RELAXATION PROCESSES AND AMPLIFICATION OF RADIATION IN A DENSE PLASMA

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Some features of relaxation in a dense low-temperature highly ionized plasma of a many-electron chemical element are discussed. It is shown that the decisive role played by the collision transitions and the competition of various relaxation channels cause (under certain conditions) a number of levels of such a medium to acquire a population inversion that is practically independent of the either the probabilities for spontaneous radiative transitions involving frequencies in the infrared or visible spectra, or of the reabsorption of the radiation. This is an important property of lasers with a dense, highly ionized plasma (plasma lasers). The negative absorption coefficient in such a medium is very large. Numerical calculations for the case of recombining lithium plasma illustrate the feasibility of the conditions formulated in the paper.

WHEN searching for new media that amplify electromagnetic radiation effectively, attention is being called more and more frequently to the possibility of using a non-equilibrium highly-ionized dense plasma. The following advantages of such a plasma have already been noticed earlier: 1) conservation of the plasma state at a high energy density, 2) existence of inverted population at an appreciable of the average particle energy, 3) broad range of frequencies (from infrared to soft x-rays) capable of being amplified in different types of plasma, 4) the possibility of obtaining in principle a high efficiency. Unfortunately, in the theoretical analysis of the amplifying properties of an essentially non-equilibrium dense highly-ionized plasma, the difficulties in calculating the kinetics of the many-level system are aggravated by the lack of reliable data on the probabilities of a number of elementary acts. Therefore a detailed analysis of the populations has been carried out so far only for a hydrogen plasma and for hydrogen like ions[1-4] (less detailed calculations were performed also for cesium and helium[5-6]). It should be noted that during the last few years progress has been made in the experimental work from the continuous "gas laser" regime to increasingly higher degrees of ionization.

The calculations made in[1,2], with recombining highly-ionized hydrogen (or hydrogen-like ions) as an example, have shown that even such a plasma can amplify radiation at a number of frequencies with technically realizable parameters of the experimental apparatus. It is also noted there that plasma of either hydrogen or of hydrogen-like ions is by far not the best medium for this purpose, owing to a large number of reasons (level degeneracy leading to the absence of branching of the relaxation electron flux through the discrete states, relatively high energy of the first-excited level, large broadening of the spectral lines as a result of the linear Stark effect). In the present paper, we calculate the relaxation processes in a dense highly ionized plasma using lithium as an example[7]; the choice of this element is due to the relative simplicity of the electron structure (one electron beyond the filled shell), which differs at the same time, as will be shown later, greatly from the hydrogen structure. The results make it possible to conclude that plasmas of a number of other elements, particularly alkali and alkali-earth metals, and also of similar ions, can be used as an active medium.

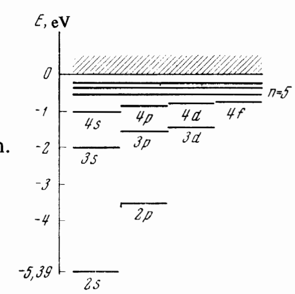

FIG. 1. Level scheme of the Li atom.

The level scheme of the lithium atom is shown in Fig. 1. We used in the calculations the balance equations for the populations N_{nl} of the levels (nl), where n is the principal quantum number and l the orbital quantum number, in the form

$$\frac{dN_{nl}}{dt} = -N_{nl}\Big\{\Big[\sum_{n'l'\neq nl} U(nl,n'l') + B'(nl)\Big]N_e + A(nl)\Big\} + \dot{N}_e \sum_{n'l'\neq nl} U(n'l',nl)N_{n'l'} + \sum_{E(n'l')>E(nl)} A(n'l',nl)N_{n'l'} + B(nl)N_e^3 + A(nl)N_e^2. \quad (1)$$

