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Lasing on the $5s^2\,^2D_{5/2} \rightarrow 5p\,^2P_{3/2}$ Cd II ion transition upon sputtering of metal cadmium by nuclear particles

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***Abstract*. A basically new method for direct conversion of the energy of nuclear reactions into laser radiation is considered. The method is based on the use of the emission of excited ions (or atoms) produced by bombarding a solid by charged particles produced in nuclear reactions. The results of an experimental study of the emission of excited Cd II ions and Cd I atoms are presented for metal cadmium irradiated by α particles from ^{238}Pu. When cadmium was heated to a temperature $T > 160\,^{\circ}$C (at least $0.5T_m$, where T_m is the melting temperature), the emission of Cd II ions, which are predominantly in the $4d^9 5s^2\,^2D_{5/2,3/2}$ states, and resonantly excited Cd I atoms was observed.**

Keywords: *nuclear pumping, ion emission, stimulated emission.*

Mixtures of light and heavy inert gases and mixtures of helium with metal vapours (Cd, Zn, Hg) and molecular gases (N_2, H_2) are used as laser media excited by products of the ^{235}U (n, f), ^{3}He (n, p) ^{3}T, ^{10}B (n, α) ^{7}Li nuclear reactions [1 – 3]. The energy of nuclear reactions is converted in these media into laser radiation in a multistage process, which includes the moderation of charged particles (products of nuclear reactions) in a buffer gas with the formation of ions and metastable atoms of this gas, which undergo transitions to excited states in three-body collisions. A further energy transfer to active particles contained in the buffer gas at a low concentration proceeds through collisions involving the excited states of the buffer gas in the course of numerous plasmochemical reactions (their number may amount to several hundreds) [4].

To ensure an efficient operation of a nuclear-pumped laser, the nuclear particles should be fully moderated in the active medium. High kinetic energies of particles (~ 0.1 – 100 MeV) impose certain requirements on the cross-sectional size of the active element and pressure of the buffer gas inside it. In particular, the relation $pd \geqslant 1$ should be fulfilled (p is the buffer-gas pressure in atmospheres, and d is the diameter of the laser tube in centimetres). Most of nuclear-pumped lasers are characterised by the values $p \sim 1$ atm and $d \sim 3$ cm.

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The main drawbacks inherent in this pumping scheme are as follows: a low conversion efficiency of the energy of nuclear reactions caused by a nonselective population of the working levels of the active gas, the appearance of a thermal lens in the laser active medium, and a significant collision broadening of the laser-transition line. Certain difficulties arise in the reactor – laser system [5] using the ^{235}U(n, f) nuclear reaction: a small amount of ^{235}U that may be contained in the laser tube causes a significant contamination of the active gas medium by uranium fission fragments.

Below, we describe a new method for direct conversion of the energy of nuclear reactions into laser radiation. The method uses the emission of excited ions (or atoms) upon the bombardment of a metal foil by charged particles produced in nuclear reactions ([6], p. 590). In principle, the laser operation does not require a buffer gas, because nuclear particles are slowed down and lose all their kinetic energy in a metal layer $\sim 10^{-3}$ cm thick, sputter this layer, and, avoiding all the intermediate stages, directly form excited ions (or atoms) in the states that are the upper levels of laser transitions. For example, when a cadmium film is bombarded by fast electrons ($E_e \sim 150$ keV), α particles ($E_\alpha \sim 5.3$ MeV), or products of the ^{3}He (n, p) ^{3}T nuclear reaction ($E_p = 0.57$ MeV, $E_T = 0.19$ MeV), excited Cd^+ ions ($5s^2\,^2D_{5/2,3/2}$) are immediately produced and make it possible to obtain lasing at three wavelengths of 441.6, 325, and 353.6 nm [7, 8]. To prevent the mirrors and windows of the laser tube from making dusty, the laser active element must contain a certain amount of a buffer gas. Such an ion-emission nuclear-pumped laser can operate within a range of buffer-gas pressures satisfying the condition $0 < pd \leqslant 1$.

