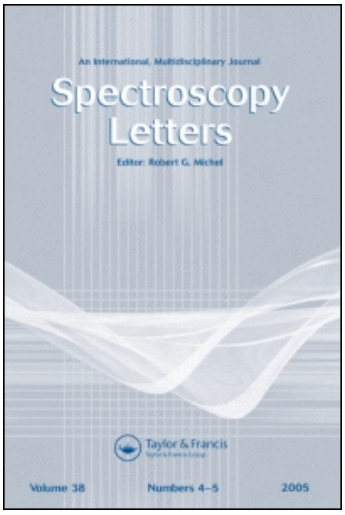

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EXCIMER MEDIA GAIN

G. Gerasimov[a]
[a] S.I. Vavilov State Optical Institute, St. Petersburg, Russia

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EXCIMER MEDIA GAIN

G. Gerasimov

S.I.Vavilov State Optical Institute, 199034,
St. Petersburg, Russia

ABSTRACT

The experimental and theoretical investigation of the hydrogen gas discharge plasma illuminated by an external narrow-spectrum source shows that the gain coefficient of the hydrogen plasma can be increased if external source spectrum overlaps with the hydrogen continuum. This effect can be used in the laser process of excimer molecules.

Key Words: Ultraviolet; Hydrogen; Gas discharge; Gain; Stimulated emission.

INTRODUCTION

Laser action of ultraviolet (UV) and vacuum ultraviolet (VUV) radiation is presently one of the major objectives in quantum electronics. The low temperature plasma of hydrogen with spectral transition on the repulsive states has been discussed as the first candidate for short-wavelength laser action [1]. Similar spectral transitions in argon, krypton and xenon were used at first for VUV laser action [2–7]. According to reference [3], the gain coefficient κ (for example, of Xe_2) for an amplified small signal is:

$$\kappa = \sigma[R_2^*] \quad \sigma = \lambda^2 A/8\pi\Delta\nu \tag{1}$$

here σ is the photodissociation cross section of a stimulated transition on the low repulsive term, λ is the transition wavelength, $[R_2^*]$ is the concentration of excited molecules, $\Delta\nu$ is the spectral transition width, which was estimated in [3] as the energy difference between two projections of the vibration turn point on the low repulsive level. The xenon spectrum width quantitative estimate [3] yields 10^{15} s^{-1}. This means that the concentration of excited molecules should be not less than 10^{15} cm^{-3} for a gain coefficient $> 10^{-3}$ cm^{-1} (this value is necessary for laser implementation in practice). It is an extremely difficult problem to obtain the excited molecules concentration of 10^{15} cm^{-3}. According to references [3,8], this is an essential reason why the continuous wave (cw) VUV laser action by the inert gas excimers is basically impossible, and pumping parameters should be very high in the case of a pulsed laser action.

Spontaneous Emission

We will revise the molecular spontaneous emission on the repulsive level by the quantum mechanical before explaining stimulated emission. The simulation of dimer spontaneous emission continua was described in many papers [9–11]. According to (11), the theoretically predicted continua involves the equation

$$I_\upsilon(\lambda) = 1/\lambda^6 \left\{ \int \Psi_\upsilon(R)\mu(R)\phi_\varepsilon(R)\,\mathrm{d}R \right\}^2 \tag{2}$$

and summing I_υ over discrete vibrational levels υ of the electronic state Ψ, taking into account the population of the vibration levels. Equation (2) determines, to within a constant, the wavelength dependence of the energy density of the radiation from excimer molecules. Here λ is the wavelength of the transition $\upsilon \to \varepsilon$, $\Psi_\upsilon(R)$ is the vibration wave function of the upper bound electronic state Ψ, $\phi_\varepsilon(R)$ is the vibration wave function of the repulsive electronic state ϕ, and $\mu(R)$ is the dipole moment of the electronic transition $\Psi \to \phi$. Omitting details of the calculation of the vibration wave functions by Eq. (2), we note that, to obtain the spontaneous emission probability at frequency ν_I, one should calculate the integral (2). We keep in mind that $\Psi_\upsilon(R)$ is the vibration wave function of upper bound electronic state Ψ, and $\phi_\varepsilon(R)$ is the vibration wave function of the repulsive electronic state ϕ at internuclear distance R, corresponding to transition frequency ν_I of the emission continuum. It is possible to obtain the probability distribution of spontaneous emission by calculation of the integral (2) for each frequency ν_I of the emission continuum. Thus, the full emission spectrum of a transition between the bound and repulsive levels appears as a broad continuum, even allowing for the population of only one vibration level. This continuum arises from combining many equivalent "elementary" overlapping profiles. Each profile width depends, according to the Heisenberg uncertainty principle, on the lifetime of the upper levels which have