The coefficients $U(nl, n'l')$, $B'(nl)$, $B(nl)$, and $A(n'l', nl)$ describe, when multiplied by suitable factors, the probabilities of inelastic collisions of the Li atom with electrons $Li_{nl} + e \to Li_{n'l'} + e$, three-particle recombination $Li^+ + 2e \to Li_{n'l'} + e$, ionization by electron impact $Li_{nl} + e \to Li_{nl}^+ + 2e$, and radiative spontaneous transitions between the discrete levels $Li_{n'l'} \to Li_{nl} + h\nu$. The quantity

$$A(nl) = \sum_{E(n'l')<E(nl)} A(nl, n'l')$$

corresponds to the total probability of radiative decay of the state (nl), and $E(nl)$ is the energy of this state. The probabilities of the radiative transitions from the continuous spectrum are small, and their influence on their relaxation processes in the considered dense low-temperature plasma was disregarded. The values of the probabilities of the radiative spontaneous transitions, needed for the calculations, were taken from[8] and are apparently the most accurate ones. There are practically no experimental data for the cross section of the collision processes, and therefore the problem was solved using the probabilities of the collision act, which we have calculated in two ways: starting from the data of[9] in accordance with the Dravin formula, and from the data of Vaĭnshteĭn and Sobel'man[10]; in the former case, the cross sections of the collisions for the optically forbidden transitions were assumed equal to zero, while in the latter all the transitions entering into (1) were taken into account.

The solution of the system (1) for the populations of eight excited states of Li was obtained in the "stationary flow" approximation[2-4] at different electron densities $N_e \sim 10^{11}-10^{16}$ cm^{-3} and energies kT_e = 0.05, 0.1, 0.2, and 0.5 eV and different populations of the ground state 2s of the Li atom. The effective recombination coefficient $\gamma = -N_e^{-2}dN_e/dt$ was calculated from the formula

$$\gamma = \frac{1}{N_e}\Big[-\sum_{nl} B(nl)N_{nl} + N_e^2\sum_{nl} B'(nl) + N_e\sum_{nl} A(nl)\Big]. \quad (2)$$

The calculation was made both for an optically thin plasma, and with allowance for reabsorption of the radiation by the lines of the principal series (transitions to the ground state). Reabsorption was taken into account approximately by the method of Holstein and Biberman[11] for cylindrical tubes of 6 and 25 mm diameter.

The dependence of the populations N_{nl} of the excited states and of the effective recombination coefficient γ on the density of the Li atoms in the ground state 2s for an optically thin plasma can be represented in the form[3]

$$N_{nl} = N_{nl}' + r_{nl}N_{2s}, \quad \gamma = \alpha - SN_{2s}/N_e, \quad (3)$$

where N_{nl}' and α are the level populations and the recombination coefficient calculated at N_{2s} = 0; r_{nl} and S are positive quantities that depend on the density and temperature of the free electrons. Calculation has shown that the population of the ground state in an optically thin lithium plasma has practically no influence on the populations of the excited levels and the effective recombination coefficient γ up to ionization degrees ~ 0.1% when $kT_e \lesssim 0.1$ eV. When kT_e = 0.2 eV, the population of the ground state influence only the resonant level 2p, and when kT_e = 0.5 eV the predominant process in the plasma is ionization, and the populations of all the levels, as well as the coefficient γ (which in this case is negative and determines the ionization rate) depend on the degree of ionization. Table I lists the quantities r_{2p}, the recombination coefficients α, and the ionization coefficient S, calculated for kT_e = 0.2 and 0.5 eV at different values of the electron densities, using collision probabilities obtained in accordance with[10].

Figure 2 illustrates the influence of the reabsorption of radiation on the population of the 2p level at kT_e = 0.05, 0.1, 0.2, and 0.5 eV at different n_e. The

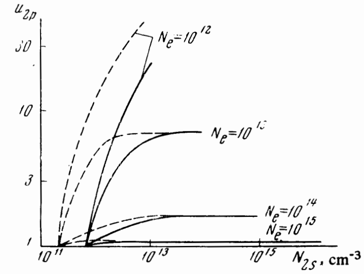

FIG. 2

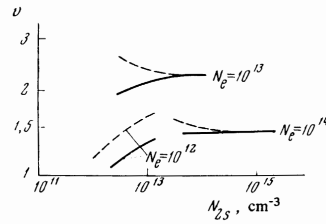

FIG. 3

FIG. 2. Influence of reabsorption of radiation on the population of the 2p level of Li at different densities N_e (cm^{-3}) of the free electrons. Here u_{2p} is the ratio of N_{2p} (with reabsorption) $2N_{2p}$ (without reabsorption); the solid curve corresponds to a tube of radius 3mm, the dashed ones to 12.5 mm.

