

ACTIVE MEDIA

PACS numbers: 42.55.Lt; 42.60.Lh
DOI: 10.1070/QE2000v030n07ABEH001767

Analysis of the possibility of lasing on the third continuum in argon

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Abstract. The possibility of lasing on the third continuum in argon was studied theoretically. This possibility is considered from the point of view of two hypotheses on the nature of the third continua, which have survived through the mid-1990s. The gain was calculated under conditions when the oscillation is most probable, namely, in the pressure range between 1 and 30 bar for specific pump power in the range 10^{-3} – 10 MW cm^{-3} bar^{-1}. The gain is shown to be negative under the above conditions and, hence, lasing cannot be achieved.

1. Introduction

It is known that upon excitation of dense rare gases, molecular emission bands (continua) appear in the range of wavelengths exceeding the wavelength corresponding to atomic resonance lines. These bands are commonly numbered sequentially as their distance from the resonance emission line is increased. Thus, the continuum that is located immediately behind the resonance line is referred to as the first continuum, and then the second and the third continua follow [1].

The first continuum arises from the radiative transitions from highly excited vibrational levels of the first electronic excited states $^{1,3}\Sigma_u^+$ of rare gas molecules R_2^* to the ground repulsive state $^1\Sigma_g^+$ (Fig. 1). The second continuum corresponds to the same electronic transition, although from lower vibrational states [2, 3]. The known Xe (174 nm), Kr (146 nm), and Ar (126 nm) excimer lasers operate on the transitions corresponding to the second continua [4 – 8].

The third continua in rare gases have been known since the mid-1950s [9, 10]. The hypotheses about their nature are reviewed in Ref. [11]. Boichenko et al [38] analysed Refs [11 – 37], which were devoted to the study of third continua during the last decade. Only two concepts about their nature still existed by the mid-1990s.

According to Langhoff [12, 13], the third continua are related to the transitions in doubly charged molecular ions from the $R_2^{++}(R_3^{++})$ states to the $R^+ + R^+(R_2^+)$ states. As follows from Ref. [14], the third continua are formed by the transitions of singly charged molecular ions from the R_2^{+*} states [which asymptotically correspond to the ion ground R^+ state and the excited atomic state R^* (^{1,3}P)] to the R_2^+ states (they correspond asymptotically to the ground states of the ion R^+ and the atom R) (see Fig. 1). Trimers R_3^{+*} of singly charged ions can also contribute to the emission of the third continua [11].

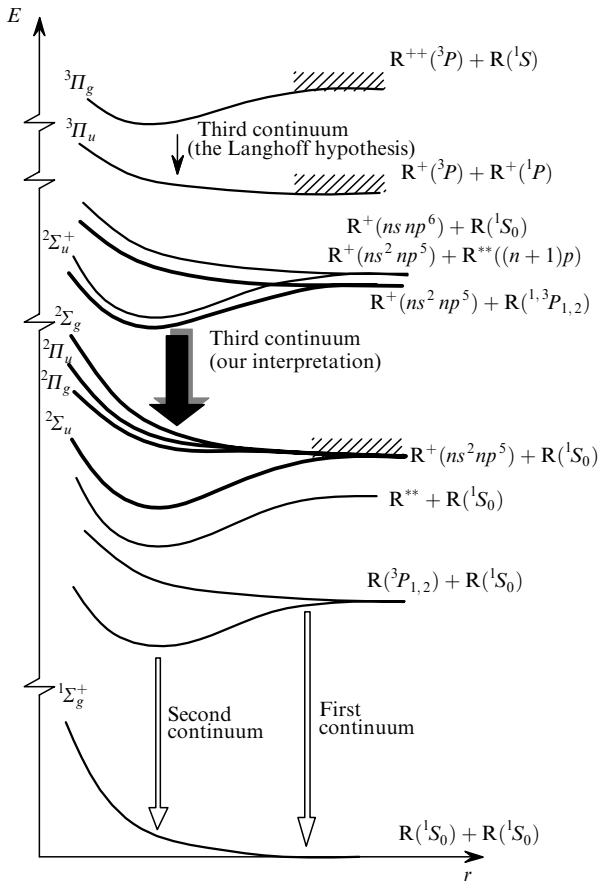

Figure 1. Terms of rare gas molecules with transitions explaining the nature of the first, second, and third continua.

