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# Increase in the power of lasing on atomic and ion transitions in chemical elements

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***Abstract*. A method for increasing the power of pulsed lasing on atomic and ion transitions in chemical elements obtained by the conversion of the UV radiation of excimer lasers in cells with metal vapours is studied. A part of UV radiation transmitted through a cell with metal vapour is used for pumping a dye solution in such a way that the cell converter with metal vapour represents a master oscillator, while the dye cell represents an amplifier. The study is performed by the example of amplification of weak spectral components of radiation from a $XeCl^*$ laser converted in mercury and barium vapours. In the amplifying stage the longitudinal pumping of the dye is used and a scheme for suppressing self-excitation is employed. It is found by selecting dyes that the alcohol solution of uranin is nearly optimal for amplification of the 546.1-nm laser line of mercury, while the best results in amplification of the 533-nm and 648.2-nm laser lines of barium were obtained by using alcohol solutions of rhodamine 6G and oxazine 17, respectively. The power of the 546.1-nm mercury line was increased by an order of magnitude, while the power of the 533-nm and 648.2-nm lines of barium – almost by a factor of twenty-five.**

***Keywords***: *metal vapour lasers, dye lasers, mercury and barium vapour lasers, UV laser radiation conversion.*

## 1. Introduction

Sources of laser radiation resonant to atomic spectral transitions can be used for studying the properties of excited states of atomic particles such as radiative lifetimes, quenching constants, photoionisation cross sections and for investigation of energy transfer in atomic – molecular reactions, etc.

Such sources can be based, in particular, on dye lasers. However, tunable dye lasers that can be used in such investigations are quite complicated, multistage opto-mechanical devices containing additional intracavity elements for the laser line narrowing and stabilisation, the background radiation suppression, etc.

Lasing observed in vapours of chemical elements upon the propagation of a UV laser pulse in them [1 – 6] opens up new possibilities for solving the above-mentioned problems.

In this paper, we studied the possibility of increasing the pulsed laser power at the atomic 546.1-nm line of mercury and the 533-nm and 648.6-nm lines of barium, which are generated upon the propagation of UV radiation in vapours of these elements. These laser lines lie in the visible spectral region and are of interest as objects for studying the possibility of increasing their power in amplifiers based on organic dyes.

## 2. Conditions for the observation of lasing in vapours of chemical elements and laser radiation properties

The propagation of quasi-resonance and, in the general case, high-power nonresonance laser radiation in vapours of chemical elements is accompanied by interesting physical effects of conversion of laser radiation to different spectral regions. In particular, the propagation of laser radiation through a column of chemical elements is often accompanied by stimulated electron Raman scattering (SERS) [7]. In the presence of quasi-resonances, SERS becomes a highly efficient process. Upon propagation of short-wavelength radiation, for example, pulsed UV radiation of excimer lasers in a column of metal vapours, the spectrum of converted radiation contains, except high-power stimulated Raman scattering components, many weak spectral components caused by stimulated and, possibly, superradiance Dicke [8, 9] transitions between the inverted states of atomic particles [1 – 6]. Studies of the temporal characteristics of converted radiation showed that radiation pulses converted by atoms of ions were observed, as a rule, on the leading edge of the UV pump pulse.

In the presence of a quasi-resonance in the spectrum of an atom, the propagation of UV laser radiation through an extended vapour column results in the production of an extended medium containing the first ions. Analysis of the emission spectra of vapours shows that the ionisation of atoms is accompanied by the excitation of ions and proceeds via the shifted states of atoms [5, 6]. In this case, the ionisation of atoms and excitation of ions are so efficient and selective that superluminescence of ions appears at the resonance lines and transitions from the resonance to metastable states (see, for example, [5, 6]).

