [17, 18] containing neon as a buffer gas.

The effect of ambipolar diffusion on the electron density was taken into account by adding the corresponding term to the balance equation:

$$\frac{\mathrm{d}N_{\rm e}}{\mathrm{d}t} = \ldots - \frac{0.929\mu T_{\rm e}}{R^2 T_{\rm g}^{1/2}}\, N_{\rm e} + \ldots .$$

Here, $N_{\rm e} = N_{\rm i\,Cu} + N_{\rm i\,Ne}$ is the electron density and $N_{\rm i\,Cu}$, $N_{\rm i\,Ne}$ are the copper and neon ion densities (in $\mathrm{cm}^{-3}$); $t$ is the time (in s); $\mu = 7$ cm$^2$ (V s)$^{-1}$ is the mobility of copper ions in neon; $T_e$, $T_g$ are the electron and gas temperatures (in eV); and $R$ is the tube radius (in cm).

### 2.2 Radiative transitions

The spontaneous radiative transition probabilities for excited states of copper were calculated from the data of Refs [10, 19]. The radiation reabsorption for transitions to the ground state was taken into account by introducing escape factors (see, for instance, Ref. [8]).

In accordance with the experimental widths of the $Cu(P_{3/2}) \rightarrow Cu(D_{5/2})$ ($\Delta\nu = 6$ Hz, $\lambda = 510.6$ nm) and $Cu(P_{1/2}) \rightarrow Cu(D_{3/2})$ ($\Delta\nu = 9$ GHz, $\lambda = 578.2$ nm) transitions given in Ref. [10], we used the following amplification cross sections for laser transitions at 510.6 and 578.2 nm, respectively: $\sigma_{ph} = 3.37 \times 10^{-14}$ and $2.81 \times 10^{-14}$ cm$^2$.

We considered two equations for the intensities $I(\lambda)$ of two laser lines in the zero-dimensional approximation (for more details, see Refs [7, 9]):

$$\frac{dI(\lambda)}{dt} = \sigma_{ph}(\lambda)c\left(N_b - \frac{g_b}{g_a}N_a\right)I(\lambda) - \gamma I(\lambda) - wI(\lambda) + AN_b\frac{\hbar c}{2\pi\lambda}\frac{\Delta\Omega}{4\pi},$$

where $N_b$ and $N_a$ are the populations of the upper and lower laser levels, respectively; $\gamma = (c/2l)\ln(1/r_1 r_2)$ is the reciprocal lifetime of a photon in the cavity; $c$ is the velocity of light; $l$ is the length of the excited medium; $r_i$ are the reflectivites of the cavity mirrors; $w$ is the coefficient of radiation losses in the cavity arising from the absorption in the optical elements; the last term represents the laser radiation seeding due to the spontaneous emission into a solid angle $\Delta\Omega$.

To consider the population inversion, provision was also made for the regime in which stimulated radiation has no effect on level populations.

### 2.3 Thermal balance

We described the electron thermal balance by the equation

$$\frac{d}{dt}\left(\frac{3}{2}N_e T_e\right) = -Q_{i\,Cu} - Q_{i\,Ne} - Q_{wall} - Q_{\Delta T} + \rho j^2(t). \quad (1)$$

Here, $Q_{i\,Cu}$ and $Q_{i\,Ne}$ are the power densities spent for the ionisation of copper and neon, respectively (these quantities are expressed in terms of the populations of excited atomic states [7–9], and they take into account the excitation and deexcitation of the levels included in the model);

$$Q_{wall} = \frac{5.41 \times 10^4 T_e^{1.5}}{R^2 \sigma_{e\,Ne}}\frac{N_e}{N_{Ne}}\ \mathrm{W\ cm^{-3}}$$

is the power of heat removal to the walls (where $T_e$ is in eV, $\sigma_{e\,Ne}(T_e)$ is the transport cross section for elastic collisions of electrons with neon atoms in units of $10^{-16}$ cm$^2$, which weakly depends on the temperature in the 2 eV range and is approximately equal to $1.5 \times 10^{-16}$ cm$^2$ [20], and $R$ is the tube radius in cm);

$$Q_{\Delta T} = 2\left[\frac{m_e}{m_{Ne}}k_{Ne}N_{Ne} + \frac{m_e}{m_{Cu}}k_{ei}N_e\right]N_e(T_e - T_g)$$