The nature of the third continua was studied often in the expectation that lasing would be obtained [11, 12, 17, 29 – 31]. This hope was reinforced particularly after obtaining lasing on the second continuum transitions. Lasing on transitions of the third continua would be attractive for the simple reason that the active medium of such lasers consists of a rare gas

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Received 29 December 1999
Kvantovaya Elektronika **30** (7) 567 – 572 (2000)
Translated by E N Ragozin; edited by M N Sapozhnikov

and is not aggressive. Moreover, the production of optical elements (primarily mirrors) is simpler and more reliable for the 210 – 300 nm wavelength ranges corresponding to the third continua, whereas the second-continuum transitions are located in the spectral region from 126 to 174 nm. Because the width of the radiative transitions in the third continua is significantly greater than this of the second-continuum transitions and the positions of the emission peaks depend on the medium pressure, a possibility would arise to tune the oscillation wavelength in the 10 – 30 nm range near the radiation wavelength.

A gain of 0.08 cm^{-1} was obtained in the experiment [20] upon high-power proton pumping of xenon. A theoretical study performed in this work revealed that the gain should be negative throughout the pressure and pump power ranges corresponding to the experiments conducted, which suggests that lasing on the third continuum is unfeasible under these conditions. Subsequent experiments confirmed the occurrence of a negative gain.

The third continua in xenon and krypton share a common property that inhibits lasing. The point is that the absorption bands of Kr_2^+ and Xe_2^+ ions, which are among the major absorbers, overlap significantly with the emission bands of the third continua for krypton and xenon [39]. According to current concepts of the nature of emitting states of the third continua, this fact renders lasing in krypton and xenon impossible. The reason is that the population density and the absorption cross sections of the ground state of singly charged molecular ions always exceed the population density and the stimulated-emission cross sections for the molecular excited states of singly charged ions and for the ground state of doubly charged molecular ions.

In comparison with krypton and xenon, argon proves to be a special case. The emission intensity maxima of the third continua of argon lie in the 200 – 240 nm range when the pressure is changed from 1 to 17 bar. The absorption edge of Ar_2^+ molecular ions corresponds to approximately 200 nm [39]. It is precisely this fact that could be of decisive importance in obtaining lasing in argon. Lasing on the third continuum of argon was reported in Ref. [40]. However, since then, no one has been able to reproduce this result which, as noted by the authors themselves, unstable [41].

To elucidate the cause of this instability, several investigations were recently pursued [41, 42]. In Ref. [42], the gain in the 215 – 260 nm range was measured; the gain attained maxima of approximately 0.03 cm^{-1} at about 230 and 244 nm. In Ref. [41] a 'resonator effect' was discovered. The effect was as follows: when the second mirror was mounted in the resonator, the radiation intensity at 220 nm increased by about a factor of 10 and at 240 nm by more than a factor of 10 in comparison with the case when only one mirror was mounted. The experiments were conducted at a pressure of 3 bar. We are nevertheless reminded that nobody has managed to reproduce lasing since 1994 [40]. Since the parameters of the electron pump beam are not given in the papers under discussion, a direct calculation to verify the gain reported in these papers is hindered.

In connection with the foregoing, argon holds the greatest promise as an active medium for obtaining lasing on the third continuum. This paper is dedicated to a theoretical quest for the conditions whereby lasing is possible. The search was performed on the basis of a kinetic description of the processes corresponding to the two hypotheses for the origin of the third continua considered above.

2. On the nature of the emitting states of the third continuum

The two competing viewpoints on the nature of the third continua will be referred to as hypotheses of the emission on the transitions of singly [14] and doubly [12] charged ions (see Fig. 1).

The analysis showed that the features of the third continua can be explained by the emission of singly charged ions. The hypothesis that the emission is due to doubly charged ions [12, 13] was found to be inconsistent with experimental data on the third continua and also with theoretical considerations.

The point is that this hypothesis contradicts the experimental pressure dependences of the emission energy of the third continua; the modern notions of the rate constants of the key plasmachemical reactions occurring in the active medium of rare gases; the experiments on quenching the third continua of argon, krypton, and xenon by different reagents; the experiments on the duration of the emission of the third continua upon a high-power pumping of the medium; and also the experiments on excitation of crystal rare-gas samples.

