Experimental investigation of the possibility of constructing a cw electron-beam-excited recombination laser

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A report is given of a systematic experimental investigation of the possibility of constructing an electron-beam-excited laser in which the upper active level is filled as a result of recombination of a supercooled stationary plasma and the lower level is depopulated by ionization of an impurity gas [L. I. Gudzenko, M. V. Nezlin, and S. I. Yakovlenko, Sov. Phys. Tech. Phys. **18**, 1218 (1974)]. The presentation of the experimental data is preceded by an analysis of a laser system differing from other treatment of the same subject [L. I. Gudzenko, M. V. Nezlin, and S. I. Yakovlenko, *ibid*.; Yu. I. Syst'ko and S. I. Yakovlenko, Sov. J. Quantum Electron. **5**, 364 (1975); L. I. Gudzenko, L. A. Shelepin, and S. I. Yakovlenko, Sov. Phys. Usp. **17**, 848 (1975)] by complete and correct allowance for the principal elementary processes and by solution of the self-consistent problem. Systematic experiments give a clear negative result. The fundamental correctness of this result is demonstrated. A comparison is made of the present results with the calculations carried out by other authors. The reported experiments have been made possible by the generation [S. V. Antipov, M. V. Nezlin, E. N. Snezhkin, and A. S. Trubnikov, Sov. Phys. JETP **38**, 931 (1974); E. N. Snezhkin and M. V. Nezlin, Sov. Phys. JETP **46**, No. 3 (1977)] of a quasistationary supercooled "beam" plasma with an electron temperature $T_e \lesssim 0.2$ eV, helium plasma density $N_e > 10^{14}$ cm^{-3} and helium–hydrogen mixture concentration $N_0 = 10^{18}$–10^{19} cm^{-3}.

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INTRODUCTION

A quasistationary supercooled (recombining) "beam" weakly ionized helium plasma, characterized by a high electron density N_e and a low electron temperature T_e ($N_e \gtrsim 10^{14}$ cm^{-3}, $T_e \lesssim 0.2$ eV), has been produced experimentally.[1,2] A plasma with these parameters is very far from equilibrium. In particular, the concentration of excited helium atoms in such a plasma exceeds the thermodynamic-equilibrium value (given by the Saha formula) by more than 20 orders of magnitude (!). [This departure from equilibrium appears because a gas is ionized by a beam of fast electrons which does not participate in the (reverse) process of three-particle recombination.]

It would seem that such a plasma is a very suitable medium for inverting the population of excited atomic states and building a quasi-cw (or cw) electron-beam-excited recombination laser. The idea of this laser was first put forward by Gudzenko *et al.*[3] who also carried out preliminary calculations. An experimental investigation of the possibility of realization of this idea was the main task of our investigation. However, our experiments gave a clear negative result. The validity of this result became clear from our analysis of the proposed laser, which was much more comprehensive and rigorous than the earlier analyses. We found that a beam cw recombination laser, proposed by Gudzenko *et al.*,[3] and considered by Syts'ko and Yakovlenko as well as by Gudzenko *et al.*,[4] was not realistic.

A discussion of the aspects considered in our analysis also helps in understanding the properties of such an interesting physical object as a quasistationary supercooled plasma and in finding its more realistic applications, for example, in the plasma chemistry of excited atoms and molecules[5] and in experimental studies of the recombination kinetics.[6,7]

§1. GENERAL ANALYSIS OF A CW RECOMBINATION LASER UTILIZING A SUPERCOOLED BEAM PLASMA[3]

We shall now consider in detail a beam laser of the kind described above. Let us assume that a high-intensity beam of fast electrons (energy 10–20 keV, current density ~10 A/cm^2) propagates in a dense neutral gas (to be specific, we shall consider helium with an atomic concentration $N_{He} = 10^{18}$–10^{19} cm^{-3}). This beam ionizes the gas and produces a plasma. The length of the beam is assumed to be approximately equal to its ionization range and the gas density sufficiently high to ensure that the collisions of the plasma electrons with the gas atoms effectively damp out the Langmuir electron oscillations excited by the beam in the plasma.[1,2] The beam dissipates almost all its energy in "ionization losses," i.e., in transferring electrons from the ground state of the helium atoms (principal quantum number $n=1$) to the ionization continuum and to the nearest—to the ground state—excitation level with the principal quantum number $n=2$ (Fig. 1). From all the excitation levels of the helium atom, we shall consider the two active levels: a ($n=2$) and b ($n=3$); to be specific, we shall assume that these levels are nearest to the ground state (the fundamental problem of the structure of the levels will be considered a little later). The higher of these levels is filled by the recombination flux of electrons from the continuum between the excitation levels and the lower level by the flux of electrons from the upper level (recombination) and from the ground state (excitation by the electron beam and by the secondary electrons).

Population inversion can be established if the rate of depopulation of the lower level is sufficiently high. This can be ensured by adding a small amount of an impurity gas (hydrogen) in a molecular concentration N_{H_2} such

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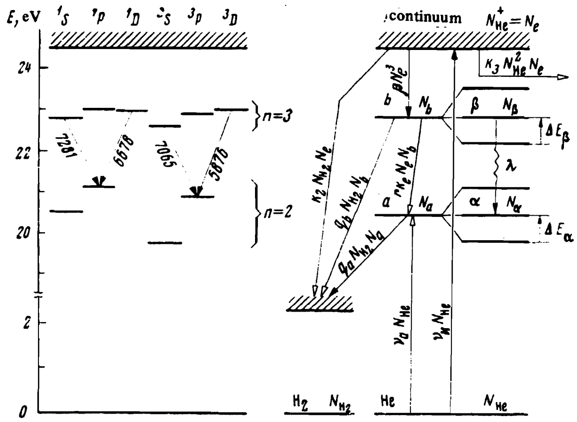

FIG. 1. Schematic representation of the processes governing the populations of the energy levels of the helium atom (on the right) and the splitting scheme of these levels (on the left).

that the ionization energy of the molecules is less than the excitation energy of the lower active level of the He atoms.[1)] The excited He atoms then ionize the H_2 molecules and these atoms then go to the ground state. We shall assume that the levels a and b are normally split into systems of sublevels $\{\alpha\}$ and $\{\beta\}$ (Fig. 1). We shall adopt a very optimistic assumption (see elsewhere for details[8–10]) that, in spite of the lack of a thermodynamic equilibrium with the plasma electrons in the system of levels $n=3$ and $n=2$, the distribution of the populations of the sublevels within each of the levels satisfies the Boltzmann law with the distribution temperature $T_d = T_e$ for $n=3$ and $T_d \geq T_e$ for $n=2$.