is power density spent for cooling electrons through elastic collisions with neon atoms and copper ions; $k_{Ne}$ and $k_{ei}$ are the rates of elastic collisions of electrons with neon atoms and ions; $N_{Ne}$ is the density of neon atoms; $m_e$ is the electron mass; $m_{Ne}$ and $m_{Cu}$ are the masses of neon and copper atoms;

$$\rho = \frac{1}{\sigma} \approx \frac{7.5 \times 10^{-2}}{T_e^{1/2}}\left(\frac{1}{T_e} + 5.2 \times 10^{-3}\frac{N_{Ne}}{N_e}T_e\right)$$

is the specific plasma resistance [21] in $\Omega$ cm; $\sigma$ is the plasma conductivity; and $j$ is the electric current density.

The balance equation for the gas temperature was not considered, because the equation integration time does not exceed the interval between the pulses, during which the gas temperature is almost constant.

The density $j$ of electric current through the active medium in the model was described in two ways. One involved the solution of the Kirchhoff equations for the electric circuit simultaneously with the solution of the kinetic equations for the densities of different reagents and the balance equations for the electron temperature. The other employed directly the experimental time dependence of the current. This was especially significant in the testing of the kinetic model with the aid of experimental data.

The transient equations for the concentrations of different reagents, the balance equations for the electron temperature, and (where necessary) the Kirchhoff equations for the electric circuit (22 equations in all) were solved employing the PLAZER code package [9, 16]. The model took 107 kinetic reactions into account.

### 2.4 On the model testing

The model tested by comparing with the calculations performed in Ref. [10]. A good agreement was observed, despite some distinctions between the models noted above.

We encountered several difficulties when testing the model with the aid of experimental data. The matter is that, although the total number of experimental papers on CVLs is very large (see, for instance, books [22, 23]), combined measurements of the parameters of the active medium were performed only in the works of Petrash and collaborators [1, 24–28], Piper and collaborators [10], and in Hogan's thesis [29]. However, the data provided by these papers are also inadequate for a full analysis. For instance, the initial prepulse concentrations of electrons and copper atoms in the ground state were not measured in Ref. [10]. By comparing our calculations with the calculations and the experimental data from this work, we took these concentrations equal to the estimative values employed in the calculations [10]. The time dependences of output laser power were also not measured in [10]. In Refs [27, 28], no measurements were made of the time dependences of the densities of the upper and lower states for the yellow line; furthermore, measurements were performed only in the small-signal gain mode.

Nevertheless, by using the experimental time dependences of the current, we obtained good agreement between the copper level populations, the electron density, and the output power at the 510 and 578-nm lines and the available experimental data [10, 27–29].

## 3. Effect of initial electron density and metastable states of copper on the quenching of lasing

### 3.1 Electric circuit

To clarify the question of whether there exists a critical initial electron density, we selected one of the versions of a three-circuit electric circuit (Fig. 1, see also Ref. [3]). This circuit is described by the following equations for currents and voltages:

$$-(L_1 + L_2)\frac{\mathrm{d}I_1}{\mathrm{d}t} = I_1 R_{\mathrm{T}} - U_1, \quad \frac{\mathrm{d}U_1}{\mathrm{d}t} = -\frac{I_1 + I_2}{C_1},$$

$$-L_4 \frac{\mathrm{d}I_2}{\mathrm{d}t} = -U_1 + U_3 - U_2, \quad \frac{\mathrm{d}U_2}{\mathrm{d}t} = -\frac{I_2}{C_2},$$

$$-L_3 \frac{\mathrm{d}I_3}{\mathrm{d}t} = I_3 R_{\mathrm{d}} - U_3, \quad \frac{\mathrm{d}U_3}{\mathrm{d}t} = \frac{I_2 - I_3}{C_{\mathrm{c}}}.$$

Here, $I_1$, $I_2$, and $I_3$ are the currents in the first, second, and third circuits, respectively [in this case, the current $I_3$ flows through the gas-discharge tube with a resistance $R_{\mathrm{d}}(N_{\mathrm{e}}, T_{\mathrm{e}})$]; and $U_1$, $U_2$, and $U_3$ are the voltages across the corresponding capacitors.

