Quantum Electronics **36** (12) 1176 – 1180 (2006)

LASER APPLICATIONS AND OTHER TOPICS IN QUANTUM ELECTRONICS

PACS numbers: 42.55.Lt; 34.80.Gs
DOI: 10.1070/QE2006v036n12ABEH012790

On the possibility of pumping Xe_2^* lasers and VUV lamps in the afterglow of a background-electron multiplication wave

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***Abstract*. It was shown earlier that the ionisation propagation in a gas at about the atmospheric pressure may proceed due to the multiplication of the existing electrons with a low background density rather than the transfer of electrons or photons. We consider the feasibility of using the plasma produced in the afterglow of this background-electron multiplication wave for pumping plasma lasers (in particular, Xe_2^* xenon excimer lasers) as well as excilamps. Simulations show that it is possible to achieve the laser effect at $\lambda \approx 172$ nm as well as to substantially improve the peak specific power of the spontaneous radiation of xenon lamps.**

Keywords: *xenon lamp, excimer laser, afterglow, VUV radiation.*

1. Introduction

It is known that beam-pumped lasers operating on photodissociation xenon transitions were the first-realised excimer lasers [1 – 4] (see also recent reviews [5, 6] and collection [7]). However, numerous attempts to pump Xe_2^* lasers by a gas discharge instead of a beam have not been successful. Lasing by Kr_2^* dimers in a discharge has been reported [8] and amplification by Ar_2^* has been observed [9]. However, so far these results have not been reproduced by other scientific groups.

The difficulty consists primarily in the high requirements imposed on the specific pump power. The matter is that the temperature increase to several tenths of an electron-volt leads to the quenching of lasing due to the increase of absorption in photoassociation reactions (for more details, see Refs [5, 7]). As for the ordinary discharge, for a high energy input it overheats the gas. There are grounds to believe that the pumping of rare-gas dimers can be achieved by using the recently realised (see review Ref. [10]) volume nanosecond discharges with a subnanosecond rise time. We will primarily consider the feasibility of pumping dense xenon.

2. On the prospect for employing the afterglow of a background-electron multiplication wave to pump plasma lasers

2.1 Background-electron multiplication wave

In Refs [11, 12], attention was drawn to the fact that the ionisation in a gas at about the atmospheric pressure can propagate due to multiplication of the existing electrons with a low background density instead of the transfer of electrons or photons. Simply stated, the difference in electron multiplication times at different points in space, which arises from the nonuniformity of the field intensity magnitude, may give rise to the motion of ionisation front not related directly to electron transfer. The preliminary background ionisation may be provided by a small number of runaway electrons as well as by the natural background of radioactivity and cosmic radiation. The background formation is comprehensively discussed in Ref. [12].

The multiplication wave* emerges at conducting irregularities, its front travels in the direction opposite to the gradient of the electric intensity modulus and its speed is proportional to the gas density. In this case, the direction of multiplication wave propagation is independent of the direction of the electric field. This allowed abandoning the photon hypothesis of streamer formation and making an assumption that both anode- and cathode-directed streamers are a multiplication wave [11, 12], the streamer arising from the instability of the multiplication wave front [13].

As indicated by experiments, when the rise time of the high-voltage pulse (of the order of one hundred kilovolts) is short enough (ranges into the subnanoseconds), the multiplication wave produces volume ionisation and the streamer has no time to develop. In particular, it is precisely this volume ionisation that accounts for the mechanism of high-power subnanosecond beam generation in dense gases [10]. The beam generation takes place when the multiplication wave approaches the anode and the nonlocal runaway-electron criterion is fulfilled [10, 14, 15].

It was also shown that the spatial distribution of the plasma glow between a plane electrode and a spherical (as well as pointed) one occupied a substantial volume for a subnanosecond rise time of the high-voltage pulse [16, 17].

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Received 30 June 2005; revision received 26 April 2006
Kvantovaya Elektronika **36** (12) 1176 – 1180 (2006)
Translated by E.N. Ragozin

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* We use the term ‘multiplication wave’ to distinguish this process from the commonly considered ionisation wave caused by transfer processes (electron drift and electron heat transfer).

In this case, the geometry of glow regions is nearly the same on changing the sign of the supplied voltage, which may only be attributed to the existence of the multiplication wave, which is independent of the sense of the field.

Note that the above conception of volume-discharge and high-current electron-beam production in a dense gas [10, 14, 15] was called into question in [18], where it was stated that the theory is incorrect from all aspects and all the experiments are doubtful. However, in Refs [19, 20] it was shown that the key critical concepts of Ref. [18] are erroneous.