If the electron temperature T_e is sufficiently low compared with the energy splitting ΔE of the levels a and b (or one of them), the distribution of the populations between the sublevels is nonuniform: the highest population within the level b can be expected for the lowest of the sublevels of the system $\{\beta\}$ and the lowest population within the level a can be expected for the highest of the sublevels $\{\alpha\}$. We can easily see that the strong Boltzmann factor $\exp(\Delta E/T_d)$ makes the population inversion condition between such a pair of sublevels very much easier to satisfy than the condition for population inversion between the average a and b levels.

We shall now adopt the following assumptions.

1. The plasma electrons have a Maxwellian distribution of energies with a temperature T_e.

2. All the heavy particles—atoms, molecules, and ions—have the same temperature T and act as a thermostat (reservoir). In all the calculations given below, we shall assume that $T=0.1$ eV, as given in our earlier paper.[2]

3. The contribution of the secondary ("cascade") electrons to the ionization and excitation of the helium atoms is manifested by an effective increase in the rates of these processes by a factor of about 1.5. In all the calculations given below, we shall assume that $\nu_i = 200$ sec^{-1} and $\nu_a = 100$ sec^{-1}, which correspond to the electron beam energy of 10–20 keV and current density of 10–15 A/cm^2 [11a,11c]; here, $\nu_i = N_1\langle\sigma v\rangle_i$ is the frequency of ionization of a gas by a beam of electrons of density N_1 and velocity v, $\nu_a = N_1\langle\sigma v\rangle_a$ is the frequency of excitation of the gas atoms by the beam, σ_i and σ_a are the ionization and excitation cross sections. (It should be noted that a reduction in ν_i to 100 sec^{-1} as a result of a proportional reduction in ν_a does not greatly affect the results of calculations—see below.) These values of ν_i and ν_a correspond to our experimental conditions.[2] The proposed laser was investigated (§2) using the same apparatus as in our earlier study.[2]

4. When a helium–hydrogen mixture is ionized by a beam, formation of the hydrogen ions can be ignored because these ions are rapidly converted to the H_3^+ species, which—in its turn—recombines "instantaneously" by a dissociative mechanism.[2]

5. Of all the mechanisms which can result in the loss of the He^+ ions, only one is helpful in the establishment of population inversion: this is the three-particle recombination, in which the third body is a plasma electron:

$$He^+ + e + e \xrightarrow{\beta} He(n) + e, \qquad (1)$$

where He(n) is an excited atom in the state whose principal quantum number is n. We shall assume that the rate of this recombination is $\beta = 3\times10^{-27} T_e^{-9/2}$ cm^6/sec,[7] where T_e is in electron-volts. The presence of any other channel of disappearance of the He^+ ions will result in a reduction in the rate of filling of the upper active level with electrons.

6. The undesirable He^+ ion loss channels are:

a) charge exchange with the H_2 molecules:

$$He^+ + H_2 \xrightarrow{k_2} \begin{cases} H^+ + He + H \\ H_2^+ + He \\ HeH^+ + H \end{cases}, \qquad (2)$$

whose rate constant at $T=0.1$ eV is $k_2 \approx 1.5\times10^{-13}$ cm^3/sec (Ref. 12a);

b) conversion into the molecular ion:

$$He^+ + 2He \rightarrow He_2^+ + He \qquad (3)$$

whose rate constant at $T=0.1$ eV is $k_3 = 4\times10^{-32}$ cm^6/sec.[13a] The ionized products of the reactions (2) and (3) interact chemically with hydrogen forming the very rapidly recombining ion H_3^+ and, therefore, cease to participate in the processes of interest to us (details are given in our earlier paper[2]). Consequently, the only type of ion which remains in the plasma is the atomic species He^+, whose concentration is equal to the electron density N_e. Thus, these processes remove helium ions and, at a given rate of ionization of helium by the electron beam, strongly reduce the rate of the filling of the upper active level by recombination. It is of fundamental importance that these processes be ignored in the initial proposal[3] of the laser in question; moreover, the charge exchange process is ignored in the later calculations[4a] and a basic error is made in allowance for the conversion (see our earlier paper[2]) so that the negative influence of this process on the laser is greatly underestimated.

7. The rate of loss of the helium ions as a result of the three-particle recombination reaction (1) represents a fraction p of the total ion-formation rate and the rest, $(1-p)\nu_i N_{He}$, represents the sum of the charge-exchange and conversion processes.

We shall now consider the balance of the populations of the active levels (Fig. 1). As pointed out earlier, electrons reach the upper active level of the helium atom ($n=3$) practically only as a result of the three-particle recombination process. They are transferred from this level to the lower level as a result of collisions with the plasma electrons

$$\mathrm{He}(n=3)+e\xrightarrow{k_e}\mathrm{He}(n=2)+e+E_{\mathrm{kin}} \tag{4}$$

("electron shunting" of the lasing transition, whose rate increases on increase in the plasma density) and collisions with the helium atoms

$$\mathrm{He}(n=3)+\mathrm{He}(n=1)\xrightarrow{k_{He}}\mathrm{He}(n=2)+\mathrm{He}(n=1)+E_{\mathrm{kin}} \tag{5}$$

("gas shunting" of the lasing transition, whose rate increases with the gas density). It is important to stress that the latter process, which increases the rate of shunting of the lasing transition by a factor r ($r>1$) is also ignored in the earlier treatments.[3,4a]

Collisions with the H_2 molecules (ionization of the impurities) transfer the excited He atom to the ground state:

$$\mathrm{He}(n=3)+\mathrm{H}_2\xrightarrow{q_b}\mathrm{He}(n=1)+\mathrm{H}_2^{+}+e+E_{\mathrm{kin}} \tag{6}$$

The processes (1)–(6) govern the population of the upper active level. The lower level is filled from above by the processes (4) and (5) and from below by the excitation of He ($n=1$) by the beam and cascade electrons. The lower level is depopulated, as pointed out earlier, practically only as a result of collisions with the H_2 molecules (ionization of the impurities):

$$\mathrm{He}(n=2)+\mathrm{H}_2\xrightarrow{q_a}\mathrm{He}(n=1)+\mathrm{H}_2^{+}+e+E_{\mathrm{kin}}. \tag{7}$$