FIG. 3. Influence of radiation reabsorption on the ionization rate of a Li plasma at energy kT_e = 0.5 eV and at different densities of the free electrons. Here v – ratio of γ (with reabsorption) to γ (without reabsorption); solid curves correspond to a tube of radius 3mm, dashed – 12.5 mm.

population N_{2p} increases with increasing N_{2s} the stronger, the smaller the electron density, this being connected with the decisive role of the radiative transitions in the population of this level at small N_e. When $N_e \gtrsim 10^{15}$ cm^{-3}, the reabsorption has practically no influence on the level population. It is seen from Fig. 2 that at sufficiently large values of N_{2s}, the influence of the reabsorption is the same for tubes with different radii. The influence of the dragging of the radiation on the plasma ionization rate is illustrated by Fig. 3, from which it is seen, for example, that for $N_e \sim 10^{13}$ cm^{-3} the ionization rate in the case of reabsorption increases by 2–3 times. Comparison of Figs. 2 and 3 makes it possible to assess the role of the stepwise ionization, since an increase of the ionization rate in reabsorption is the result of the growth of the population of the excited states (primarily the 2p level), meaning also of the additional ionization from these levels. It is seen from these figures that at densities $N_e \lesssim 10^{12}$ cm^{-3} the influence of the reabsorption u_{2p} on the level population 2p exceeds approximately by one order of magnitude the analogous factor for the ionization rate. With increasing N_e, the ratio v/u_{2p} increases, and when $N_e \gtrsim 10^{14}$ cm^{-3} these characteristics become equalized: $v = u_{2p}$. This means that when $N_e \gtrsim 10^{14}$ cm^{-3} the stepwise ionization makes the main contribution to the total ionization rate.

We now turn to the question of the existence of inverted population and the possibility of obtaining generation in the case of a lithium plasma. The results of the calculation[7] offer evidence of the presence of an inverted population for a number of Li levels in a recombining plasma at $kT_e \lesssim 0.2$ eV. When kT_e = 0.5 eV, and the predominant process in the plasma is ionization, there is no inversion[1)]. We emphasize that at large electron densities the reason for the

[1)]The conditions necessary to produce inversion in a lithium plasma during the course of rapid ionization are not discussed here.

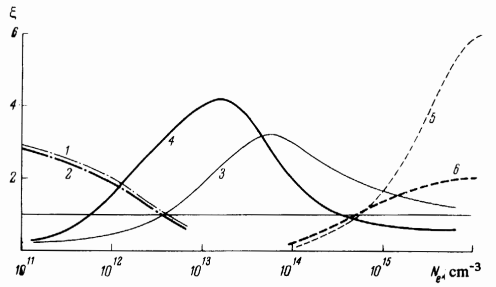

FIG. 4. Dependence of the relative level population ξ on the concentration of the free electrons at energy $kT_e = 0.2$ eV (curves 1 and 2 correspond to levels 3p-3s; 3, 4 – to levels for s-3p; 5, 6 – to levels 3s-2p; curves 1, 3, and 5 were obtained using the collision probabilities calculated in accordance with [9], while curves 2, 4, and 6 were calculated in accordance with [8].

formation of the inverted population differs from the case of hydrogen[1,2].

Figure 4 shows the dependence of the ratio

$$\xi = \frac{N_{nl}}{g_{nl}} \Big/ \frac{N_{n'l'}}{g_{n'l'}}$$

on the concentration of the free electrons at $kT_e = 0.2$ eV. For the levels 2p – 3s, the best inversion conditions occur at low densities N_e, and the inversion vanishes at $N_e \approx 4 \times 10^{12}$ cm^{-3}. The inverted population of this pair of levels at low densities of the free electrons is due to a more intense radiative decay of the lower level: A(3s) > A(3p). At the same time, the probability of the collision transitions of the lower level 3s is smaller than the collision probability for 3p, and therefore with increasing N_e the collision processes affect adversely the amplification conditions, until the inversion vanishes completely. It is clear that in such a mechanism of inversion (which is analogous to the case of a recombining hydrogen plasma) it is impossible to increase N_e strongly, meaning that it is impossible to obtain large values of the level populations or the gain; it is necessary also here that there be no reabsorption of the radiation corresponding to the radiative decay of the lower working level.

