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The Effect of Super-Radiance on the C-A Transition of the Excimer Molecule XeCl* at 352 nm Excited Only by ^{235}U Fission Fragments

Aleksey Iustinovich Mis'kevich[1], Jin Bo Guo[2], Yurii Alekseevich Duyzhov[3], Anton Viktorovich Podkopaev[3]

[1]Department of Laser Physics, National Research Nuclear University MEPHI, Moscow, Russia

[2]School of Physical Science and Technology, Tianjin Polytechnic University, Tianjin, PR of China

[3]Department of Cosmic Energy System, Institute of Physics and Power Engineering, Obninsk, Russia

Email address:
aimiskevich@mephi.ru (A. I. Mis'kevich)

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Abstract: The work is devoted to the study of spectral, temporal and generation characteristics of plasma generated in dense gas mixtures by the products of neutron nuclear reaction $^{235}U(n, f)$. This plasma differs in its properties from the discharge plasma, as has the track structure and low temperature of electrons, and the presence in gas mixtures of electronegative gas can become without electrons. On the basis of such nuclear-excited plasma excimer gas lasers with the nuclear pumping, carrying out direct transformation of nuclear energy to the laser radiation UV range of lengths of waves, can be created. We found out high efficiency of formation of excimer molecules of XeCl* in dense (~ 760 Topp) Ar - Xe - CCl_4 of gas mixture with low (~ 10^{15} mol/см^3) concentration of CCl_4 at the nuclear pumping. Here we report the first clear observation of super-radiance at 352 nm on C-A transition of excimer molecule $XeCl^*$ excited solely by nuclear pumping Ar-Xe-CCl_4 gas mixture at a pressure of 760 Torr with a low concentration of CCl_4 (0.15Torr). The mixture was excited by products of neutron nuclear reaction $^{235}U(n,f)$ pumped at a rate of 2kW/cm^3. A laser cell with a length of 100cm was placed inside an optical resonator tuned at 352 nm. Super-radiance had a very sharp increase and low neutron threshold (about $2\div4.10^{14}$ neutron/cm^2· s) when we used the tuning resonator at 352 nm wavelength with 10.8% reflection of output and 62.1% reflection of back spherical mirrors, and super-radiance disappeared if the laser output mirror was replaced by a quartz window.

Keywords: Excimer, Super-Radiance, Nuclear Pumping, XeCl, Fission Fragments

1. Introduction

Super-radiance emission at 352 nm on the C-A transition XeCl* excimer molecule was observed in a nuclear-excited plasma for the first time. The plasma was created by the ^{235}U (n,f) reaction induced by thermal neutrons from the pulse nuclear reactor BARS-6.

Halogen and xenon-containing mixtures excited by products of nuclear reactions allow to receive UV light emission with high (more than 10%) efficiency on B-X or C-A transitions of XeF*, XeCl*, XeBr* and XeI* excimer molecules at the wavelengths of 351nm, 308nm, 282nm and 253nm respectively. Experimental studies of nuclear-pumped excimer media were started immediately after the appearance of an excimer laser pumped by an electron beam and were mainly carried out for the excimer XeF * molecule [1-6] because the calculations predicted a low threshold for lasing (about 10 kW/cm^3) for XeF*excimer only [7-9]. In works [1, 2] weak amplification (~ 10^{-4} cm^{-1}) of the spontaneous emission of the B – X band of the XeF * molecule (λ = 351 nm) was reported when pumping $^{10}B(n, \alpha)^7Li$ nuclear products with a specific pumping power ~ 50 W/cm^3. For the gas mixture ^3He-Xe-NF_3 excited by the products of the nuclear reaction $^3He(n, p)^3T$, the gain was measured at a specific pumping power of 5 kW /cm^3 and amounted to ~ $7\cdot10^{-3}$ cm^{-1} [3], and when pumping a Ne(Ar) -Xe-NF_3 gas

mixture with uranium fission fragments with a specific pumping power of ~ 2 kW /cm^3 the gain is ~ $2 \cdot 10^{-3}$cm^{-1} [4, 5]. Magda et al. [6] observed the unusual shape of luminescence pulse in XeF* system pumped by ^{235}U fission fragments at 1.5-2.5 kW/cm^3. In spite of the several attempts of direct nuclear pumping excimer lasing has not been received yet [1-6].