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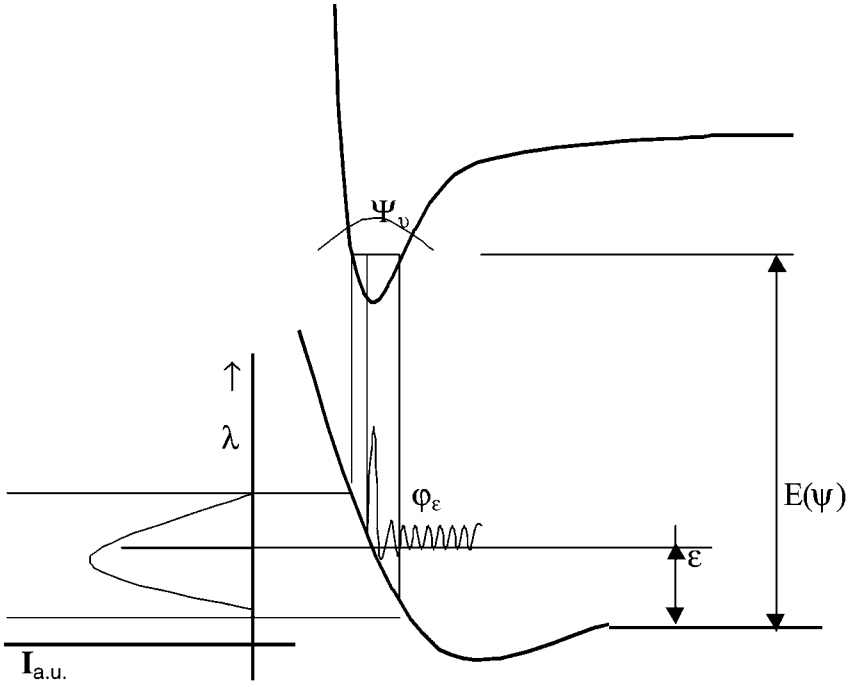

Figure 1. Diagram of transition in excimers.

been repeatedly measured experimentally. According to a simple estimate, the ratio of the experimental continuum width to the Heisenberg width can differ by 5–6 orders of magnitude. The full VUV spectrum width depends on the repulsive ground state slope and as the elementary profile width on the transition probability. Figure 1 shows a diagram of the full spectrum arising from a transition from vibration level $\upsilon = 0$ of the excited upper state on the repulsive ground state. This spectrum width is several nm for inert gases and for hydrogen about ten nm. The spontaneous emission spectrum is built up into a continuum from a set of the unlimited overlapped elementary profiles being randomly stimulated.

Stimulated Emission

Consider the case when an excimer media is illuminated by an external light source with spectral density ρ_1 and width $\Delta\nu_1$. Let us assume that ρ_0 and $\Delta\nu_0$ are the spectral density and the width of the excimer media spectrum, respectively. We bear in mind that Eq. (1) for the gain coefficient was obtained for a small

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signal passing through an inverted media. Suppose that spectral density ρ_1 of the external light source is much higher than ρ_0. Let us again suppose that width $\Delta\nu_1$ is limited by the elementary profile width. It should be noted that the notion of small signal is quite different in atomic and molecular media if a media is illuminated by a source with a narrow-spectrum of width $\Delta\nu_1$. Actually, if one supposes that spectrum density ρ_1 is the small signal for the width $\Delta\nu_{at}$ in atomic media, the same signal will be also small for width $\Delta\nu_{mol}$ in molecular media. The ratio of the atomic spectral density to the small signal density will be much larger than the same ratio for the molecular media. The difference between two ratios can be roughly estimated as $\Delta\nu_{mol}/\Delta\nu_{at}$ and can be as high as 10^6. The spectral probability distribution of stimulated emission will have a maximum at frequency ν_1 of the external light source. It can be readily demonstrated that the number of molecules which take part in the stimulated emission at frequency ν_1 can be estimated as $[R_2^*]n/(n+k-1)$, where n is the ratio of the spectral density of the external source to the spectral density of excimer media, k is the ratio the molecular width to the width of the external source spectrum. According to the Franck-Condon principle, the ratio of the stimulated emission probability at the frequency ν_1 and that near to ν_1 will equal n. Equation (1) for gain κ_1 within frequency band $\Delta\nu_1$ will read:

$$\kappa_1 = \{\lambda^2 A/8\pi\Delta\nu_1\}[R_2^*]n/(n+k-1) \tag{3}$$

Equation (3) can be transformed after multiplying by $\Delta\nu$ both the numerator and the denominator.

$$\kappa_1 = \kappa nk/(n+k-1) \tag{4}$$

According to Eq. (4) the excimer gain can be increased by a factor of $nk/(n+k-1)$ upon illuminating of the excimer media. The value of $nk/(n+k-1)$ can reach the several order of magnitudes even in the case of a incoherent source. Actually, if the spontaneous emission probability of excimers and atoms in the external source are approximately equal, the ratio of the spectral densities of the molecular and atomic sources will differ as the widths of excimer and atomic spectra. For example, a simple estimate shows that the ratio of the width of the hydrogen molecular continuum to that of the mercury resonant line is 10^4–10^6.