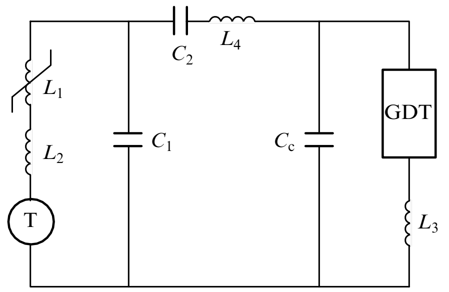


**Figure 1.** The electric circuit used in the calculations: (GDT) gas-discharge tube; (T) thyratron; the inductances $L_1 = 27.2$ μH, $L_2 = 1.5$ μH, $L_3 = 3.2$ μH, and $L_4 = 20$ nH; the capacitances $C_1 = 1.5$ nF, $C_2 = 1$ nF, $C_{\mathrm{c}} = 0.235$ nF.

The equations for currents and voltages in the electric circuit were solved simultaneously with the kinetic equations, from which we calculated the tube resistance $R_{\mathrm{d}} = \rho l/S$ from the resultant temperature and electron density, where $l = 40$ cm is the tube length and $S = \pi R^2$ is its cross section (the tube radius is $R = 1$ cm). The voltage across the tube was calculated by the Ohm law: $U_{\mathrm{d}} = R_{\mathrm{d}} I_3$.

The initial voltage across the capacitors $U_1 = U_2 = 14$ kV was taken as the initial conditions for the circuit; after actuation of the thyratron T and change of sign of the voltage across the capacitor $C_1$, the total voltage across the two capacitors was about 28 kV. It was assumed that the currents were absent at the initial instant of time: $I_1(0) = I_2(0) = I_3(0) = 0$.

The time dependence of the thyratron resistance was expressed in terms of the time dependence of the current $I_1(t)$ flowing through the thyratron,

$$R_{\mathrm{T}}(t) = R_0 + R_1 \exp\left[-\frac{4I_1(t)}{I_0}\right],$$

in which the following parameters were used: $R_0 = 5\ \Omega$, $R_1 = 2$ M$\Omega$, and $I_0 = 1$ A. The time dependence of the variable inductance was also expressed in terms of the time dependence of the current $I_1(t)$,

$$L_1(t) = L_1 \exp\left[-0.1\left(\frac{I_1(t)}{I_0}\right)^4\right].$$

The following values were taken for fixed-value inductances and capacitances: $L_1 = 27.2$ μH, $L_2 = 1.5$ μH, $L_3 = 3.2$ μH, $L_4 = 20$ nH, $C_1 = 1.5$ nF, $C_2 = 1$ nF, and $C_{\mathrm{c}} = 0.235$ nF. In the results outlined below, it was assumed that $N_{\mathrm{Cu}}(0) = 0.6 \times 10^{15}$ cm$^{-3}$ and $N_{\mathrm{Ne}} = 1.62 \times 10^{18}$ cm$^{-3}$.

### 3.2 Quenching of lasing with increasing initial electron density

The critical electron density was expressed in terms of the experimental value of the peak current $j_{\max}$:

$$N_{\mathrm{e\,cr}} = N_{\mathrm{e\,cr}}^{(0)}\left[a + \left(a^2 + 1\right)^{1/2}\right]. \quad (2)$$

Here,

$$N_{\mathrm{e\,cr}}^{(0)} = \frac{j_{\max}}{e}\left(\frac{m_{\mathrm{e}} k_{\mathrm{Ne}}(T_{\mathrm{e\,cr}}) N_{\mathrm{Ne}}}{J_{\mathrm{Cu}} k_{\mathrm{i\,Cu}} N_{\mathrm{Cu}}}\right)^{1/2}$$

is the critical electron density in the case when the conductivity is determined by collisions with neutrals; $a = k_{ei}(T_{\mathrm{e\,cr}}) N_{\mathrm{e\,cr}}^{(0)} / (k_{\mathrm{Ne}}(T_{\mathrm{e\,cr}}) N_{\mathrm{Ne}})$ is a dimensionless quantity that is significant when Coulomb collisions make an appreciable contribution to the conductivity; $k_{\mathrm{i\,Cu}}$ (in cm$^3$ $\times$s$^{-1}$) is the ionisation rate for copper atoms; and $J_{\mathrm{i\,Cu}} = 7.73$ eV is ionisation energy of copper. In this case, lasing was assumed to terminate if the electron temperature during the pulse does not exceed the critical value $T_{\mathrm{e\,cr}} \approx 2$ eV.

