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LASER PROCESSES AND ACTIVE MATERIALS FOR SUSTAINABLE ENERGY PRODUCTION

V. LUPEI

National Institute of Lasers, Plasma and Radiation Physics Magurele-Bucharest, Romania
E-mail: lupei_voicu@yahoo.com

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Abstract. The paper discusses several laser materials and processes that could be of interest for sustainable energy production, in two main directions, inertial nuclear fusion and the solar-magnesium-hydrogen cycle. It is inferred that the transparent ceramics with large size, suitable spectroscopic and thermo-mechanical properties and composition could offer a large basis for selection of such laser materials.

Key words: inertial nuclear fusion, laser materials, laser emission, transparent ceramics.

1. INTRODUCTION

The development of human society and the increase of global population put great pressure on the energy supply, in all its forms (centralized, local, mobile). The actual methods of production and transportation electricity are based mostly on the use of fossil fuels, with two major consequences, the exhaustion of the fuel reserves and severe environmental threat, including the global warming. Production of electricity based on nuclear fission will be limited to less than 10% of demand, either because of limited supply of fuel and by the need to replace the old reactors or because the accumulation of large amounts of radioactive wastes. Identification of new energy resources is thus one of the most important tasks of the scientific research and development. A major requirement for these new energy sources would be the sustainability. Despite of the great effort and important progress in identification and development of new processes and renewable resources for sustainable production of electricity or for development of non-conventional sources (biofuels, photovoltaic, wind, hydro, geothermal, tidal and so on), their contribution to the actual energy production is limited. Moreover, the foreseen development of these energy sources does not offer confidence that they

will be able to solve the increasing demand for energy when the traditional fuel reserves will be exhausted or their use limited because of pollution, a situation foreseen to install in the fourth-fifth decades of the 21st century.

Most actual resources for energy production are based on the stored or real-time supply of energy by the Sun. The role of the Sun as the main supplier of primary energy suggests two main directions of research: energy production by similar processes as in Sun and a more complete use of the solar energy, particularly the transformation of its intermittent energy in a new form of storable energy resource. In both these directions the lasers could play an important role.

Generaton of energy in the Sun is based mainly on the nuclear fusion of plasma of the Hydrogen isotopes Deuterium and Tritium to produce Helium, neutrons and energy. The fusion requires short distance between the fusing nuclei, so that the nuclear force overcomes the Coulomb repulsion (confinement), followed by ignition, i. e. initiation and self-propagation of reaction in the whole body of fuel. The oceans on Earth contain practically inexhaustible amounts of Deuterium, whereas Tritium can be produced in the fusion plant by reacting part of the neutrons released in the fusion with Lithium. Reproduction on Earth of the conditions for confinement is a very difficult task and two main directions of action are envisaged: magnetic confinement of plasma of Deuterium and Tritium in a Tokamak-type installation and inertial confinement of D-T pellets under rocket-jet ablation by direct intense nanosecond laser or laser-produced X-ray irradiation. The energetic neutrons produced by the fusion irradiate a blanket surrounding the reactor and the generated heat can be further used for generation of electricity. There are two main approaches of the inertial fusion: the central fusion, when the pellet is compressed by a MJ nanosecond laser to the level able to initiate and sustain the nuclear reaction and fast ignition, with less compression but with a second, short-pulse (picosecond) hundred kJ laser to initiate the ignition of the compressed fuel. It is estimated that a inertial nuclear fusion electric plant with continuous operation would require 10-15 Hz laser repetition rates. Both these methods of confinement (magnetic and inertial) require high energy consumption so a major condition for production of energy would be an energy multiplication factor large enough to exceed by far all consumption of energy involved in the process as well as in the production of all parts and consumables. The scientific and technical solution of the problems involved in the controlled nuclear fusion requires dedicated long-term research and intensified present-day effort.

The second direction of using the lasers in production of energy is transformation of the intermittent low-density solar energy in concentrated energy sources that could be used in the energy production cycle. Of particular interest would be the utilization of the solar pumped lasers for production of transportable resources that can be used for further production of energy where and when needed, such as in the solar-magnesium-hydrogen cycle. The metallic magnesium can react with water at ~70^{o}C to produce hydrogen, MgO and hot steam. The

hydrogen produced by this reaction can be used for powering cars, eventually by the electricity produced in fuel cells. MgO oxide can be subsequently thermally decomposed to metallic Mg and oxygen at temperatures ~ 4,000 K and it was found that such condition can be realized by irradiation with kW lasers. If scientific and technical solution for production of kW laser power under solar pumping can be found, the only consumable in this cycle would be the solar radiation: the crucial point would be the identification of laser materials able to absorb as much as possible from the solar radiation and transform it efficiently into laser radiation.

