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On the Mechanisms of Excitation of Laser States in Singly Ionized Argon*

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In this work a discussion of the excitation mechanisms of electronic states in a Ar+ laser is presented. Comparing the experimental and the theoretical results for population densities for upper and lower laser states, a two-step excitation mechariism is proposed for a $CwAr^+$ laser. The idea presented in this work can be used to study excitation mechanisms of the other noble gas laser.

Discutem-se neste trabalho os mecanismos de excitação de estados eletrônicos em um *laser* de Ar^4. Comparando-se os resultados experimentais e teóricos, para as densidades de população relativas aos estados mais altos e mais baixos do *laser*, propõe-se um mecanismo de excitação a dois estágios para um *laser* de $CwAr^+$. A idéia aqui apresentada pode ser utilizada para investigar mecanismos de excitação do outro *laser* de gás nobre.

1. Introduction

Recently, we have obtained a parametrization for the cross section in terms of integrals over radial functions'; it was shown that the convergence of the expansion for the total cross section is fast enough so that only a small number of parameters must be kept in this expansion for the case of electron scattering by Ar^+.

The convenience of such pararnetrization is remarkable because the more complicated the atomic spectra, the better will be the chance that such a parametrization be reliable.

In this work the results of Ref. 1 are used to propose a two-step excitation mechanism for C.w. ion lasers.

Since the advent of the ionized rare gas lasers, much interest has arisen in these laser systems, because they are some of the most intense con-

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tinuous source of coherent radiation in the visible spectrum. The understanding of the excitation mechanisms, in the extremely high-current discharges used, depends upon the calculations of the absolute value and energy dependence of the cross sections for reactions leading to the upper laser states.

Bennet [2,3], has suggested that a possible mechanism of inversion in the pulsed ionized noble gases could be explained by the sudden perturbation method[4]. This method is essentially a one-step process; the neutral atom is simultaneously ionized and excited by impact with a fast moving electron. Koozekanani[5] has performed extensive numerical calculations based on Hartree-Fock wave functions and intermediate coupling in the sudden perturbation method, and Bennet[6], Clout and Heddle[7], Latimer and St. John[8], obtained experimental results for some of the excitation cross sections for the process

$$A + e \rightarrow (A^+)^* + 2e.$$

The results obtained by Bennet[6] were shown to be in good agreement with those obtained by Koozekanani[5], for energies near the peak of the total ionization cross section, or at energies typically involved in Argon ion lasers. But there was serious disagreement between these results and those or Ref. 7. Assuming the sudden perturbation method, only excitation of states which exhibit the same parity as the ground state of the ion, which is $(p)^5\ {}^2P_{3/2}$, can occur. For the case of singly ionized noble gases, the only states which the sudden approximation mechanism would predict to be excited would be given by either a ${}^2P_{1/2}$ or a ${}^2P_{3/2}$ state, since these states have the same spin and orbital angular momentum as the ground state of the ion[5].

Rudko and Tang[9] obtained results for the intensity of lines for transitions from the upper to the lower laser states. From these results, it is clear that the most important contributions to the total power output, are not transitions from the ${}^2P_{1/2}$ and ${}^2P_{3/2}$ upper to the lower laser states for a C.W. process.

Almost all theoretical and experimental efforts have been concentrated on the one step excitation mechanism. In Section 2 we discuss a two step excitation and in Section **3** we present our conclusions.

2. Two Step Excitation Mechanism

The two step processes are

$$\begin{aligned} &e + A \rightarrow 2e + A^+ \\ &e + A^+ \rightarrow e + (A^+)^*. \end{aligned}$$

We will be interested in the process,

$$e + A^+ \rightarrow e + (A+)^*.$$

In other words, we assume that there will be a considerable amount of singly ionized Argon in the ground state after starting the discharge.