Calculations performed using our detailed model illustrate the fact that the enhanced initial electron density lowers the maximum electron temperature during the pump pulse, resulting in the quenching of lasing (Figs 2 and 3). Indeed, for a relatively low initial electron density $N_{\mathrm{e}}(0) = 4 \times 10^{13}$ cm$^{-3}$, the maximum electron temperature is significantly greater than $T_{\mathrm{e\,cr}}$, and the population inversion takes place (Fig. 2). In Fig. 2, $g(P_{3/2}) = 4$ and $g(D_{5/2}) = 6$ are the statistical weights of the corresponding states, the initial electron density is $N_{\mathrm{e}}(0) = 4 \times 10^{13}$ cm$^{-3}$, the initial population density of the metastable $D_{5/2}$ level is $N_{D_{5/2}}(0) = 2.1 \times 10^{13}$ cm$^{-3}$, the initial population density of the metastable $D_{3/2}$ level is $N_{D_{3/2}}(0) = 0.62 \times 10^{13}$ cm$^{-3}$, the initial density of copper atoms in the ground state is $N_{\mathrm{Cu}} = 0.6 \times 10^{15}$ cm$^{-3}$, and the neon density is $N_{\mathrm{Ne}} = 1.62 \times 10^{18}$ cm$^{-3}$. The initial densities of the metastable levels were prescribed in accordance with the Boltzmann distribution corresponding to an electron temperature of 0.31 eV. This temperature was realised in the afterglow at the instant of time when the electron density was $4 \times 10^{13}$cm$^{-3}$. For a higher initial density [$N_{\mathrm{e}}(0) = 4\times 10^{14}$ cm$^{-3}$], the electron temperature even at its peak was noticeably lower than $T_{\mathrm{e\,cr}}$, and the population inversion does not occur (Fig. 3).

To determine the critical electron density, we performed a series of calculations with different initial electron densities. The initial density of the metastable states was assumed zero. Different initial electron densities correspond to different pump pulse repetition rates (for more details, see

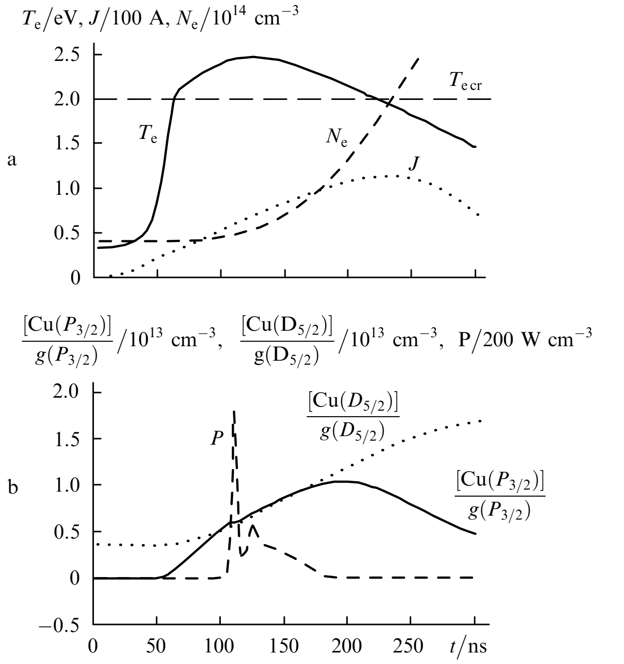


**Figure 2.** Time dependences of the electron temperature $T_e$, the current through the discharge tube $J$, and the electron density $N_e$ (a), and also of the population density of the resonance level $[Cu(P_{3/2})]/g(P_{3/2})$, the population density of the metastable level $[Cu(D_{5/2})]/g(D_{5/2})$, and the specific total output laser power in two lines $P$ (b).

