

ACTIVE MEDIA

https://doi.org/10.1070/QEL16418

Measurement of the gain of an optically pumped Xe_2Cl active medium

S.B. Mamaev, L.D. Mikheev, E.V. Polyakov, A.A. Rogashevskii, A.P. Shirokikh, V.I. Yalovoi

Abstract. **We have observed for the first time the gain in a Cl_2–Xe gas mixture optically excited by radiation of a XeCl laser (308 nm), which lead to the photoassociation Xe + Cl + $h\nu$ → XeCl(B,C), accompanied by the formation of a Xe_2Cl^* active medium in the recombination of XeCl(B,C) with Xe. The small-signal gain at a wavelength of 532 nm in a mixture of Cl_2 (1 Torr) with Xe (2 atm) is measured to be $(1.1 \pm 0.3)\times10^{-3}$ cm^{-1} in an active volume measuring 0.5×1×80 cm for an excitation energy of 170 mJ in a pulse having a duration of 60 ns. Being recalculated to the maximum of the gain band of Xe_2Cl located at a wavelength of 504 nm, the gain amounted to $(1.3 \pm 0.3)\times10^{-3}$ cm^{-1}. The prospects of further development of the present method for exciting the Xe_2Cl^* active medium are discussed.**

Keywords: *photoassociation, laser active media, petawatt facilities, visible spectral range.*

1. Introduction

Broadband active media of photochemical lasers, e.g., XeF(C-A), Kr_2F, Xe_2Cl, are of great interest for amplifying femtosecond pulses in hybrid laser facilities [1–4]. The Xe_2Cl active medium is particularly attractive for amplifying such pulses to high peak powers due to the broad (100 nm) gain band in the spectral region near 500 nm and the long (250 ns) lifetime of the excited state. Besides that, in the process of optical pumping of the working chlorine + xenon mixture one can implement the repetitively pulsed regime without replacement of the working mixture.

The essence of the considered method of photochemical excitation of the Xe_2Cl active medium consists in the photoassociative light absorption with excitation at 308 nm:

$$Xe + Cl + h\nu \rightarrow XeCl(B,C)$$

followed by the three-body recombination

$$XeCl(B,C) + 2Xe \rightarrow Xe_2Cl^* + Xe.$$

S.B. Mamaev, E.V. Polyakov, A.A. Rogashevskii, A.P. Shirokikh, V.I. Yalovoi P.N. Lebedev Physical Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia; e-mail: alsir44@mail.ru;
L.D. Mikheev P.N. Lebedev Physical Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia; National Research Nuclear University 'MEPhI', Kashirskoe sh. 31, 115409 Moscow, Russia; e-mail: mikheev@sci.lebedev.ru

Received 10 May 2017
Kvantovaya Elektronika **47** (8) 701–704 (2017)
Translated by V.L. Derbov

For the first time the small-signal gain and laser oscillation in the Xe_2Cl^* active medium was observed under e-beam pumping of the CCl_4–Xe–Ar mixture [5]. However, due to absorption induced in the plasmas, arising under the impact of the electron beam, the amplification appeared only in the afterglow after the termination of the pump. The small-signal gain at a wavelength of 514.5 nm achieved 6% per pass, or nearly 3×10^{-2} cm^{-1}. The presence of induced absorption lead also to the narrowing of the gain spectral band, which decreased the potentialities of the active medium for the amplification of femtosecond pulses.

With optical excitation, which is practically free of the drawbacks caused by the induced absorption, the laser oscillation in Xe_2Cl^* was observed for the first time in Ref. [6]. The mixture of molecular chlorine with xenon and nitrogen was excited by radiation of an open strong-current discharge initiated by the electric explosion of a thin tungsten wire.