So, based on the data of Refs [11, 38], one can conclude that the third continua in rare gases are determined primarily by the emission of singly charged ions. Although the emission of doubly charged ions can contribute to the emission of the third continua, this contribution is small and has only a slight effect at pressures $p \geqslant 1$ bar. The emission of doubly charged ions can make a major contribution only for $p < 0.3$ bar, when the absolute radiant energy of the third continua is low in comparison with the energy radiated at optimal pressures (of the order of 1 bar and over).

3. Kinetic model

3.1. Population kinetics of a singly charged molecular ion

We used the same model as in Ref. [25]. Boichenko et al. [25] used this model, based on the hypothesis that the emission is due to singly charged ions [14, 11], to analyse the quenching of the third continua of argon by different rare-gas impurities pumped by the electron beam.

The model comprises 13 reagents: The ground and the first two excited atomic states Ar, Ar^*, Ar^{**} (the centres of the 4*s* and 4*p* levels); the corresponding molecular states Ar_2^* and Ar_2^{**}, Ar^+ ions in the ground state; Ar^{+*} ions in the $3s3p^6$ state; the ground Ar_2^+ state of the molecular ion; the $Ar_2^{+*}(1)$ states which correspond asymptotically to the $Ar^+ + Ar^*(^{1,3}P)$ states; the $Ar_2^{+*}(2)$ states corresponding to the $Ar^{+*} + Ar$ and $Ar^+ + Ar^{**}$ states; as well doubly ionised atomic (Ar^{++}) and molecular (Ar_2^{++}) ions and electrons. The model incorporates about 80 reactions. The equations for the time-dependent reagent densities and the electron and gas temperatures were solved numerically. The calculations were performed employing the PLASER code pack-age [43].

We briefly outline the main channels of population of the states that contribute to the radiation of the third continuum [11, 38]. Upon a weak pumping of the medium, the excited states of a singly charged molecular ion are populated through the following reaction chain. Excited atomic ions are produced in collisions of a neutral particle with a fast electron:

$$Ar + e\,(beam) \rightarrow Ar^{+*} + e + e\,(beam). \qquad (1)$$

Next, the excited ion enters into the conversion reaction

$$Ar^{+*} + 2Ar \rightarrow Ar_2^{+*}(2) + Ar. \quad (2)$$

These states lie approximately 1 – 2 eV higher than those that form the emission of the third continuum. The transition to the working states occurs through de-excitation of $Ar_2^{+*}(2)$ in collisions with rare gas atoms or by electron impact:

$$Ar_2^{+*}(2) + Ar \rightarrow Ar_2^{+*}(1) + Ar,$$
$$Ar_2^{+*}(2) + e \rightarrow Ar_2^{+*}(1) + e. \quad (3)$$

The third continuum is formed by the radiative transitions:

$$Ar_2^{+*}(1) \rightarrow Ar^+ + Ar + \hbar\omega \,(\text{the third continuum}). \quad (4)$$

Upon a strong pumping of the medium (see, e. g., Ref. [20], where the medium was pumped by a high-power proton beam) the main contribution to the population of excited states of singly charged molecular ions is made by the conversion reactions involving excited atomic states [11, 20]:

$$Ar^+ + Ar^{**} + Ar \rightarrow Ar_2^{+*}(2) + Ar,$$
$$Ar^+ + Ar^* + Ar \rightarrow Ar_2^{+*}(1) + Ar. \quad (5)$$

In this case, naturally, the reactions (3) of de-excitation of the $Ar_2^{+*}(2)$ states by neutral particles and electrons may play a significant role as before.

The strong pump differs from the weak one in that the degree of ionisation in the active medium is $\alpha \geqslant 0.1$. This corresponds to specific pump powers $w > 10 - 100$ MW cm^{-3} bar^{-1} [11, 38]. Below, we consider the model of pumping by a hard ioniser (electron and ion beams, short-wavelength radiation, etc.), which is characterised by a universal quantity — the ionisation frequency ν [43]. For instance, upon an electron beam pumping,

$$\nu \approx 2\sigma(E)j/e,$$

where $\sigma(E)$ is the ionisation cross section by electrons with energy E for rare gas atoms; j is the electron current density; and e is the electron charge.

