Mechanism of the population inversion formation at infrared transitions in SrI atom and SrII ion

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Lasing at the **r–m** transitions in atomic strontium at λ = 6.45 μm holds very good promise for medical applications. This lasing is known to occur together with that at the **r–m** transitions in ion strontium at λ = 1.03 and 1.09 μm and, under certain conditions, with the lasing at the transitions from the $4d^3D_{3,2,1}$ levels to the $5p^3P^0_{2,1,0}$ levels in atomic strontium at λ = 3.01, 2.92, 2.69, and 2.60 μm. The possible mechanisms of the population inversion formation for the transitions in atomic and ion strontium are discussed with the allowance made for specific features of the atomic strontium structure and the excitation functions of the singlet and triplet SrI states.

Introduction

Soldatov with co-authors in Ref. 1 has presented the results of investigation of a strontium-vapor laser operating at several SrI and SrII transitions. The longitudinal-type gas-discharge laser operates at a high pulse repetition frequency thus providing for conditions of efficient lasing at the SrI transition from the resonant level to the metastable one at λ = 6.45 μm ($5s5p^1P^0_1$–$5s4d^1D_2$). During the electric current pulse, the lasing at λ = 6.45 μm is accompanied by the lasing at the SrI transitions from $4d^3D_{3,2,1}$ levels to $5p^3P^0_{2,1,0}$ levels (λ = 2.69, 2.92, and 3.01 μm), as well as at the transitions in SrII ion at λ = 1.09 ($5p^2P^0_{1/2} - 4d^2D_{3/2}$) and 1.03 μm ($5p^2P^0_{3/2}$–$4d^2D_{5/2}$) from the resonant levels to the metastable ones.

The lasing at the transitions in SrII at λ = 1.03 and 1.09 μm, and in SrI at λ = 6.45 μm, as well as at λ = 3.01 and 3.06 μm was earlier reported in Refs. 2 and 3, while the lasing at the transitions from the triplet 4^3D level to the triplet 5^3P^0 level (λ = 2.60, 2.69, and 2.92 μm) was reported in Ref. 4. In this paper, we discuss the possible mechanisms of formation of the population inversion at these transitions and their relation with lasing at the line λ = 6.45 μm.

Experimental conditions and results

The sealed-off active element of the strontium-vapor laser was characterized by ∅ = 15 mm, the interelectrode separation of 500 mm, and the following discharge parameters:

Rectifier voltage – 7.4 kV;
Mean current – 180 mA;
Mean total output power – 1.5 W;
Pulse repetition frequency – 18 kHz;
Buffer gas – helium;
He pressure – 100 Torr;
Evaporator operating temperature – 700°C;
Cavity – unstable, telescopic type.

Figure 1 depicts the shapes of the current and voltage pulses. The voltage amplitude applied to the tube was 12 kV. These conditions are optimal for the strontium-vapor laser under consideration. The time behavior of the lasing pulses is shown in Fig. 2. If the lasing at the lines of the **r–m** transitions in SrI and SrII begins almost simultaneously, then that at the triplet transitions lags behind by about 40 ns. To get the spectral resolution of the laser transitions from the triplet $^3D_{3,2,1}$ levels to the triplet $^3P^0_{2,1,0}$ levels, a McPherson grating spectrometer with the 150 grooves/mm grating and a pulse chopper transmitting the light for 40 μs at the pulse repetition frequency of 5 Hz were used.[1]