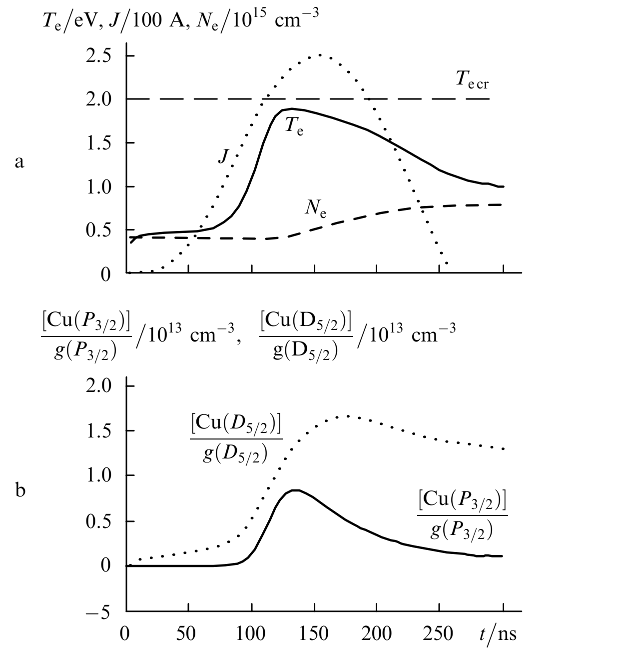


**Figure 3.** The same as in Fig. 2 for an above-critical initial electron density, $N_e = 4 \times 10^{14}$ cm$^{-3}$. The initial densities of the metastable states were assumed to be zero.

Ref. [2]). As expected, according to Ref. [2], the lasing terminates when the initial electron density exceeds $N_{e\,cr} \approx 2 \times 10^{14}$ cm$^{-3}$ (see Fig. 4).

We will compare this result with the estimate obtained from expression (2). Under the conditions assumed in the calculation, we have $j_{max} = 67.5$ A cm$^{-2}$, $N_{Cu} = 0.6 \times 10^{15}$ cm$^{-3}$, and $N_{Ne} = 1.62 \times 10^{18}$ cm$^{-3}$. Hence, expression

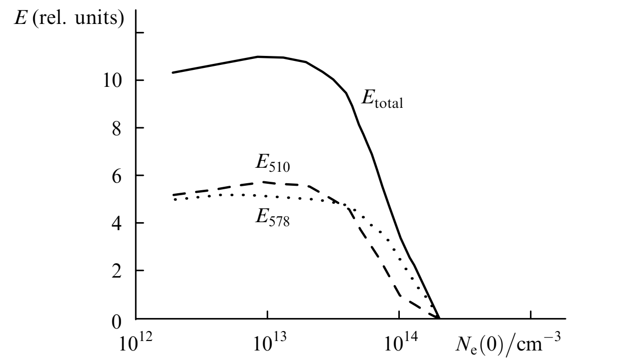


**Figure 4.** Output laser pulse energy $E$ at the 510 and 578-nm lines and the total output laser energy $E_{total}$ as functions of the initial electron density. The initial populations of the metastable states were assumed to be zero.

(2) gives $N_{e\,cr} = 2.27 \times 10^{14}$ cm$^{-3}$. This value is in good agreement with the value ($N_{e\,cr} = 2 \times 10^{14}$ cm$^{-3}$) obtained using the detailed model.

Thus, the above results confirm the conclusion of Ref. [2] that there exists a critical value of the prepulse electron density above which the quenching of lasing occurs. Moreover, the estimate of the critical electron density is consistent with the results of calculations based on the detailed model.

### 3.3 Quenching of lasing with increasing the initial density of metastable states

In another series of calculations, we changed the initial populations of the metastable states $N_{D_{5/2}}(t=0)$ and $N_{D_{3/2}}(t=0)$ in the $D_{5/2}$ and $D_{3/2}$ states. For simplicity they were assumed to be $N_{D_{5/2}}(t=0) = N_{D_{3/2}}(t=0) = N_m$. The initial electron density was $N_e(0) = 4 \times 10^{13}$ cm$^{-3}$.

The calculations show that the lasing terminates at a rather high initial density of the metastable states $N_m \approx (2-3) \times 10^{13}$ cm$^{-3}$ (Fig. 5), which is close to the initial electron density. The calculations also revealed that the critical density of the metastable states remained invariable upon a ten-fold decrease in the initial electron density [down to $N_e(0) = 4 \times 10^{12}$ cm$^{-3}$] and hence far exceeded $N_e(0)$.

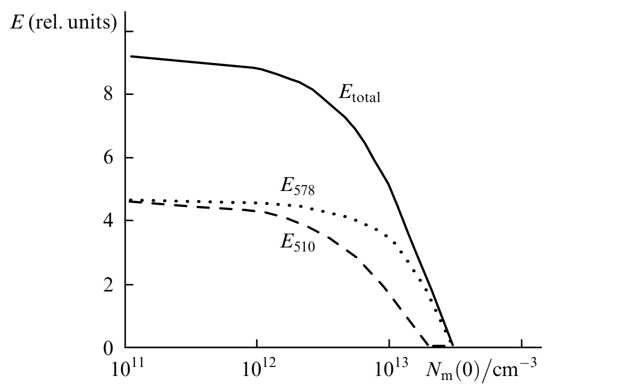


**Figure 5.** Output laser energy $E$ at the 510 and 578 nm lines and the total output laser energy $E_{total}$ as functions of the initial population density $N_m$ of the metastable states. The initial electron density was assumed to be $4 \times 10^{13}$ cm$^{-3}$.