Figure 1 shows laser transitions observed during the propagation of radiation from a $XeCl^*$ laser through a cell filled with barium vapour [2, 3, 5]. In this case, many single-

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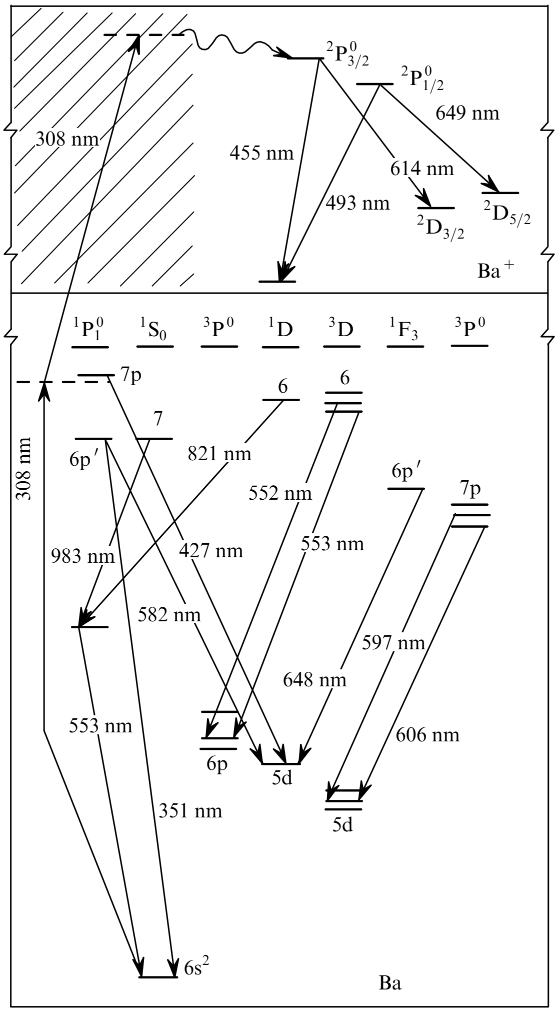

**Figure 1.** Stimulated transitions observed in the barium atom and ion upon pumping of barium vapour by a XeCl$^*$ laser at 308 nm.

pass lasing lines of the barium atom and ion are observed. In particular, lasing occurs at the 553-nm and 351-nm lines of the barium atom and at the 455-nm line of the barium ion. Note that for most of the transitions observed in vapours of barium and other chemical elements, no other pumping methods, except optical pumping discussed here, have been found so far. However, as mentioned above, the conversion efficiency and energy of the spectral components discussed above are small compared to those of the SERS components. At least, to use these components in practice, it is necessary to find the method for increasing their power and energy.

## 3. Selection of the method for increasing the power and energy of converted radiation

There exist several methods for solving this problem. For example, a multipass vapour cell can be used. In this case, the cell should have a large aperture and, hence, the vapour flow rate should be high. In this paper, we study the possibility of increasing the pulsed power of spectral components of converted radiation in the visible region by amplifying them in a dye amplifier.

The principle of the method is as follows. Lasing at atomic and ion transitions in vapours of most metals is not accompanied by a complete absorption of pump radiation. Therefore, UV radiation transmitted through a vapour converter can be further used to pump other active media, in particular, organic dye solutions. The dye solution can be used in turn to amplify weak converted radiation. Of course, the pumping of the dye and injection of radiation into it should be synchronised. In this case, the cell converter with metal vapour will be a master oscillator, while the dye cell will be an amplifier. This scheme successfully combines the high monochromaticity and automatic frequency-locking of the converted component generated in metal vapours to the frequency spectrum of atoms and ions with the high conversion efficiency and broad amplification band of dye amplifiers.

To implement this scheme in practice, a number of conditions should be fulfilled. First, it is necessary to select a dye having the amplification band in the required spectral range. Second, the pump pulse of the dye and the injection of the amplified spectral component to it should be synchronised. The first condition is simply fulfilled because there are many dyes that can amplify visible radiation upon UV pumping. The second condition can be provided in the general case by using optical delay lines either in the pump channel of the dye or in the radiation injection channel. It is obvious that optical delay lines complicate the pump system and introduce undesirable energy losses. However, as mentioned above, lasing at atomic and ion transitions is observed on the leading edge of the pump pulse, so that it can be expected that the synchronisation condition will be automatically fulfilled in this case.

