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Evaluating the prospects of exciting the Xe_2Cl active medium by laser radiation for amplifying femtosecond pulses

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***Abstract*. The possibility of exciting the Xe_2Cl active laser medium produced upon photoassociation $Xe + Cl + h\nu \rightarrow XeCl(B)$ caused by absorption of XeCl laser radiation at 308 nm followed by a three-particle recombination to $Xe_2Cl(4\,^2\Gamma)$ is considered. The main pump parameters determining the small-signal gain are estimated, which is of practical interest for amplifying femtosecond laser pulses.**

Keywords: *photoassociation, active laser media, femtosecond pulse amplifiers.*

Broadband active media of photochemical lasers on the 405-nm $Kr_2F(4\,^2\Gamma - 1, 2\,^2\Gamma)$, 490-nm Xe_2Cl $(4\,^2\Gamma - 1, 2\,^2\Gamma)$ and 485-nm XeF (C–A) are attractive for amplifying femtosecond pulses in hybrid laser systems [1, 2]. This approach is based on the idea of using these media for amplification of Ti : sapphire laser radiation converted by nonlinear optical techniques (e.g., by frequency doubling) into the desired spectral range [3]. This allows a several orders of magnitude increase in the contrast of multiterawatt pulses in Ti : sapphire laser based systems. Photochemical method of exciting these media has been developed upon optical pumping of open discharges by VUV radiation (see, for example [2, 4]) and used for constructing femtosecond optical pulse amplifiers [3, 5, 6].

Here, we consider an alternative approach involving optical excitation of the Xe_2Cl^* active medium by laser radiation. In this method, the B state of XeCl is excited by photoassociation upon absorption of XeCl laser radiation at 308 nm by the weakly bound ground state of XeCl(X) followed by recombination of XeCl(B) with Xe, which leads to the formation of Xe_2Cl^*. The XeCl(X) concentration sufficient for realisation of such an approach can be obtained in a Xe–Cl mixture with atomic chlorine of concentration $10^{16} - 10^{17}$ cm^{-3} produced by electric discharge, electron beam or optical technique. This process efficiently occurs, for example, in mixtures of Cl_2 with Xe. The excitation mechanism considered here is described by the reactions:

$$Cl + 2Xe \leftrightarrow XeCl(X) + Xe, \tag{1}$$

$$XeCl(X) + h\nu \rightarrow XeCl(B,C), \tag{2}$$

$$XeCl(B,C) + 2Xe \rightarrow Xe_2Cl^* + Xe. \tag{3}$$

The rate constants of dissociation of XeCl(X) molecules ($k_{-1} = 5.6 \times 10^{-12}$ cm^3 s^{-1} [7]) and recombination of XeCl(B, C) with xenon ($k_3 = 1.3 \times 10^{-30}$ cm^6 s^{-1} [8]), as well as the cross section of the XeCl(X→B) transition ($\sigma_1 = 2.6 \times 10^{-16}$ cm^2 [9]), are known from the literature. The recombination rate constant k_1 of chlorine and xenon atoms can be obtained by estimating the equilibrium constant K_b between the bound and free states of chlorine and xenon atoms from the detailed balancing principle using the formula [10]

$$K_b = \frac{g_{RG}}{g_R g_G} h r_e^2 \left(\frac{8\pi}{kT\mu}\right)^{1/2} \left[1 - \exp\left(-\frac{hc\omega_e}{kT}\right)\right]^{-1} \exp\left(\frac{D_e}{kT}\right),$$

where $g_{RG}/(g_R g_G) = 1/2$ is the ratio of statistical weights in the bound and free states of atoms (RG = XeCl, R = Xe, G = Cl), $r_e = 3.23$ Å [11] is the equilibrium internuclear distance in XeCl(X), $\mu = 4.7 \times 10^{-23}$ g is the reduced molecular mass, $\omega_e = 26.2$ cm^{-1} [11] is the vibrational frequency, and $D_e = 281$ cm^{-1} [11] is the molecular dissociation energy. At room temperature, $K_b(XeCl) \approx 4 \times 10^{-22}$ cm^3. This leads to the value $k_1 = 2.2 \times 10^{-33}$ cm^6 s^{-1} for the recombination rate constant.

If the rate of depletion of the XeCl(X) state due to pumping exceeds the rate of its dissociation, the excitation rate of XeCl(B, C) is determined by the rate of recombination (1) of chlorine atoms. For this purpose, it is necessary to maintain the pump photon flux density I at a level exceeding $k_{-1}[Xe]/\sigma_1 = 5.4 \times 10^{23}$ photons cm^{-2} s^{-1}, where $[Xe] = 2.5 \times 10^{19}$ cm^{-3} is the concentration of xenon atoms corresponding to the xenon pressure of 1 atm, which ensures a nearly maximum efficiency of formation of Xe_2Cl^*.