The method proposed for the direct nuclear-to-light energy conversion makes it possible to increase the uranium content in the laser active element by a factor of 2 – 4 (or to decrease correspondingly its cross-sectional size), to reduce the buffer-gas pressure by a factor of ten and more, to increase the lasing efficiency by a factor of 5 – 7 due to the single-stage character of the energy conversion and a selective population of the laser level, and to reduce the gas component of the fragment radiation activity of the laser. The ion-emission laser efficiency is higher than that of conventional-type nuclear-pumped lasers but is lower than the efficiency of excimer lasers.

Attempts to use high-energy electron beams for obtaining vapours of refractory metals (Mo and W) were made earlier [9]. Using the cathode sputtering of these metals by electrons in an electric discharge at a room temperature, the concentration of Cu atoms sufficient for the operation of a

copper vapour laser was obtained [10]. A laser action on transitions of ionised Cd, Zn, Cu, and Ag atoms in tubes with a hollow cathode without external heating was reported in papers [11, 12].

Substances are sputtered under their bombardment by ions and nuclear particles. The sputtering is accompanied by the emission of photons, because a certain number of the escaping ions and neutral atoms are in excited states [13]. When high-energy products of the $^3He(n,p)^3T$ nuclear reaction were used in [7, 14, 15], a high-intensity emission was observed during the sputtering of Cd, Zn, Pb, Ag, Bi, and Cs films deposited on silica substrates. The intensity of this emission significantly increased, as the substrates were heated up to 70 °C (for Cs atoms), 140 – 240 °C (for Zn, Pb, Bi, Cd), and 560 °C (for Ag). A similar effect was observed in [8] under the bombardment of a Cd foil heated up to 200 – 240 °C by α particles.

We performed spectroscopic studies of the emission of excited ions upon the bombardment of metal cadmium by the 5.3-MeV α particles and analysed the application of this method of nuclear-to-light energy conversion for obtaining a stimulated emission on the $5s^2\,^2D_{5/2} \rightarrow 5p\,^2P_{3/2}$ transition of Cd II ions ($\lambda = 441.6$ nm). In practice, it is preferable to use thick metal foils instead of thin films deposited on quartz substrates. The spectroscopic investigations were carried out using the setup described in [16]. A metal Cd sample in the form of a disk 40 mm in diameter and 1 mm thick was mounted on a flat heater and placed inside an evacuated chamber facing an α source (^{238}Pu) with an activity of $\sim 10^7$ Bq. The chamber had a quartz window that ensured the detection of light with an MDR-23 monochromator using a FEU-100 photomultiplier operating in the photon counting mode. The temperature of Cd was varied from 20 to 320 °C and was monitored with a chromel – copel thermocouple. The chamber was evacuated to a high vacuum and filled with pure helium up to a pressure of a few to 2280 Torr.

The lasing experiments were carried out on a VIR-2M pulsed nuclear reactor, where the products of the $^3He(n,p)^3T$ nuclear reaction were used for laser pumping, and using a specially designed laser pumped by a high-current electron beam with a pulse duration of ~ 3 ns and an energy of ~ 150 keV (the electron beam current density was 500 A cm^{-2}).

A typical luminescence spectrum observed upon the bombardment of a Cd foil by α particles in a He buffer-gas atmosphere is shown in Fig. 1. The spectrum contains Batler lines of Cd II ions at 441.6, 325.0, and 353.6 nm and lines of cascade transitions at 214.4 and 226 nm related to them. Among the atomic lines, only the Cd I resonance lines at 326.1 and 228.8 nm are present. As to other ionic lines, only the transitions from the Cd II $6s^2S_{1/2}$ level (at 274.9 and 257.3 nm) are observed. Despite the large lifetime of the upper level (~ 830 and 300 ns for the lines at 441.6 and 325 nm, respectively [17]), the Batler lines have the maximum brightness in the spectrum. The energy level diagram of a Cd atom and ion is presented in Fig. 2 (the observed transitions are denoted by the arrows).