The luminescence and gain were also observed in liquid and solid phases of the Cl_2–Xe mixture under optical excitation at a wavelength of 308 nm [7, 8]. At a pump density of 2.1×10^{17} photons cm^{-3} the gain was 11% per pass or nearly 0.15 cm^{-1} in a liquid [7] and 13 cm^{-1} in a 200-μm-thick solid-state layer [8]. In Ref. [9] the prospects of exciting the Xe_2Cl^* active gas medium with radiation of a XeCl laser for amplification of femtosecond pulses are assessed and the possible small-signal gain in this medium is calculated.

This paper presents the results of the study of gain properties of the Xe_2Cl* active medium, optically excited at 308 nm and the measurement of the small-signal gain at a wavelength of 532 nm.

2. Experimental results and discussion

The optical scheme of the experimental setup is presented in Fig. 1. The working mixture, consisting of chlorine and xenon, was placed in a cuvette (*15*) measuring 80 cm in length and 2.5 cm in diameter. The pressure varied within 0.5–10 Torr for chlorine and 0.5–2 atm for xenon. The maximal pressure of 2 atm was determined by the cuvette mechanical strength. The molecular chlorine is a donor of atomic chlorine that arises because of photolysis under the action of laser radiation.

The working mixture was excited using radiation of a XeCl laser (*3*) at a wavelength of 308 nm with an output energy of 0.35 J in a pulse having a duration of $\tau_p \sim 60$ ns. The energy was measured using a calorimeter (*4*), to which part of pump radiation was directed by means of a plate (*5*). The output energy of the XeCl laser differed from pulse to pulse by no more than 10%.

