

# Metallization of cells in pulsed lasers. Causes and effects

V.M. Klimkin

*Institute of Atmospheric Optics,  
Siberian Branch of the Russian Academy of Sciences, Tomsk*

Received November 1, 2000

Appearance of a metal coating on the inner surfaces of gas-discharge channels causes contraction of discharges in pulsed metal lasers due to weakening of the skin effect.

## Introduction

A repetitively pulsed mode is the most interesting for practical applications of pulsed gas-discharge lasers operating at electronic transitions of chemical elements and some molecules. The repetition frequency of laser pulses in this mode is  $10^4$ – $10^5$  Hz.

Repetitively pulsed metal-vapor lasers have a long history of development, and the processes proceeding in the discharge plasma, supply circuits, switches, etc. have been extensively studied. However, there are some open problems in the physics of such lasers. Two of them: scaling of laser systems and extending of the list of active media capable of operating in the pulsed mode with a somewhat long service life, are of principal character.

Such a character of these problems is caused by practical needs, on the one hand, and the complexity of physical problems to be solved, on the other hand. They involve, in particular, the following questions:

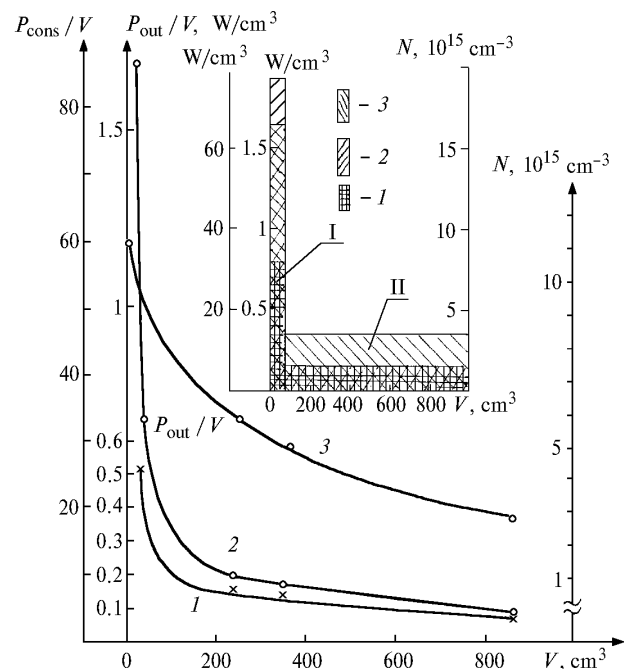
– Why does the longitudinal high-energy pulsed discharge in gas-discharge channels up to 10 cm in diameter not contract?

– Why is the long repetitively pulsed lasing characteristic of only Cu and Au, whereas the Mendeleev table includes about 20 elements with a suitable structure of energy levels?

## 1. Two problems in physics of repetitively pulsed lasers

1. The physical essence of the problem of scaling the pulsed lasers was studied using the Cu-vapor laser as an example. In this paper, we consider only two aspects of the problem. The first (energetic) aspect is that the specific characteristics of the active media, such as specific output energy, density of active particles, and specific input power, decrease significantly as the diameter of the discharge channel increases. At the same time, the existing models of an active medium do not predict such a behavior. It was found experimentally<sup>1</sup> that for the Cu-vapor laser with the discharge channel  $\varnothing > 1$  cm the values of the specific characteristics mentioned above are roughly five times less than those for the same laser with the discharge channel less than 1 cm in diameter. The dependence of the mentioned characteristic on the channel diameter has some peculiarities: as the diameter

increases from 0.3 to 1 cm, the specific characteristics drop down significantly, whereas they change only slightly for channels with the diameters from 1 to 10 cm. For example, in Fig. 1 borrowed from Ref. 2 it is clearly seen that the specific characteristics of the Cu-vapor laser change sharply near  $\varnothing \sim 1$  cm. This allows us to speak about the existence of two lasing modes: for narrow and wide channels. It is clear that lasing in narrow and wide channels differs in the mechanisms of limiting the most important parameters of the active medium, such, for example, as the Cu vapor density. If the problem of conservation of the active medium properties will be solved, for example, keeping the vapor density in large-diameter channels at the level of that in channels  $\varnothing \leq 1$  cm, then the output power of the Cu-vapor laser of about 500–1000 W/m could be expected.