We also conducted a series of calculations in which the initial electron density was varied, while the initial density of the metastable states was taken in accordance with the Boltzmann distribution for the initial electron density $T_e(0)$. The latter was assumed equal to the temperature $T_e(t)$ at the instant of the afterglow $t$ at which the given electron density $N_e(t)$ corresponds to the initial electron density for the pump pulse $N_e(0)$. The dependences $N_e(t)$ and $T_e(t)$ in the afterglow were given in Ref. [2]. The afterglow calculations conducted within the framework of the model presented in our work agree with the results of Ref. [2]. In this case, the contribution of ambipolar diffusion and electron heat conduction to the dependences $N_e(t)$ and $T_e(t)$ was less than 5 % for tubes with a radius of about 1 cm.

The results of calculations with a nonzero initial density of the metastable states are presented in Fig. 6. The quenching of lasing occurs for lower (by a factor of 2.5 – 3) initial electron densities than for a zero initial density of the metastable states. For the given density of copper atoms and the given pumping conditions, the limitations related to the initial density of the metastable states prove to be more significant.

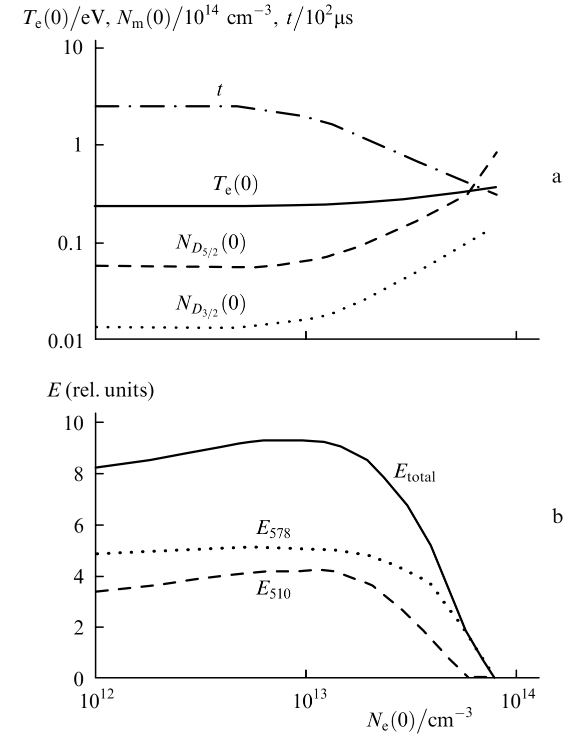


**Figure 6.** Initial electron density is plotted as abscissa. Plotted as ordinate are initial electron temperature $T_e(0)$, initial population density of the metastable $D_{5/2}$ state for the 510 nm line $N_{D_{5/2}}(0)$, initial population density of the metastable $D_{3/2}$ state for the 578 nm line $N_{D_{3/2}}(0)$, and point in time $t$ at which the electron density plotted as abscissa is realised in the solution of the equations for the electron temperature and density during the afterglow (a), and also output laser energy $E$ at the 510 and 578 nm lines and the total output laser energy $E_{total}$ (b).

# 4. Discussion of results

## 4.1 On the limitation mechanisms

The mechanism of limitations of the initial density of the metastable states is evident. It is related to the fact that the population of the resonance level has a maximum during the pump pulse. If even the initial population of the metastable state is high enough, its population throughout the pulse can also exceed the population of the resonance level. Note, however, that the critical density of the metastable states has not been estimated or calculated previously.