Two changes were introduced in comparison with the model of Ref. [25]. First, the reaction

$$Ar^{+*} + Ar \rightarrow Ar^+ + Ar^*$$

was included, with a rate of 10^{-11} cm^3 s^{-1} (for more details, see Ref. [28]). Second, the frequency of ionisation of the $Ar(^1S_0)$ state with formation of the Ar^{+*} state was changed. Boichenko et al. [25] assumed that $\nu(Ar^{+*}) = 0.17\nu$. A detailed analysis of the Ar^{+*} excitation cross sections is given in Ref. [44]. It follows from this analysis that, according to different papers, the frequency $\nu(Ar^{+*})$ is in the range between 0.025ν and 0.25ν for an electron energy $E = 1$ keV, where the data of Ref. [45] were used for ν for $E = 1$ keV. Most likely [44], it is reasonable to adopt the relationship $\nu(Ar^{+*}) = 0.05\nu$. We will also use this relationship for $E > 1$ keV.

We will now elaborate upon the issue of the cross sections of stimulated-emission and radiation absorption by different plasma components. The stimulated-emission cross section at the third continuum of argon can be estimated from the relationship

$$\sigma^{(3)} = \frac{\lambda^2}{4\Delta\omega} A,$$

with a reasonably high accuracy, where λ is the characteristic radiation wavelength; $\Delta\omega$ is the width of the emission band; and A is the inverse lifetime of the $Ar_2^{+*}(1)$ state. The inverse lifetimes of the states $Ar_2^{+*}(1)$ and Ar_2^* are approximately equal. Hence, taking into account that the width of the third continuum $\Delta\lambda^{(3)}$ for $p > 1$ bar exceeds the width of the second continuum $\Delta\lambda^{(2)}$ by approximately a factor of 6.5 [46], and that, for the $Ar_2^*(^1,^3\Sigma_u^+)$ states, the stimulated-emission cross sections for the second continuum are $\sigma(^1\Sigma_u^+) = 1.2 \times 10^{-17}$ cm^2 and $\sigma(^3\Sigma_u^+) = 9.8 \times 10^{-20}$ cm^2 [47 – 49], we obtain

$$\sigma^{(3)} = \left(\frac{\lambda^{(3)}}{\lambda^{(2)}}\right)^4 \frac{\Delta\lambda^{(2)}}{\Delta\lambda^{(3)}} \left[\frac{1}{4}\sigma^{(2)}(^1\Sigma_u^+) + \frac{3}{4}\sigma^{(2)}(^3\Sigma_u^+)\right] = 3.0 \times 10^{-18} \text{ cm}^2.$$

In deriving this expression, we used the relation $\Delta\omega = 2\pi c\Delta\lambda/\lambda^2$. In addition, we took advantage of the fact that, in the ionisation frequency range under study, the population densities of the $Ar_2^{+*}(1)$ and Ar_2^* states, which correlate with the $Ar^*(^3P_{1,2})$ state, will correspond approximately to the statistical weights of these states.

The absorption cross section of Ar_2^+ increases both with gas temperature and radiation wavelength in the 200 – 240 nm range [39]. As noted above, the emission peak of the third continuum lies in the 210 – 240 nm range when the pressure is changed from 1 to 17 bar. The absorption cross section increases rather steeply with radiation wavelength (Table 1). It is taken as 2×10^{-19} cm^2. This cross section corresponds to the left edge (210 nm) of the emission maximum of the third continuum, being substantially lower than the cross section for the remaining emission domain. Note also that the model takes into account the fact that the rate of the conversion reaction

$$Ar^+ + 2Ar \rightarrow Ar_2^+ + Ar$$

decreases with increasing gas temperature. Specifically, it is equal to $T^{-3/4}1.6 \times 10^{-32}$ cm^6 s^{-1}, where T is expressed in

Table 1. Wavelength and temperature dependences of the Ar_2^+ ion absorption cross sections taken from Ref. [39].

Wavelength (nm)	σ/cm^2	
	300 K	600 K
200	2.73×10^{-20}	2.84×10^{-19}
210	1.60×10^{-19}	9.44×10^{-19}
220	6.73×10^{-19}	2.34×10^{-18}
230	2.17×10^{-18}	4.91×10^{-18}
240	5.38×10^{-18}	8.57×10^{-18}
250	1.11×10^{-17}	1.34×10^{-17}
260	1.89×10^{-17}	1.85×10^{-17}

in electron-volts. For $T = 0.026$ eV (300 K), the reaction rate coincides with the value 2.5×10^{-31} cm^6 s^{-1} obtained experimentally.