The sputtering mechanism of populating Cd II ion levels ensures a higher efficiency of the nuclear-to-light energy conversion compared to that in vapour – gas media. As an example, Fig. 3 shows the temperature dependence of the Batler line intensity of Cd ions ($\lambda = 441.6$ nm). The data were obtained by irradiating a quartz cell filled with a

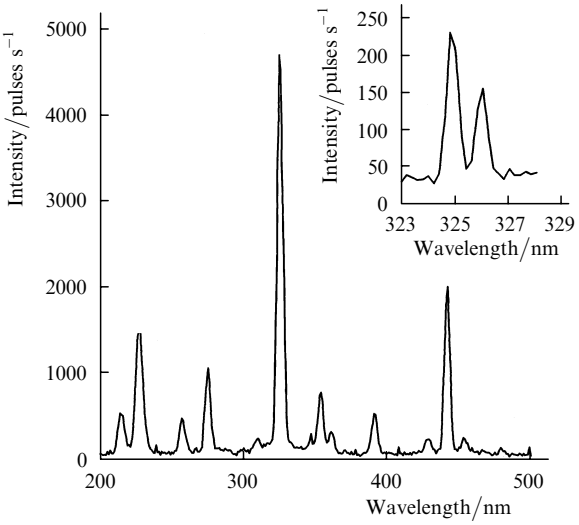

Figure 1. Luminescence spectrum measured with a resolution of 2.8 nm upon bombardment of a Cd foil by 5.3-MeV α particles. The inset shows a part of the spectrum near 325 nm measured with a resolution of 0.39 nm. The foil temperature is 240 °C, and the helium pressure is 600 Torr. The spectrum was recorded with an MDR-23 monochromator with a 1220 lines mm^{-1} diffraction grating and a FEU-100 photomultiplier without filters.

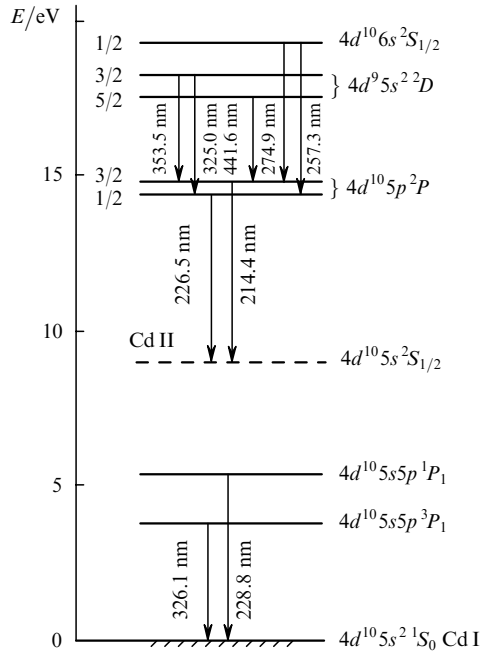

Figure 2. Energy level diagram of a Cd I atom and a Cd II ion. The arrows denote the transitions observed upon sputtering of the Cd foil by α particles.

^3He – Cd mixture (at a ^3He pressure of 650 Torr) placed in the core of the IRT-MIFI reactor. A metal Cd film was deposited on the side surface of the cell by sputtering products of the $^3He(n,p)^3T$ nuclear reaction. When the temperature of the cell walls exceeded the Cd melting temperature ($T_m = 320.9$ °C), the film was destroyed and an ordinary vapour – gas mixture was formed, which was then excited using a conventional mechanism involving atoms and molecules of the buffer gas. One can see that

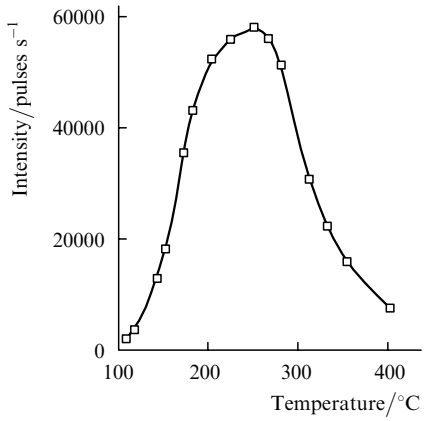

Figure 3. Intensity of the 441.6-nm Batler line of Cd II ions versus the Cd temperature. The Cd film was sputtered by products of the ^{3}He (n, p) ^{3}T nuclear reaction in an atmosphere of ^{3}He at a pressure of 650 Torr.