In both these directions of energy production using lasers, the main actual problem is the absence of reliable laser solutions. Strong development effort is underway and many solutions are investigated: identification of new laser materials and solutions for enhancement of efficiency and for scaling of power or energy as well as of the repetition rate can be crucial. The paper discusses the state of the art of this development, including the results of the authors' own research, as well as some new directions of action.

2. LASER SYSTEMS FOR INERTIAL NUCLEAR FUSION

2.1. THE NUCLEAR D-T FUSION REACTION

The most efficient nuclear fusion reaction takes place between the nuclei of D and T, leading to formation of the intermediate nucleus ^{5}He that decays rapidly to ^{4}He by ejection of a neutron, $^2D + {}^3T \rightarrow {}^5He \rightarrow {}^4He + n$. The ~17.6 MeV energy released in reaction is shared by the ^{4}He nucleus, in fact a α-particle (3.5 MeV), and the neutron (14.1 MeV). An essential condition for reaction is a very short distance between the reacting nuclei, such as the nuclear forces would overcome the ~ 0.01 MeV electrostatic repulsion barrier, much smaller than the released energy. The condition for reaction is established by the Lawson criterion [1] that relates the density of interacting nuclei to the time of confinement: the D-T plasma must be confined to densities hundred times the density of lead to grant confinement times of tens of picoseconds. It was estimated that such densities can be obtained by the action of short-time high-energy particle or photon beams on the surface of a capsule containing a D-T mixture. A symmetric configuration of such beams ablates the surface of capsule causing the material to implode inwards at high speed. The compression produced by this inward movement together with the convergent shock-waves creates the necessary density as well as temperatures in excess of hundred millions of Kelvin degrees. The convergence of the shock waves requires highly symmetric confinement, and ignition in such systems could be possible only when a large fraction of the fuel is confined. The neutrons released in fusion can be used for heating a mantle surrounding the reaction chamber and are thus the main mechanism by which the energy released in fusion can be used for

production of electricity. By contrary, the ^{4}He nuclei loose the energy inside the reaction chamber, leading to further heating of the fuel and propagation of reaction in the whole volume of fuel, i.e. to ignition that consumes the largest part of fuel.

2.2. EARLY EXPERIMENTS

Idea of using lasers for nuclear fusion arouse in the 1960 decade, but in fact, the fusion experiments began only in the early 1970s when stronger lasers become available; most of these experiments used Nd:glass lasers. The D-T fusion reaction under laser irradiation was demonstrated by various groups of research; however, despite of constant progress, no break-even point, i.e. the point when the released energy overcomes the spent laser energy was demonstrated. These experiments, together with the modeling of the fusion process, evidenced the enormous complexity of laser-driven nuclear fusion [2]. It became evident that compression and ignition of a D-T capsule would require MJ nanosecond laser beams, either by direct action on its surface, or indirectly, irradiating the inner walls of a high-Z hohlraum containing the fuel capsule, in order to create a bath of X-rays that act on the fuel surface. The direct irradiation could be more effective in transferring the laser energy to the fuel, but due to the Rayleigh-Taylor instabilities the creation of a symmetric confinement and convergence of the shock waves necessary to grant ignition is very difficult. By contrary, in case of indirect action with creation of X-rays, the confinement can be central and symmetrical and, at high enough energy, it can grant the conditions of ignition. Under these circumstances there are two basic variants of a fusion equipment: (i) the central confinement and ignition under the action of a unique laser; (ii) fast ignition [3, 4] in which a high energy laser confines part of the fuel and a second, fast laser ignites the confined part and grants condition for propagation of the reaction in the rest of the fuel.