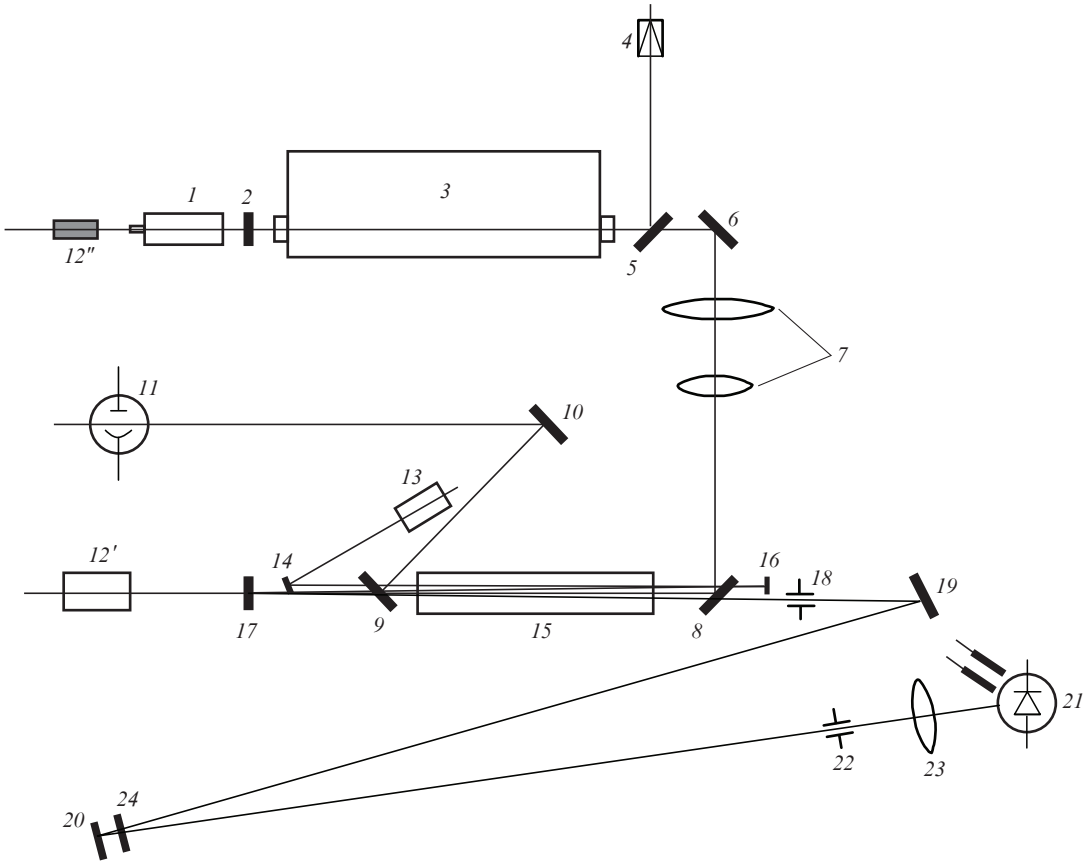

Figure 1. Optical scheme of the experiments:
(*1*) collimator; (*2*, *6*, *8*–*10*) dielectric mirrors for the wavelength 308 nm; (*3*) pump laser (XeCl laser); (*4*) calorimeter; (*5*) silica plate; (*7*) telescope; (*11*) vacuum photoelectric cell FEC; (*12′*, *12″*) alignment lasers; (*13*) probe laser with the radiation wavelength 532 nm; (*14*, *16*, *17*, *19, 20*) dielectric mirrors for the wavelength 532 nm; (*15*) cuvette with the Cl_2–Xe working mixture; (*18*, *22*) apertures; (*21*) photodiode; (*23*) lens; (*24*) attenuating filter for 308 nm.

In the experiments, the scheme of longitudinal pumping of the gain medium was applied. To increase the flux density of radiation and to match the apertures of the laser and the cuvette, a telescope (*7*) with fourfold reduction was used. The pump energy entering the cuvette (*15*) amounted to 170 mJ and the radiation intensity I_{max} at a maximum of the pump pulse was nearly equal to 10^{25} photons cm^{-2} s^{-1} in the beam having a rectangular cross section of 1.0 cm×0.5 cm. The pump radiation was directed along the cuvette axis by means of dielectric mirrors (*6*, *8*) and after the reflection from mirrors (*9*, *10*) was detected by a photoelectric cell (FEC) (*11*). The degree of the Cl_2 decomposition $\alpha \approx \sigma_p I_{max} \tau$ amounted to 10%, where $\sigma_p = 1.6\times10^{-19}$ cm^2 is the Cl_2 absorption cross section at a pump wavelength of 308 nm [10].

The active medium luminescence pulse, arising due to the pump action, was measured using a photodiode (*21*) (DET210). The signals from the photodiode and FEC were recorded using a double-beam digital Tektronix TDS 1002 oscilloscope. The oscillograms of the luminescence and pump pulses are presented in Fig. 2.

The onset of luminescence of the working mixtures coincided with that of the pump pulse, but the signals of the pump and the luminescence possessed different shapes. As compared to the pump signal, the luminescence one had an overextended rising edge. The luminescence maximum was observed at the trailing edge of the pump pulse. The delay of the luminescence maximum with respect to the pump pulse is due to the integral dependence of the atomic chlorine and Xe_2Cl^* formation on the pump pulse. The decay of luminescence corresponds to the exponential law with the characteristic time 30–50 ns. The observed decay rate is nearly by two times higher than the expected one, assuming the Xe_2Cl^* quenching by the molecular chlorine with the quenching constant $k_{Cl_2} = 4\times10^{-10}$ cm^3 s^{-1} [10], which can be explained by the presence of uncontrollable impurities or products that arise in the pump process in the working mixture. The maximal luminescence intensity was observed at the chlorine pressure about 1 Torr and

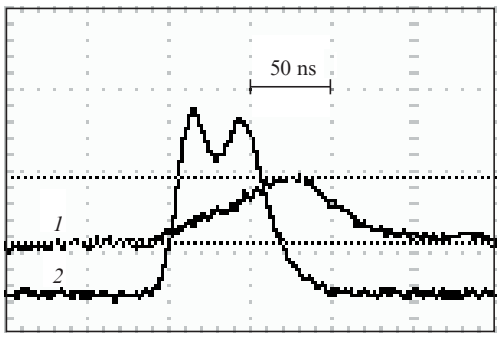

Figure 2. Oscillograms of (*1*) the Xe_2Cl^* luminescence pulse in the mixture of Cl_2 (1 Torr) and Xe (2 atm) and of (*2*) the pump pulse at 308 nm.

increased with increasing xenon pressure up to 2 atm at a fixed pressure of chlorine.