Thus, the volume discharges on the basis of background-electron multiplication wave at about an atmospheric pressure have been realised. There is good reason to consider the prospect of using them for pumping lasers and high-pressure lamps.

3. Multiplication wave and plasma lasers

There are many discharge types: glow, arc, corona, spark discharges, etc. A wealth of papers, reviews, and books are devoted to them. However, a pulsed discharge, which is underlain by the background-electron multiplication wave in a nonuniform field, until now has not been considered as a separate form of discharge. At the same time, it possesses several distinguishing features. Of significance to laser physics is the fact that the plasma produced by this discharge is overcooled in the degree of ionisation (recombination-nonequilibrium plasma), and such media serve as active media of plasma lasers [7, 21, 22].

In this discharge, kinetic processes take place in the following way. Electrons are overheated at the front of multiplication wave. They accelerate in the electric field and ionise the gas. The plasma screens the field behind the multiplication wave front, with the effect that the field-induced heating of electrons is severely decreased. Electrons begin to cool down due to collisions with gas atoms. Since the degree of ionisation is not high for a high gas density, the electrons are cooled down to a low temperature limited only by recombination heating. In many respects the situation is similar to that which takes place when a dense gas is pumped by a pulsed electron beam. These questions are considered in sufficient detail in the theory of plasma lasers [7, 21 – 24].

The electron density N_e behind the multiplication wave can be estimated from the expression

$$E = 2\pi e r_D N_e,$$

where E is the electric field intensity; $r_D = [T_e/(4\pi e^2 N_e)]^{1/2}$ is the Debye radius; e is the electron charge; and T_e is the electron temperature. For instance, for $E \sim 10^5$ V cm^{-1} and $T_e \sim 1$ eV we have $N_e = E^2/(\pi T_e) \sim 2 \times 10^{16}$ cm^{-3}. Close values result from calculations of the multiplication wave propagation between a wire and a cylinder in the atmosphere of xenon [7, 10, 14, 15, 25], which were made on the basis of the diffusion-drift model for xenon elaborated in Ref. [26].

Note that producing a plasma of such density with the use of an electron-beam pump requires highly intense electron beams with a current density $j \sim 100$ kA cm^{-2}. Lasing by Xe_2^* excimers was obtained even for $j \sim 100$ A cm^{-2}.

The dimensions of the overcooled plasma region are controlled by the velocity of the multiplication wave. According to calculations by analytical formulas proposed in Refs [11, 12], to simulations in the framework of the diffusion-drift model [7, 10, 14, 15, 25], and experimental data [7, 10], for a voltage of ~ 100 kV the multiplication wave traverses a distance $d \sim 1$ cm in less than 1 ns. Therefore, one would expect a transverse dimension of the active medium of about 1 cm.

Electrons are cooled due to inelastic and elastic collisions. The time taken to cool down to a temperature $T_e \sim 0.1$ eV in a helium atmosphere is equal to ~ 10 ns and in the xenon atmosphere to ~ 100 ns (for more details, see below the results of calculations). When employing xenon as the active laser medium it is therefore expedient to add helium in order to speed up the cooling and intensify recombination. However, the main reason for the proposed dilution of xenon by helium is that a uniform discharge in xenon is more difficult to realise.

From the aforesaid it follows that the afterglow plasma of background-electron multiplication wave holds promise for obtaining lasing by those transitions which were observed to lase in a dense gas pumped by an electron beam and in the afterglow of a pulsed discharge [7]:

– by transitions of atomic xenon (this was realised in Ref. [27]);

– by transitions of atomic neon in Penning mixtures with hydrogen and argon;

– by transitions of metal ions (cadmium, zinc, strontium, calcium) mixed with dense rare gases;

– by transitions of exciplex molecules (KrF, XeCl, etc.);

– by transitions of rare-gas dimers (Xe_2, Kr_2, Ar_2).

Note at the same time that gas-discharge lasers operating in the gas – plasma transit regime (i.e. in ionisation-nonequilibrium plasma), lasers utilising self-terminating transitions in particular, are unpromising for pumping by a discharge on the basis of multiplication wave, unlike plasma lasers operating in the plasma – gas transit regime (i.e. in recombination-nonequilibrium plasma; for more details, see Refs [7, 21 – 24]). To take one example, an attempt to put in operation a nitrogen laser made in Ref. [27] using a discharge with a multiplication wave did not meet with success.