Special experiments with active media based on XeF [4, 10], KrF [11], XeCl [12] molecules were carried out at pulsed nuclear reactors to generate lasing. ^{3}He-Xe-NF_3 [4], Ne-Xe-NF_3 [4], Ne(Ar) -Xe-NF_3 (SF_6) [10], ^{3}He-Kr-NF_3 [11], Ar-Xe-CCl_4(HCl) [12] were used. Here only Ar-Xe-CCl_4(HCl) gas mixtures have a low content of the donor. These experiments also did not give a positive result except for the work [12], where it was reported about obtaining weak generation of the XeCl molecule on the B-X transition (λ = 308 nm) with uranium fission fragments at a specific pumping power of ~ 1 kW / cm^3.

We conducted experimental studies of luminescence of halogen-containing rare-gas molecules upon the excitation of a dense (~ 1 atm) gas mixture by fast electrons with the energy of 150 keV and uranium fission fragments (E_f~ 70 MeV). It was found that to obtain an effective action of an excimer laser on the B-X, C-A or $4^2\Gamma$-$1^2\Gamma$ transitions of molecules XeCl*, XeBr*, XeI*, KrF*, Xe_2Cl*, Kr_2F, Xe_2I*, it is necessary to maintain a low concentration of halogen-containing molecules in the gas mixture compared to the concentration of electrons in a track of a nuclear particle [13-17]. This is due to the properties of a track structure of plasma created by high-energy charged particles: all the kinetic energy of a charged particle in a dense medium stands out in a narrow region of space along the particle's trajectory with characteristic dimensions of about 1 micron in diameter and about several centimeters long [18]. The structure of the formed track plasma (concentration and temperature of electrons in the track, concentration of the positive ions) only depends on the total pressure and composition of the gaseous medium and depends weakly on specific power contributions to the gas. The characteristic concentration of electrons in the track is 10^{13}-10^{14} cm^{-3} [18, 19]. The characteristics of the track plasma start to change due to specific power contributions to the gas when the tracks begin to overlap.

2. Nuclear Excited Plasma and Formation of the Excimer Molecules

The presence of electronegative gas (for example CCl_4, SF_6, C_3F_7I), makes the track plasma electron-free due to the effective attachment of electrons to donor molecules of halogen atoms. The formation of negative halogen ions at atmospheric pressure of the gas mixture and partial pressure of the electronegative donor of about 0.03 Torr occurs in a few nanoseconds [20]. Plasma-chemical processes of energy conversion into light radiation occur within the track volume, the main formation channel of the excimer molecules XeCl*, XeBr*, XeI*, KrF*, Xe_2Cl*, Kr_2F, Xe_2I*, Xe_2Cl* is via the ion-ion recombination of Xe^+, Xe_2^+, Kr^+, Kr_2^+ positive ions and negative halogen ions Br^-, Cl^-, F^-, I^- In this case the optimal concentration of a donor halogen molecules should not exceed the concentration of electrons in the track significantly, since the number of negative ions formed is determined by the available electron concentration in the track. Excess of the donor in the mixture will cause strong quenching of the excimer luminescence. Ion-ion recombination should be carried out due to mutual diffusion of ions of different signs within the volume track. At high pressure of the gaseous medium the diffusion coefficient is small.

Our research has shown that Ar-Xe-CCl_4 and Xe-CCl_4 gas mixtures of a high pressure with a low concentration of CCl_4 molecules are of particular interest. When these gas mixtures were pumped by nuclear particles, an effective population was observed in the B, C and $4^2\Gamma$ states of the excimer molecules XeCl* and Xe_2Cl* in this condition [13-17].

3. Experimental Setup

For laser experiments with these media we constructed a stainless steel laser cell (5-cm-i.d. x 100cm long). Inside this cell we inserted a tube (2.7-cm-i.d.x 70 cm long) on the inner surface of which a 5 mg/cm^2 layer of ^{235}U was applied. The optical cavity consisted of 2 multilayer dielectric 200-cm-radius spherical mirrors on quartz substrates with a diameter of 40 mm at a distance of 1 meter from one an other and having about 0.3% transmission at 308 nm (rear mirror) and 0.4% front. These mirrors are intended to work at λ= 308 nm (B-X transition of XeCl*). At a wavelength of 352 nm these mirrors had a transmission of 37.9% (rear mirror) and 89.2% (front) and had produced a low-quality optical resonator at a wavelength of 352 nm. Due to the lack of high-quality mirrors at 352 nm, we used this resonator in experiments at a wavelength of 352 nm.