The Ar_2^* absorption cross section is approximately equal to 10^{-18} cm^2 [50] or 4×10^{-18} cm^2 [51], and the Ar^{**} absorption cross section, to 3×10^{-18} cm^2 [51]. These cross sections are often calculated from the Kramers formula [52] $\sigma = (J/\hbar\omega)^3 8.32 \times 10^{-18}$ cm^2, where J is the ionisation potential of the excited level; for our data, this formula leads to about the same value. The absorption cross section of Ar^* is 10^{-20} cm^2 [51]. We will use the cross sections given in Table 2. The absorption cross section for Ar_2^{**} is taken to be the same as for Ar^{**}.

One can see that we have adopted, where possible, the underestimated absorption cross sections, which favours lasing. However, if the gain proves to be negative even under these lasing-promoting circumstances, this will mean that lasing is undeniably impossible.

Table 2. Absorption cross sections used in the modeling.

Reagent of the active medium	σ/cm^2
Ar_2^+	2×10^{-19}
Ar_2^{**}	3×10^{-18}
Ar_2^*	10^{-18}
Ar^{**}	3×10^{-18}
Ar^*	10^{-20}

3.2. Kinetics of production of doubly charged ions

Although, as noted in Section 3, doubly charged ions make only a minor contribution to the emission of the third continua in the pressure range of interest, we examined the possibility of lasing even in this hypothetical case.

According to Refs [12, 37], the states of doubly charged ions that emit in the third continuum are populated as follows. Atomic doubly charged ions are produced in a single event upon the fast beam electron ionisation:

$$Ar + e\,(beam) \rightarrow Ar^{++} + 2e + e\,(beam). \quad (6)$$

Then, the conversion

$$Ar^{++} + 2Ar \rightarrow Ar_2^{++} + Ar \quad (7)$$

and the emission

$$Ar_2^{++} \rightarrow 2Ar^+ + \hbar\omega, \quad (8)$$

which was assigned to the third continuum in Refs [12, 13], should occur.

Moreover, Schumann and Langhoff [37] noted that the experimental pressure dependences of the emission intensity can be explained only if it is assumed that the Ar^{++} state is destroyed by the reaction

$$Ar^{++} + 2Ar \rightarrow 2Ar^+ + Ar\,. \quad (9)$$

In the opinion of Schumann and Langhoff [37], this reaction should have an extremely high rate of 1.3×10^{-30} cm^6 s^{-1}, which is virtually equal to the conversion rate. However, so high a reaction rate cannot be realised owing to a large energy defect (for more details, see Ref. [28]). According to both experimental and theoretical results, the rate of decay of doubly charged atomic ions in the interaction with two argon atoms is 1.5×10^{-30} cm^6 s^{-1} and is attributed to the conversion reaction. At the same time, for the total rate of reactions (7) and (9) not to exceed 1.5×10^{-30} cm^6 s^{-1}, Schumann and Langhoff [37] lowered the reaction rate (7) down to approximately 2×10^{-31} cm^6 s^{-1}.

To verify the possibility of lasing under the assumption that the third continuum is formed by the emission of doubly charged molecular ions, reactions (6) – (8) were also included in the model. Neglect of reaction (9) is simply a circumstance beneficial for lasing. There is good reason to emphasise once again that the hypothesis of doubly charged ion emission fails to account for the wealth of experimental data on the third continua, as shown in Ref. [38].

According to Krishnakumar and Srivastava [45], the cross section of a doubly charged ion production for $E = 1$ keV amounts to 5% of the cross section of a singly charged ion production, so that

$$\nu(Ar^{++}) = 0.05\nu.$$

This relationship was used for reaction (6). The rate of reaction (7) was taken to be 1.4×10^{-30} cm^6 s^{-1} (for more details, see Refs [25, 28]). The radiative lifetime of the Ar_2^{++} ion was taken as 5 ns — the radiative lifetime of the third continuum determined experimentally.