Let us take for definiteness the concentration of chlorine atoms at $[Cl] = 10^{16}$ cm^{-3}, for which the rate of Xe_2Cl^* quenching by atomic chlorine is relatively low and comparable to the radiative decay rate for Xe_2Cl^* if we assume that the rate constant of Xe_2Cl^* quenching by atomic chlorine is equal to the corresponding value of 4×10^{-10} cm^3 s^{-1} for molecular chlorine [13]. Neglecting losses due to quenching and radiative decay of excited

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states, we can estimate the concentration $k_1[\mathrm{Xe}]^2[\mathrm{Cl}]\tau_\mathrm{p}$ of excimers Xe_2Cl^*, which, for example, amounts to $\sim 1.4 \times 10^{15}$ cm^{-3} for a pump pulse duration of $\tau_\mathrm{p} = 10^{-7}$ s. For the cross section of the induced transition $\mathrm{Xe_2Cl}(4\,{}^2\Gamma \rightarrow 1, 2\,{}^2\Gamma)$ $\sigma_\mathrm{st} = \lambda^4/(8\pi c \tau_\mathrm{2sp} \Delta\lambda) = 3.5 \times 10^{-18}$cm^2 ($\tau_\mathrm{2sp} = 242$ ns is the radiative lifetime of Xe_2Cl^*, $\Delta\lambda = 80$ nm is the amplification bandwidth [14]), the corresponding small-signal gain k_g is $\sim 5 \times 10^{-3}$ cm^{-1}. Under our conditions, about 1 % of chlorine atoms are in the bound state, which ensures the absorption of pump photons over a length of about 40 cm, which is convenient for realisation of the longitudinal pump scheme in the amplifier.

Almost the same results were obtained in a more exact solution of the rate equations under steady-state pumping and stationary profile of XeCl(X) distribution along the direction of pump beam propagation, the profile formation time being $\sim(\sigma_1 I)^{-1}$. For the sake of definiteness, we take Cl_2 as a donor of atomic chlorine and assume that molecular chorine dissociates to such an extent during the preparation of the initial Xe – Cl mixture by the above methods, that it cannot noticeably affect the relaxation of excited states. Then the complete pattern of kinetic processes in the case of laser excitation of the Xe – Cl mixture can be described by the following rate equations:

$$\frac{\mathrm{d}n_1^*}{\mathrm{d}t} = \sigma_1 I n_1 - k_3 n_1^* N^2 - n_1^* \tau_1^{-1}, \tag{4}$$

$$\frac{\mathrm{d}n_1}{\mathrm{d}t} = k_1 m N^2 - k_{-1} n_1 N - \sigma_1 I n_1 + n_1^* \tau_1^{-1}, \tag{5}$$

$$\frac{\mathrm{d}n_2^*}{\mathrm{d}t} = k_3 n_1^* N^2 - n_2^* \tau_2^{-1}. \tag{6}$$

Here, we take into account the fact that the XeCl(C) state lies just 98 cm^{-1} below the XeCl(B) state [8], and these states under our conditions can be treated as a single XeCl(B, C) state with the radiative lifetime $\tau_\mathrm{1sp} = 25$ ns due to strong collisional mixing [8]. In the equilibrium state, the ratio of populations of the XeCl(C) and XeCl(B) states amounts to 1.6 [8]. For simplicity, we assume that the XeCl(B, C) molecules undergo transitions to the ground state upon quenching. In rate equations (4) – (6), n_1^*, n_1, n_2^*, N, and m are the concentrations of XeCl(B, C), XeCl(X), Xe_2Cl^*, Xe, and Cl, respectively; $\tau_1^{-1} = \sigma_1 I (n_\mathrm{1B}^*/n_1^*) + k_{1m} m + (\tau_\mathrm{1sp})^{-1} + k_{1N} N$ is the decay rate for XeCl(B, C) due to parasitic processes of quenching by atomic chlorine and xenon, as well as due to spontaneous and induced radiation to the ground state; n_1B^* is the XeCl(B) concentration; k_{1m} is the rate constant for XeCl(B, C) quenching by atomic chlorine, which is assumed to be equal to the value 4.3×10^{-10} cm^3 s^{-1} for molecular chlorine [15]; $k_{1N} = 5 \times 10^{-12}$ cm^3 s^{-1} is the rate constant of quenching by xenon [8]; $\tau_2^{-1} = k_{2m} m + \sigma_2 I + (\tau_\mathrm{2sp})^{-1}$ is the decay rate for Xe_2Cl^* due to quenching by atomic chlorine, absorption of pump radiation by triatomic excimers, and their radiative decay (the rate constant k_{2m} is assumed to be equal to the corresponding value of 4×10^{-10} cm^3 s^{-1} for molecular chlorine [13], and the absorption cross section is $\sigma_2 = 1.3 \times 10^{-17}$ cm^2 [16]).

If the pump time $\sim 10^{-7}$ s is much longer than the characteristic times of all processes taken into account except τ_2, we can put $\mathrm{d}n_1^*/\mathrm{d}t = \mathrm{d}n_1/\mathrm{d}t = 0$. In this case, the solution of Eqns (4) – (6) for n_2^* has the form

$$n_2^* = \frac{\alpha k_3 k_1 m N^4 \tau_2}{\tau_1^{-1} + (1 + \alpha) k_3 N^2} [1 - \exp(-t/\tau_2)],$$

where $\alpha = \sigma_1 I (k_{-1} N)^{-1}$ is the parameter determining the excess of the rate of XeCl excitation over the rate of XeCl(X) dissociation.

