

ACTIVE MEDIA

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Luminescence of Xe_2Cl^* upon laser excitation of Cl_2–Xe mixtures in a wide pressure range

A.P. Shirokikh, S.B. Mamaev

Abstract. **The luminescence of Xe_2Cl^* excimer upon optical excitation of high-pressure Cl_2–Xe mixtures by XeCl laser pulses (λ = 308 nm) with different energies is investigated. It is shown that small-signal gain of the Xe_2Cl amplifier may reach ~1.3 × 10^{-1} cm^{-1} in a mixture of Cl_2 and Xe with partial pressures of 4 Torr and 9 atm, respectively, and an excitation fluence of 10^{26} photons cm^{-2} s^{-1}.**

Keywords: *active laser media, visible spectral range, gain, femtosecond systems, subexawatt power, repetitively pulsed regime.*

Good prospects of an optically excited Xe_2Cl^* active medium for amplification of femtosecond pulses in hybrid laser systems were substantiated in [1]. This medium is attractive because it possesses a wide luminescence spectrum (100 nm) in the vicinity of 500 nm, which corresponds to a 4-fs-long transform limited pulse, and a high excited-state lifetime (250 ns), providing a high saturation energy (0.15 J cm^{-2}) for the laser transition.

The optical excitation method was implemented in [2], where a mixture of chlorine with xenon was excited by an XeCl laser and a small-signal gain of 1.3×10^{-3} cm^{-1} was obtained at an Xe_2Cl^* concentration of 4.1×10^{14} cm^{-3}. In this work, we investigated the Xe_2Cl^* excimer luminescence at different energies of excitation of chlorine–xenon mixtures by XeCl laser pulses in a wide range of partial pressures of mixture components in order to estimate the possibility of increasing the active-medium gain.

The optical scheme of the experimental setup is presented in Fig. 1. The excitation source for the working mixtures was a 308-nm XeCl laser (*4*) with maximum output energy of ~0.15 J in a 60-ns FWHM pulse. The excitation energy was varied by attenuating the laser beam using glass light filters (*11*). Part of the beam was diverted by a plate (*9*) to a measuring complex, where the excitation pulse energy and shape were detected, as well as the energy distribution over the beam cross section. To increase the excitation fluence, the laser beam was focused by a lens (*7*) with a focal length of 26 cm along the axis of the cell (*1*) with a working mixture. The energy distribution over the beam cross section at different distances from a similar lens (*16*) was monitored by a profilometer (*5*). The distribution in the focal waist had the form of an asymmetric sharp peak because of the beam astigmatism. A necessary condition for correct determination of the excitation density is a uniform energy distribution over the beam cross section.

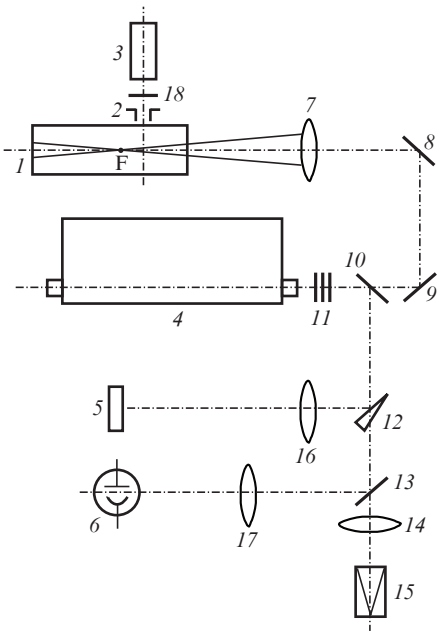

Figure 1. Schematic of the experimental setup: (*1*) cell with a working mixture; (*2*) diaphragm; (*3*) PEM FEU-36; (*4*) XeCl laser; (*5*) profilometer SP620U; (*6*) vacuum photoelectric cell; (*7*, *16*) lenses (focus F = 26 cm); (*8*, *9*) mirrors; (*10*, *13*) beam splitters; (*11*, *18*) attenuating light filters; (*12*) optical wedge; (*14*, *17*) lenses; (*15*) calorimeter; F is the focus of lens *7*.