In the present paper we restrict our consideration to the feasibility of pumping Xe_2^*.

3.1 Simulations of Xe_2^* afterglow

As noted above, estimates suggest that a relatively high electron density should be produced after passage of a multiplication wave through the gas. In this case, simulations may be performed by either specifying the required initial electron density or modelling an external hard ionisation source which affords the requisite electron density in a short time. The latter approach has been adopted in our work, i.e. the initial electron density is provided by an imaginary electron beam.

We considered the afterglow of a xenon plasma with the initial electron density corresponding to the above estimates for the discharge involving the background-electron multiplication wave. Use was made of the kinetic model of a xenon – helium mixture described in Ref. [28]*. An inves-

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* It is noteworthy that the Xe_2^* radiation in the afterglow of a discharge was also considered in a recent paper by Lo et al. [29].

tigation was made of the regime whereby the amplified spontaneous emission exerted a weak effect on the population density of the Xe_2^* dimer levels. It was assumed that the initial electron density was produced by the pulse of an external electron beam. The beam action was characterised by the ionisation frequency $\nu = 2\sigma j/e$, where σ is the ionisation cross section. The frequencies for the beam excitation from the ground state were introduced in a similar way. In this case, the energy input to the corresponding gas component is given by the expression

$$W = E_p \nu N,$$

where E_p is the energy expended to produce an electron–ion pair ($E_p = 22$ eV for xenon and 46 eV for helium) and N is the ionised atom density. For simplicity, the time dependence of the ionisation frequency was assumed to be bell-shaped. The beam pump duration was taken to be short enough in comparison with the characteristic afterglow development times. The current density was selected in such a way as to afford the production of the specified electron density.

The kinetic equations for particle number balance included 17 components: ions and the excited states of atomic and molecular xenon and helium. In addition, the thermal balance equations were considered for the electron and gas temperatures.

In the description of spontaneous emission we considered two states of the xenon dimer: the resonance $[Xe_2(^1\Sigma_u^+)]$ and metastable $[Xe_2(^3\Sigma_u^+)]$ states, which make contributions to the emission band under discussion. In this case, to calculate the specific powers of spontaneous emission, we used the expressions

$$Q_1 = A_1\hbar\omega[Xe_2(^1\Sigma_u{}^+)], \; Q_2 = A_2\hbar\omega[Xe_2(^3\Sigma_u^+)],$$

$$Q = Q_1 + Q_2,$$

where $A_1 = 1.82 \times 10^8$ s^{-1}, $A_2 = 10^7$ s^{-1} are the spontaneous decay rates; $[Xe_2(...)]$ are the population densities of the corresponding dimers. The efficiency η of conversion of the energy inputted into the medium to spontaneous radiation was defines as

$$\eta = \frac{\int_0^\infty Q\mathrm{d}t}{\int_0^\infty W\mathrm{d}t}.$$

In our model, advantage was taken of the plasma chemical reaction rate coefficients which had earlier been employed in the simulation of XeCl and XeF laser kinetics, the Kr–Xe binary mixture kinetics, as well as the kinetics of the third continua of xenon (for more details, see Ref. [28]). The kinetic model of the Xe–He mixture took into account 132 reactions. The system of kinetic equations was formed and numerically solved with the use of PLASER code package [7, 22–24, 30].

4. Results of simulations

4.1 Xe_2^* lasing conditions

The calculated characteristics of Xe- and He–Xe-plasma afterglow are represented in Figs 1–3. The simulations were carried out for different gas pressures p for the same energy input (~ 74 mJ cm^{-3}), the base duration of a discharge pulse was ~ 0.1 ns. Under these conditions, the highest electron density was equal to $\sim 2 \times 10^{16}$ cm^{-3}.

One can see that gain in pure xenon arises only for $p > 1$ atm (Fig. 1a). With increasing pressure, the peak value of the gain rises (Fig. 2), but the characteristic lifetime