Bearing in mind these points, we obtain the following system of equations for calculating the conditions for the appearance of population inversion and of the necessary parameters of the medium:

$$\left.\begin{aligned}&(1-p)\nu_i N_{He}=k_2N_eN_{H_2}+k_3N_eN_{He}^2,\quad p\nu_iN_{He}=\beta N_e^3,\\&\beta N_e^3=rk_eN_eN_b+q_bN_{H_2}N_b,\quad (r-1)k_eN_eN_b=k_{He}N_{He}N_b,\\&rk_eN_eN_b+\nu_aN_{He}=q_aN_{H_2}N_a,\quad I_{\beta\alpha}=I_\beta-(g_\beta/g_\alpha)N_\alpha,\\&N_\beta=\frac{g_\beta}{U_b(T_e)}N_b\exp\left(-\frac{\Delta E_\beta}{T_e}\right),\quad N_\alpha=\frac{g_\alpha}{U_a(T_a)}N_a\exp\left(-\frac{\Delta E_\alpha}{T_a}\right).\end{aligned}\right\} \tag{8}$$

Here and later, N_a, N_b, N_α, and N_β are the concentrations of the excited atoms at the levels a, b and at the sublevels α, β; g_a, g_b, g_α, and g_β are the statistical weights of the levels a, b and of the sublevels α, β; $I_{\beta\alpha}$ is the inversion density; ΔE_α (or ΔE_β) is the energy gap between a given sublevel α (or β) and the lowest sublevel in the system $\{\alpha\}$ (or $\{\beta\}$); T_a is the temperature of the distribution between the sublevels $\{\alpha\}$; $T_b=T_e$ is the temperature of the distribution between the sublevels $\{\beta\}$;

$$U_a(T_a)=\sum_\alpha g_\alpha\exp\left(-\frac{\Delta E_\alpha}{T_a}\right),\quad U_b(T_e)=\sum_\beta g_\beta\exp\left(-\frac{\Delta E_\beta}{T_e}\right)$$

are the partition functions of the levels a and b (if $\Delta E_{\alpha,\beta}\ll T_e, T_a$, we have $U_a=g_a$, $U_b=g_b$, $N_\alpha=g_\alpha N_a/g_a$, $N_\beta=g_\beta N_b/g_b$); k_e, k_{He}, q_b, and q_a are the rate constants of the deexcitation of the atoms of the main gas (helium) by the processes (4)–(7), respectively, which are summed over all the sublevels, for example:

$$\left.\begin{aligned}&q_a=\frac{1}{U_a(T_a)}\sum_\alpha g_\alpha q_\alpha\exp\left(-\frac{\Delta E_\alpha}{T_a}\right),\\&k_{He}=\frac{1}{U_b(T_e)}\sum_\beta g_\beta k_{He\,\beta}\left(-\frac{\Delta E_\beta}{T_e}\right).\end{aligned}\right\} \tag{9}$$

The solution of the system (8) for the inversion density is

$$I_{\beta\alpha}=\frac{g_\beta p\nu_iN_{He}}{N_{He}^{1/3}N_{H_2}(p\nu_i/\beta)^{1/3}rk_e+N_{H_2}{}^2q_b}$$
$$\times\left\{\left[\frac{1}{U_b(T_e)\exp(\Delta E_\beta/T_e)}-\frac{\nu_a}{p\nu_i}\frac{q_b}{q_a}\frac{1}{U_a(T_a)\exp(\Delta E_\alpha/T_a)}\right]-N_{He}^{1/3}\left(\frac{p\nu_i}{\beta}\right)^{1/3}\frac{rk_e}{q_a}\frac{(1+\nu_a/p\nu_i)}{U_a(T_a)\exp(\Delta E_\alpha/T_a)}\right\}. \tag{10}$$

Equation (10) yields the necessary (but not sufficient) condition for the appearance, in principle, of population inversion ($I_{\beta\alpha}>0$):

$$R'=\frac{\exp(\Delta E_\alpha/T_a)}{\exp(\Delta E_\beta/T_e)}\frac{U_a(T_a)}{U_b(T_e)}p\frac{\nu_i}{\nu_a}\frac{q_a}{q_b}>1. \tag{11}$$

We shall now consider Eq. (11) in greater detail. We shall do this by first estimating the ratio q_a/q_b of the rates of depopulation of the lower and upper active levels of the helium atom by the ionization of the hydrogen molecules. Each of these rates depends on the partial rate constants $q_{\alpha,\beta}$ and on the distribution of the populations between the sublevels. For example, of all the sublevels of the state $n=2$, the highest value of q_α is exhibited by the resonance sublevel $2\,^1P$ but, in the case of a Boltzmann distribution of populations, its population is very low compared with the lowest sublevel $2\,^3S$ [the energy gap is $\Delta E_{2\,^1P}\approx 1.4$ eV and, at the distribution temperature $T_a\approx 0.3$ eV, we have $\exp(-\Delta E_{2\,^1P}/T_a)\approx 10^{-2}$)]. Therefore, the $n=2$ level is depopulated primarily via the $2\,^3S$ sublevel (the sublevels $2\,^1S$ and $2\,^3P$, like $2\,^1P$, make only a small contribution to q_a—see Table I) but this level is characterized by a relatively small value of q_α (approximately one-third of the "resonance" value). According to Eq. (9),

$$U_a(T_a)\,q_a\approx g_{2^3S}q_{2^3S}=3q_{2^3S}.$$

We shall now estimate the value of R' for the most convenient $3\,^3S-2\,^3P$ transition (the sublevel $3\,^3S$ is the lowest in the $n=3$ state and, consequently, has the highest population); in this case, we have $\exp(\Delta E_{3\,^3S}/T_e)=1$ (Fig. 1). Bearing in mind that in the temperature range of interest to us ($T_e\approx 0.2$–0.3 eV) we have $U_b(T_e)\geqslant 10$ and that $\nu_i/\nu_a\approx 2$,[11a] $T_a=0.3$ eV, $\Delta E_{2\,^3P}=1.14$ eV, we find from Eq. (11) and the table of the rates of the processes involved that

$$R'\leqslant\exp\left(\frac{\Delta E_{2^3P}}{T_a}\right)p\frac{\nu_i}{\nu_a}\frac{g_{2^3S}}{10}\frac{q_{2^3S}}{q_3}\approx 4p. \tag{12}$$

TABLE I.