EXPERIMENT

The discharge molecular hydrogen plasma was used as the amplified media in our experiments. The mercury resonant line ($\lambda = 253.7$ nm) and laser radiation ($\lambda = 244$ nm and $\lambda = 628$ nm) were used as the external sources. The molecular hydrogen continuum ($^3\Sigma_g^+ - {}^3\Sigma_u^+$) (see Fig. 2) lies from 160 nm to 500 nm with the intensity maximum at 240–260 nm [14]. The experiments were repeated in five series of the independent studies (with different experimental setup). The essential result will be presented further.

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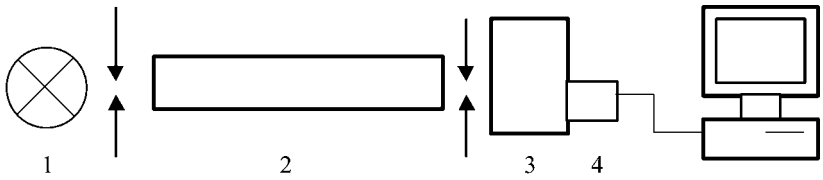

Figure 2. The main setup of the experiment: 1 – light source, 2 – discharge tube, 3 – Jarrel Ash monochromator, 4 – CCD detector.

Figure 2 shows a typical experimental setup. External source 1 was illuminated by capillary discharge tube 2, the light was recorded with detector 4 at the exit slit of spectrometer 3, and the result was processed with computer. The 30 cm long discharge tube was filled with hydrogen flowing at a pressure of 0.5 torr. The discharge current was varied from 10 to 600 mA. A Jarrel-Ash spectrometer with a CCD detector was used in the experiments.

In each experiment, successively we recorded the spectra of the external source with the pumped out and filled discharge tube, with and without the discharge. The capillary discharge spectrum was also recorded without the external source. The intensity of the external source increased when the capillary discharge was switched on and when the mercury lamp ($\lambda = 253.7$ nm), and the laser light ($\lambda = 244$ nm) were used as the external sources.

Figure 3 demonstrate typical result of the experiments. The intensity difference of the external source with and without the hydrogen discharge is about 50%. In the best experiments the difference could reach 70% [13]. The result was

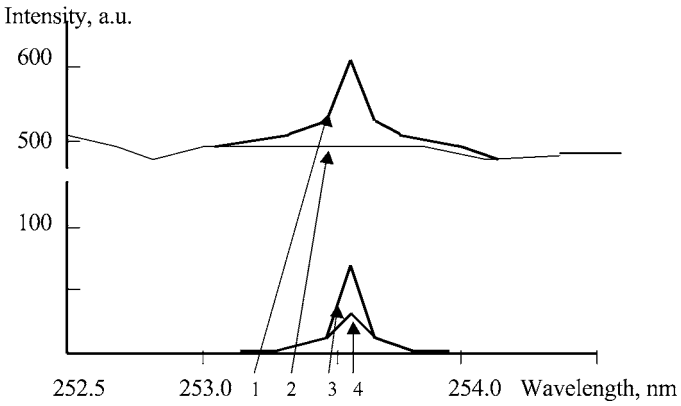

Figure 3. Amplification of 244 nm – laser light passing through an H_2 – discharge: 1 – H_2 continuum and laser, 2 – H_2 continuum, 3 – laser line, 4 – gain of intensity if the laser light passes through the discharge.

completely negative under any experimental conditions if the helium-neon laser ($\lambda = 628$ nm) was used as the external source.

The estimate of the gain from this experiment is 0.015 cm^{-1}. The theoretical estimate from Equation (1) yields a value by 3–5 order of magnitude smaller than the experimental result.

CONCLUSION

Thus, the experiments with hydrogen plasma illuminated by an external narrow spectrum source shows that the excimer media gain coefficient can be increased if the external source spectrum overlaps the excimer continuum. According to our study, several new excimer media can now be used for laser action, provided that a narrow-band of external sources are employed.

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