The stimulated-emission cross section is estimated by the formula

$$\sigma_{++}^{(3)} = \frac{\lambda^2}{4\Delta\omega} A = \frac{\lambda^4}{8\pi c \Delta\lambda} A$$

with the same value of $\Delta\lambda^{(3)}$ which was used above to estimate the stimulated-emission cross section for singly charged molecular ions. For $A = 1/(5 \times 10^{-9}\ \text{s}) = 2 \times 10^8$ s^{-1} we obtain $\sigma_{++}^{(3)} = 5.3 \times 10^{-18}$ cm^{-2}. However, among the states of Ar_2^{++}, only the $2\,^3\Pi_g$ states have radiative lifetimes of about 5 ns ($\tau > 6.4$ ns [35]); the remaining states [$2\,^3\Pi_u$ ($\tau > 15$ ns) and $1\,^3\Sigma_u^-$ ($\tau \sim 250$ ns)] have far longer lifetimes [35]. Also taking into account the relative statistical weight of the $2\,^3\Pi_g$ state among all of the Ar_2^{++} states, we deduce that the effective stimulated-emission cross section should, in the consideration of the total population of the Ar_2^{++} levels, be at least two times less than that given above. We, therefore, assume that

$$\sigma_{++}^{(3)} = \sigma^{(3)} = 3 \times 10^{-18}\,\text{cm}^2.$$

The models discussed above include the reactions of production of only the dimer states of excited ions. At high pressures, trimers will also be produced efficiently; nevertheless, this approach is valid for high pressures, too. Including the kinetics of production and decay of trimers will result in modifications of $Ar_2^{+*}(Ar_2^{++})$-producing reactions: $Ar_2^{+*}(Ar_2^{++})$- and $Ar_3^{+*}(Ar_3^{++})$-producing reactions would come into play. However, if we do not separate the contributions of dimer and trimer states of excited singly (doubly) charged ions to the emission of the third continuum, the inclusion of trimers will not change the results significantly: The rates of reactions of dimers with some reagents and of similar reactions of trimers with the same reagents are approximately equal.

Table 3. Gain and absorption coefficients (in cm^{-1}) given for the points in time when the $Ar_2^{+*}(1)$ density is maximum ($\varkappa^- = \varkappa^-(Ar_2^+) + \varkappa^-(Ar_2^{**}) + \varkappa^-(Ar_2^*) + \varkappa^-(Ar^{**}) + \varkappa^-(Ar^*)$). The pump intensity is related to the ionisation frequency as $W = 112pv$, where W is expressed in W cm^{-3}, v in inverse seconds, and p in atmospheres.