Quantity	Dimensions	Numerical value at $T=0.1$ eV	Reference	Remarks
q_{2^3S}	cm^3/sec^{-1}	3.0×10^{-10}	[14]	Rises weakly with increasing T
q_{2^1S}	"	$\approx q_{2^3S}$	[13c]	—
q_{2^1P}	"	8.5×10^{-10}	[13b]	$\propto T^{3/10}$
$q_{n=3}$	"	2.1×10^{-9}	[15]	Practically independent of T or T_e
k_e	"	5.6×10^{-7}	[10]	$T_e \leqslant 0.3$ eV
$k^*_{He(3^3S)}$	"	3.0×10^{-10}	[16]	Rises weakly with increasing T
k_2	"	1.5×10^{-13}	[12a]	$\propto T$
k_3	cm^6/sec^{-1}	4.0×10^{-32}	[13a]	$\propto T^{-3/4}$
β	"	$3\times10^{-17}\,T_e^{-9/2}$	[7]	—
ν_i	sec^{-1}	200	[11a, c]	$j=10-15$ A/cm^2, $V_{acc}=10-20$ kV
ν_a	"	100	[11a]	

*Makes the dominant contribution to k_{He}.

The necessary condition for population inversion $R'>1$ can be satisfied only for $p>1/4$ and the sufficient condition (for $I_{\beta\alpha}>0$) can only be satisfied if $p\gtrsim 1/2$, i.e., in the range of the parameters of the helium–hydrogen mixture in which the three-particle recombination is the main channel for the disappearance of the charged particles.

We can easily see that, in the absence of a strong splitting of the active levels (corresponding formally to the condition $T_e, T_a \gg \Delta E_{\alpha,\beta}$), the necessary inversion condition is far from being satisfied. In fact, we have

$$R'\approx p\frac{\nu_i}{\nu_a}\frac{g_{2^1P}}{g_3}\frac{q_{2^1P}}{q_3}, \tag{13}$$

because—as pointed out earlier—the highest value of q_α is exhibited by the resonance sublevel $2\,^1P$. Substituting in Eq. (13) the data from Table I, we obtain

$$R'\approx {}^1/_{15}p\ll 1 \tag{13a}$$

instead of the necessary condition $R'>1$. This means that, on the average, population inversion between the helium atomic levels $n=3$ and $n=2$ cannot be achieved, as already mentioned.

It is appropriate to mention here that the condition $R'>1$ is physically analogous to the criterion $R>1$ used earlier,[3] which has a very clear physical meaning. In fact, if we ignore the charge exchange, conversion, and shunting of the $b\to a$ lasing transition, we obtain from Fig. 1 the following upper limit for the ratio of the concentrations of atoms in the states b and a:

$$\frac{N_b}{N_a}=\frac{\nu_i/q_b}{\nu_a/q_a},$$

and, from the inversion requirement $N_b/N_a>g_b/g_a$, we obtain the necessary (but, naturally, not sufficient) criterion

$$R=\frac{\nu_i q_a g_a}{\nu_a q_b g_b}>1.$$

Going over to the condition $R'>1$, we see that, in the situation described by Eq. (13a), the upper level of the investigated lasing transition is filled too slowly (because of the charge exchange and conversion) compared with the lower level and it is depopulated too rapidly (because of the shunting of the lasing transition and because $q_b\gg q_a$); consequently, if there is no splitting of the levels a and b, population inversion is impossible.

It follows that the inversion ($I_{\beta\alpha}>0$) can only be established because of the splitting of the active levels and, as indicated by the necessary condition $R'>1$, this requires a Boltzmann distribution of the populations between the sublevels with a fairly low plasma electron temperature $T_e<0.3$ eV because $T_a\approx 0.3$ eV and $T_e<T_a$.

If the necessary inversion condition $R'>1$ is satisfied, it follows from Eq. (10) that the inversion ($I_{\beta\alpha}>0$) is possible only if the impurity concentration exceeds a certain threshold. The physical meaning of this threshold is associated not only with the depopulation of the lower active level by the impurity (which depopulates even more rapidly the upper active level!) but with the need to reduce T_e by the excitation of the vibrational degrees of freedom of the impurity molecule.[2] However, if the impurity concentration is increased too much, the inversion density again increases because then not only is the depopulation of the lower level accelerated but also that of the upper level[3] and the charge exchange with the ions of the main gas (helium) is speeded up; moreover, the ionization losses of the beam electrons increase,[11] their range decreases, and the rate of cooling of the plasma electrons slows down. Therefore, we shall consider only such He–H_2 mixtures in which the H_2 concentration does not exceed that of He.

If the inverted state ($I_{\beta\alpha}>0$) is reached, the optical gain $\varkappa_{\beta\alpha}$ can be found from the following relationship (see, for example, Heard's handbook[17]):

$$\varkappa_{\beta\alpha}=2.15\cdot10^{-8}\frac{g_\alpha}{g_\beta}\frac{\lambda}{T^{1/2}}f_{\alpha\beta}I_{\beta\alpha}, \tag{14}$$

where $\varkappa$ is in reciprocal centimeters, λ is the wavelength of the $\beta\to\alpha$ transition in centimeters, $f_{\alpha\beta}$ is the oscillator strength of the same transition in the absorption case, and the temperature T is in electron-volts.

We shall now present graphically the results of an analytic solution of the self-consistent system of equa-