In the gain study, the active region of the cuvette was transilluminated with cw radiation of a 532-nm probe laser (*13*). The power of the probe beam amounted to 20 mW, its diameter being 2–3 mm. The beam entered the cuvette at a small angle to its axis, which was provided by a mirror (*14*). To increase the sensitivity of the measurement method, the probe beam passed through the active medium three times, being reflected by mirrors (*16*) and (*17*). After reflection from a mirror (*19*), the beam was directed to the photodiode (*21*). The photodiode detected the signal of the amplified probe radiation together with the luminescence signal (Fig. 3). The system of apertures (*18*) and (*22*) allowed these signals to be obtained at nearly the same level of magnitude.

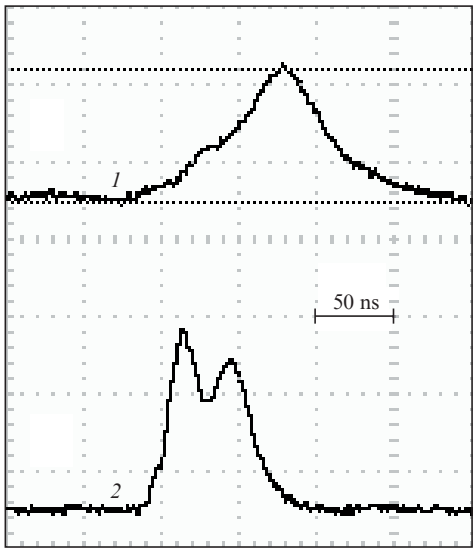

Figure 3. Oscillograms of (*1*) the luminescence signal together with the amplified probe radiation after three passes through the active medium and of (*2*) the pump signal. The mixture is the same as in Fig. 2.

The recorded signal with the amplitude I consists of the constant probe signal I_0, the increment ΔI produced by the gain, and the luminescence signal I_L:

$$I = I_0 + \Delta I + I_L,$$

I_L being determined in the experiment without the probe beam (Fig. 2). Then the total gain is $K = \exp(gL) = 1 + \Delta I/I_0$, where g is the small-signal gain and $L = 80$ cm is the path length of the beam propagation through the active medium.

The maximal amplification of probe radiation is recorded in the mixture of Cl_2 and Xe at the pressure 1 Torr and 2 atm, respectively. Figure 4 presents the time dependence of the gain K at a wavelength of 532 nm. The maximal value $K = 1.27$ corresponds to the small-signal gain $g = (1.1 \pm 0.3) \times 10^{-3}$ cm^{-1} at the above wavelength.

The 532-nm wavelength of probe radiation is shifted towards the long-wave region from the maximum of the gain cross section spectral dependence. This is seen from Fig. 5, illustrating the experimentally measured luminescence spectrum of Xe_2Cl^* and the spectral dependence of the gain cross section calculated on its base. The luminescence spectrum is plotted using the experimental values of intensity, obtained at

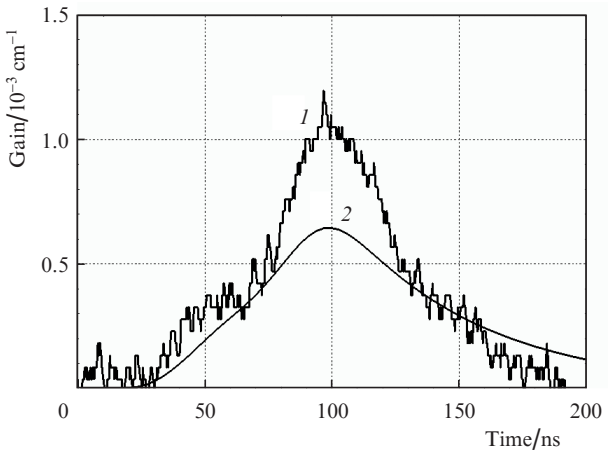

Figure 4. Time dependences of (*1*) measured and (*2*) calculated gain at $\lambda = 532$ nm.