at the operating temperatures ($\sim 320 - 360\,^\circ$C), which are typical for nuclear-pumped Cd vapour lasers, the efficiency of populating the upper laser level is 2.5 – 5.0 times lower than that achieved by sputtering a metal film.

The luminescence intensity arising upon the Cd sputtering by nuclear particles depends on the temperature of the Cd foil surface. The intensity of all the lines drastically increased with increasing temperature but the shape of the spectrum did not change, and the ratio between the intensities of individual lines remained constant. Fig. 4 shows the increase in the intensities of the 441.6-nm line and two unresolved lines at 325 and 326.1 nm with temperature. An exponential increase in the luminescence intensity begins above 150 – 160 °C and is well approximated by the expression

$$I \sim \exp(-A/kT), \qquad (1)$$

where A is the activation energy of this process, $k = 8.6153 \times 10^{-5}$ eV K^{-1} is the Boltzmann constant, and T is the absolute temperature of the Cd foil. For the unresolved lines at 325 and 326.1 nm, the activation energy is $A =$ 0.85 eV, which corresponds to 85.51 kJ mol^{-1} per 1 mole of Cd. For the 441.6-nm line, we have $A = 0.749$ eV or 75.33 kJ mol^{-1}. These data agree well (with allowance for the measurement error) with the activation energy of the self-diffusion coefficient for metal Cd equal to 76 – 80 kJ mol^{-1} ([6], p. 383). Therefore, we can assume that changes in the intensity of Cd II ion lines upon sputtering are caused by an increase in the self-diffusion rate of Cd atoms in the cadmium foil.

Using a movable diaphragm placed inside the chamber and making it possible to change the boundaries of the light-collection region, we measured the spatial distributions of Cd-foil sputtering products (excited ions) emitting at 325, 441.6, and 274.9 nm depending on the distance to the surface of the Cd disk for various pressures of helium in the chamber. Despite great differences (by a factor of almost 300) in the luminescence lifetimes of the levels corresponding to the wavelengths mentioned above, all the three distributions were close to each other (Fig. 5). Because the 274.9-nm line (the $6s^2S_{1/2} \rightarrow 5p^2P_{3/2}$ transition in Cd II) is not related to long-lived Batler levels by cascade transitions, these experimental facts can be explained only by assuming that there exists a two-step ion-emission mechanism: the ejection of microscopic metal droplets from the foil bombarded by nuclear particles and the subsequent self-diffusion-assisted emission of excited Cd II ions in the $5s^2{}^2D$ ($\lambda = 325$ and 441.6 nm) and $6s^2S_{1/2}$ ($\lambda = 274.9$ and 257.3 nm) states, resonantly excited atoms, and neutral Cd atoms from flying droplets.

We estimated the size of microscopic droplets by determining the mass of the metal sputtered by α particles for various energies of α particles. The Cd foil was weighed

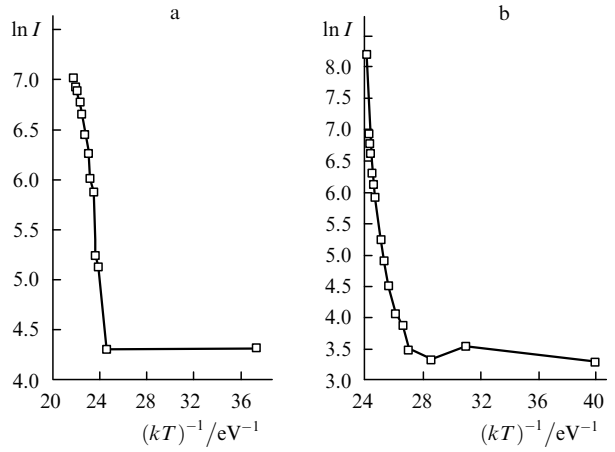

Figure 4. Intensities I of (a) two unresolved Cd II lines at 325 and 326.1 nm and (b) the Cd II line at 441.6 nm versus $(kT)^{-1}$ upon sputtering of the Cd foil by 5.3-MeV α particles at a helium pressure of 54 Torr.