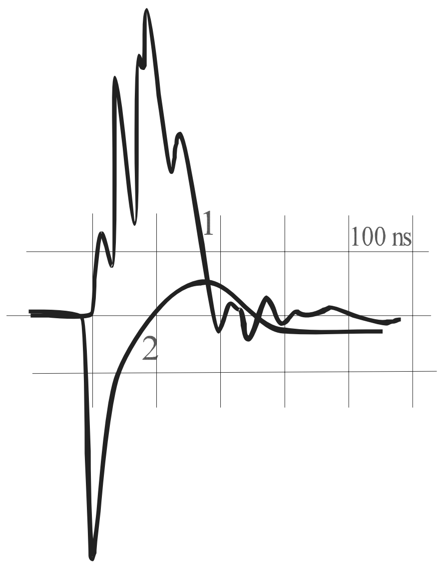
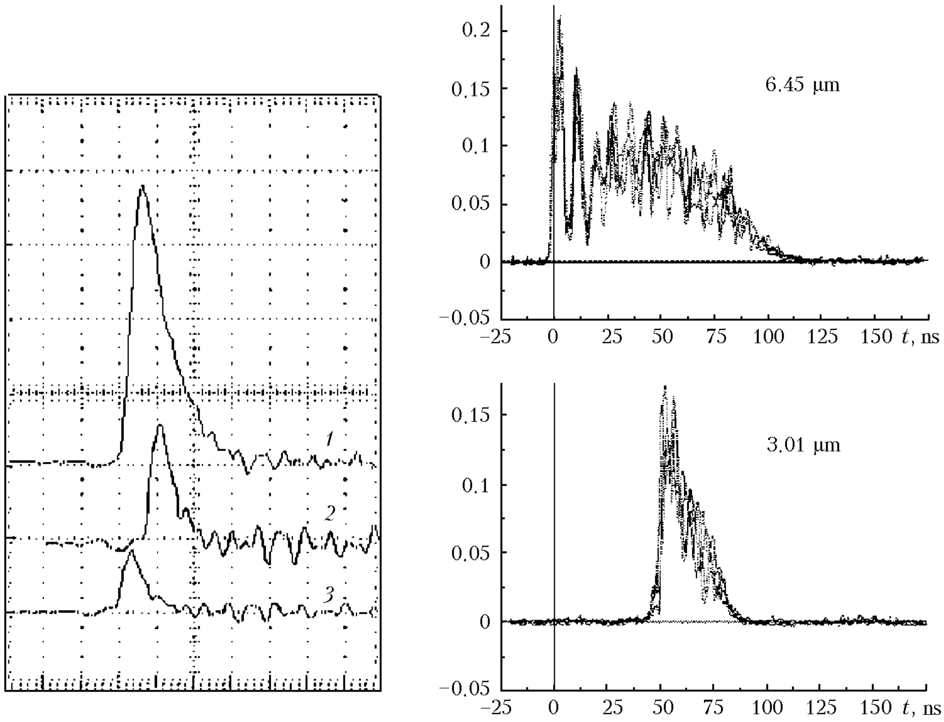


Fig. 1. Current (*1*) and voltage (*2*) pulses.



a b

Fig. 2. Time dependence of lasing (in rel. units): total pulse (*a*): λ ~ 1, 3, 6.45 (*1*), 3 (*2*), and 1 μm (*3*); lasing pulses at the lines λ = 6.45 and 3.01 μm (*b*); the scale graduation of the oscilloscope is 50 ns.

Figure 2*b* depicts the time behavior of the intensity of the 6.45 and 3.01-μm laser lines in arbitrary units. The power meter has measured the following average power at the monochromator output:

2.69 μm – 12 mW; 3.01 μm – 31 mW;
2.92 μm – 28 mW; 6.45 μm – 170 mW.

Discussion

From the diagram of the SrI energy levels (Fig. 3), we can see that the $^3D_{3,2,1}$ levels are metastable (not connected optically with the SrI ground state $5s^1S_0$).

The $^3P^0_{2,0}$ levels are metastable as well. The level $^3P^0_1$ is connected with the strontium ground state through a very weak optical transition (forbidden according to the selection rules). However, the experiments have shown[5] that the effective cross section of excitation of the $^3P^0_1$ level from the ground state is large ($Q \sim 7.5 \cdot 10^{-16}$ cm^2 at $E_{max} \sim 4.6$ eV). Note, for a comparison, that the effective cross section of excitation of the resonant $^1P^0_1$ level achieves the value of about $20 \cdot 10^{-16}$ cm^2 (Ref. 6) at the same energy. According to Ref. 5, the triplet states are populated by electron impact. If we assume that under our experimental conditions the maximum value of T_e is about 4–5 eV, then at that time the expected population of the $^3P^0_1$ level achieves $\sim 10^{12}$ cm^{-3}. As can be seen from Fig. 4, the population inversion at the 3.01 and 3.06 μm transitions can be expected even at the equal population of the triplets as a whole, if we assume that the populations of the sublevels in the triplets mix efficiently due to atom–atom collisions.[3]