The energy distribution over the beam cross in the observation region at a distance of 24 cm from the lens is presented in Fig. 2. The uniformity deteriorates while approaching the focus, and the fluence decreases while moving away from the focus. The distribution has the form of a plateau in the middle of the beam cross section. The beam sizes were determined at a level of 0.5 of the average energy in the plateau region. The beam cross-sectional area was S = 0.04 cm^2. The excitation fluence was found as $I = E/(h\nu S\tau)$, where E is the excitation energy at the input of the working medium, $h\nu$ is the photon energy, and τ is the excitation pulse duration. The maximum excitation fluence was I_{max} = 10^{26} photons cm^{-2} s^{-1} (~60 MW cm^{-2}).

Luminescence was observed in the direction perpendicular to the cell axis through a diaphragm (*2*) using a photo-

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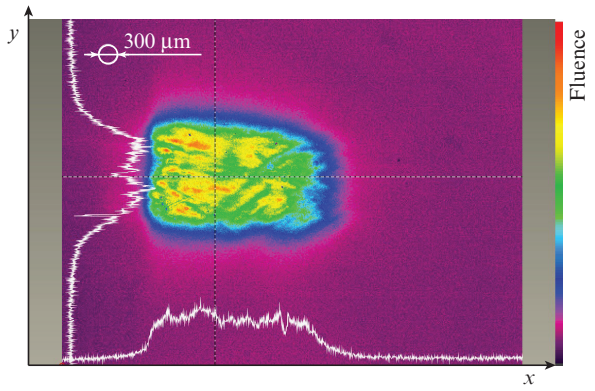

Figure 2. (Colour online) Energy distribution over the beam cross section in the observation region. The profilograms correspond to the dotted straight lines in the beam cross section.

electron multiplier (PEM) (*3*) with light filters (*18*), which provided PEM operation in the linear regime and blocked 308-nm radiation. The characteristic shape of excitation and luminescence pulses was presented in [2]. In this study, a PEM was used instead of photodiode to increase sensitivity. Luminescence pulses whose amplitudes corresponded to the maximum concentration of Xe_2Cl^* excimer for mixtures of certain composition were recorded with a step change in excitation energy.

The dependences of the amplitude of Xe_2Cl^* luminescence pulses on excitation fluence were recorded at a constant (2 atm) xenon pressure in mixture and a chlorine pressure varying in the range of 1–8 Torr (see Fig. 3a) and at a constant chlorine pressure (1 Torr) and a xenon pressure changing from 1 to 9 atm (Fig. 3b). The experimental values are approximated by polynomials of the order no higher than 3, plotted using the least-squares method. The dependences form fanlike sets of divergent curves, tending to saturation. These curves are similar in shape to the calculated dependence of Xe_2Cl^* concentration on excitation power, obtained within the kinetic model of the active medium for a Cl_2–Xe mixture at chlorine and xenon pressures of 1 Torr and 2 atm, respectively [2].

It follows from Fig. 3 that an increase in excitation fluence by an order of magnitude leads to an increase in the luminescence pulse amplitude for a Cl_2–Xe mixture by a factor of ~7 at chlorine and xenon pressures of 4 Torr and 2 atm, respectively [Fig. 3a, curve (*3*)], and by a factor of ~8 at corresponding pressures of 1 Torr and 9 atm [Fig. 3b, curve (5)]. With an increase in partial chlorine pressure in mixture, as well as with an increase in xenon pressure, the dependences saturate at higher excitation fluences.

The dependences of luminescence pulse amplitude on chlorine pressure in a mixture with Xe (2 atm) at excitation intensities $0.1I_{max}$, $0.4I_{max}$, and $0.7I_{max}$ are presented in Fig. 4. An increase in chlorine pressure from 1 to 4 Torr leads to a rise in the luminescence pulse amplitude by a factor of 1.8–2.4, depending on the excitation energy.