different wavelengths by means of the monochromator and the photodiode, followed by the approximation of the obtained values with a Gaussian curve $f(\nu) \sim \exp[-4\ln 2(\nu - \nu_0)^2/\Delta\nu^2]$, where $\Delta\nu = 4000$ см$^{-1}$ ($\Delta\lambda = 100$ nm) is the luminescence bandwidth at the half-maximum level. The maximum of fluorescence corresponds to the wavelength $\lambda_0 = 496$ nm. The spectral dependence of the gain cross section (Fig. 5) was calculated using the relation [11]

$$\sigma_\nu = \frac{\lambda^2}{4\pi\tau c\Delta\nu}\left(\frac{\ln 2}{\pi}\right)^{1/2} f(\nu),$$

where $\tau = 245$ ns is the lifetime of Xe_2Cl^* [12]. The maximal value of the gain cross section corresponds to the wavelength $\lambda_0 = 504$ nm and amounts to 3.2×10^{-18} cm^2. The presented spectral dependence of σ_ν and the experimentally obtained value $g = (1.1 \pm 0.3) \times 10^{-3}$ cm^{-1} at the wavelength 532 nm make it possible to calculate the maximal value of the gain at the wavelength $\lambda_0 = 504$ nm, equal to $g_0 = 1.3 \times 10^{-3}$ cm^{-1}. The corresponding concentration of Xe_2Cl^* amounts to 4.1×10^{14} cm^{-3}.

An important characteristic of the considered mechanism of exciting the Xe_2Cl^* active medium is the excitation efficiency that can be defined as a ratio of the energy of active molecules stored in the active medium at the time of reaching

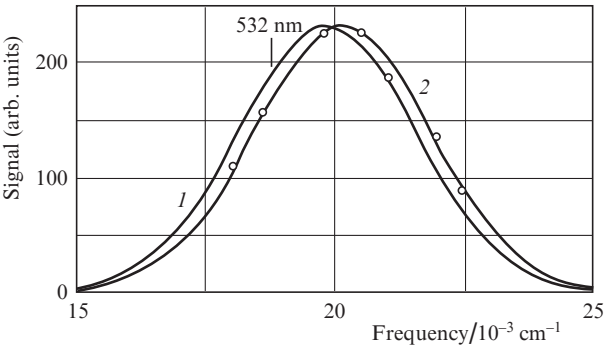

Figure 5. Experimental values of the luminescence intensity (points), (*1*) calculated spectral dependence of the gain and (*2*) approximation of the spectral dependence of the luminescence intensity with a Gaussian curve.

the gain maximum to the absorbed energy of the pump pulse (~70 mJ). The latter was determined by measuring the pump laser radiation energy at the input and output of the cuvette with the working mixture. The above value of the maximal concentration of Xe_2Cl^* corresponds to the stored energy ~6 mJ, which yields the excitation efficiency of about 9%.