One may compare the cross sections obtained by the distorted wave close coupling approximation, given in Table 1 for the process

$$A+ + e \rightarrow (A+)^* + e,$$

with the results of Bennet[6] for the process,

$$A + e \rightarrow (A^+)^* + 2e,$$

State	Energy (Ryd.)						
	2.0	2.5	3.0	3.5	4.0	4.5	5.0
$^4P_{5/2}$	5.68	6.53	8.58	8.85	10.50	14.60	20.10
$^4P_{3/2}$	4.39	4.87	6.14	6.30	7.43	10.10	9.39
$^4P_{1/2}$	5.85	6.60	8.44	8.55	10.10	13.90	18.60
$^4D_{7/2}$	0.003	0.004	0.0047	0.0047	0.0053	0.0061	0.0064
$^4D_{5/2}$	16.80	18.50	22.40	22.50	25.80	30.70	31.80
$^4D_{3/2}$	3.57	3.88	5.52	6.00	7.47	13.20	10.10
$^4D_{1/2}$	2.31	2.70	3.58	3.71	4.44	6.17	5.73
$^2D_{5/2}$	4.78	5.36	6.67	6.77	7.88	9.89	9.77
$^2D_{3/2}$	1.87	2.01	2.49	2.55	2.98	4.09	3.71
$^2P_{1/2}$	4.11	4.56	5.56	5.59	6.43	7.71	7.85
$^2P_{3/2}$	1.31	1.42	1.73	1.75	2.01	2.53	2.46
$^4S_{3/2}$	9.00	9.90	12.00	10.20	14.00	17.20	17.10
$^2S_{1/2}$	2.39	2.64	3.17	3.17	3.61	4.18	4.36

Table 1 - Total excitation cross section from the ground state $^2P_{3/2}$ to states arising from configuration $3p^4 4p$ of A^+: (^3P core) – (Units of $10^{-2}\,\pi a_0^2$)

at the same incoming electron energies. It is seen that the cross sections for some of the transitions in the former process are one order of magnitude greater than those occuring in the latter; therefore, the first process can be thought as a mechanism of excitation of laser states. Furthermore, it has been shown in a previous work that, under the approximation $1 = l'$, excitations from the G.S. to configurations where the parity of the running electron wave function is different from that for the artificially constructed running electron in the ground state, do not occur[1]. [A more detailed investigation shows that, generally speaking, there is a preference for transitions without change in 1 (Ref. 10). We have shown that the contribution to the total cross section from the terms with a change in l is small for transitions between states arising from the $3p^5$ and $3p^4\,4p$ configurations. The same can be expected for transitions between the ground state and the $3p^4$ $4s$ configurations].

This provides an explanation for the population inversion mechanism in a rare gas laser, since excitations from the G.S. to configurations $(3p^4)4s$, and $(3p^4)3d$ cannot occur in this approximation, while states arising from configuration $(3p^4)4p$ are strongly populated. Therefore, a large transition probability, between states arising from this configuration and those arising from configuration $(3p^4)4s$ and $(3p^4)3d$, can be expected on the basis of this theory. As we commented previously, laser action is observed between most of these states[8].

Unfortunately, no experimental measurement of the cross section for the process, $e + A^+ \rightarrow e + (A')^*$, can be found in the literature, and only indirectly we can compare the results of these calculations with the experimental data available. Knowing the excitation cross section, we will calculate the rate of excitation of atoms to any of the allowed states. If we assume only electron excitation, this rate is given by,

$$\frac{dN_B}{dt} = \langle \sigma_B(E)\, v \rangle \left(\frac{I}{e v_d} \right) N - \frac{N_B}{T_B}, \qquad (1)$$

here, N_B is the number of excited atoms in state B per cm^3, N is the number of non excited atoms/cm^3, e is the electronic charge, $\sigma(E)$ is the cross section. The notation $\langle\,\rangle$ means taking the average, v is the thermal velocity, v_d the drift velocity, and T_B the lifetime of the excited state B, given by,

$$T_B = (\sum_{B'} A_{BB'})^{-1}, \qquad (2)$$

where

$$A_{BB'} = (2J_B + 1)^{-1}\left(\frac{64}{3h}\pi^4\right)\frac{S_{BB'}}{\lambda_{BB'}} \tag{3}$$

In Eq. (3), $(2J_B + 1)$ is the multiplicity of the upper level, $\lambda_{BB'}$ the transition wavelength between levels B and B', $S_{BB'}$ the line strength, and the summation is over all the possible lower levels, to which the transition is not forbidden.