However, in the case of a lithium plasma, at high densities, the mechanism of collision relaxation becomes decisive; this mechanism differs from that in the hydrogen plasma and gives rise to a different law governing the formation of the inverted population. It is seen from Fig. 3 that at small values of N_e there is no inverted population of the level pair 4f-3p. With increasing electron density, inversion occurs at $N_e \sim 6 \times 10^{11} - 3 \times 10^{12}$ cm^{-3} and increases subsequently, with the maximum ratio $(N_{4s}/g_{4s})/(N_{3p}/g_{3p})$ obtained at $N_e \sim 10^{13} - 10^{14}$ cm^{-3}. For the pair of levels 3s – 2p, at ionization degrees $\gtrsim 1\%$, the inversion occurs at even larger electron densities, $N_e \sim 6.5 \times 10^{14}$ cm^{-3}, and exists in any case up to $N_e \sim 10^{16}$ cm^{-3} (at $N_e > 10^{16}$ cm^{-3} and $kT_e > 0.2$ eV it is impossible to solve the system (1) in the "stationary flow" approximation). The reason for the formation of the inverted population consists in these cases in the fact that the probabilities of the collision decay of the upper working levels 4s and 3s is smaller than that of the corresponding lower 3p and 2p[2)]. The radiative probabilities and the optical density of the plasma at sufficiently large N_e practically play no role in all these processes. Thus, the considerable deviation of the level populations from equilibrium values and the formation of inverted population are due in a dense recombining plasma not to the radiation yield, but to the presence of unilateral recombination electron flux. The advantage of this mechanism over the method of producing inversion via the difference between the radiative decay times (the only mechanism in the case of hydrogen and hydrogen-like ions) lies in the possibility of obtaining a large inversion by increasing the electron density, and also in the fact that the dragging of the radiation is negligible, so that it becomes possible to use for the discharge large-diameter tubes, meaning also a large volume of the active medium. The performance of the experiments is also facilitated to some degree, since it is not necessary to obtain an optically thin plasma.

Let us discuss briefly the possibilities of preparing a highly ionized recombining plasma with inverted population of the lithium levels by rapidly cooling the free electrons. Theoretical estimates for several methods of cooling are given in[12]. Gol'dfarb et al.[13] observed experimentally inverted population in a cooled recombining hydrogen plasma, while Johnson[6] quotes the results of an unpublished paper by Hinnov, from which it is possible to assess the population inversion of a number of levels during the process of ionization and recombination of a helium plasma.

We stop here to discuss the conditions for obtaining nonequilibrium population of Li in a Li + He plasma by rapidly terminating the electric heating field (the helium serves as a thermostat for maintaining the low temperature of the heavy particles). We assume that during the heating time the Li is fully and singly ionized, while He remains practically non-ionized. Then, after the field is turned off, the electron energy drops rapidly to $kT_e \sim 0.4$ eV and a decay of the plasma begins. The system of equations describing the time variation of the concentration and temperature of the free electrons has the following form (we disregard here ambipolar diffusion, which plays no role at the considered plasma parameters):

$$\frac{dN_e}{dt} = -\gamma(N_e, T_e) N_e^2,$$

$$\frac{dT_e}{dt} = -\frac{2m_e}{M_{\mathrm{He}}}\nu_{\mathrm{He}}(T_e - T) - \frac{2m_e}{M_{\mathrm{Li}}}\nu_i(T_e - T_i) - \frac{T_e}{N_e}\frac{dN_e}{dt} + \frac{2}{3k}E^*\gamma(N_e, T_e) + \frac{2}{3k}E_{\mathrm{met}}RN_{\mathrm{met}} \quad (4)$$

Here m_e, M_{He}, M_{Li}—masses of the electrons and of the He and Li atoms; ν_{He}, ν_i—frequencies of electron collisions with the He atoms and Li^+ ions; T, T_i—temperatures of the He atoms and Li^+ ions; E^*—average energy transferred to the free electron in

2)The existence of an optimal density of free electrons for the population inversion of the level pair 4s-3p and the deterioration of the inversion with further increase of N_e are due to the fact that the flux of electrons to the 3p level increases rapidly with increasing population of the 3d level.