p/bar	v/s^{-1}	$\varkappa^+(Ar_2^{+*}(1)) - \varkappa^-$	$\varkappa^+(Ar_2^{++}) - \varkappa^-$	$\varkappa^+(Ar_2^{+*}(1))$	$\varkappa^+(Ar_2^{++})$	$\varkappa^-(Ar_2^+)$	$\varkappa^-(Ar_2^{**})$	$\varkappa^-(Ar_2^*)$	$\varkappa^-(Ar^{**})$	$\varkappa^-(Ar^*)$
1	10^5	-9.51×10^{-3}	-9.34×10^{-3}	2.99×10^{-5}	6.19×10^{-5}	-3.31×10^{-4}	-3.53×10^{-4}	-4.61×10^{-4}	-8.35×10^{-3}	-7.71×10^{-5}
	10^4	-8.63×10^{-3}	-8.60×10^{-3}	1.27×10^{-5}	2.77×10^{-5}	-3.25×10^{-4}	-3.14×10^{-4}	-5.90×10^{-4}	-7.33×10^{-3}	-6.81×10^{-5}
	10^3	-1.90×10^{-3}	-1.82×10^{-3}	2.99×10^{-7}	7.49×10^{-6}	-1.22×10^{-4}	-4.35×10^{-5}	-3.64×10^{-4}	-1.28×10^{-3}	-1.48×10^{-5}
	10^2	-1.46×10^{-4}	-1.23×10^{-4}	1.71×10^{-8}	8.83×10^{-7}	-2.72×10^{-5}	-1.87×10^{-6}	-2.79×10^{-5}	-6.60×10^{-5}	-9.52×10^{-7}
	10	-8.50×10^{-6}	-8.05×10^{-6}	1.56×10^{-9}	9.08×10^{-8}	-3.54×10^{-6}	-5.51×10^{-8}	-2.46×10^{-6}	-2.01×10^{-6}	-7.58×10^{-8}
3	10^5	-4.74×10^{-2}	-4.81×10^{-2}	1.76×10^{-4}	1.66×10^{-4}	-1.84×10^{-3}	-4.58×10^{-3}	-6.99×10^{-3}	-3.45×10^{-2}	-3.10×10^{-4}
	10^4	-2.54×10^{-2}	-2.53×10^{-2}	1.52×10^{-5}	7.82×10^{-5}	-9.40×10^{-4}	-1.64×10^{-3}	-1.13×10^{-2}	-1.13×10^{-2}	-1.42×10^{-4}
	10^3	-3.11×10^{-3}	-2.96×10^{-3}	6.82×10^{-7}	1.11×10^{-5}	-2.20×10^{-4}	-6.88×10^{-5}	-2.10×10^{-3}	-5.75×10^{-4}	-1.01×10^{-5}
	10^2	-3.19×10^{-4}	-2.94×10^{-4}	5.86×10^{-8}	1.20×10^{-6}	-5.96×10^{-5}	-3.69×10^{-6}	-1.91×10^{-4}	-4.05×10^{-5}	-7.17×10^{-7}
	10	-2.47×10^{-5}	-2.41×10^{-5}	5.61×10^{-9}	1.23×10^{-7}	-1.05×10^{-5}	-1.19×10^{-7}	-1.22×10^{-5}	-1.42×10^{-6}	-4.07×10^{-8}
10	10^5	-2.12×10^{-1}	-2.11×10^{-1}	2.49×10^{-4}	6.27×10^{-4}	-5.84×10^{-3}	-2.85×10^{-2}	-1.26×10^{-1}	-5.05×10^{-2}	-7.42×10^{-4}
	10^4	-5.51×10^{-2}	-5.43×10^{-2}	1.38×10^{-5}	1.10×10^{-4}	-1.64×10^{-3}	-1.94×10^{-3}	-4.78×10^{-2}	-2.98×10^{-3}	-7.12×10^{-5}
	10^3	-8.16×10^{-3}	-8.03×10^{-3}	1.08×10^{-6}	1.32×10^{-5}	-4.16×10^{-4}	-8.42×10^{-5}	-7.37×10^{-3}	-1.72×10^{-4}	-3.91×10^{-6}
	10^2	-9.05×10^{-4}	-8.67×10^{-4}	1.00×10^{-7}	1.39×10^{-6}	-1.15×10^{-4}	-4.57×10^{-6}	-7.34×10^{-4}	-1.36×10^{-5}	-2.52×10^{-7}
	10	-7.73×10^{-5}	-7.70×10^{-5}	9.79×10^{-9}	1.41×10^{-7}	-2.77×10^{-5}	-2.37×10^{-7}	-4.83×10^{-5}	-8.36×10^{-7}	-1.54×10^{-8}
30	10^5	-4.47×10^{-1}	-4.48×10^{-1}	1.92×10^{-4}	8.97×10^{-4}	-1.08×10^{-2}	-3.17×10^{-2}	-3.92×10^{-1}	-1.40×10^{-2}	-3.77×10^{-4}
	10^4	-1.18×10^{-1}	-1.18×10^{-1}	1.46×10^{-5}	1.24×10^{-4}	-3.07×10^{-3}	-2.24×10^{-3}	-1.11×10^{-1}	-1.02×10^{-3}	-2.97×10^{-5}
	10^3	-2.11×10^{-2}	-2.07×10^{-2}	1.24×10^{-6}	1.41×10^{-5}	-7.64×10^{-4}	-1.08×10^{-4}	-1.97×10^{-2}	-6.47×10^{-5}	-1.60×10^{-6}
	10^2	-2.40×10^{-3}	-2.33×10^{-3}	1.18×10^{-7}	1.46×10^{-6}	-2.02×10^{-4}	-5.19×10^{-6}	-2.12×10^{-3}	-4.77×10^{-6}	-8.83×10^{-8}
	10	-2.31×10^{-4}	-2.18×10^{-4}	1.16×10^{-8}	1.47×10^{-7}	-5.53×10^{-5}	-3.33×10^{-7}	-1.62×10^{-4}	-3.85×10^{-7}	-5.75×10^{-9}

