

LASER APPLICATIONS AND OTHER TOPICS IN QUANTUM ELECTRONICS

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An ultraviolet barrier-discharge OH molecular lamp

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***Abstract*. The energy and spectral parameters of a barrier discharge in a mixture of argon with hydroxyl $^{\bullet}$OH are studied experimentally. A sealed lamp with the radiation intensity maximum at $\lambda = 309.2$ nm, an emitting surface area of ~ 700 cm^2, and a radiant excitance of 1.5 mW cm^{-2} has been fabricated. The radiant power of the lamp is 1.1 W.**

Keywords: *barrier discharge, OH molecules.*

1. Gas mixtures containing a buffer gas (Ar, Ne, Xe) and an easily ionisable admixture (Hg, Cd, Zn, Rb, Cu, Tl) are widely used in modern low- and medium-pressure optical radiation sources [1].

It has long been found that upon excitation of an inert gas containing impurity water, the $A^2\Sigma \rightarrow X^2\Pi$ emission band of $^{\bullet}$OH is observed at $\lambda \sim 309$ nm [2, 3]. It is known that the presence of water can quench lasing, for example, in a XeCl laser. For an incoherent radiation source, however, this adverse effect can be turned into an advantage and used for developing an optical radiation source in which water plays the role of an easily ionisable impurity. According to [3], upon excitation of inert gases by an electron beam, the 309-nm emission band was most intense in argon.

The authors of [4 – 7] studied a glow discharge and a low-pressure rf discharge (down to 30 Torr) in a mixture of hydroxyl $^{\bullet}$OH and buffer gases, and determined the conditions under which the radiation from the $^{\bullet}$OH molecule (the $A^2\Sigma \rightarrow X^2\Pi$ band) dominates in the discharge spectrum, while the emission from the inert gas in the interval 200 – 700 nm is almost completely suppressed. Such a situation is possible because the ionisation and excitation potentials of $^{\bullet}$OH radicals are much lower than these potentials for inert gas atoms.

However, several questions that are important for developing new types of lamps remain unanswered in the above publications. For example, the energy parameters of the $A^2\Sigma \rightarrow X^2\Pi$ band of the $^{\bullet}$OH radical, the emission efficiency and the service life of the lamp were not determined, and the structure of the hydroxyl emission band was not presented. The aim of our study is to fill these gaps and fabricate sealed lamps with high energy parameters.

Unlike [4 – 7], we used a barrier discharge (BD) for exciting a working mixture. In this discharge, the passage of current is confined to at least one dielectric layer and the characteristic sizes of the electrodes are substantially larger than the electrode gap. Such a discharge has a high parametric flexibility. By varying the extrinsic BD parameters such as voltage (from hundreds of volts to kilovolts), working mixture pressure p (from fractions of a torr to 10^3 Torr), and discharge gap d (in the range 1 – 10 mm), we can change the parameter E/n (from 1 to 500 Td), where E is the field strength and n is the gas concentration. In this case, the characteristic parameters of the BD plasma can vary over wide ranges (the mean electron temperature can vary from 1 to 10 eV, the degree of ionisation from 10^{-4} to ~ 1, and the electron concentration from 10^{11} to 10^{15} cm^{-3}). Depending on pressure, a BD is either a homogeneous discharge or randomly distributed microscopic discharges (filaments) in which the duration of current does not exceed a few tens of nanoseconds [8, 9]. In addition, the contact between the electrode and the working mixture is ruled out in lamps with two dielectric barriers, which ensures a long service life of the device [10].

For this reason, a BD is an interesting candidate for obtaining the efficient emission of $^{\bullet}$OH radicals.

2. Figure 1 shows the construction of the OH lamp. Bulb (*1*) is made of two 11-cm long coaxial KU-1 quartz tubes of diameters 2.2 and 4.6 cm. External electrode (*2*) has the

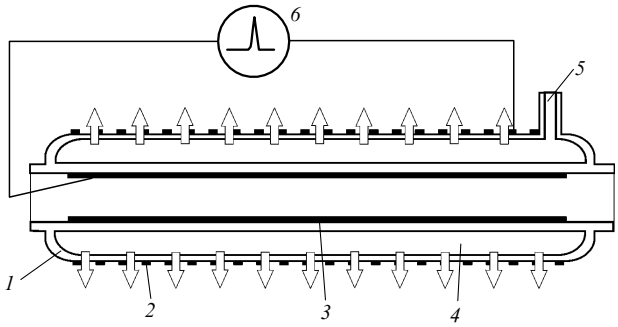

Figure 1. Construction of the OH lamp: (*1*) bulb; (*2*) perforated electrode; (*3*) solid electrode; (*4*) discharge gap; (*5*) branch pipe for replacing the working mixture; (*6*) generator. The arrows show the direction in which the radiation emerges from the lamp.