## 4. Experimental

Figure 2 shows the scheme of the experimental setup assembled to verify the method described above. The 308-nm radiation of pump excimer laser (*1*) propagates through a spatial selection and formation system and cell converter (*2*) with metal vapour. A part of radiation generated in the metal vapour cell and directed forward was injected into dye cell (*3*). A part of UV radiation of the

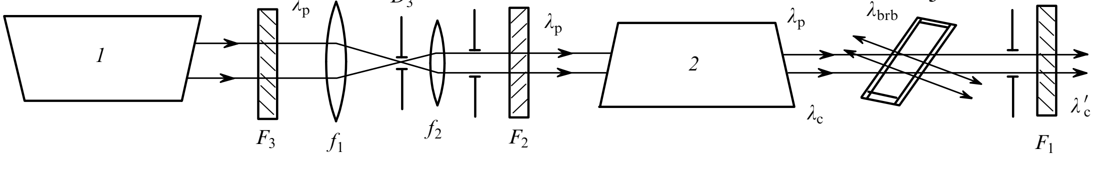

**Figure 2.** Optical scheme of the experimental setup: (*1*) pump laser; (*2*) metal vapour cell (converter); (*3*) dye cell; $F_1 - F_3$ – optical filters; $D_1 - D_3$ – apertures; $f_1, f_2$ – lenses; $\lambda_p = 308$ nm – pump radiation; $\lambda_c$ – converted radiation; $\lambda_{brb}$ – broadband dye emission; $\lambda'_c$ – radiation amplified in the dye.

excimer laser that was not used in the metal vapour was directed into the dye cell coaxially with injected radiation.

Dye solutions were studied in a 1-cm-thick quartz cell. The cell windows were tilted with respect to the optical axis of the system to avoid self-excitation of the amplifier and for the spatial separation of amplified radiation and dye emission in the case of self-excitation. Because the cell had no circulation system, the pulse repetition rate did not exceed 2 Hz and the pump pulse energy was lower than 10 mJ. The dye concentration was selected experimentally within limits typical of amplifying stages in dye lasers.

The incident radiation and radiation transmitted through the dye were analysed in intensity, temporal dependence, and spectrum.

In particular, to determine the influence of the amplification process on the spectrum of the injected component, we measured the spectral widths of injected and amplified radiation by using an IT-51-30 Fabry–Perot interferometer with the 8-mm base crossed with an MDR-3 monochromator. Radiation transmitted through the interferometer was projected with an MTO-1000 objective on the slit of the monochromator. Spectral scan was performed by using an additional narrow aperture mounted across the entrance slit of the monochromator and displaced along the slit. Radiation transmitted through the interferometer, aperture, and monochromator was detected with a photomultiplier. The output pulses of the photomultiplier were integrated with a Brookdeal stroboscopic detection system and recorded on a chart recorder. The spectral resolution of the detection system was controlled by recording the emission line of a single-frequency He–Ne laser.

The emission spectra of vapours in a broad wavelength range were recorded by using an MDR-3 monochromator and a Brookdeal detection system. The temporal parameters of emission were measured with a coaxial photocell or an LFD-2 photodiode and were recorded with a fast oscilloscope.

## 5. Experimental results and discussion

We studied the properties of our setup by amplifying the spectral components of radiation of a XeCl$^*$ laser converted in mercury and barium vapours. The amplification of the long-wavelength 546.1-nm component of the mercury triplet was studied in most detail. This component is convenient because it can be efficiently amplified in a number of organic dyes. In addition, the design of mercury vapour cells is simpler than that of cells for vapours of refractory metals.