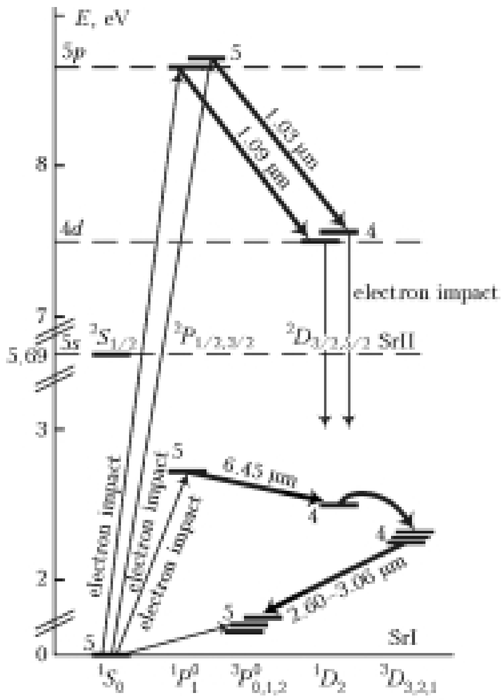


Fig. 3. Partial scheme of the energy levels of the SrI atom and SrII ion: the dashed lines are for convergence limits of the series of one-electron and two-electron excitation of the strontium atom.

If the separation between the closest sublevels is $\Delta E \leq 0.1\, k\, T_{\text{gas}}$, then the levels in the multiplets are re-populated according to the Boltzmann law depending on the gas temperature. The effective cross section of this process is large ($Q \cong (3–5) \cdot 10^{-15}$ cm^2) (Ref. 7). At the medium pressure (60–100 Torr), the populations of the neighboring levels are mixed even under less strict condition $\Delta E \leq kT_{\text{gas}}$, and they are not necessarily the levels of the multiplets.[8] Under conditions of our experiment, the characteristic time of spin-orbital mixing of the levels in the triplets is 1 ns.

It can also be seen from Fig. 4 that to obtain the population inversion at other transitions, the "center of gravity" of the $^3D_{3,2,1}$ triplet must lye higher than that for $^3P^0_{2,1,0}$, that is, the population inversion of the upper triplet as a whole with respect to the lower one must take place.

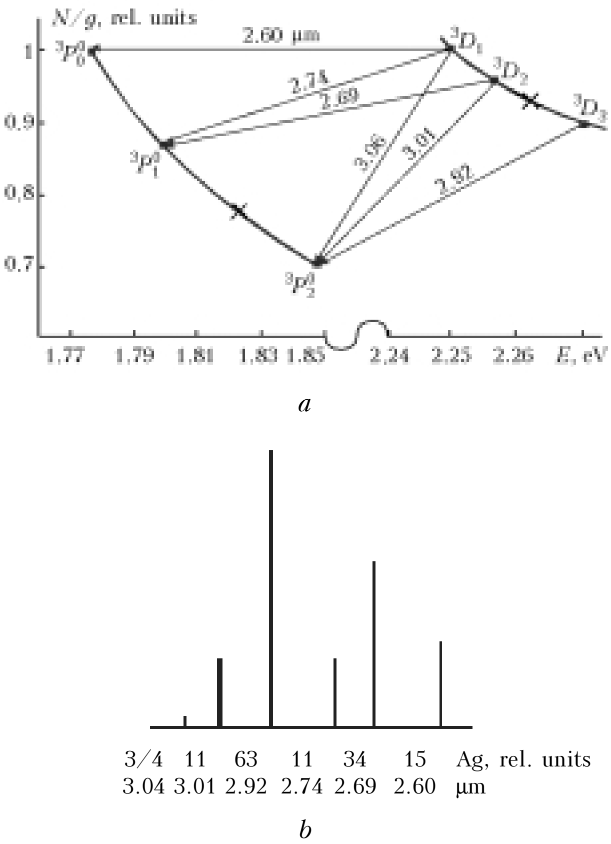


Fig. 4. Qualitative pattern of population inversion (*a*) and relative probabilities of $^3D_{3,2,1}$ – $^3P^0_{2,1,0}$ transitions (*b*); "centers of gravity" of the upper and lower triplets are marked by crosses; $T_{\text{gas}} \sim 0.2$ eV.

Let us discuss the possibility of selectively populating the $4^3D_{3,2,1}$ triplet and the resonant levels of SrII.