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form of a spiral and a transmittance above 95 %. Solid electrode (*3*) is made of aluminium – magnesium foil. The gas mixture in gap (*4*) was excited by a meander-shaped pulsed voltage applied to the electrodes with an amplitude up to 5 kV and a duration of 1.5 μs. The pulse repetition rate could be varied from 26 to 96 kHz. The power supplied to the plasma depended on the amplitude and repetition rate of voltage pulses and achieved 70 W at the maximum repetition rate. The discharge formed in gap (*4*) could be diffusive or filamentary in shape.

The radiation power of the lamp in absolute units was measured with a C8026 photodetector (Hamamatsu Photonics KK) with a H8025-222 head. The radiation spectrum of the discharge and its time dependence were recorded with a high-resolution HR4000 spectrometer (Ocean Optics B.V.). The input power was determined from the oscillograms of voltage and current pulses by the method developed in [11]. These oscillograms were recorded with a TDS 224 oscilloscope (Tektronics Inc.) with the help of a current shunt and a voltage divider. The water vapour pressure at input (*5*) was monitored with a HIH-4000 humidity sensor (Honeywell Int. Inc.).

3. The pressure of argon and water vapour was varied in the experiments, and the emission power, the power supplied into the discharge and the emission spectra were measured. The upper limit of the pressures employed was limited by the increase in the breakdown voltage across the discharge gap with increasing the pressure.

As in [5], the addition of hydroxyl to an inert gas suppresses the emission of the inert gas, and the A – X emission band of $^{\bullet}$OH dominates in the range 200 – 680 nm. According to [12], the spectrum may also exhibit the B – A transition at 269.6 nm; however, we did not observe this transition in our experiments.

Figure 2 shows the detailed spectrum of the A – X band of $^{\bullet}$OH, in which some high-intensity lines could be identified. The spectrum (the line maxima and intensities) is close to the spectrum of flames containing a hot gas, oxygen and hydrogen ($T = 3000$ K) [2], the Balmer lamp spectrum ($T \sim 1200$ K) with a small admixture of water in the bulb [13], and the numerically calculated spectrum of the $A^2\Sigma \rightarrow X^2\Pi$ band of $^{\bullet}$OH radical [13].

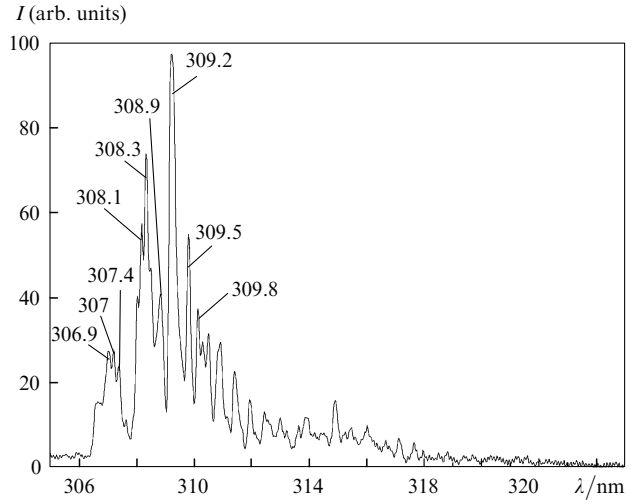

Figure 2. Emission spectrum of the A – X band of the $^{\bullet}$OH lamp (water vapour pressure ~ 1 Torr and argon pressure 300 Torr).

The integrated intensity of the A – X band depended on the argon pressure (Fig. 3) and the water concentration in the working mixture. The concentration of water at low pressures ($p < 0.2$ Torr) could not be accurately monitored due to the limitations of our humidity detector. We could not also detect any part of the water forming a water-hydroxyl complex [$(H_2O)_m$, $(OH)_n$, or $(H_2O)_m(OH)_n$] at the walls and in the bulb. However, the following remarks can be made after taking into account this limitation of the effect of water concentration on the intensity of the A – X band. The emission power is low for a vapour pressure lower than a fraction of a torr, and its value increases sharply with increasing the pressure to a few tenths of a torr. A further increase in the vapour pressure to several torr reduces the emission power by 1 – 1.5 orders of magnitude. This can be attributed to an increase in the self-absorption of radiation from the $^{\bullet}$OH radical, which is manifested in a relative flattening of the A – X band profile at low hydroxyl concentrations.