We studied the amplification of the 546.1-nm emission line of mercury in the alcohol solution of uranin, which was most convenient for this purpose among the solutions available. Figures 3a, b show the absorption and luminescence bands of uranin and the $^3S_1-^3P_2$ line of mercury being amplified. Figure 3b also shows the pump line and components of the mercury triplet. One can see from Fig. 3 that the long-wavelength 546.1-nm ($^3S_1-^3P_2$) component of the mercury triplet lies within the amplification band of uranin, whereas the short-wavelength 435.8-nm ($^3S_1-^3P_1$) and 404.7-nm ($^3S_1-^3P_0$) components lie within the absorption band of uranin. Therefore, only the long-wavelength component can be amplified. The short-wavelength components are absorbed in the uranin solution. The alcohol solution of uranin serves as a rejecting filter for the blue and violet emission lines of mercury, which are injected to the dye cell together with the green line.

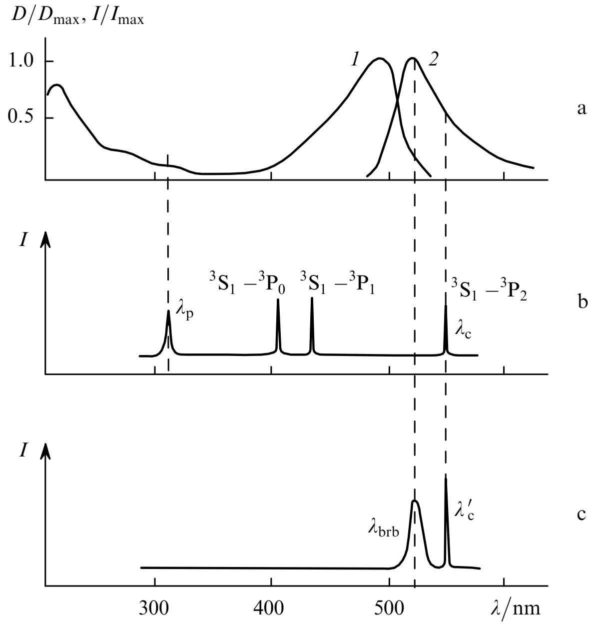

**Figure 3.** Normalised absorption $D(\lambda)$ (*1*) and luminescence $I(\lambda)$ (*2*) spectra of uranin (a), the spectrum of radiation injected into the dye (b), and the spectrum of radiation amplified in the dye (c); $D(\lambda)$ – optical density.

The emission spectrum at the output of the dye cell contains, along with the amplified 546.1-nm line, the broadband laser emission at 530 nm despite the precautions taken to suppress it (Fig. 3c). We found that, when the dye cell was rotated around the axis perpendicular to the pump beam, the broadband generation was always observed in the direction of the normal to the cell windows (see Fig. 2), while the propagation direction of the amplified component preserved, experiencing only parallel displacements from the pump beam. This circumstance allows us to separate spatially these components and to study their spectral and temporal properties.

Figure 4 shows the oscillograms of pump pulses, the injected component of radiation converted in the mercury vapour, and the radiation pulse amplified in the dye cell. These data correspond to the pump energy $E_p = 4.3$ mJ and the injected radiation energy $E_c = 3$ μJ. A comparison of the amplitudes of injected and amplified pulses shows that, despite self-excitation of the amplifier, the intensity of injected radiation is increased by an order of magnitude. We also found the two specific features of amplification: the broadband lasing pulse of the dye was shorter than the amplified pulse and the broadband radiation was delayed by a few nanoseconds with respect to the amplified radiation component. It seems that for this reason the broadband lasing does not compete considerably with the amplification process. However, such a competition can take place, for example, in the case of shorter pump pulses. In this case, it is necessary to suppress broadband lasing more efficiently.