Table I. The coefficient r_{2p}, the recombination coefficient α, and the ionization coefficient S for different values of the plasma parameters N_e and kT_e

N_e, cm^{-3}	kT_e, eV	r_{2p}	α, cm^3/sec	S, cm^3/sec	N_e, cm^{-3}	kT_e, eV	r_{2p}	α, cm^3/sec	S, cm^3/sec
10^{11}	0.2	3.5^{-8}	1.1^{-11}	—	10^{15}	0.2	$2,1^{-5}$	5.1^{-10}	—
	0,5	$8,8^{-6}$	6.3^{-13}	5^{-12}		0,5	5.6^{-3}	$6,9^{-12}$	$2,7^{-10}$
10^{12}	0,2	$3,4^{-7}$	1.2^{-11}	—	10^{16}	0,2	2.3^{-5}	$4,9^{-9}$	—
	0,5	8.7^{-5}	6.7^{-13}	5.4^{-12}		0.5	6^{-3}	$2,5^{-11}$	$3,4^{-10}$
10^{13}	0,2	3.1^{-6}	1.6^{-11}	—	10^{17}	0.2	2.3^{-5}	4.9^{-8}	—
	0,5	7.7^{-4}	9.8^{-13}	9.5^{-12}		0.5	6.0^{-3}	2^{-10}	$3,4^{-10}$
10^{14}	0,2	$1,4^{-5}$	$6,1^{-11}$	—					
	0,5	$3,6^{-3}$	2.7^{-12}	$7,9^{-11}$					

triple recombination; E_{met}, N_{met}—energy and density of the metastable He; R—probability of collision of second kind between the electron and an He atom in the metastable state. Simple estimates show that if $N_{met} \lesssim 10^{-5} N_{He}$, the influence of the metastable states on the lithium recombination kinetics can be neglected.

The calculated recombination coefficient γ in the region $N_e \approx 3 \times 10^{13}$—$10^{16}$ cm^{-3}, $kT_e \approx 0.1$—0.3 eV can be approximately represented by the formula $\gamma = cT^{-1/2} N_e$. If $N_H \gg N_{Li^+}$, then we can assume that $T = T_i$ = const during the plasma decay process. Under such assumptions, the system (4) was solved with initial values $kT_e = 0.4$ eV, $N_e(0) = 10^{14}$—10^{16} cm^{-3} at He pressures 1—30 Torr, $kT_i = T = 0.05$ eV and 0.1 eV, and at various values of E*. Some of the results are shown in Fig. 5, which gives the time dependences of N_e (solid curve) and kT_e (dashed curve).

As shown in the calculation, the production (by cutting off the field heating the He + Li) of a nonequilibrium plasma with $kT_e \lesssim 0.2$ eV and $N_e \sim 10^{14}$ cm^{-3} is perfectly realistic. Furthermore, at these parameters there exists a sufficiently large inverted population of the 4s—3p levels. Interpolating the results of Table I to the range $kT_e = 0.1$—0.3 eV and Ne = 3 $\times 10^{13}$—10^{15} cm^{-3} we obtain, from the running values of $N_e(t)$ and $T_e(t)$, the time variation of the inverted population of the 4s—3p levels. The corresponding results are shown in Fig. 3 (dash-dotted curves). We see that the inverted population reaches at a certain

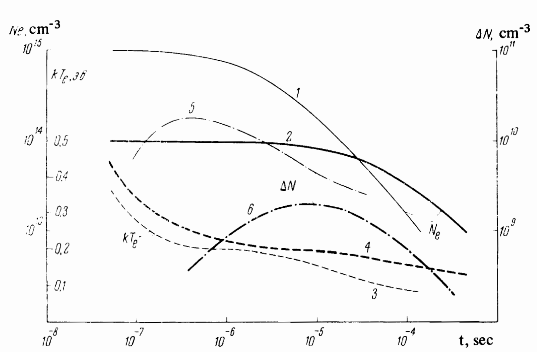

FIG. 5. Time variation of the electron concentration N_e (solid curves 1 and 2), the electron energy kT_e (dashed curves 3, 4) and the inverted population $\Delta N = N_{4s}/g_{4s} - N_{3p}$ (dash-dot curves 5, 6) in a decaying lithium plasma (curves 1, 3, 5 were obtained using the probabilities calculated from [9], at a helium density $N_{He} = 3 \times 10^{17}$ cm^{-3} E* = 5.3 eV and $kT_e = kT_i = 0.05$ eV, while curves 2, 4 and 6 were obtained at $N_{He} = 3 \times 10^{16}$ cm^{-3}, E* = 5.39 eV, kT = kT_i = 0.1 eV with the probabilities calculated in accordance with [8].