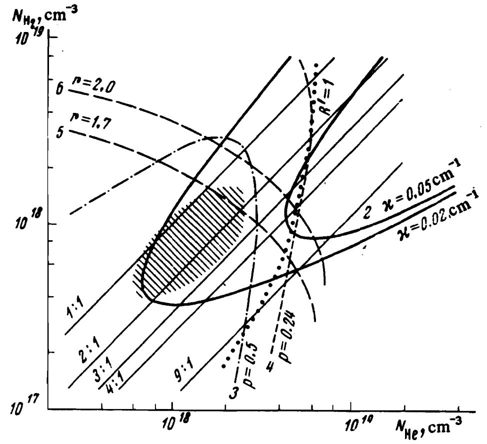

FIG. 2. Lines of constant values of p (chain), r (dashed), R' (dotted), and $\varkappa$ (continuous) for the $3\,^3S \rightarrow 2\,^3P$ (7065 Å) transition plotted in the plane of the composition of helium–hydrogen mixtures, assuming that $\nu_i = 200$ sec^{-1} and $T = 0.1$ eV. Curves 1 ($\varkappa = 0.02$ cm^{-1}) and 2 ($\varkappa = 0.05$ cm^{-1}) represent the solution of a partly self-consistent problem: $T_e = T_e(N_{He}, N_{H_2})$; $T_a = T_e$ for $T_e \geqslant 0.3$ eV, $T_a = 0.3$ eV when $T_e < 0.3$ eV; $p = 1$; $r = 1$. Curves 3 ($p = 0.5$), 4 ($p = 0.24$), 5 ($r = 1.7$), and 6 ($r = 2$) represent the solution of a self-consistent problem: $T_e = T_e(N_{He}, N_{H_2})$; $T_a = T_e$ for $T_e \geqslant 0.3$ eV, $T_a = 0.3$ eV for $T_e < 0.3$ eV; $p = p(N_{He}, N_{H_2})$; $r = r(N_{He}, N_{H_2})$. The same solution corresponds to the shaded region of the parameters of the mixture, where $\varkappa \approx 0.01$ cm^{-1}. Thin straight lines are the proportions of the components in the mixtures investigated experimentally.

tions (8) and (14) for the optical gain together with Eqs. (9) and (11) from our earlier paper[2] describing the plasma electron temperature. Table I gives the values of the rates needed in our calculations. We shall plot all the quantities of interest to us in the plane of the parameters of the working mixture N_{He} and N_{H_2}. Figure 2 shows the lines of constant p and r, the $R' = 1$ curve (on which and to the right of which there is no population inversion, i.e., $I_{\beta\alpha} < 0$), curves of constant gain $\varkappa$ (plotted without allowance for the charge exchange, conversion, and gas shunting of the lasing transition: $p = 1$, $r = 1$), as well as lines corresponding to various proportions of the helium–hydrogen mixture: 1:1; 2:1; 3:1; 4:1; 9:1. Clearly, the working region lies somewhere between the $R' = 1$ line and the $\varkappa = \varkappa_{min}$ curve, where $\varkappa_{min}$ is the minimum necessary (in accordance with the experimental conditions) value of $\varkappa$; it is also clear that this region is located in the range of the lowest possible r and the largest possible p.

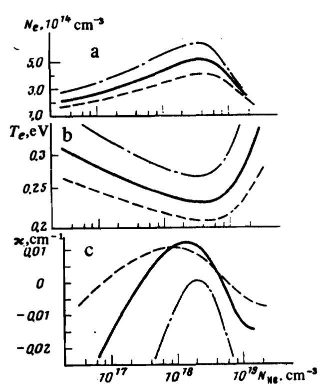

FIG. 3. Plasma density (a), electron temperature (b), and gain in the $3\,^3S \rightarrow 2\,^3P$ transition (c) plotted as a function of the helium concentration for different amounts of H_2 in helium–hydrogen mixtures (calculations). The dashed lines correspond to $N_{H_2} = N_{He}$, the continuous lines to $N_{H_2} = N_{He}/2$, and the chain lines to $N_{H_2} = N_{He}/4$. In all cases, it is assumed that $\nu_i = 200$ sec^{-1} and $T = 0.1$ eV. The self-consistent solution is $T_e = T_e(N_{He}, N_{H_2})$; $T_a = T_e$ for $T_e \geqslant 0.3$ eV, $T_a = 0.3$ eV for $T_e < 0.3$ eV; $p = p(N_{He}, N_{H_2})$; $r = r(N_{He}, N_{H_2})$.

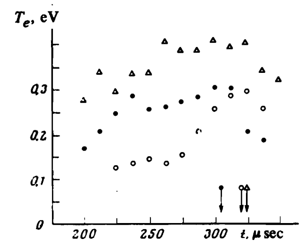

FIG. 4. Time dependences of the plasma electron temperature for different amounts of H_2 in helium–hydrogen mixtures (experimental results): ○) $N_{H_2} = N_{He}$; ●) $N_{H_2} = N_{He}/3$; △) $N_{H_2} = N_{He}/9$. In all cases it is assumed that $N_{He} \approx (2-4) \times 10^{18}$ cm^{-3} and $\nu_i = 100-200$ sec^{-1}. Time is measured from the beginning of an electron-beam pulse and the moments corresponding to the end of such a pulse are identified by arrows.

The required region can be found sufficiently reliably by calculating the dependences of the gain $\varkappa$ on the parameters of the helium–hydrogen mixture in the course of displacement along the intersecting constant-proportion lines: 1:1; 2:1; 4:1 (Fig. 2). The results obtained in this way are plotted in Fig. 3c. The variation of N_e and T_e is shown in Figs. 3a and 3b on the basis of our calculations[2] and in Figs. 4 and 5 on the basis of our earlier experiments.[2] We can see that there is a definite (relatively narrow) range of mixture parameters in which the amplification (stimulation) of light is possible: $0 \leqslant \varkappa \leqslant \varkappa_{max} \approx 1 \times 10^{-2}$ cm^{-1}; outside this range, we have $\varkappa < 0$ and light is absorbed strongly; for example, if $N_{He} \leqslant 10^{17}$ cm^{-3}, we find that $\varkappa \approx -2 \times 10^{-2}$ cm^{-1}. It must be particularly stressed that, throughout the N_{He}, N_{H_2} plane, the value of $\varkappa$ never exceeds 1×10^{-2} cm^{-1} and the maximum $\varkappa$ for a given proportion of the gases in the mixture corresponds to near-minimal T_e.

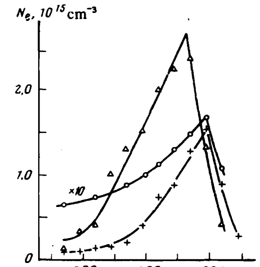

FIG. 5. Time dependences of the plasma density for different amounts of H_2 in helium–hydrogen mixtures (experimental results): ○) $N_{H_2} = N_{He}$; ●) $N_{H_2} = N_{He}/2$; △) $N_{H_2} = N_{He}/9$. In all cases it is assumed that $N_{He} \approx (2-4) \times 10^{18}$ cm^{-3} and $\nu_i = 100-200$ sec^{-1}. Time is measured from the beginning of an electron-beam pulse and the end of a pulse corresponds to the density maximum.