The laser cell was filled with gas mixture composed of Ar-760 Torr, Xe-40 Torr, CCl_4- 0.15 Torr.

The laser cell was installed inside a polyethylene moderator of a fast neutrons with the wall thickness of 5 cm and the length of 95 cm and was placed directly between two reactor cores of a pulse nuclear reactor BARS-6. The shape of the neutron pump pulse was recorded by a fission chamber KNT-5 installed inside the neutron moderator on the outer surface of the middle part of the laser cell. The specific power contribution of the fission fragments into the gas medium during the whole time of the neutron pump pulse with the duration of $2 \cdot 10^{-4}$ s was 180 mJ/cm^3.

The optical detection apparatus was located about 18m away, beyond the reactor shielding using 3 mirrors with Al coating. It consisted of a MAYA-2000Pro diffraction spectrometer with a fiber light guide and a photomultipliers FEU100 and FEU106 working in a current regime. UV optical filters at λ =308 nm and λ =352 nm were installed before the photomultipliers. The signals of each photomultiplier were recorded using a two-channel TDS220 and TDS1012 digital oscilloscope. At the same time one of

the channels recorded the shape of the neutron burst by the KNT-5 fission camera.

4. Experimental Results

Strong super- radiance emission was obtained at 352nm on the C-A transition of an excimer molecule XeCl* (figure 1). The upper trace is the thermal-neutron pulse from the fission chamber KNT-5, the lower trace is a neutron-excited Ar-Xe-CCl_4 super-radiance pulse from FEU-100, showing very rapid growth of the amplitude of a super-radiance emission. Despite the strongly non-optimal parameters of the laser mirrors (small reflection coefficient at λ =352 nm, small length of the laser tube), the threshold of the super-radiance emission was low (about $2\div4\cdot10^{14}$ n/cm^2.s) which corresponded to 15÷30% of the maximum neutron flux in the moderator. The super-radiance emission with the duration of about 90 microseconds occurred at the leading edge of the pump pulse and stopped after the maximum values of the neutron flux density had been reached (figure 1).

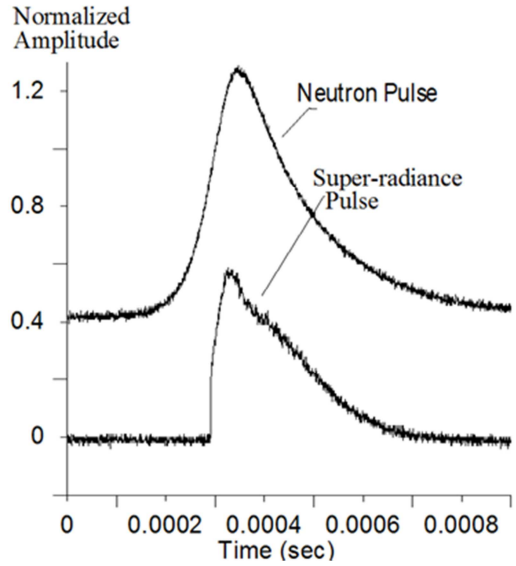

Figure 1. *Typical data trace of the super-radiance signal (using the BARS-6 nuclear reactor). This trace was taken using Ar-Xe-CCl_4 gas mixture (Ar-760 Torr, Xe-40 Torr, CCl_4- 0.15 Torr) pumped by the products of $^{235}U(n,f)$ nuclear reaction.*

Upper trace: fission chamber KNT-5
Lower trace: super-radiance pulse recorded by a FEU106 photomultiplier with UV optical filters at λ=352 nm.
The optical resonator had 10.8% reflection of the output and 62.1% reflection of the back mirrors at 352 nm.