instant of time a maximum value, after which it drops off, in spite of the continuing cooling of the electrons, as a result of the rapid decrease of N_e. The values 0.75 and 0.06 cm^{-1} of the coefficient of negative absorption near the maximum are fully sufficient to obtain generation in tubes of relatively short length. At the same time, a nonequilibrium plasma with parameters $N_e \gtrsim 10^{15}$ cm^{-3} and $kT_e \lesssim 0.2$ eV could not be obtained, owing to the slow cooling of the electrons resulting from the strong energy release in collision recombination and from the rapid decrease of the electron density at large N_e. Therefore inverted population of the levels 3s—2p is not realized by the given method of cooling the free electrons. However, it is perfectly realistic to expect a nonequilibrium recombining plasma with large electron densities and to obtain thereby an inverted population in the case of other substances, in which it is not necessary to cool the electrons to such low temperatures (for example, for lithium-like ions), and where extraneous electrons can be used; the source of the "extraneous flux" of electrons may be a high concentration of an easily ionized substance, such as cesium.

These calculations show that it is possible in practice to produce a laser on the basis of a highly-ionized lithium plasma. We have analyzed the possible inaccuracy and performed a number of controlled calculations. It has turned out that the qualitative conclusions are not connected with the employed approximations. In particular, allowance for the fact that the upper levels ($n \geq 5$) are not in equilibrium with the continuous spectrum effects only the absolute values of the populations and the magnitude of the recombination coefficient. In order to clarify the role of the collision processes for optically forbidden transitions, a control calculation was made both with and without allowance for these probabilities, as calculated in accordance with[10]. In both cases, the populations of the discrete levels and the recombination coefficient for $kT_e = 0.05$, 0.1, and 0.2 eV turned out to be close (the difference did not exceed 20%). However, at $kT_e = 0.5$ eV, when the predominant process in the plasma is ionization, the populations may differ by a factor 4—6 and the ionization rates by a factor 2—3.

An analysis of the results of the calculation and clarification of the mechanism of inverted-population formation for lithium make it possible to draw certain general conclusions with respect to plasmas of other atoms and ions. The inverted population of the levels in a dense plasma is due to the relations between the probabilities of the collision transitions. These proba-

Table II. The factor A, the oscillator strength f_{ki}, and the energy factor $k(\Delta E)$ for certain transitions in Li and He atoms

Element	Transition		A	f_{ki}	$k(\Delta E)$
	i	k			
Li	$4s$	$3p$	0,128	0,223	78
	$3p$	$3s$	1,03	1,23	109
	$3s$	$2p$	0,356	0,115	11.6
	$2p$	$2s$	3,24	0,753	9.5
He	5^3S	4^3P	—	0,223	$5.6\cdot10^2$
	4^3P	4^3S	—	1,21	$1,06\cdot10^3$
	4^3S	3^3P	—	0,145	56
	3^3P	3^3S	—	0,9	$2,53\cdot10^2$
	3^3S	2^3P	—	0,069	8
	2^3P	2^3S	—	0.54	27,4

bilities can be estimated from products of two factors[9,10]: the oscillator strengths of the corresponding transition (or a certain function A connected with the line strength) and an energy factor $k(\Delta E)$ that depends on the distance ΔE between the levels. If the change of the orbital quantum number on going to a lower energy level is $\Delta l = +1$, then the oscillator strength is smaller than for the transition with $\Delta l = -1$. For lithium, and also other alkali elements, helium, alkali-earth elements, and the corresponding ions we can separate the main channel of the relaxation $np \rightarrow ns \rightarrow (n-1)p \rightarrow (n-1)s$. Therefore the rate of relaxation of the levels ns is lower than the corresponding rate for the levels $(n-1)p$, and inverted population of the level pairs $ns - (n-1)p$ may be realized. A similar analysis points to the possible existence of inversion of the levels $np - (n-1)d$.

To prevent deequalization of the populations it is necessary that the energies of the free electrons do not exceed the corresponding distances between levels. Table II lists for certain transitions in Li and He the values of the oscillator strength, the factor A, and

$$k(\Delta E) = \left(\frac{\mathrm{Ry}}{\Delta E}\frac{E_{nl}}{E_{n'l'}}\right)^{3/2},$$

and also of their products. It can be concluded from these data that inversion exists between the states 4s – 3p, 3s – 2p for Li and $5^3S - 4^3P$, $4^3S - 3^3P$, and $3^3S - 2^3P$ for He. The experimental data given in[6] confirm the presence in helium of the indicated inversion. The inversion between the S and P states upon recombination is the characteristic feature also of lithium- and helium-like atoms and ions (for example, in Hg II). The approach considered here facilitates the search for the possible active media and operating transitions for lasers using a dense plasma.

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