**Fig. 1.** Experimentally observed dependence of specific characteristics of Cu-vapor laser on the volume  $V$  of the discharge channel: consumed power  $P_{\text{cons}}$  (1), specific output power  $P_{\text{out}}$  (2), and density of Cu atoms<sup>1</sup> (3);  $V$  is the volume of gas-discharge channel,  $N$  is the vapor density. The fragment interprets this dependence within the hypothesis on two lasing modes: for narrow (I) and wide (II) discharge channels.<sup>2</sup>

The second (gas-discharge) aspect is connected with the absence of a model describing the discharge behavior. For metal-vapor lasers, typical discharges with energy deposition of 3–4 kW/m in tubes  $\varnothing$  2–10 cm at gas pressure of 300–500 Torr are subjected to ionization-overheating instability. Therefore, they necessarily must contract, what is experimentally observed in pure inert gases, for example, at the stage of cell heating. However, in the presence of metal vapor, such discharges automatically decontract. We call this phenomenon, basic for metal-vapor lasers, the Petrash effect.<sup>2</sup> The effect of automatic decontraction is experimentally found to be rather stable at entering the vapor into the discharge, since it is observed at wide variations of gas-discharge conditions. Contraction of discharge in the mixture of inert gases and metal vapor was observed in only one case, which will be considered below.

2. The second problem – extending the list of active media – has not been adequately studied, and, in my opinion, can be formulated in the following way.

First, chemical elements having self-limited laser transitions, including those elements, which have laser transitions from resonance to metastable levels, can be divided into three groups: *A*, *B*, and *C* as follows. Two elements (Cu and Au), for which repetitively pulsed mode of lasing (in the mixture of their vapor with inert gases) has a long life (> 1000 h), form the group *A*. A number of elements, in whose vapor only short-term (1–10 h) pulsed lasing was observed by now, are included in the group *B*. Finally, the group *C* incorporates gases (Ar, Ne, Xe, N<sub>2</sub>, H<sub>2</sub>, etc.) and atoms of some elements, in which the pulsed lasing was not observed at all. Within the framework of this classification, the problem of extending the list of active media is connected with the search of physical and technical ways of transferring the elements from the groups *B* and *C* to the group *A*.

Second, for some metals from the group *B* it was unambiguously found that the service life of active elements operating in the pulsed mode is limited due to some unusual cause: covering of the inner surface of gas-discharge channels with the working metal.

Below it will be shown that a barely noticeable phenomenon – metallization of cell walls – is of great significance for understanding the processes proceeding in pulsed lasers, and it is closely connected with the problems of scaling and extending the list of active media.

## 2. Experimental observation of metallization

A typical experiment on observing the cell metallization in pulsed lasers was the following. A BeO-ceramic gas-discharge cell with the discharge channel 1 cm in diameter and 40 cm in length was filled with He. A weighted sample of metal Eu lied freely on the inner surface of the channel inside the cell. As the cell was switched on, the laser output power at the transition  $\lambda = 1.0019 \mu\text{m}$  in the Eu<sup>+</sup>

spectrum was, as a rule, 2–3 W at the repetition frequency of excitation pulses  $f = 10 \text{ kHz}$ . However, during 1–3 h of operation, the power decreased gradually down to  $\sim 0.1 \text{ W}$  and lower. As this took place, we visually observed through the cell wall that the discharge constricted.

An almost homogeneous metal layer was seen on the inner surface of the used gas-discharge ceramic channel and on polished sections of ceramic channel samples.

As a result, it was found that, to restore the efficiency of cells of pulsed lasers operating at elements of the group *B*, the cells should be dismantled and the inner metal layer should be removed by washing.

## 3. Causes and effects of metallization

According to the above-said, the vapor of Eu, as a typical element of the group *B*, forms a homogeneous metal layer on the inner surface of oxide-ceramic discharge channels. However, it is well-known that the vapor of Cu and Au deposit on the walls of gas-discharge ceramic channels in the form of drops. So, the following conclusions can be drawn:

1. Separation of elements into the groups *A* and *B* has a simple physical meaning: elements of the group *A* do not moisten BeO and Al<sub>2</sub>O<sub>3</sub> ceramic, and elements of the group *B* to a greater or lesser degree possess the property of moistening the material of gas-discharge channels. For some metals of the group *B*, for example, rare earth elements, the moistening of the channel walls is so efficient that liquid metals run down the close-grained ceramic as if it was a fuse. It should be noted that the inner metal layer on the walls of gas-discharge channels is formed at evaporation of both liquid- and solid-state metals (sublimation).

2. The list of the elements of the group *A* can be extended at the expense of the elements of the group *B* in the following ways:

- search for and working out of new materials for gas-discharge channels;
- design of cells with controlled vapor density;
- design of specialized profiles of walls in the channels made of traditional materials.