It is known that the general property of the radiative decay of strongly inverted transitions (with the total gain per passage exceeding 20) [10] is the presence of several superradiance components located within the spectral interval determined by the type of broadening of the transition under study. The intensities of these components change

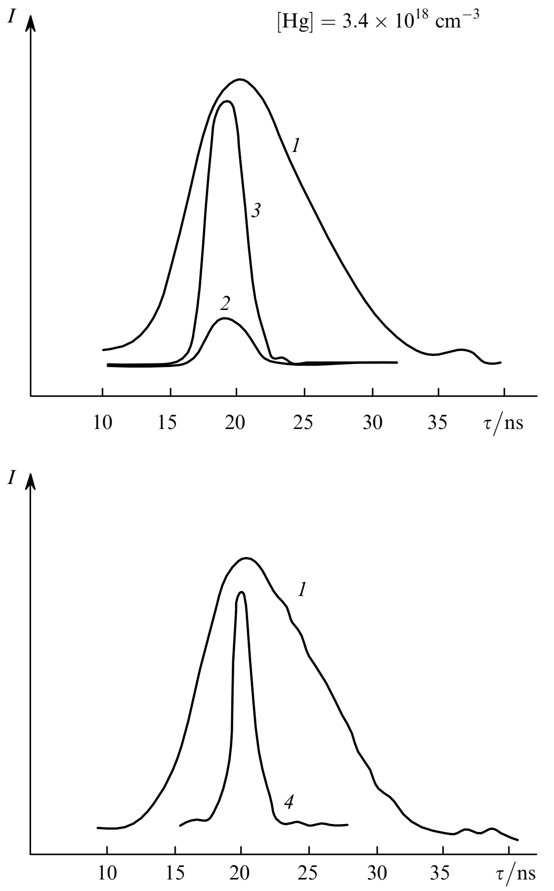

**Figure 4.** Oscillograms of the pump pulse (*1*), injected radiation at 546.2 nm (*2*), radiation at 546.1 nm amplified in the dye cell (*3*), and broadband dye emission at 530 nm (*4*).

chaotically from pulse to pulse. Figure 5 shows the interferograms of the pulse injected to the dye and the amplified pulse of the long-wavelength 546.1-nm component of the mercury triplet, which demonstrate the difference in the intensities of the fine-structure components of injected and amplified radiations. These differences are observed because our system for recording interferograms cannot control simultaneously the spectra of each of injected and amplified pulse. Therefore, interferograms presented in Fig. 5 do not illustrate the real intensity ratio of the components because they were recorded at different instants and demonstrate only that additional amplification in the dye occurs at least within the entire spectral contour of injected radiation.

Similar studied were performed with two components of the XeCl* laser radiation converted in barium vapour (at different pressures of the buffer helium gas). Stimulated radiation at the 553-nm resonance transition of barium (Fig. 6) and the 648.2-nm atomic transition was amplified in solutions of rhodamine 6G and oxazine 17, respectively. In the first case, the amplification of the injected signal by an order of magnitude was achieved, while in the second case the gain was almost 25. The latter result demonstrates that a large gain is achieved when the injected radiation line is located near the maximum of the amplification band of the dye. Therefore, the choice of rhodamine 6G for amplification of the resonance line of barium is not fortunate because the injected component falls on the short-wavelength wing of the amplification band of rhodamine 6G. (The better amplification of the 533-nm line of barium can be expected

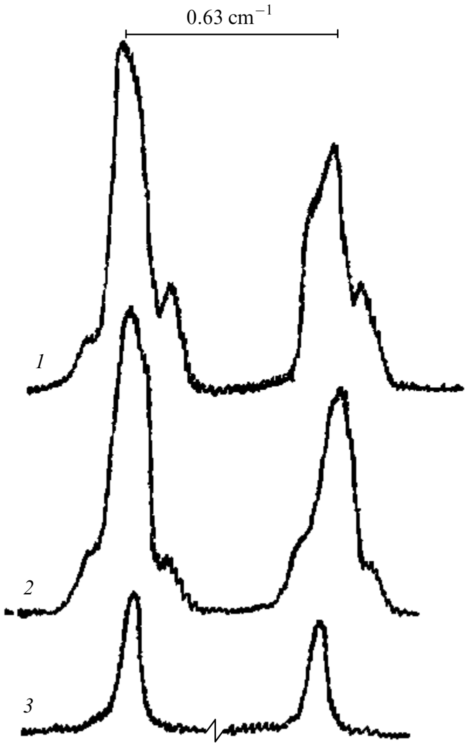

**Figure 5.** Interferograms of the 546.1-nm laser line of mercury amplified in the dye (*1*) and injected into the dye (*2*), and of the He–Ne laser line (*3*). The line intensities are presented in arbitrary units.