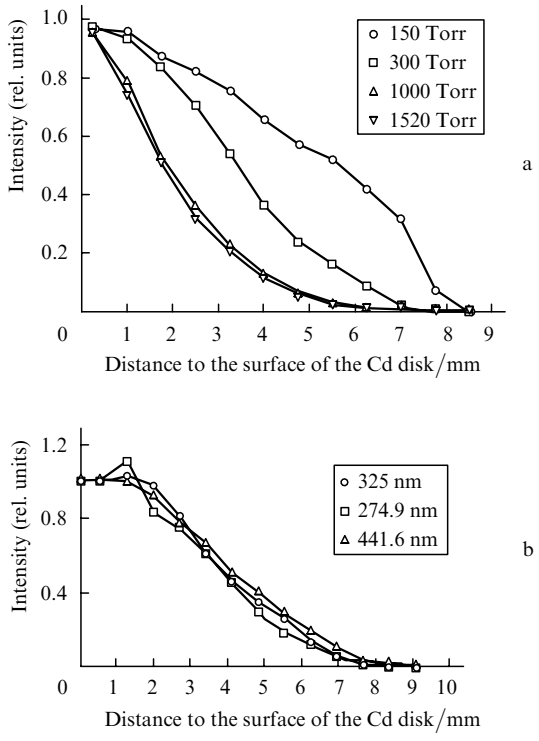

Figure 5. Spatial distributions of the luminescence intensity at a wavelength of 325 nm (a) for various pressures of helium in the chamber with a Cd foil sputtered at $T = 240\,^\circ$C and (b) for separate spectral lines at a constant helium pressure of 300 Torr and $T = 240\,^\circ$C.

on an analytical balance before and after its irradiation in the chamber filled with helium at various pressures. The irradiation by α particles was performed at a constant temperature of 240 °C for a fixed time. Because the flux and energy of α particles incident on the target are known, we could find the metal mass knocked out by one α particle under such conditions. The metal loss caused by the thermal evaporation was taken into account experimentally by weighing the sample before and after the exposure in the chamber for a fixed time in the absence of a source of α particles in it. We found that, at a substrate temperature of 240 °C, a single α particle with an energy of ~ 5 MeV produced a microdroplet with a mass of 10^{-14} g containing $\sim 10^8$ Cd atoms. Note for comparison that, at room temperature, the sputtering ratio for metal Cd measured in the evacuated chamber for a two-month exposure was as low as 4×10^{-17} g per α particle.

The study of microphotographs of the irradiated Cd foil surface has shown that the metal is ejected from deep layers of the foil. For an individual α particle, the ejection region has a shape close to a cylinder with a diameter of ~ 1 μm and a length of the order of the α-particle transit in the substance. Thus, depending on the initial energy of α particles, microdroplets of different dimensions and temperatures are formed. Both these factors influence the self-diffusion rate of Cd atoms and their yield from the metal.

The fact that long-lived Batler Cd^+ ions in the $5s^2\,^2D_{5/2,3/2}$ states and excited Cd atoms in the $4d^{10}\,5s5p$ state are mainly emitted upon the foil sputtering by nuclear particles can be explained using the concepts of a thermal droplet model, self-diffusion, and decay of autoionisation states of excited Cd atoms.

When an α particle is moderated in Cd, it interacts with atoms of the crystal lattice, knocks out some of them to interstitial sites, and forms the so-called temperature wedge [18] within which the entire kinetic energy of this particle is contained in the form of vibrations of the lattice and displaced atoms. The temperature within this wedge increases considerably and may exceed the melting temperature of the metal. According to [18], in this case, the metal structure differs from an amorphous structure of a real liquid and it is actually an overheated solid. A microdroplet of the heated metal that contains Cd atoms displaced from lattice sites is ejected due to thermal stresses in the foil. The microdroplet size and the velocity of its ejection depend on the α-particle energy, foil temperature, and the angle of incidence of the α particle onto the metal surface.