1. Destruction of the singlet metastable state $4d^1D_2$ is most probable at a hyperelastic impact with a free electron, at which the spin exchange occurs between the atomic electron and the free one; this exchange brings the atom into the state ^3D (in Fig. 3 this process is marked by the arcuate arrow). By analogy with the 2^1S and 2^3S states of helium,[9] the effective cross section of this process is large $\sim 3 \cdot 10^{-14}$ cm^2. Assuming that this process can proceed at a rather high population of the $4d^1D_2$ level due to lasing at $\lambda = 6.45$ μm, that is, about ~20 ns after its beginning, we can easily estimate the characteristic time of the hyperelastic impact and compare it with the experimental value (~20 ns). Assuming $T_e \sim 3$ eV and $N_e \sim 10^{14}$ cm^{-3}, we actually obtain $\Delta t_{\text{he}} \sim 20$ ns.

Figure 4*a* explains the lasing at the transitions between the $^3D_{3.2,1}$ and $^3P^0_{2,1,0}$ levels in selectively populating the upper triplet. The fact that the line with $\lambda = 2.92$ μm can have higher gain at a smaller difference ($N_{\text{up}}/g_{\text{up}} - N_{\text{low}}/g_{\text{low}}$, where N_{up}, g_{up}, N_{low}, and g_{low} are the populations and the statistical weights of the upper and lower levels),[4] is explained by its higher transition probability. The ratio of the transition probabilities in the lines of the multiplets is shown in Fig. 4*b* and is calculated by the rule of sum.[10]

2. The results obtained confirm high efficiency of pumping the SrII resonant levels in the process of their direct excitation from the ground state of the strontium atom.[2,6,11,12] This process proceeds in a single event of electron impact with an atom as ionization with simultaneous excitation. For strontium, this process has not been studied theoretically, though in Ref. 11 it was assumed that the wave function of the SrI atomic ground state $5s^2(^1S_0)$ is considerably mixed with the $5p^2$ configuration, from which the SrII ion levels $5p(^2P^0)$ are excited by the direct electron impact.

The population of the lower (metastable) states of the SrII transitions ($\lambda = 1.03$ and 1.09 μm) by the direct electron impact is unlikely, which provides for the population inversion at these transitions for ~70 ns.

It should be noted that, in the excitation of the SrI atom and the SrII ion by the electron impact, the important factors are the configurations of the SrI atom, corresponding to the two-electron excitation of the s^2 electrons. The first electronic states of these configurations are $4d5p$, $5p^2$, $4d^2$. As was shown in Ref. 6, the cross sections of excitation of the levels by the electron impact resulting in the simultaneous change of the states of two electrons are large and comparable (for the same n_{eff}) with the single-electron ones. Besides, they strongly interact with the neighboring levels of the single-electron excitation, and, as was mentioned above, from them the SrII levels $5p(^2P^2)$ and $4d(^2D)$ can be populated through the direct electron impact (see Fig. 3).

In its turn, by the end of the current pulse (approximately $N_e \sim 10^{14}$ cm^{-3}, $T_e \sim 1–1.5$ eV), as the lasing at $\lambda = 1.03$ and 1.09 μm is terminated, efficient de-excitation of the metastable SrII $4d(^2D)$ states at collisions with electrons and re-population of all the lasing levels of the SrI atom from above through the states of the two-electron excitation are possible. Since the mechanism of selective destruction of the SrI $^1D_2$ level is still in force, the lasing at $\lambda = 6.45$ μm lasts actually by the end of the electric current pulse.

Conclusions

The lasing at several SrI and SrII transitions in the mixture of a buffer gas with the strontium vapor in a longitudinal repetitively pulsed discharge is observed in any current pulse. Along with the lasing at the **r–m** transitions of SrI ($\lambda = 6.45$ μm) and SrII ($\lambda = 1.03$ and 1.09 μm), the lasing at the transitions between the levels in the triplets ($4^3D_{3,2,1} - 5^3P^0_{2,1,0}$), $\lambda = 3.01$, 2.92, and 2.69 μm, arises with the lag of about 40 ns. The significant role of the collisional mixing of levels, depending on the gas temperature has been demonstrated. The mechanism has been proposed for the selective population of the ^3D triplet after termination of self-terminating lasing at $\lambda = 6.45$ μm, as well as for de-population of the metastable SrII levels $4d(^2D)$ that continue the lasing at $\lambda = 6.45$ μm till the end of the electric current pulse.

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