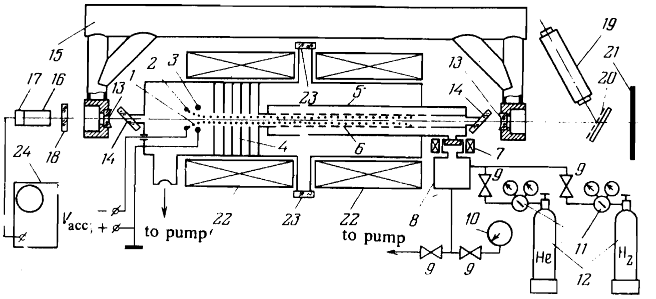

FIG. 6. Schematic diagram of the apparatus: 1) electron beam; 2) ring cathode; 3) acceleration electrode; 4) gas delay line; 5) outer tube; 6) inner tube defining the working zone; 7) electromagnetic valve; 8) reservoir below the valve; 9) gas valves; 10) manometer; 11) reducing valves; 12) gas cylinders; 13) spherical resonator mirrors; 14) Brewster windows; 15) resonator base; 16) FÉU-27 photomultiplier; 17) emitter follower; 18) interference filter; 19) OKG-11 alignment laser; 20) semitransparent mirror; 21) screen; 22) magnetic field coils; 23) windows for spectroscopic diagnostics; 24) S1-37 oscillograph.

We can also see that the He–H_2 mixture with the 4 : 1 proportion is clearly unsuitable for the amplification of light: practically everywhere we have $\varkappa < 0$; this is accounted for by the far too high value of T_e: the H_2 impurity is insufficient for the effective cooling of the electrons (see our earlier paper[2]).

If we assume (see §3) that, in the working range of the parameters, the minimum gain sufficient for a successful experiment is 1×10^{-2} cm^{-1}, we can then use Fig. 3c to plot the region shown shaded in Fig. 2. Roughly speaking, this is the required working region. At the center of this region, we have $T_e = 0.2$ eV, $N_e \approx 4\times10^{14}$ cm^{-3}, $N_{He} \approx 1.2\times10^{18}$ cm^{-3}, and $N_{H_2} \approx 0.8\times10^{18}$ cm^{-3}.

Thus, the position of the working region is largely influenced by the undesirable channels of the He^+ ion loss—the charge exchange of Eq. (2) and the conversion of Eq. (3)—which reduce directly the population of the upper active level (Fig. 1) and restrict the electron temperature from below.[2]

§2. APPARATUS AND EXPERIMENTAL RESULTS

Our experiments were carried out with the aim of detecting stimulated emission of light and we used the same apparatus (Fig. 6) as that employed earlier[2] to obtain a dense quasistationary supercooled helium–hydrogen plasma ($T_e \lesssim 0.2$ eV, Figs. 3–5), whose temperature was low enough for a systematic check of the practicality of the proposed recombination laser. At the ends of the apparatus, along the electron beam axis, there were rigidly located Brewster windows (K8 glass disks, 6 cm in diameter and 1 cm thick). The lines of force of the magnetic field diverged quite strongly near the windows and the electron beam density in this region was negligible. The windows were protected from inside by metal screens which were moved away only during a beam pulse. The windows were replaced periodically (after 200–300 shots).

Spherical mirrors with a radius of curvature 10 m and diameter of 4 cm had multilayer dielectric coatings and were placed outside the vacuum part of the apparatus; they formed on optical resonator. For each investigated line, we used a separate pair of mirrors whose transmission coefficient was less than 0.5% for the line in question. The resonator base was a steel tube, 22 cm in diameter, with walls 1 cm thick and 4.3 m long, to which angle brackets made of steel tubes (15 cm in diameter with walls 1 cm thick and 60 cm long) were attached; these brackets carried the alignment mechanisms The alignment was performed by the usual method, employing an OKG-11 helium–neon laser whose beam was injected from outside along the axis of the apparatus by a semitransparent mirror. The rigidity of the resonator was sufficient to maintain the alignment for several days; nevertheless, the alignment was regularly checked after each 20–30 shots. The distance between the resonator mirrors was 3.8 m and the distance from the mirrors to the Brewster windows was about 20 cm. The aperture of the optical system was limited by the tubes (internal diameter 2.2 cm) to which the Brewster windows were attached.

Behind one of the mirrors, there was a narrow-band interference filter, which selected the required line, and an FÉU-27 photomultiplier. The signal from this photomultiplier was recorded with an S1-37 persistent-image oscillograph. The working mixture consisted of helium and hydrogen in the proportions 1 : 1, 2 : 1, 3 : 1, 4 : 1, and 9 : 1 (the straight lines corresponding to these proportions are plotted in Fig. 2). This range of proportions was known to include the probable laser action region calculated in §1.

We used a helium–hydrogen mixture of high purity. The total impurity content of the helium did not exceed 0.0055% (0.002% N_2, 0.002% Ne, 0.0005% O_2, 0.0005% H_2O, and 0.0005% of hydrocarbons) and the impurity content of the hydrogen was 0.0036% (0.003% N_2, 0.0005% O_2, 0.0001% H_2O, and practically no other impurities). A reduction in the number of impurities (other than hydrogen) was essential because, otherwise, the charge exchange between the helium ions and the molecules of the impurity gases occurring four orders of magnitude faster (!) than with hydrogen[12] could alter decisively the properties of the medium (see §1 and our earlier paper[2]).

In all cases, the plasma regime was set by the conditions of our earlier experiments,[2] modified specially for the present task. The beam electron energy was 10–20 keV and the current was about 10 A; the intensity of an external longitudinal magnetic field in the working volume was 1200 Oe and the beam diameter was 1 cm. The duration of the beam pulses amounted to a few hundreds of microseconds (the characteristics of the beam current oscillogram were reported earlier[1]). The electron gun cathode (2 in Fig. 6) was a ring, 2.2 cm in diameter, made of a tungsten wire, 1.2 mm in diameter. The gun was outside the solenoid, where the magnetic field was approximately one-fifth of that in the working zone. This gun produced a converging electron beam which diverged again outside the working zone. Thus, the whole plasma filament allowed free longitudinal escape of light in both directions. The diameter of the plasma filament in the working zone was about 1 cm.