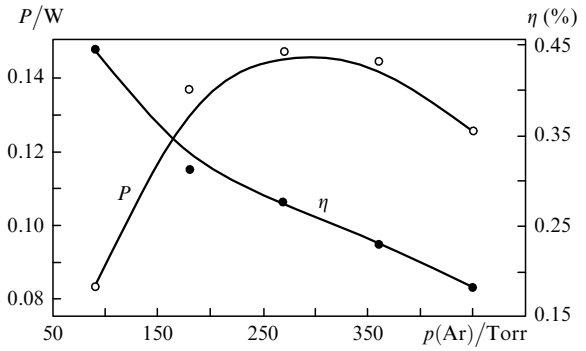

Figure 3. Average power and emission efficiency of the A – X band of $^{\bullet}$OH under various values of the argon pressure and an H_2O pressure of about 1.3 Torr.

At low pressures of water vapour (lower than 0.5 Torr) and moderate argon pressures (lower than 180 Torr), the discharge is diffusive and its plasma parameters are close to those of a glow discharge and the parameters obtained in [5]. As the pressure of water vapour and/or argon is increased, the discharge is completely or partially transformed into a filamentary type.

Thus, by using the parametric flexibility of a barrier discharge, we determined the effect of elevated pressures on the spectral and ultimate energy parameters of radiation. A distinguishing feature of a barrier discharge is also the ease with which the bulbs can be scaled. This fact and the data obtained enabled us to develop a sealed lamp with a working zone length of 55.5 cm (Fig 4).

The emitting surface area and the radiant excitance of this lamp are ~ 700 cm^2 and 1.5 mW cm^{-2} respectively,

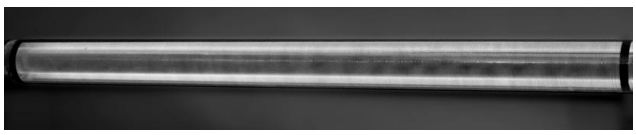

Figure 4. Photograph of the emitter of a sealed barrier-discharge OH lamp.

while its radiant power is 1.1 W. The efficiency of the lamp is 0.4 %.

Figure 5 shows the results of the first tests on the service life of the OH lamp. One can see that the emission power of the lamp slightly decreases during the first 10 hours of its operation (region I in the figure), after which it is stabilised (region II). According to [14], the initial decrease in the power of gas-discharge devices is due to absorption (mainly due to absorption of the working gas or, in our case, water) by the inner shell of the quartz lamp. As the water and argon molecules are accumulated, the internal pressure in the micropore channels of the quartz shell increases considerably. The absorption is also accompanied by desorption as a part of the gas is retrieved from the micropores of the quartz shell to the volume of the bulb. When these processes are balanced, the loss of water molecules is minimised and the emission power of the lamp is stabilised.

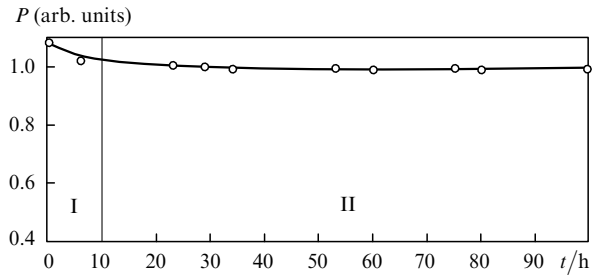

Figure 5. Service life of a barrier-discharge OH lamp ($Ar : H_2O = 270 : \sim 0.2$ Torr).

It can be expected that an increase in the size of the OH lamps will result in an increase in the duration of segment I, and the service life of the emitter in segment II will be not worse than that of excilamps filled with inert gas halides [9, 10].

Other mechanisms of water loss are also indicated in the literature, e.g., (i) proton and electron exchange with the detachment of an oxygen atom from the quartz surface; (ii) reaction involving the formation of $Si(OH)_4$ [15]; and (iii) formation of inert gas–water clusters, e.g., $Ar_n(H_2O)_m$ [16]. However, the formation of $Ar_n(H_2O)_m$ requires an average electron energy of 15–70 eV which is hard to realise in a barrier discharge, while the accumulation of $Si(OH)_4$ requires a much longer time than the duration of our experiments. The role of the former mechanism can be judged from the emission spectrum, but we did not observe the oxygen line in the spectra recorded in our experiments.

4. Thus, our investigations led to the development of the first barrier-discharge lamp operating on the $A^2\Sigma \rightarrow X^2\Pi$ band of the hydroxyl $^{\bullet}OH$ with an area of ~ 700 cm^2 and a radiant power of 1.1 W. The lamp has a long service life owing to the use of electrode-free discharge and is an ecologically safe source of optical radiation since it does not contain cadmium or mercury vapour.

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