The causes of lasing contraction in the process of metallization of the walls of gas-discharge channels are most interesting from the practical point of view. Natural assumption that the contraction is caused by discharge shunting with metal films is not justified, because the integral conductivity of the discharge plasma in far afterglow is  $1\text{--}10 \Omega^{-1} \cdot \text{cm}^{-1}$ , whereas the calculated longitudinal integral (over the cross section) conductivity of a film is  $10^{-5} \Omega^{-1} \cdot \text{cm}^{-1}$ . Consequently, the cause of lasing breakdown at metallizing the cell walls should be sought for in violation of stability (contraction) of the discharge. In this connection, it is worth comparing differential, rather than integral, conductivity of the film and plasma. The differential conductivity of plasma is here the plasma conductivity

in a layer, whose cross section is equal to the cross section of the film. Within the framework of this concept, the differential conductivity of plasma is in any case lower than that of metal. It is obvious herefrom that the effect of the film on the discharge is connected with shunting only the outer plasma layer adjacent to the film, rather than the entire discharge volume. Coming back to the above-described experiments with the He-Eu laser, it should be concluded that the breakdown of gas-discharge channels is caused by violation of discharge stability (re-contraction) due to shunting the outer plasma layers of the pulsed discharge by a metal film.

As is well-known, stabilization of high-energy gas discharges requires preionization. The purpose of preionization is to generate a great number of free electrons providing the volume mechanism of ionization of the gas-discharge gap. One of the methods of the discharge preionization is to apply a short high-voltage pulse preceding the main excitation pulse to the discharge gap. It is also known that the efficiency of preionization increases significantly, if some easily ionized addition is added to the working mixture.

It is now commonly accepted that the preionization of the gas-discharge gap is not needed in pulsed discharges of atomic and molecular lasers, because the residual ionization of a gas is rather high. However, taking into account the above-said, there are three arguments in favor of automatic preionization in pulsed discharges. The automatic preionization is close, in its essence, to two-pulse preionization systems. Because the initial conductivity of the gas-discharge gap is due to the previous current pulse, the process mentioned above should be called the process of additional ionization.

The arguments in favor of additional ionization are the following. First, the residual ionization is radially inhomogeneous, and there must exist the process compensating for this heterogeneity. Second, automatic decontraction of the contracted pulsed discharge in pure inert gases is observed as some easily ionized addition (metal vapor) enters the plasma. As was noted above, this is a traditional method to intensify the preionization process. Third, the discharge stability is lost (repeated irreversible contraction) at metallization of the cell walls, what can be treated as a consequence of shunting the additional preionization and weakening its effect on the plasma.

The leading role of the outer plasma layers in stabilization of the pulsed discharge is indicative of the skin effect as a process of automatic additional ionization.

The role of the skin effect as a physical factor improving the radial homogeneity of the energy deposition into the gas, on the one hand, and breaking the synchronism of excitation of the active medium over the channel radius, on the other hand, was earlier discussed in Ref. 3. Figure 2 (Ref. 3) illustrates the influence of the skin effect on the radial distribution of the deposited power. The obtained curves are the results of computer simulation of the processes proceeding in the gas-discharge cells of large diameter.

The computer model<sup>3</sup> ignored the ionization-overheating instability of the discharge. Nevertheless, one can see from Fig. 2 that the homogeneity of the energy deposition into the gas improves with allowance made for the skin effect. This is a weighty argument in favor of the skin effect as one of the components of decontraction of pulsed discharges.

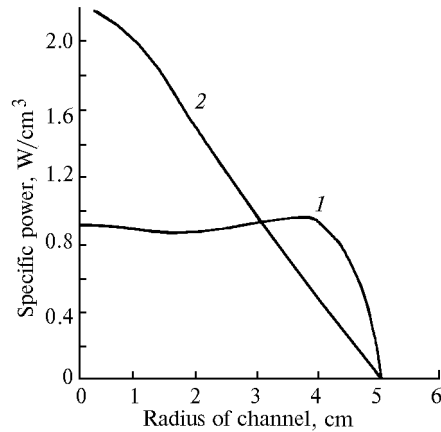


Fig. 2. Calculated distribution of the energy over the radius of the discharge channel 10 cm in diameter with (1) and without (2) allowance for the skin effect.<sup>3</sup>