The displaced Cd atoms diffuse inside the droplet in search for vacancies. The self-diffusion coefficient for Cd at $T \sim 160 - 200$ °C increases by many orders of magnitude and promotes an accelerated emission of the displaced atoms out of the metal. Obviously, this process strongly depends on the droplet size. It is precisely this factor whose action can account for an experimental increase in the intensity of Batler lines observed with increasing the buffer-gas pressure, because the depth of penetration of α particles into the metal decreases due to a reduction in their energy in the gas and the formation of smaller droplets.

A specific shape of the luminescence spectrum observed upon metal sputtering by high-energy particles is associated with the predominant excitation of electrons of the inner shells of metal atoms. When Cd I atoms with the filled $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 5s^2\,^1S_0$ electronic shells are sputtered, the displaced atoms, which have one excited 4*d*-($4d^9 5s^2 5p$) or 5*s*-electron ($4d^{10} 5s5p$), can be formed in the region of the temperature wedge inside the metal. When such atoms are ejected (due to self-diffusion) to vacuum, atoms in the autoionisation state and resonantly excited atoms are produced. Their decay occurs in the transitions

$$Cd^*(4d^9 5s^2 5p) \rightarrow Cd^+(4d^9 5s^2) + e + \Delta E, \quad (2)$$

$$Cd^*(4d^{10} 5s5p) \rightarrow Cd(4d^{10} 5s^2) + h\nu. \quad (3)$$

Reaction (2) leads to the formation of Batler ions and emission of lines at 325, 353.6, and 441.6 nm by them. Reaction (3) leads to the emission of Cd I resonance lines at 228.8 and 326.1 nm.

The efficiency of populating the Batler Cd II levels upon the metal-foil sputtering was evaluated by comparing the Cd spectra with the luminescence spectra of the He–Ne–Ar mixture (the pressures of He, Ne, and Ar were 760, 15, and 7.5 Torr, respectively) and the Ar–Xe–CCl_4 mixture (the pressures of Ar, Xe, and CCl_4 were 760, 76, and 3.8 Torr, respectively). The compositions and pressures of gas mixtures corresponded to the compositions and pressures of active media of nuclear-pumped lasers described in [19, 20]. Comparison of the intensities of the 585.2-nm line (the $2p_1 \rightarrow 1s_2$ transition in Ne I atoms), the 308-nm line (the $B \rightarrow X$ transition in $XeCl^*$ molecules), and 441.6-nm line (the $5s^2\,^2D_{5/2} \rightarrow 5p^2P_{3/2}$ transition in Cd II ions at a foil temperature of 240 °C) measured under identical conditions has shown that the ratio of the upper-level populations of the these transitions in Ne, Cd, and $XeCl^*$ is 0.011 : 1 : 18.2.

These experimental data allow us to estimate the threshold characteristics of the nuclear-pumped Cd laser utilising the sputtering of a metal film by products of nuclear reactions and operating on the $5s^2\,^2D_{5/2} \rightarrow 5p^2P_{3/2}$ transition in Cd II ions. The lasing threshold of the Cd laser can be determined using the technique described in [16], by comparing it to the lasing threshold of a nuclear-pumped ^{3}He–Ne–Ar laser [19] known from reactor experiments. Its value for thermal neutrons is $\sim 10^{15}$ neutrons cm^{-2} s^{-1}. Because the energy stored due to the inversion at the $5s^2\,^2D_{5/2}$ level of Cd II is almost 100 times higher than the energy stored at the $2p_1$ level of Ne I and is exceeded only by the energy stored at the *B* level of $XeCl^*$ excimer molecules, high energy characteristics of such a Cd laser can be expected in the *Q*-switched or amplification modes.