The line strength is given by

$$S_{BB'} = \sum_{M_B M_{B'}} |\langle J_B M_B| \bar{P} |J_{B'} M_{B'}\rangle|^2 \tag{4}$$

and results for the probability per unit of time are given in Refs. 9 and 11.

For the steady-state case,

$$\frac{dN_B}{dt} = 0, \tag{5}$$

and we obtain

$$N_B = \left(\frac{I}{ev_d}\right) N\langle\sigma_B(E)\, v\rangle T_B\,. \tag{6}$$

Using Ref. 9, and Eq. 2, we calculate the lifetime of some states arising from configuration $(3p^4)$ 4p, neglecting transitions to the configuration $(3p^4)$3d. These results are given in Table 2.

States	Lifetime (10^{-9} sec.)
$^4D_{5/2}$	7.7
$^4D_{3/2}$	7.4
$^4P_{3/2}$	9.9
$^4S_{3/2}$	4.8
$^4D_{1/2}$	7.4
$^4P_{1/2}$	9.9
$^4P_{5/2}$	9.9
$^2D_{5/2}$	9.7
$^2D_{3/2}$	9.8
$^2P_{3/2}$	9.9
$^2P_{1/2}$	10.5
$^2S_{1/2}$	8.2

Table 2 - Calculated lifetimes for certain states of the $3p^4 4p$ configuration of A^+. The transitions of the $3p^4$ 3d configuration are neglected. (Core ^3P). [From Ref. 9].

To calculate the average of the cross section over the thermal velocities, we must know the velocity distribution of the electrons in the Argon gas laser. Much effort has been put on experiments to determine this distribution, but because of the complexities involved on these experiments, no data is available in the literature. Therefore, any theoretical calculation must be made assuming a certain velocity distribution[12]. In this work, we chose a Boltzman eriergy distribution, and for the electron temperature, $KT = 0.5$, or $T \simeq 0.8 \times 10^5$ °K. This temperature is in the range of the electron temperatures in the electric discharges used for the Argon ion laser[12].

Here we are only interested in comparing the various population densities of states of configuration $3p^4 4p$, by considering excitation from the G.S..

Furthermore, we do not know v_d nor N in **Eq.** 6. Therefore, our results will be given in terms of $\langle\sigma(E)\,v\rangle T$, and v_d, N, **I** are assumed to be the same for excitations of all those states.

Because there is a certain disagreement between the lifetimes given by Refs. 9 and 11, we will calculate $\langle\sigma(E)\,v\rangle T$ for lifetimes given by both references. These results are given in Table 3, where we have deiined,

$$J = \int \sigma(E)\,\boldsymbol{E}\,\exp(-E/KT)\,dE, \tag{7}$$

and we have $\langle\sigma(E)\,v) \sim \boldsymbol{J}$.

State	J	$J \times T_B$ (arbitrary units)	
	arbitrary units	Rudko and Tang	Statz et al.
$^4P_{5/2}$	1.69	6.91	11.57
$^4P_{3/2}$	0.48	2.91	4.84
$^4P_{1/2}$	0.66	3.94	6.55
$^4D_{7/2}$	0	0	0
$^4D_{5/2}$	1.82	11.39	14.06
$^4D_{3/2}$	0.41	2.47	3.09
$^4D_{1/2}$	0.27	1.56	2.01
$^2D_{5/2}$	0.53	4.64	5.16
$^2D_{3/2}$	0.20	1.88	1.98
$^2P_{1/2}$	0.45	3.98	4.72
$^2P_{3/2}$	0.14	1.31	1.40
$^4S_{3/2}$	0.96	4.43	4.64
$^2S_{1/2}$	0.25	1.89	2.12

Table 3 - Average of the cross section over thermal velocities, and population densities in arbitrary units. [States from the core ^3P].