By way of an example, let us consider a Xe – Cl mixture under a pressure of 1.5 atm and an atomic chlorine concentration of 5×10^{16} cm^{-3}; the mixture is excited by XeCl laser pulse of duration 10^{-7} s, which ensures $\alpha = 2$. Under our conditions, $I = 1.6 \times 10^{24}$ photons cm^{-2} s^{-1}, $\tau_1^{-1} = 4.1 \times 10^8$ s^{-1}, $\tau_2^{-1} = 4.5 \times 10^7$ s^{-1}, $n_2^* = 2 \times 10^{15}$ cm^{-3}, and $k_\mathrm{g} = 7 \times 10^{-3}$ cm^{-1}. In this case, the gain varies insignificantly along the direction of propagation of pump radiation. For example, in the cross section in which the radiation flux decreases by a factor of e, the gain is $\sim 5.5 \times 10^{-3}$ cm^{-1}. This is due to an increase in the population of the XeCl(X) state and a decrease in the rate of decay of Xe_2Cl^* due to pump radiation with decreasing flux of this radiation.

The value obtained for k_g is much higher than the small-signal gain obtained at present in the photochemical XeF(C – A) amplifier (2×10^{-3} cm^{-1} [5, 6]) and indicates high potentialities of the method of exciting the active medium Xe_2Cl^*. A considerable advantage of this medium is also the broader amplification width and thrice as large saturation energy (0.15 J cm^{-2}) on the working transition, which determines the output energy per unit aperture area of the amplifier.

The main advantage of the optical excitation of gaseous active media is that, unlike in the case of electron pumping, there is no induced absorption in such media, which could reduce the effective width of the amplification band. This advantage is easily realised in the method of Xe_2Cl excitation considered here since, even if atomic chlorine is obtained by the electric discharge or electron beam method, the characteristic plasma recombination times are much smaller than the recombination times for chlorine atoms, which are of the order of $\sim 10^{-4}$ s under the conditions considered by us.

As for the methods for obtaining the required concentrations of atomic chlorine, the most obvious at present is the possibility of using an electron beam and optical pumping, although the latter is an order of magnitude less efficient from the energy point of view. However, the electric discharge is the most attractive method due to its simplicity and compactness. The only problem is that it is difficult to obtain a homogeneous discharge occupying a volume of ~ 1 L in a mixture with a high xenon concentration and special approaches are required for obtaining such a discharge.

Photoassociation of collision-induced atomic pairs (in particular, xenon and chlorine atoms) has been used in spectral and kinetic studies for a relatively long time (see, for example, [13, 16 – 18]). The above estimates show that this method can also be of practical significance, since it makes it possible to excite active laser media (in particular, in designing femtosecond pulse amplifiers for obtaining high-contrast femtosecond radiation at a multiterawatt power level, which can be used in the physics of ultrastrong laser fields).

As for the other two transitions [$\mathrm{Kr_2F}(4\,{}^2\Gamma - 1, 2\,{}^2\Gamma)$ and XeF(C – A)] mentioned at the beginning of this paper, their associative excitation in mixtures of krypton or xenon with fluorine [followed by three-particle recombination of KrF(B) into Kr_2F or collisional relaxation of XeF(B) into the C state] is low-efficient since KrF does not possess a bound ground state. In the case of XeF, the C state is strongly quenched by xenon (the quenching rate constant is 1.2×10^{-10} cm^3 s^{-1} [19]). However, the existence of a high binding energy (1175 cm^{-1} [20]) for XeF(X), which exceeds the binding energies of other diatomic halogenides of noble gases, makes it possible to realise other methods for exciting the XeF(C – A) transition. As a matter of fact, due to the high binding energy, the lifetime of the XeF(X) molecule under atmospheric pressure amounts to about 10^{-7} s (the dissociation rate constant is 1.1×10^{-11} cm^3 s^{-1} [21]). This makes it possible to obtain XeF(X) using a short-time discharge or an electron beam with subsequent optical excitation in the B state by radiation of an excimer XeF laser with a time delay sufficient for relaxation of states responsible for induced absorption.

It is also interesting to obtain XeF in the ground state with the help of photodissociation of XeF_2 by ArF or KrF laser radiation. In this case XeF(X) molecules are mainly formed at vibrational levels $v'' = 3$, 4 [22], which are the lower levels of the XeF (B – X) laser-induced transition. This circumstance compensates to a considerable extent higher energy expenditures characteristic of the optical method for obtaining XeF(X). A more detailed analysis of the proposed methods of pumping is required for estimating the prospects of laser excitation of the active medium on the XeF(C – A) transition with a view to obtain the gain that is of practical interest for amplifying femtosecond pulses.

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