The possibility of obtaining stimulated radiation upon the sputtering of a metal Cd film (by products of the ^{3}He(n, p)^{3}T nuclear reaction) and a metal foil (by an electron beam) was demonstrated in experiments on the VIR-2M pulsed nuclear reactor and an Arina pulsed electron accelerator, respectively.

The laser active element with a Cd film was made of a quartz tube 34 mm in diameter and 800 mm in length with two internal mirrors. Multilayer dielectric spherical mirrors with a radius of curvature of 10 m and a reflectivity of 99.8 % at 441.6 nm were used. The mirrors had an aperture of 22 mm, which was determined by the design of adjustment heads. A thin Cd layer was deposited on the inner surface of a part of the quartz tube 600 mm long. The laser active element was filled with ^{3}He up to a pressure of 1.1 atm and was placed close to the VIR-2M reactor core in a water moderator of neutrons inside an electrical heater. The water moderator was 50 cm long and had 5-cm-thick walls. The electrical heater was equipped with measuring

thermocouples and an electronic regulator, which ensured the maintenance of a constant temperature along the entire length of the Cd film. The fluence of thermal neutrons at the axis of the laser active element was 8×10^{12} neutrons cm^{-2}, corresponding to the maximum flux density of pump thermal neutrons $\Phi = 2.8 \times 10^{15}$ neutrons cm^{-2}s^{-1}. The experiments were performed simultaneously with two active elements operating at temperatures of 162 and 206 °C.

Quasi-cw lasing at 441.6 nm was obtained with each active element. The output power was 2.83 and 2.0 mW for the lasers operating at 162 and 206 °C with pulse durations of 2.04 and 1.88 ms, respectively (Fig. 6). As the fluence of pump thermal neutrons increased by a factor of five (up to 4×10^{13} neutrons cm^{-2}), the output power (pulse duration) increased to 3.01 mW (to 3.01 ms at 162 °C) and 2.99 mW (to 2.15 ms at 206 °C). However, the laser pulse shape was modified to a certain degree in this case (Fig. 7). In all cases, the lasing threshold measured was within $(1.5 - 6.0) \times 10^{15}$ neutrons cm^{-2}s^{-1}.

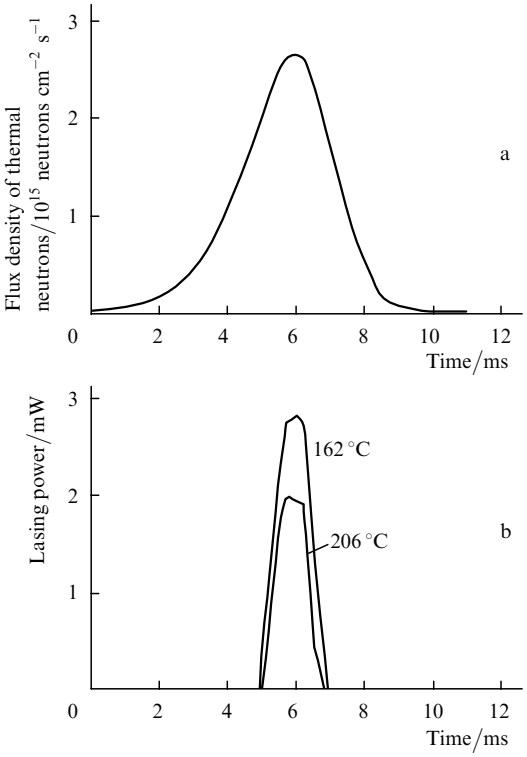

Figure 6. Oscillograms of the (a) pump neutron pulse from the VIR-2M reactor and (b) laser pulses at 441.6 nm upon sputtering of the Cd film by products of the ^{3}He(n, p)^{3}T nuclear reaction for various temperatures of the laser cell, a ^{3}He pressure of 825 Torr, and a maximum flux density of pump thermal neutrons of 2.83 × 1015 neutrons cm^{-2} s^{-1}.