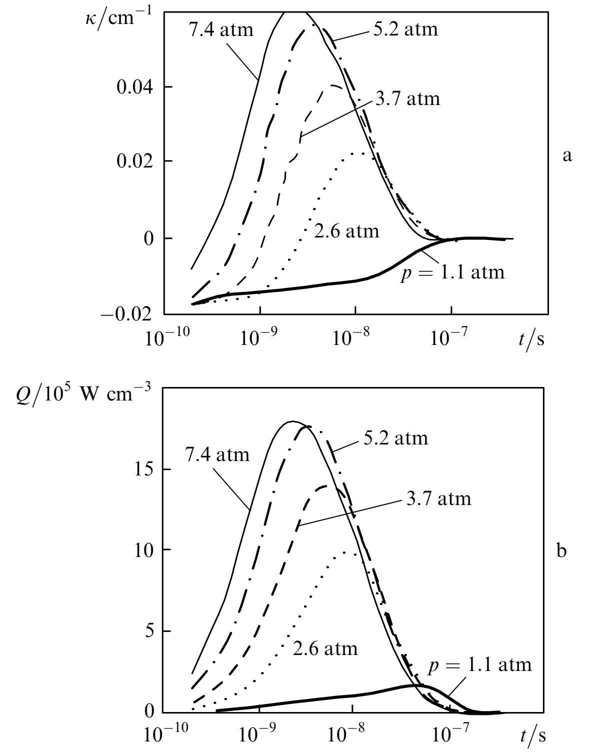

Figure 1. Time dependences of the gain κ (a) and the specific power of spontaneous emission Q (b) at the ~ 172-nm Xe_2^* photodissociation transition for different pressures of pure xenon.

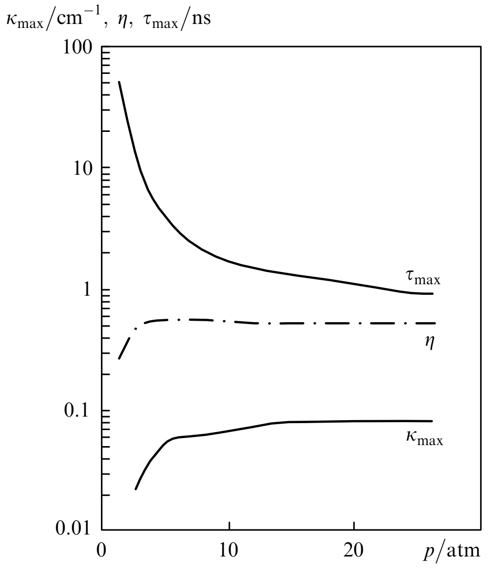

Figure 2. Peak gain κ_{max}, efficiency η of inputted-energy conversion to spontaneous radiation, and time τ_{max} to attainment of the peak spontaneous emission power for the ~ 172-nm Xe_2^* transition as functions of pure xenon pressure.

of the gain and the time to attainment of the peak gain ($\sim 1 - 10$ ns) shorten in this case. For $p \approx 2$ atm, the gain is ~ 0.02 cm^{-1}, which is sufficient for the onset of laser oscillation with an active medium of length ~ 10 cm.

It has been known that lasing in pure xenon under beam pumping takes place for $p > 13$ atm. The point is that a relatively long (50 – 100 ns) pump by a high-power electron beam is responsible for the heating of the active medium. This substantially strengthens absorption of laser radiation due to photoassociative transitions, which are the reverse of the laser transition. This issue was investigated in sufficient detail in Refs [7, 21 – 24]. It is possible to substantially lower the threshold pressure by taking advantage of a discharge with a subnanosecond rise time.

It goes without saying that the production of a volume discharge in a dense gas is an intricate task because of the demanding requirements on the steepness of the rise time of the voltage pulse. However, Alekseev et al. [27, 31] reported that they managed to achieve this for a helium pressure below 6 atm and a nitrogen pressure below 3 atm. It has also been possible to produce a specific energy input of ~ 1 J cm^{-3} in a time of 5 ns [10]. That is why the production of a volume nanosecond discharge for a xenon pressure of 2 atm and an energy input of ~ 0.1 J cm^{-3} also appears to be a realistic task. To avoid energy input into the discharge upon the shorting of the electrodes, advantage can be taken of a dielectric barrier at the anode, as is done in excilamps (see below).

To lower the working xenon pressure, it is expedient to employ helium as the buffer gas. Simulations show (Fig. 3) that even the addition of 1 atm of helium to xenon at the same pressure gives rise to a positive gain. It is pertinent to note that the gains in pure xenon are higher than in xenon – helium mixtures for the same partial xenon pressures. However, in pure xenon it is more difficult to realise the volume discharge wave.