The mechanism of limitations in initial electron density is also clear enough [2]. It is due to the fact that population inversion necessitates a relatively high electron temperature $T_{e\,cr} \approx 2$ eV defined by the resonance-to-metastable state excitation cross section ratio. For $T_e < T_{e\,cr}$, the population of the metastable states prevails. Since the energy inputted into the medium is limited, for instance by the capacities of the pumping system in use, the critical temperature may not be attained when the initial electron density $N_e(0) > N_{e\,cr}$ is high enough. This is clear even from the fact that the heat capacity of the electron gas increases with electron density. Even if it is assumed that all of the energy inputted into the medium is spent for electron heating, the electron temperature would not exceed some maximum value

$$T_{e\,max} \sim \rho j_{max}^2 \Delta t N_e^{-1} \quad (3)$$

for a limited energy of the pump source, where $\Delta t$ is the characteristic time of a pump pulse. In this case, the maximum temperature would be lower than the critical one when the electron density is high enough,

$$N_e > \rho j_{max}^2 \Delta t T_{e\,cr}^{-1} \,. \quad (4)$$

Of course, energy is mainly spent to excite and ionise atomic copper, rather than to heat the electrons. That is why the electron density limitation (4) is not rigorous enough. It merely implies the existence of the critical electron density for a limited energy of the pump source. For more exact estimates, expression (2) should be used.

The foregoing is clear enough. Ambiguities may arise only in determining specific domains of medium and pump parameters dominated by one or other mechanism. However, the author of a recent paper [6] casts doubt on the very existence of critical electron density. Below, we show that the main critical statements of Ref. [6] arise from misunderstanding.

## 4.2 On the estimate of critical electron density

As already noted, in Ref. [2] the critical electron density was estimated from the experimental value of the peak current $j_{max}$. In Ref. [6], the thermal balance equation, which is the initial equation for this estimate, was written in a different form to express the current density in terms of the field intensity employing the Ohm law $E = j/\sigma$. Then, the following statement was made: 'One can see that Eqn (11) does not contain $N_{e\,cr}$ at all and this equation can be written for any instant of time. This means that in the model adopted by us, the power introduced for any $N_e$ may exceed the ionisation loss as well as any other losses proportional to $N_e$ for an appropriate choice of $E$, which in turn indicates that $N_{e\,cr}$ does not exist and inversion can be obtained for any value of $N_e(0)$. Thus, without changing the approach, we arrive at the opposite conclusion concerning $N_{e\,cr}$ taking into account the dependence of $j$ on $N_e$ and expressing the introduced power in terms of an extrinsic parameter.'

The confusion in the statement of Ref. [6] cited above was caused by the following. In the context of the estimates of Ref. [2], the value of peak current $j_{max}$ is taken from experiment and therefore is an independent variable from the mathematical standpoint. From the experimental value $j_{max}$, we can judge whether the temperature could exceed the critical value $T_{e\,cr}$ during the pumping. To a given $j_{max}$ there corresponds a critical electron density $N_{e\,cr}$ such that, when $N_e(0) > N_{e\,cr}$, the electron temperature will not exceed the critical temperature during the pumping. Within the framework of the model considered above, this conclusion cannot be invalidated by any mathematical transformations.

Meanwhile, by expressing $j$ in terms of $E$ the author of Ref. [6] passed to *a different physical model*. Within the framework of his model, the pump source can have *an arbitrarily high power*. Indeed, for a given $E$, the power density introduced into the medium, $\sigma E^2 \propto N_e E^2$, is in fact arbitrarily high if $N_e$ is arbitrarily high. This means that if this approach is used even in the limiting estimate (3), the electron density is also absent,

$$N_e T_{e\,max} \sim \sigma E^2 \Delta t \propto N_e E^2 \Delta t, \quad T_{e\,max} \propto E^2 \Delta t,$$

and the temperature will not decrease for an arbitrarily high electron density. This is possible only when the energy input is arbitrarily high.

Thus, a correct, though trivial, answer was obtained within the framework of the model [6]: for a pump source of arbitrarily high power there exists no critical electron density (more exactly, it is infinitely high). However, this result could be immediately obtained from Ref. [2] by letting $j_{max}$ tend to infinity in expression (2) for $N_{e\,cr}$. Therefore, the model [6] confirms the results of Ref. [2] rather than invalidates them, when the formulas of Ref. [2] are correctly interpreted.

### 4.3 On the maximum power pumped into the medium

Note that several inexact statements of a more particular type were made in Ref. [6] along with the fundamental misunderstanding discussed above. For examplem, the author of paper [6] writes: 'Note also that in the above formulas, it is apparently assumed that the quantity $j^2_{max}/\sigma(T_{e\,cr})$ is the maximum power introduced into the medium over the excitation pulsed duration. However, the peak of the current does not correspond at all to the peak of the power introduced under typical operation conditions for CVL (see, for example, the oscillograms for current, voltage, and introduced power pulses in Refs [24, 28, 10]).'