The active medium was numerically modelled by solving the system of kinetic equations, analogous to that of Ref. [9] with the additional equation describing the photodissociation of the molecular chlorine. For numerical computations we used the programs written in C^{++} (Bulirsch–Stoer and Runge–Kutta methods) and Mathcad (Bulirsch–Stoer and Rosenbrock methods) that yielded similar results. The numerical modelling yields the peak value of the gain smaller by the factor of 0.6 and, as already mentioned above, by two times slower temporal decrease in the gain (see Fig. 4) than those observed experimentally. The difference of the peak values can be related to the following circumstance. Among the processes involved in the formation of the active medium an important role is played by the absorption of the pump radiation by Xe_2Cl^* excimers [13], which is formally considered as quenching with decay of the excimer into xenon and chlorine atoms. However, according to Ref. [13], in the process of this absorption $XeCl^-$ and Xe^+ ions appear, which can provide an additional channel of Xe_2Cl^* excitation as a result of plasmochemical reactions. Therefore, in the future the kinetic model should be supplemented with the above processes. Figure 6 presents the calculated dependence of the small-signal gain on the pump intensity in a 60-ns pulse, demonstrating the prospects of essential gain enhancement by increasing the pump energy. Due to the above reasons, the dependence in Fig. 6 should be considered as the lower estimate.

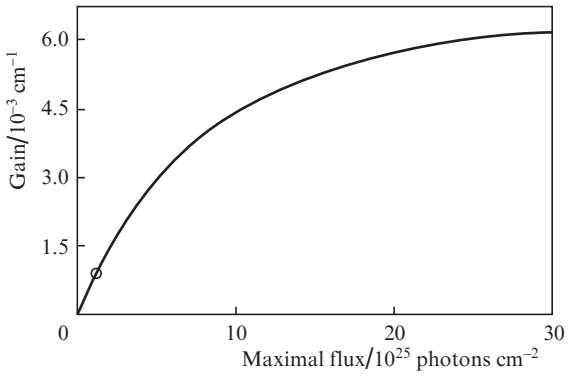

Figure 6. Calculated dependence of the maximal gain at a wavelength of 504 nm in the mixture of Cl_2 (1 Torr) and Xe (2 atm) on the photon flux for the pump pulse duration 60 ns. The circle denotes the calculated value for the actual pump used in the experiment.

3. Conclusions

Thus, we have observed for the first time the gain in the gaseous active medium Xe_2Cl^* under optical excitation of the Cl_2–Xe mixture by radiation of a 308-nm XeCl laser due to the mechanism of photoassociation of Xe and Cl atoms with subsequent recombination of the produced excimers XeCl(B,C) with xenon. The attained gain in the maximum of the amplification band located at a wavelength of 504 nm amounted to $g_0 = 1.3\times10^{-3}$ cm^{-1}, which is comparable with the gain characteristic for a XeF(C–A) amplifier, which at present is used in hybrid multiterawatt femtosecond systems operating in the visible range [3, 4]. There are prospects of considerable enhancement of the gain in the Xe_2Cl^* active medium by increasing the pump energy. The efficiency of the active medium excitation, defined as a ratio of the energy of active molecules stored in the active medium at the time of attaining the maximal gain to the pump energy absorbed at the same time, is close to 10%.

The advantage of the Xe_2Cl^* active medium as compared to XeF(C–A) is the higher value of the radiative lifetime of Xe_2Cl^* (245 ns) and, therefore, the saturation energy flux (0.15 J cm^{-2}) for the working transition in comparison with the transition in XeF(C–A) (100 ns and 0.05 J cm^{-2}) [3, 4]. Besides that, the advantage of the Xe_2Cl^* active medium consists also in the simplicity of implementing repetitively pulsed regime of operation for the amplifier of femtosecond pulses, since the working mixture of molecular chlorine and xenon is not consumed as a result of pumping and does not require replacement after each exciting pulse. In this case, XeCl lasers pumped by a fast discharge that allow repetitively pulsed operation regime can be used as a pump source. According to the literature data, the maximal energy achieved in the XeCl laser pumped with a fast discharge amounts to 66 J [14].

The scaleability of this active medium due to its gaseous nature and the presently available technologies of manufacturing e-beam-pumped XeCl lasers with an energy of at least 2 kJ [15] allow one to consider Xe_2Cl^* as a promising active medium for sub-exawatt femtosecond laser facilities operating in the visible range.

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