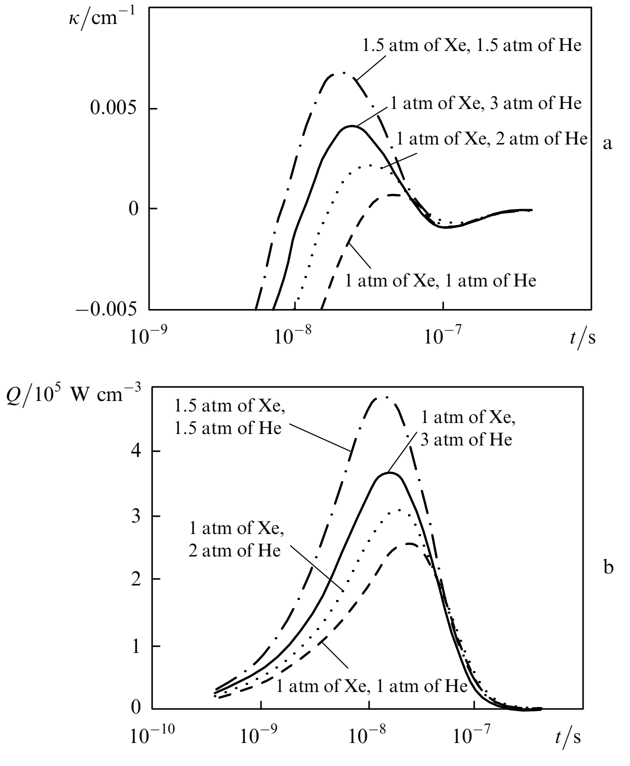

Figure 3. Time dependences of the gain κ (a) and the specific power of spontaneous emission Q (b) at the ~ 172-nm Xe_2^* photodissociation transition for different pressures and compositions of xenon – helium mixtures.

4.2 On an increase in the lamp intensity on Xe_2^* transitions

It is well known that Xe_2^* lamps involving barrier discharges exhibit efficiencies of over 50 % when employing cathodes with a small radius of curvature (for example, wires) [32, 33]. Experimental [32 – 35] and theoretical [26, 35 – 37] investigations of these excilamps allow a conclusion that the volume ionisation in them takes place by way of precisely the electron multiplication wave. It is not until the passage of the multiplication wave that the shorting of the discharge gap and the quasistationary charge accumulation at the barrier are realised [26]. However, these lamps operate at not-too-high a pressure ($p = 100 - 200$ Torr), and pump pulse duration is therefore relatively long (~ 1 μs).

To maximise the pulsed output power of these excilamps with retention of their high average efficiency is of interest for some applications. As suggested by our simulations (Fig. 1b), to do this requires going over to higher pressures ($p > 1$ atm). In this case, it is possible to attain a spontaneous emission specific power of 1 MW cm^{-3} for $p = 2$ atm. Consequently, it is required to employ voltage pulses with a subnanosecond rise time.

Note that according to the calculations of Ref. [28] the efficiency of input-energy conversion to the energy of spontaneous emission was high ($\eta \approx 50\,\% - 60\,\%$) for long pump pulses ($\sim 1$ μs) and moderate pressures ($p = 100 - 200$ Torr). For short pulses (~ 0.1 ns), as suggested by the simulations outlined above, to attain a high conversion efficiency necessitates directing attention towards higher gas pressures ($p > 1$ atm) (Fig. 2). We also mention that the optimal input power increases, according to the calculations of Ref. [28], with shortening the pump pulse duration. The output efficiency of the lamps depends only slightly on the pressure when it is higher than the optimal one.

The occurrence of the optimum for pressures of ~ 5 atm in our calculations is related to the quenching of excited excimer states by electrons. The characteristic electron quenching time for excimer states for an electron density of $\sim 2 \times 10^{16}$ cm^{-3} is equal to 5 ns, the excimer production time at a pressure of 5 atm is also about 5 ns. Furthermore, the heating of gas is also partly responsible for the shift of the optimum towards higher pressures (in comparison with the above pressures $p = 100 - 200$ Torr), because the emission efficiency lowers with increasing gas temperature, and the gas temperature lowers with increasing pressure. For a pressure of 1 atm, for instance, by the end of the pumping pulse the gas temperature rises from 300 K to a value higher than 400 K, while it remains practically invariable for a pressure of 20 atm.

5. Conclusions

We have shown that the plasma produced in the afterglow of the multiplication wave of a nanosecond-long discharge with a subnanosecond rise time may be employed to develop different plasma lasers, including rare-gas dimer lasers. In addition, these discharges hold promise for the development of excilamps delivering high-power pulsed spontaneous emission.

Acknowledgements. The authors thank V.F. Tarasenko for helpful discussions. This work was supported by the International Science and Technology Centre under Project No. 2706.

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