4. Discussion

To find the conditions under which lasing is possible, we calculated the small-signal gain in the ionisation frequency range $v = 10 - 10^5$ s^{-1} [which corresponds to a specific pump power $w = 10^{-3} - 10$ MW cm^{-3} bar^{-1}] and the pressure range $p = 1 - 30$ bar. Below the $p = 1$ bar pressure, the emission intensity of the third continua is low [38]. For an ionisation frequency $v = 10^5$ s^{-1}, the calculated gas temperature T amounts to ~ 0.2 eV by the end of the pulse. A further increase in the ionisation frequency will result in still higher gas temperatures, rendering direct use of this model impossible. Inclusion of the reactions of decay of molecular neutral and ion states by the impact of heavy particles (argon atoms) will be in order. However, these reactions would only lower the feasibility of lasing. It is not clear why the conditions for lasing for $v > 10^5$ s^{-1} would improve in comparison with those for $v < 10^5$ s^{-1}. That is why $v = 10^5$ s^{-1} was the limiting ionisation frequency in our modelling.

In our calculations, the pump pulse length (FWHM) was 100 ns, which is typical of exciplex and excimer laser excitation. The results of the calculations are given in Table 3. The small-signal gain coefficients were negative throughout the whole calculation time (up to 200 ns). Table 3 shows them for the points in time at which the density of $Ar_2^{+*}(1)$ peaked. We emphasise that, for Table 3, the times it takes to attain maximum densities of $Ar_2^{+*}(1)$ and Ar_2^{++} are different for $p = 1$ bar, $v = 10^3 - 10^5$ s^{-1} and for $p = 3$ bar, $v = 10^4 - 10^5$ s^{-1}; for the remaining p and v, these times are either equal or nearly equal.

One can see that there exist no domains where the gain $\varkappa = \varkappa^+ - \varkappa^-$ is positive. Moreover, each of the absorption coefficients arising from specific components, with the exception of Ar^*, exceeds $\varkappa^+$ throughout the domain investigated, some of $\varkappa^-$ exceeding $\varkappa^+$ by several orders of magnitude. The absence or the presence of absorption by Ar_2^+ can in no way affect the feasibility of lasing. Moreover, should the stimulated-emission cross sections be 10 times (or more) those used in the modelling, this will not result in positiveness of $\varkappa$. Recall also that the smallest absorption cross sections of the known ones were adopted for the radiation absorption by the Ar_2^+ and Ar_2^* states. Taken together, this demonstrates that lasing on the third continuum is unfeasible.

5. Conclusions

As follows from Refs [11, 38], the third continua in rare gases for $p \geqslant 1$ bar are determined primarily by the emission of singly excited molecular ions. The feasibility of lasing on the third continuum in argon was considered using a transient kinetic model. Although the third continua of rare gases under the specified pressures cannot be ascribed to the emission of doubly charged molecular ions, the possibility of lasing was also analysed in this hypothetical case.

Our study showed that lasing on the third continuum in argon is unfeasible for $p = 1 - 30$ bar and $w = 10^{-3} - 10$ MW cm^{-3} bar^{-1}. In all likelihood, lasing is not possible for $w > 10$ MW cm^{-3} bar^{-1}, either. The emission of the third continua falls in the wavelength range where absorption by neutral atomic and molecular excited states as well as by molecular ions is very high. On the other hand, owing to the large width of the third continua, the cross section for sti-mulated emission is low. For this reason, each of the reagents – Ar^{**}, Ar_2^*, Ar_2^{**}, and Ar_2^{++} – is capable to preclude

lasing.

The third continua still have not found application. Owing to a broad emission line, they will most likely find application in the calibration of spectral instruments [11]. Now that progress has been made in interpreting the third continua, the question arises of the nature of continua with wavelengths longer than those corresponding to the first, second, and third continua in rare gases. It seems that the continuum in the 230 – 500 nm range in neon is a noteworthing object to be studied (according to Ref. [53], the lower bound of this continuum can extend to 120 nm). This continuum can be easily observed upon a discharge pumping [11]. Because no other bands were observed between 90 – 100 nm (the third continuum in neon) and 230 nm in neon, this continuum should, considering the tradition of notation of the rare-gas continua [1], be referred to as the forth continuum of neon.

Acknowledgements. This work was supported by the INTAS (Grant No. 96 0351).

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