We sought stimulated emission as a result of the following transitions: $3\,^3S \to 2\,^3P$ (7065 Å), $3\,^3D \to 2\,^3P$ (5876 Å), $3\,^1S \to 2\,^1P$ (7281 Å), and $3\,^1D \to 2\,^1P$ (6678 Å). The electron beam current (1–20 A), electron energy (5–20 keV), and all the parameters governing the admission of gas and the proportions of the working mixture were altered very smoothly and systematically. A total of 1500 shots were made. A very definite negative result was obtained: none of these transitions resulted in stimulated emission lines under any conditions in any part of the oscillogram. A typical result is given in Fig. 7, showing oscillograms of the photomultiplier signals representing the intensity of the $\lambda = 7065$ Å line in the case when the opposite mirror was open (upper trace) and closed (lower trace). Very similar oscillograms were obtained for all the other lines under all the experimental conditions. The difference between the signals was practically unaffected by changes in the experimental conditions over a wide range and it did not vary with time, in spite of very great changes in the electron temperature and plasma concentration (Figs. 3–5). In view of this, we concluded that the only reasonable explanation was as follows: the difference between the signals was due to the trivial effect of a single reflection from the opposite mirror.

The experiments were concluded by checking the quality of the optical resonator. The electron gun was replaced with an OKG-11 laser, known to be in working condition; the mirrors of this laser were removed and the resonator was formed from the same pair of mirrors as that used in the search for the He line at $\lambda = 6678$ Å. Although this pair of mirrors had a much lower reflection coefficient at the $\lambda = 6328$ Å wavelength (this was the emission wavelength of the OKG-11 laser) than that at its own wavelength, alignment produced easily observed stable stimulated emission at the $\lambda = 6328$ Å wavelength. This experiment demonstrated the sufficiently good quality of the optical resonator used in the search for stimulated emission from helium.

Thus, our systematic experiments led us to the conclusion that population inversion of the excited states of the helium atom sufficient for stimulated emission could not be achieved in the investigated medium.

§3. DISCUSSION OF RESULTS AND COMPARISON WITH EARLIER CALCULATIONS OF THE INVESTIGATED LASER SYSTEM

The negative results of our search for stimulated emission, described in the preceding section, can—in our opinion—be explained basically as follows.

1. According to §1, the only hope for the stimulated emission or amplification of light in the proposed laser is based on the assumption of a Boltzmann distribution of the populations between the sublevels of the $n=3$ and $n=2$ states of the helium atom; moreover, in a stationary supercooled plasma excited by a high-power electron beam, the temperature of such a distribution should be very close to $T_e \approx 0.2$–0.3 eV, i.e., it should be sufficiently low compared with the separations between the sublevels. As pointed out in §1, this assumption is very optimistic at least in respect of the $n=2$ level. In fact, there are important reasons for doubting the validity of this assumption. First of all, it is not supported by the experimental data.[9,10] Secondly, the theoretical results (Fig. 2 in the preprint of Evstigneev and Filippov[8]) show that, for $T_e \approx 0.2$ eV and $N_e = 1\times10^{14}$ cm^{-3}, this assumption clearly does not describe the populations of the $2\,^{1,3}P$ sublevels relative to the lower $2\,^{1,3}P$ sublevels even in the absence of the electron beam: in fact, the relative population of the $2\,^{1,3}P$ sublevels is much higher than that assumed. Moreover, in the presence of an electron beam, the population of the P states increases more rapidly than that of the S states[11a] and the discrepancy becomes even greater. One should bear in mind that Evstigneev and Filippov[8] ignore the factors considered in §1, which have a very unfavorable influence on the distribution of the populations of the excited states. These factors are the atomic shunting and preferential depopulation of the upper level of the lasing transition by the impurity molecules. It should be pointed out that such factors, as well as the charge exchange and conversion of the helium ions in the proposed laser system, have been allowed by us for the first time in the present paper (in §1) and were not known to us during the initial preparations for the experiment.

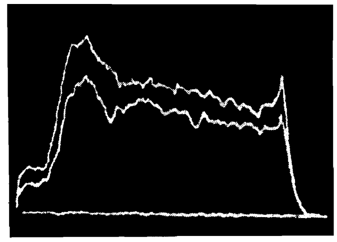

FIG. 7. Typical oscillograms of the intensity of the $3\,^3S \to 2\,^3P$ transition line ($\lambda = 7065$ Å) observed with a photomultiplier with the open (upper trace) and closed (lower trace) mirror located at the opposite end to the photomultiplier.

Thus, we may certainly conclude that the proposed assumption of a Boltzmann distribution of the impurities between the sublevels is not justified and quite likely completely incorrect. In the latter case, this means that the proposed laser scheme is unworkable, as pointed out several times in §1.

2. According to Figs. 3 and 6, over part of the plasma filament in our apparatus the density of the He–H_2 mixture is far too low (of the order of 10^{17} cm^{-3}) and, for certain proportions of the mixture, there may be strong absorption of light. Althoug the part of the filament where $\varkappa < 0$ is less than the length of the working zone, in the main part of which we have $\varkappa > 0$, the ratio of the absolute values of $|\varkappa|$ in the two parts may be unfavorable and the effect integrated over the whole length of the apparatus may correspond to very weak (unobservable) amplification or even to the absorption of light.

This relationship between the amplification and absorption of light is primarily a consequence of the action of the undesirable charge-exchange and conversion processes, of the gas shunting of the lasing transition, and of the preferential depopulation of the upper active level by the impurity molecules; all these factors reduce greatly the maximum possible gains so that it becomes $\varkappa \approx 1\times10^{-2}$ cm^{-1} (we are still ignoring the effect mentioned in subsection 1 above, as a result of which we may have $\varkappa < 0$ everywhere). It is likely that this (negative in the integrated sense) effect may occur under our experimental conditions although not for all the proportions of the mixture (for example, in the case of the 1 : 1 mixture, the effect is clearly unimportant—see Fig. 3).

Our conclusion is, therefore, that the investigated cw beam laser utilizing a supercooled plasma is unworkable, which is in conflict with the first paper on this laser[3] and with the subsequent calculations.[4] This conflict arises because the earlier work[3,4] ignores a number of basically important factors that have a radically negative effect on the proposed laser scheme which seemed to be initially (theoretically[3] and on the basis of the experiments[1,2]) so very attractive.