The active element of the electron-beam-pumped laser was made of stainless steel and represented a cylindrical heater 70 mm in diameter with a window (20 × 40 mm) on the side surface for injecting the electron beam. A metal Cd foil 1 mm thick was mounted on the inner surface of the heater along its entire length. The heater was equipped with a thermocouple and an electronic regulator that allowed us to maintain a constant temperature of the foil. The active element was located inside a vacuum chamber at a distance of 2 cm from an IMA3-150E tube, which is a pulsed injector of an electron beam with an energy of ~ 150 keV, current

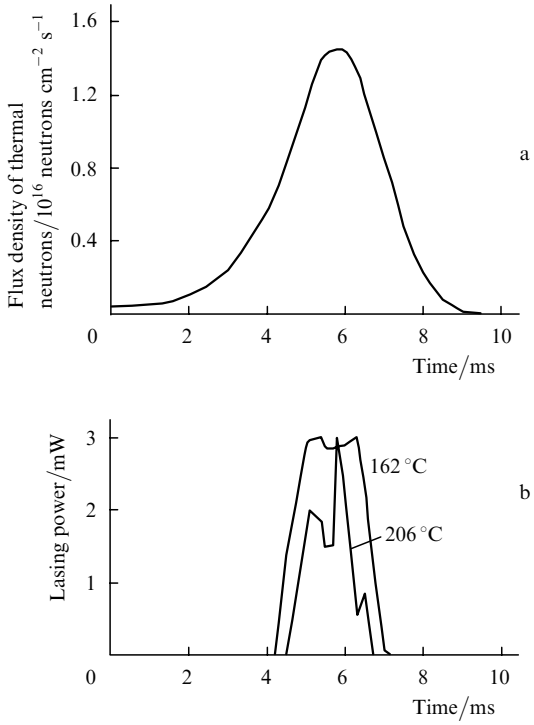

Figure 7. Oscillograms of the (a) pump neutron pulse from the VIR-2M reactor and (b) laser pulses at 441.6 nm upon sputtering of the Cd film by products of the ^{3}He(n, p)^{3}T nuclear reaction for various temperatures of the laser cell, a ^{3}He pressure of 825 Torr, and a maximum flux density of pump thermal neutrons of 1.46×10^{16} neutrons cm^{-2}s^{-1}.

density of ~ 500 A cm^{-2}, and pulse duration of 3 – 5 ns. The laser cavity was formed by two spherical mirrors 20 mm in diameter with a radius of curvature of 2 m mounted at a distance of 120 mm from each other. Their reflectivity at 441.6 nm was 99.75 %. After being thoroughly pumped out, the vacuum chamber was filled with helium, which was then continuously purified by a special gas-purification system [21] during the whole operating time. Lasing at 441.6 nm was observed at a temperature of the Cd foil of 240 °C and helium pressures of 10 to 2280 Torr. In practice, the lasing occurred near the threshold, and the emission could be observed through the exit mirror only visually in the form of a blue sickle disappearing upon misalignment of one of the cavity mirrors. The length of the laser active medium over which light was amplified was at most 4 cm. For the highly reflecting mirrors in use (reflectivities of 99.75 %), this corresponded to a medium gain of 6.25×10^{-4} cm^{-1}.

Note in conclusion that in this paper an essentially new method for direct conversion of the energy of nuclear reactions into laser radiation has been considered, which is based on the use of the emission of excited ions (or atoms) produced upon the bombardment of a solid heated to a temperature no lower that $0.5T_m$ by nuclear particles. The operation of such a laser does not require a buffer gas, because nuclear particles are decelerated in the solid within a layer ~ 10^{-3} cm thick, sputter this layer, and directly produce (avoiding all intermediate stages) excited ions (or atoms) in the states representing the upper levels of laser transitions. This method for nuclear energy conversion makes it possible to utilise low-volatile substances (e.g., metal uranium or its compounds) as an active medium of a nuclear-pumped laser. Preliminary estimates show that a

laser on transitions of ionised uranium would be able to operate in the regime of self-sputtering by fission fragments at temperatures of $\sim 600 - 700\,^{\circ}\mathrm{C}$.

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