This statement of Ref. [6] is in direct contradiction with the Joule law. According to this law, the power inputted into a unit volume is given by the expression $j^2/\sigma$, whose maximum corresponds to the maximum specific input power. Indeed, we are dealing with the power introduced directly into the active medium rather than with the power at the laser tube, to which parasitic inductances and capacitances make a contribution. The Joule law is as valid as long as the Ohm law can be applied, which is not questioned in Ref. [6].

### 4.4 On inelastic energy losses

The rate of inelastic energy losses was estimated in Ref. [2] as the rate of excitation of the resonance transition multiplied by the ionisation energy of copper $J_{i\,Cu} = 7.73$ eV [see expression (2)]. The author of Ref. [6] claims that this rather coarse approximation overstates the energy losses. Indeed, under the conditions under study it is impossible to rigorously introduce the inelastic energy loss rate, which does not depend explicitly on time. For an adequate description of ionisation and thermal balance, one should consider the balance equations for the population densities of many excited levels, as was done, for instance, in our work. It is clear, however, that the minimal energy loss rate under conditions under study is the product of the excitation rate of the resonance level and its excitation energy $E^*_{Cu} = 3.8$ eV. In other words, the ratio between the energy losses employed in Ref. [2] and the lowest possible energy losses (7.73:3.8) is only $\sim 2$. It is evident that the energy losses are overstated by no more than a factor of two, if at all. Furthermore, the quantity $J_{i\,Cu}$ appears in the estimate of the critical density (2) under the square root sign, so that the error arising from the possible overestimation of the energy losses does not exceed 40 %. This is quite acceptable for simplified models, which serve the purpose of revealing the essence of the limitation mechanism.

### 4.5 On the comparison with experiment

As mentioned above, the available experimental data are insufficient for the detailed simulation of the results of Refs [24–28]. For instance, the oscilloscope traces of currents are not given for a doubled current pulse. That is why the cause of the quenching of population inversion in the experiments of Ref. [25] so far cannot be unambiguously assigned to one or other mechanism. For this reason, the attitude towards the interpretation of these experiments is expressed in Ref. [2] rather cautiously: 'This does not mean that the high initial electron density is the only reason for the inversion breaking upon a small delay of paired pulses in experiments [4, 6, 15, 16]. The high initial population of the metastable levels also also impedes the inversion conditions.'

However, this cautious statement was so distorted in the quotation in Ref. [6] that it acquired a directly opposite meaning. The words 'The aforesaid does not imply' were omitted and replaced with the phrase 'From this follows the conclusion.' The phrase concerning the effect of metastable levels was also dropped out.

The experimental results of Ref. [25] are used in Refs [1, 6], as the main evidence that the pulse repetition rate is predominantly limited by the initial density of the metastable states rather than by the critical electron density. Indeed, according to these papers, the population inversion vanishes with increasing the initial density of the metastable states. However, it does not mean that the quenching of population inversion occurred only due to the high initial density of the metastable states. To demonstrate this directly, we performed calculations in which the initial density of the metastable states quenches the population inversion. Then, we lowered the initial electron density, with the effect that the population inversion was produced again. In particular, for an initial electron density of $8 \times 10^{13}$ cm$^{-3}$, the lasing at the 510 nm line terminates when the initial density of the metastable states becomes as high as $10^{13}$ cm$^{-3}$. But when the electron density is lowered to $4 \times 10^{13}$ cm$^{-3}$ for the same initial density of the metastable states, the lasing is resumed.

## 5. Conclusions

The results of our work can be formulated as follows:

We constructed a detailed model for the description of the CVL kinetics. The model was tested by comparison with earlier results and with the results of detailed measurements of plasma parameters.

The results of calculations confirm the conclusion of Ref. [2] that there exists a critical initial electron density for a given pumping system. When this density is exceeded, the CVL lasing terminates. The results of calculations are consistent with the estimates proposed in Ref. [2]. The calculations also demonstrate that there exists a critical initial density of metastable states. The limitations arising from the high initial densities of electrons and the metastable states are interrelated and can compete with each other.

Our analysis of the main statements of Ref. [6], which was entirely dedicated to the criticism of Ref. [2], revealed that this criticism was based on misunderstanding, and therefore the negation of the existence of critical initial electron density stated in Ref. [6] is inconsistent.

The question of existence of the critical initial electron density is important, because the limitations associated with the prepulse densities of electrons and metastable states should be made less stringent in different ways [2].

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