It should be noted that weak super-radiance emission took place on the B-X transition of a XeCl* excimer molecules with a wavelength of 308 nm. The lasing threshold was lower than the one at the C-A transition, but it stopped much earlier due to the non-stationary absorption of the radiation at 308 nm in the active medium of the laser. This was apparently due to the accumulation of the metastable Xe* atoms and the absorption due to the process (1):

$$Xe^* + hv\ (308\ nm) \rightarrow Xe^+, \quad (1)$$

and also due to the process (2) of its own self-absorption by the excimer molecules XeCl (B)* with the subsequent emission of radiation from the D-X band (λ_{max}=236 nm) (3) [17]:

$$XeCl(B)^* + hv\ (308\ nm) \rightarrow XeCl(D)^*, \quad (2)$$

$$XeCl(D)^* \rightarrow Xe + Cl + hv\ (236nm). \quad (3)$$

Figure 2 shows the fragments of the emission spectra of the Ar-Xe-CCl_4 gas mixture recorded in the presence of super-radiance (2) and without super-radiance (1) when instead of the output (10.8% reflection) and back (62.1% reflection) spherical mirrors a quartz window and a mirror with 80% transmission at 352 nm were installed. Both of the spectra are normalized by the sum intensity of the atomic lines of Ar and Xe at 696.5 ÷ 1100 nm.

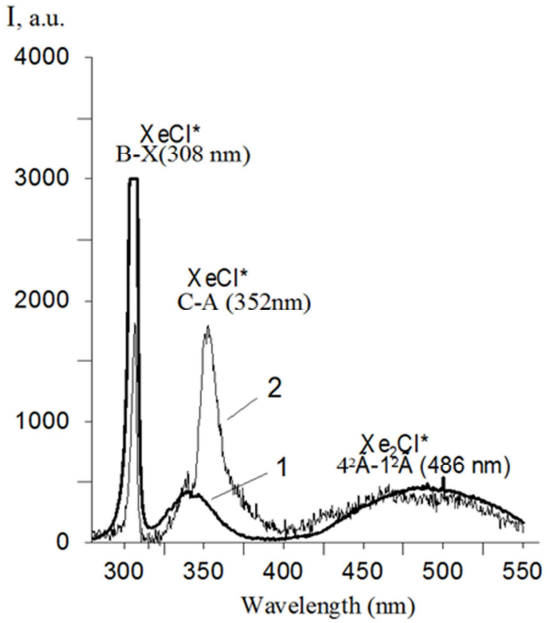

Figure 2. *The fragments of the emission spectra of the Ar-Xe-CCl_4 gas mixture (Ar-760 Torr, Xe-40 Torr, CCl4- 0.15 Torr) pumped by the products of 235U(n,f) nuclear reaction (the spectra has not been corrected for the MAYA-2000Pro spectrometer response). 1-without super-radiance. The optical resonator has a quartz output window and a back mirror with 80% transmission at 352 nm (and 98% reflection at 308 nm). The sum transmission at 352 nm is 175%. 2-super-radiance. The optical resonator has 10.8% reflection of output and 62.1% reflection of back spherical mirrors at 352 nm (and 98÷98.9% reflection at 308 nm for both mirrors).*

The super-radiance occurs in the resonator with low losses (T_{sum}=127,1%) and leads to the increase of the C-A band intensity (figure 2). The observed difference in the intensity of the B-X bands at 308 nm is associated with the large transmission of the quartz window (~ 95%) and the low transmission of the output mirror (~0.4%) at 308 nm.

5. Conclusion

Experimental researches have shown that the Ar-Xe-CCl_4

gas mixtures with low content of CCl_4 have a high efficiency of formation of excimer molecules XeCl (B)* and XeCl (C)* when excited by charge particles of high energy. Such media have high gain for B-X and C-A transitions of XeCl*. Experiments performed when pumping with fission fragments of ^{235}U detected the occurrence of the super-radiance emission on the C-A transition with the duration of 90 microseconds. Super-radiance disappeared if the laser output mirror was replaced by a quartz window and had a very sharp increase and a low neutron threshold when we used a tuning resonator at 352 nm wavelength with 10.8% reflection of output and 62.1% reflection of back spherical mirrors.

In the future, the performance of the XeCl (C)* excimer laser with nuclear pumping can be significantly improved by optimizing the design of the optical resonator, the laser tube and selecting a more optimal composition of the gas mixture.

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