The main of these factors are as follows:

1. The charge exchange between the active gas (helium ions) and the impurity (hydrogen) molecules. The role of this process (see §1 and our earlier paper[2]) increases very markedly when hydrogen is replaced with such gases as nitrogen, oxygen, or carbon dioxide: the effective cross section for the charge exchange between the helium ions and the molecules of these gases is four orders of magnitude greater (!) than that in the case of the hydrogen molecules.[12] Therefore, it is incorrect to say[3,4] that practically any gas can be used in the proposed laser as an admixture to helium. Only neon and argon can be considered, alongside hydrogen, as possible admixtures (because the cross sections for the charge exchange between the He^+ ions and these gases are also relatively small[12]). However, there are other reasons why even these gases cannot be used as an impurity in helium: first of all, their presence increases greatly (this is undesirable) the ionization losses in the electron beam (the increase is severalfold compared with hydrogen[11]); secondly, the relatively heavy monatomic gases Ne and Ar do not have vibrational degrees of freedom and cannot ensure the necessary range of electron cooling so as to obtain the required[2] sufficiently low value of the electron temperature.

2. The conversion of the atomic helium ions into molecular ions. It has been assumed[4a] that the conversion process (and the postulated slow recombination of the molecular helium ions) results in the accumulation of electrons which tends to reduce T_e and increase directly (at a constant ionization frequency in the beam) the flux to the useful channel (three-particle recombination), i.e., it results in a direct increase in the rate of population of the upper active level. However, it has been shown[2] that this assumption is incorrect: the converted ions interact chemically with hydrogen, forming very rapidly recombining products, which very soon cease to participate in the processes of interest to us. Therefore, the conversion does not increase but reduces the electron density and, consequently, increases T_e and lowers the optical gain. This process plays approximately the same role as the charge exchange.

3. The gas shunting of the lasing transition ($r > 1$ in the notation of §1).

4. The rate of depopulation of the upper active level by the impurity ionization is much faster than that of the lower level (and not conversely, as assumed erroneously earlier[3,4]). This follows from recent theoretical[15] and experimental[18] investigations.

5. The favorable Boltzmann factor, associated with the splitting of the active levels (the only hope for a workable laser!), is less important than assumed earlier.[3,4] The reduction in the population of the upper sublevels of the $n = 2$ state because of the Boltzmann distribution plays not only a positive role (facilitating inversion relative to these sublevels) but also a negative one; it reduces the rate of depopulation of the whole $n = 2$ state since the largest ionization cross section of the impurity molecules is exhibited by the upper resonance sublevel $2\,^1P$. The process 3, 4, and 5 listed above decisively prevent population inversion.

6. In contrast to §1 of the present paper, the optical gain is calculated by Syts'ko and Yakovlenko[4a] by solving a non-self-consistent problem in which the value of T_e is assumed to be an arbitrary parameter which can take any small value right down to 0.1 eV. However, we can easily see that such an arbitrary assumption is in conflict with the calculations of the authors themselves[4a] (see Fig. 1 in their paper[4a]). Figure 8 shows how unjustifiably optimistic results are obtained by the non-self-consistent solution of the problem. This figure gives the gain $\varkappa$, calculated on the assumption that $T_e = 0.25$ eV = const, $p = 1$, $r = 1$ (the graphs in this figure

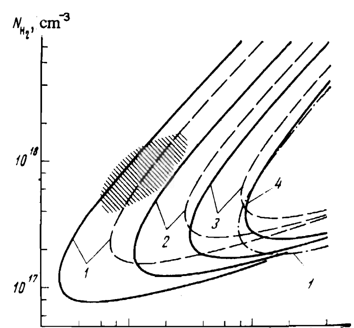

FIG. 8. Concentrations of the components of $He-H_2$ mixtures needed to achieve a specified gain for various transitions (calculations). In all cases it is assumed that $\nu_i = 200$ sec^{-1} and $T = 0.1$ eV. Non-self-consistent solution: $T_e = 0.25$ eV; $T_a = T_e$; $p = 1$; $r = 1$. Continuous lines represent $3\,^3S \rightarrow 2\,^3P$ (7065 Å); dashed curves correspond to $3\,^1D \rightarrow 2\,^1P$ (6678 Å); chain curves correspond to $3\,^1S \rightarrow 2\,^1P$ (7281 Å). Curves: 1) $\varkappa = 0.02$ cm^{-1}; 2) $\varkappa = 0.05$ cm^{-1}; 3) $\varkappa = 0.2$ cm^{-1}; 4) $\varkappa = 0.5$ cm^{-1}. Self-consistent solution: $T_e = T_e(N_{He}, N_{H_2})$; $T_a = T_e$ for $T_e \geqslant 0.3$ eV, $T_a = 0.3$ eV for $T_e < 0.3$ eV; $p = p(N_{He}, N_{H_2})$; $r = r(N_{He}, N_{H_2})$. This solution is represented by the shaded range of the mixture parameters, where $\varkappa \approx 0.01$ cm^{-1} for the $3\,^3S \rightarrow 2\,^3P$ transition.

are less optimistic than the results of Syts'ko and Yakovlenko[4a] because allowance is made for the real relationship between q_3 and q_4—see subsection 4 above). We can see that, according to the non-self-consistent solution, the working region can be practically the whole plane in Fig. 8 and the value of $\varkappa$ can be 5×10^{-2} or higher. The solution of the self-consistent problem, even without allowance for the charge exchange, conversion, and gas shunting of the lasing transition ($p=1$ and $r=1$ correspond to the partly self-consistent solution), greatly reduces the region of sufficiently high gains (compare Fig. 8 with the curves ν = const in Fig. 2). The self-consistent solution with allowance for these undesirable processes is represented by the shaded region in Fig. 8, which represents the postulated working region with a maximum gain $\varkappa\approx1\times10^{-2}$ cm^{-1}. (This region is based on Figs. 2 and 3.) A comparison of the solutions needs no comment.

7. Earlier treatments[3,4] ignore the unavoidable variation in sign of the gain $\varkappa$ along the length of the system; allowance for this factor makes for much more stringent conditions on the value of $\varkappa$ in the working region. However, since the maximum value of $\varkappa$ in the working region is relatively small (1×10^{-2} cm^{-1}) because of the unfavorable phenomena discussed above, this circumstance may also have a decisive negative effect on the proposed scheme.

Consistent allowance for all these factors leads to the conclusion that the proposed system is unsuitable for the amplification or stimulated emission of light.

We shall conclude by mentioning the possibility of more practical applications of a stationary supercooled plasma, which are referred to in the Introduction.

The authors are deeply grateful to S. V. Antipov and A. S. Trubnikov for their considerable help in the experiments.

[1)] In the absence of an impurity, the depopulation of the lower active level is very difficult: radiative transitions to the ground state are ineffective because of the reabsorption of the resonance radiation, and the collisional effect is not important because of the large energy gap (~20 eV, Fig. 1).

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