Let us now estimate the characteristic parameters of the skin layer for the pre-pulse plasma of the pulsed discharge:

the depth of the skin layer  $h$ :

$$h = \sqrt{2\varepsilon_0 c^2 / (p\omega)}$$

the frequency of the electromagnetic wave  $\tau^{-1}$ :

$$\tau^{-1} = pm / (n_e e^2) \cong (\sigma V) N;$$

the time of skin layer penetration into the plasma to the distance  $R$ :

$$T = 0.15 R^2 / (\varepsilon_0 c^2);$$

the relative energy deposition to the skin layer:

$$W = \int_0^T U(t) I(t) dt / \int_0^\infty U(t) I(t) dt,$$

where  $\varepsilon_0$  is permittivity;  $\omega$  is the upper component of the frequency spectrum of the voltage pulse;  $c$  is the speed of light;  $n_e$  is the concentration of electrons;  $e$  and  $m$  are the electron charge and mass;  $p$  is the plasma conductivity at the end of the interval between pulses;  $U(t)$  is the voltage across the gas-discharge gap;  $I(t)$  is the discharge current;  $\sigma$  is the scattering transport cross section;  $V$  is the drift velocity of electrons;  $N$  is the density of scatterers (buffer gas);  $R$  is the channel radius.

For  $n_e = 10^{13} \text{ cm}^{-3}$ ,  $p = 1 \text{ } \Omega^{-1} \cdot \text{cm}^{-1}$ ,  $\omega = 10^9 \text{ Hz}$ ,  $R = 1 \text{ cm}$ ,  $\sigma = 10^{-16} \text{ cm}^2$ ,  $V = 10^5 \text{ cm/s}$ ,  $N = 10^{17} \text{ cm}^{-3}$  we have  $h \cong 0.4 \text{ cm}$ ,  $\tau \cong 10^{-6} \text{ s}$ ,  $T \cong 2 \cdot 10^{-9} \text{ s}$ ,  $W \cong 3\text{--}5\%$ .

Let the phase of discharge evolution corresponding to the time interval from the beginning of a pulse to the time  $T$  be called the skin discharge. In view of the dependence  $T \sim R^2$ , as the channel radius  $R$  increases, the time  $T$  grows fast, and, correspondingly, the energy deposition into the skin discharge increases. For example, for  $R = 5$  cm the duration of the skin discharge is  $T \cong 50$  ns, and the relative energy deposition into the skin discharge achieves  $W \cong 25\%$ .

Obviously, redistribution of the energy deposition between the main and skin discharges is one of the components of suppressing the ionization-overheating instability in the plasma of pulsed discharges in the large-radius channels. Taking into account the results of Ref. 2, the second component of suppressing the contraction is the minimum of density of the easily ionized addition (metal vapor) along the channel axis.

### Conclusions

1. The pulsed lasing of metal lasers is sensitive to metallization of inner surfaces of gas-discharge channels. For some chemical elements, the metallization of cells limits the service life of active elements to the level of 1–10 h.

2. The influence of the wall metallization in discharge channels on the efficiency of active elements is connected with the pulsed discharge contraction.

3. One of the mechanisms of decontraction of pulsed discharges at entrance of vapors (Petrash effect) is the skin effect, which improves the radial homogeneity of the energy deposition into the discharge.

### Conclusion

The results of this work and these of Refs. 2 and 4–6 allow us to generalize the observed regularities in the contraction and decontraction of pulsed discharges as follows: discharges are decontracted, if they take place in mixtures of inert gases and metal vapor in the presence of the skin effect.

### Acknowledgments

The author is thankful to M.V. Kanaeva and A.V. Klimkin for the help in preparation of materials to printing.

This work was partially supported by the Russian Foundation for Basic Research, Project No. 99–02–17016.

### References

1. N.Ya. Lyabin, A.D. Chursin, and M.S. Domanov, *Izv. Vyssh. Uchebn. Zaved., Ser. Fizika* **42**, No. 8, 68–75 (1999).
2. V.M. Klimkin, "Problem of instability of longitudinal repetitively pulsed discharges in metal-vapor lasers," Preprint No. 1, IAO SB RAS, Tomsk (1999), 24 pp.
3. M.J. Kushner and B.E. Warner, *J. Appl. Phys.* **54**(6), 2970–2982 (1983).
4. E.P. Velikhov, A.S. Kovalev, and A.G. Rakhimov, *Physical Phenomena in Gas-Discharge Plasma* (Nauka, Moscow, 1987), 160 pp.
5. L.M. Bukshun, E.L. Latush, and M.F. Sem, *Kvant. Elektron.* **15**, No. 9, 1762–1764 (1988).
6. P.A. Bokhan and D.E. Zakrevskii, *Zh. Teor. Fiz.* **67**, No. 4, 25–31 (1997).