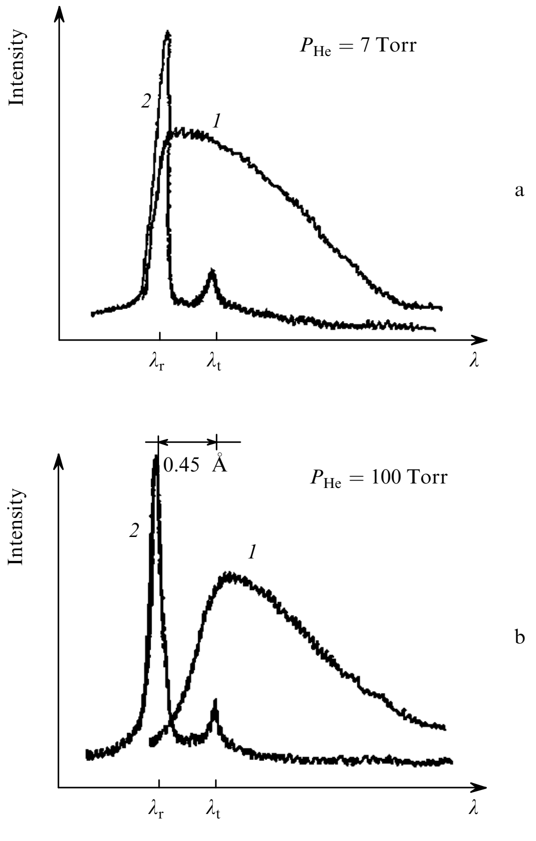

**Figure 6.** Laser emission spectra at the resonance 533-nm transition of barium (*1*) at two helium pressures and the resonance line of barium ($\lambda_{\mathrm{r}}$) observed in a hollow cathode lamp (*2*); $\lambda_{\mathrm{t}}$ is the spectral line of the triplet transition in the barium atom.

in the solution of the fluorescein 27 dye.) The amplification of radiation of atomic barium was also accompanied by broadband lasing observed in spectral regions close to the maxima of amplifications bands of dyes used in experiments, namely, at 570 nm for rhodamine 6G and at 645 nm for oxazine 17.

## 6. Conclusions

We have shown by the example of amplification of weak laser lines of mercury and barium in solutions of organic dyes that the attachment to an excimer laser consisting of a metal vapour cell and a dye solution cell can be used to obtain high-intensity stimulated radiation at many atomic and ion transitions. The radiation power at these transitions can be further increased by properly modifying amplification schemes. In particular, a change from longitudinal pumping to the transverse pumping of several amplifying dye cells may be quite successful. Such an approach permits the use of ready amplifying stages of standard dye lasers pumped by excimer lasers in which a dye master oscillator with a spectral selection system is replaced by a cell filled with the required metal isotope vapour employed as a single-frequency maser oscillator.

In conclusion, we would like to emphasise once more the simplicity of our method for obtaining laser radiation related to the atomic and ion lines of chemical elements. This method does not require the use of intracavity high-resolution dispersion elements and frequency-control and stabilisation systems. For example, of special interest for a number of practical problems is the production of frequency-stable lasing at the resonance lines of atoms and ions of some metals. This problem can be solved based on the results of this paper by using superradiance at the resonance lines of metal atoms observed upon the photodissociation of vapours of metal halides by UV laser radiation [11].

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