Based on the data of Fig. 3b, one can show how the luminescence intensity changes as compared with the case of Cl_2–Xe mixture with partial pressures of 1 Torr and 2 atm, respectively, which was used in [2]. This dependence is presented in Fig. 5. With an increase in xenon pressure the luminescence pulse amplitude increases linearly with an increase in excitation fluence, but the slope is different. At excitation

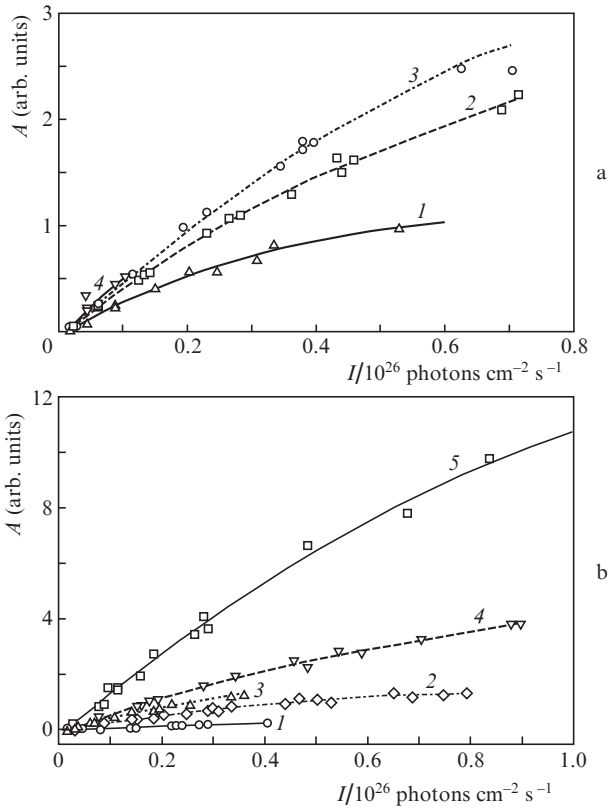

Figure 3. Dependences of the amplitude A of Xe_2Cl^* luminescence pulses on excitation fluence I for Xe–Cl_2 mixtures (a) at an Xe pressure of 2 atm and Cl_2 pressures of (*1*) 1, (*2*) 2, (*3*) 4, and (*4*) 8 Torr and (b) at a Cl_2 pressure of 1 Torr and Xe pressures of (*1*) 1, (*2*) 2, (*3*) 3, (*4*) 4, and (*5*) 9 atm. The relative measurement error does not exceed 10%.

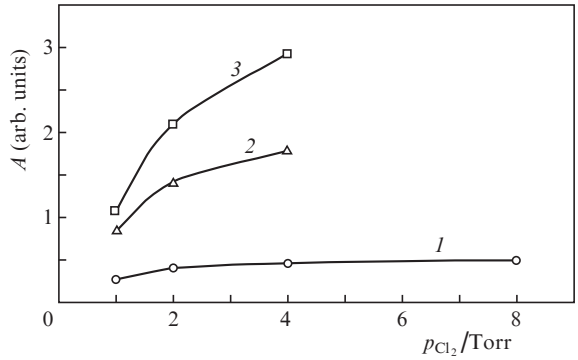

Figure 4. Dependences of the luminescence pulse amplitude A on chlorine pressure p_{Cl_2} in a mixture with Xe (2 atm) at excitation intensities (*1*) $0.1I_{max}$, (*2*) $0.4I_{max}$, and (*3*) $0.7I_{max}$.

fluences of $0.1I_{max}$ and $0.5I_{max}$, an increase in the xenon pressure to 9 atm leads to a rise in the luminescence amplitude by a factor of 5, whereas at the density I_{max} the increase is ~8 times larger; i.e., the dependence is stronger at higher energies. The linearity of the dependence indicates that even higher luminescence intensity can be obtained by increasing the xenon pressure in the mixture.