In this work we chose to compare population densities of states arising from the (^3P) core of configuration 3p^{4}4p, because in Ref. 11 only lifetimes for states arising from this core are available.

3. Conclusions

In the present section we will compare the results of Table 3 with those given by Ref. 9.

Using the results of Tahle 3, which give the population densities for direct excitation from the ground state, we can write the most populated states in order of decreasing population.

They are

$$^4D_{5/2}\,,\ ^4P_{5/2}\,,\ ^2D_{5/2}\,,\ ^4S_{3/2}\,,\ ^4P_{1/2}\,,\ ^4P_{3/2}\,,\ \text{etc}\ldots$$

From Ref. 9, the most populated states are

$$^4P_{5/2}\,,\ ^4P_{3/2}\,,\ ^4D_{5/2}\,,\ ^2D_{5/2}\,,\ ^4S_{3/2}\,,\ \text{etc}\ldots$$

If we note that the population densities in Ref. 9 are given within a certain standard deviation, that the results for the cross section in Table 1 were obtained by assuming the various approximations discussed in Refs. 1, 14 and that results of Tables (II) and (III) of Ref. 9 suggest strong cascade from higher states into the $^4P_{5/2}$ and $^4P_{3/2}$ levels, a very good qualitative agreement is obtained between the most populated states given by theory and by Ref. 8.

Also looking at Tables (II) and (III) of Ref. 9, it is difficult to understand the population of the $^2D_{5/2}$ state, because there are no strong decays from states arising from configurations $(3p^4)$ 4d and $(3p^4)$ 5s into this state. Koozekanani[5] suggested, on the basis of the sudden perturbation method, that the $(3p^4)\,4p\,^2D_{5/2}$ state could be excited via the $(3p^4)$ 5s $^2P_{3/2}$ state of Ar^+. The results of Tables (II) and (III) of Ref. 9 show that such a process is of no significance in the laser system studied there; the cascade contribution comes mainly from the (3p^4)4d states, but these contributions are of the same order of magnitude as those for the $^2D_{3/2}$ state which has a population density much smaller than the $^2D_{5/2}$ state. Therefore, direct excitation from the G.S. could account for the larger population of the $^2D_{5/2}$ state, as we see from Table 3.

All previous models proposed to explain the excitation mechanism of the laser states were able to explain only a small number of these laser states as being excited from ground state excitations. The population of the other states were explained by cascade decays from higher states into these states: however, this assumption is not proved by experiments[8,13].

Furthermore, only a few of these higher states could have their populations accounted for by direct excitation on the basis of the one step process, and cascade could not explain the population of the other states. The theory developed here can explain the population of all these states by direct excitation, and because $l = l'$ for these processes, the population for states arising from configurations $(3p^4)\,5s$ and $(3p^4)\,4d$ is expected to be smaller than those for states arising from configuration $(3p^4)\,4p$, and therefore an inversion of population does not take place.

The population of the $(3p)\,{}^4D_{7/2}$ state can be explained by cascade from higher states. Indeed, results from Rudko and Tang, show that there are very intense decays from states arising from configurations $(3p^4)5s$ and $(3p^4)4d$ into this state.

Finally, we want to compare quantitatively our theoretical results for population densities with the results of Ref. 9. Because our results are given in arbitrary units and our calculations take into account only direct excitation from the G.S., we may compare our theoretical results with those of Rudko and Tang[8] for all states where the cascade contributions from upper states are roughly the same. [See Tables II and III, of reference (8)]. This comparison is presented in Table 4.

State	Theory	Experiment
${}^2D_{5/2}$	4.64	4.78
${}^2D_{3/2}$	1.88	1.75
${}^4D_{1/2}$	1.56	1.61
${}^4D_{3/2}$	3.09	3.13
${}^4S_{3/2}$	4.43	3.38
${}^2S_{1/2}$	1.89	1.20

Table 4

From all the results we obtained, there is a strong evidence that direct excitations from the G.S., as well as cascade contributions from higher levels, are very important in the explanation of the population of laser states. These results strongly suggest that a two step process occurs in the C.w. laser system.

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