Based on the results obtained, one can conclude that an increase in the partial pressure of chlorine–xenon mixture components leads to a rise in the luminescence intensity and, correspondingly, increases the active-medium gain. This

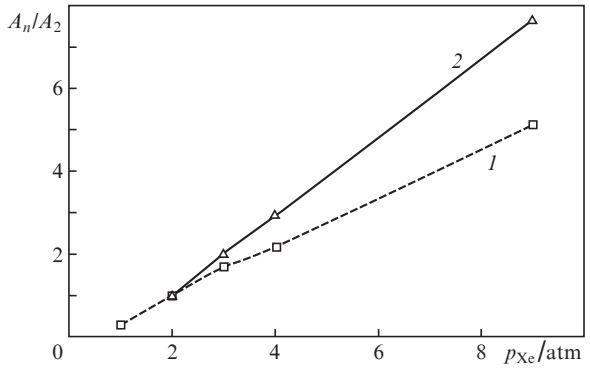

Figure 5. Relative change in the luminescence pulse amplitude A_n, normalised to the amplitude A_2 at an Xe pressure of 2 atm in the mixture, depending on the Xe pressure p_{Xe} in the mixture with Cl_2 (1 Torr) at excitation energies (*1*) $0.1I_{max}$ and $0.5I_{max}$ and (*2*) I_{max}.

dependence tends to saturate with an increase in chlorine pressure but remains linear for xenon.

Proceeding from the small-signal gain value $g_0 = (1.3 \pm 3) \times 10^{-3}$ cm^{-1}, measured in [2] for a mixture of chlorine (1 Torr) and xenon (2 atm) at an excitation fluence $I = 10^{25}$ photons cm^{-2} s^{-1}, one can conclude the following. Since an increase in the partial chlorine pressure in mixture from 1 to 4 Torr leads to a rise in the luminescence amplitude by a factor of 1.8 (Fig. 4) and an increase in the xenon pressure from 2 to 9 atm causes an increase in the luminescence amplitude by a factor of 5 (at the same excitation fluence) (Fig. 5), the gain can be increased by a factor of 9 [to a value of $(1.2 \pm 0.6) \times 10^{-2}$ cm^{-1}] by increasing the partial pressures of mixture components.

At $I_{max} = 10^{26}$ photons cm^{-2} s^{-1}, the luminescence pulse amplitude for a mixture of Cl_2 (1 Torr) with Xe (2 atm) increases by a factor of 5.2 [Fig. 3a, curve (*1*); Fig. 3b, curve (*2*)]. With an increase in the Cl_2 pressure from 1 to 4 Torr (at the same excitation level), the amplitude increases by a factor of 2.5 [Fig. 3a, extrapolation by curve (*3*)], while with an increase in Xe pressure from 2 to 9 atm it increases even more by a factor of 8 [Fig. 5, curve (*2*)]. Thus, the gain can be increased by a factor of 104, i.e., to $\sim(1.3 \pm 0.7) \times 10^{-1}$ cm^{-1}.

Electron-beam and discharge excitation of similar mixtures yielded a gain up to 3×10^{-2} cm^{-1} [3–7]. Induced absorption is absent in our case, due to which the spectral band can be used more efficiently to amplify femtosecond pulses. The possibility of changing the active-medium gain allows one to consider various designs of amplification systems.

The scalability of Xe_2Cl^* active medium in volume and energy (in view of its gaseous nature), as well as the technological progress in designing XeCl lasers excited by an electron beams with an energy of at least 2 kJ [8], give grounds to consider Xe_2Cl^* a promising active medium for developing femtosecond laser systems of subexawatt power in the visible spectral range.

Another advantage of this medium is the simplicity of implementing the repetitively pulsed regime for a femtosecond-pulse amplifier, because the chlorine--xenon mixture, used as a working medium, is not spent upon excitation, and one does not need to replace it after each pulse. Fast-discharge pumped XeCl lasers, operating in the repetitively pulsed regime, can be used as excitation sources.

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