In the central confinement the global MJ energy must be delivered in a given spatial distribution, so it can be in fact obtained from an ensemble (hundreds) of individual lasers of much lower energy (order of 10 kJ). In the fast ignition the confined part of the fuel is much smaller than in case of central confinement and it must not be necessarily centered; in this case the direct irradiation of the fuel capsule can be safely used and the energy of the confinement laser could be about an order of magnitude less than in the central confinement. Since the confined part remains in this state for several tens of picoseconds, the duration of the ignition pulse must be of the order of picoseconds, but the peak power must be high, of the order of Petawatt (the energy of tens of kJ). The necessary repetition rate to grant continuous energy generation would be 10-15 Hz. In case of central confinement the multiplication ratio i.e. the ratio between the released energy to the energy spent over the whole cycle for production of the laser energy and of the fuel is of the order of 350, whereas in case of central ignition this would be reduced to ~100.

2.3. LASER SOLUTIONS

The requirements for the laser radiation for nuclear fusion impose very careful selection of the laser solutions (laser schemes, laser materials, laser design). The lasers used in these experiments are in fact complex systems with amplification: the radiation of a high quality low-energy laser is amplified in large systems to the desired level. Due to their efficiency, compactness, reliability, controllable regime (wavelength, dynamics of emission), the solid-state lasers based on transparent solids activated with laser active ions [5] are actually the solution for the inertial fusion. The nanosecond lasers for confinement would operate in Q-switch regime, whereas those for fast ignition would use mode-locking. The high energy can be then obtained by multi-pass (regenerative) amplification, although Chirped Pulse Amplification (CPA) could be envisaged for the PW lasers for ignition.

The high-energy Q-switch emission or amplification requires the storage of inversion till the moment of restoring the Q quality factor (oscillators) or to the moment of injecting the pulse to be amplified (amplifiers). In case of CW pumping with rate W_p the population of the emitting level in presence of luminescence with the effective lifetime $\tau_{eff} = \tau_{rad}\eta_{qe}$, which accounts for the reduction of the emission quantum efficiency η_{qe} by non-radiative processes, is $n_{em} = \tau_{eff} W_p n_0$. The pumping of solid-state lasers is made traditionally in energy levels with suitable absorption properties placed above the emitting level and from which the excitation decays to the emitting level with the efficiency η_p; usually $\eta_p \approx 1$. The pump rate relates to the incident CW power P_i by $W_p = \frac{P_i}{V}\eta_a\eta_p$, V is the pumped volume and η_a the pump absorption efficiency. Thus, a high population in the emitting level will require high incident pump power and high absorption efficiency. The power of the CW pump sources is limited, so the stored inversion is low an thus Q-switching of the CW pumped lasers could provide pulses of low energy. High pump intensities can be obtained by pulse pumping: for a square pulse of duration t_p the density of ions pumped to the emitting level is $n_0 W_p (t_p / \tau_{eff})$, but due to luminescence and non-radiative de-excitation the maximum population in the emitting level is reached at the end of pump pulse,

$$n_e(t_p) = n_0 W_p \tau_{eff} \left[1 - \exp\left(-t_p / \tau_{eff}\right)\right] \tag{1}$$

and increases with t_p and with τ_{eff}. With this equation, the fraction of excited ions available at the end of the pumping pulse (the storage efficiency) is

$$\eta_{st} = \left[1 - \exp\left(-t_p / \tau_{eff}\right)\right]\left(t_p / \tau_{eff}\right)^{-1}; \quad (2)$$

high storage efficiency can be obtained for short t_p and/or long τ_{eff}. As a compromise between high $n_e(t_p)$ and high η_{st}, it is customary to use $t_p \approx \tau_{eff}$.

The theory of Q-switching indicates that a high emission cross-section is not favorable to generation of high-energy pulses; moreover, it could facilitate the amplified spontaneous emission in directions outside the laser axis. A parameter of importance for the efficiency of the laser emission is the quantum defect between the pump and laser emission quanta, which is usually characterized by the quantum defect (Stokes) ratio between the corresponding wavelengths, $\eta_{qd}^{(l)} = \lambda_p / \lambda_l$. A large quantum defect (small Stokes ratio) would reduce the laser parameters and increase the heat generation of heat. The resonator design should also grant high superposition η_v of the pumped and the laser-mode volumes. A pumped volume larger than the laser mode volume determines low η_v, and a fraction of the excited ions is left outside of the lasing process and de-excite by luminescence and non-radiative processes even after extraction of the Q-switched pulse. The Q-switching can be accomplished by different methods, for the high energy lasers electro-optic switching would be preferred. The duration of the Q-switch pulses is of several nsec to tens of nsec, and the peak power ranges from millijoules to joules.

Pulses in the range of picoseconds or femtoseconds can be generated by mode-locking (ML) techniques; the pulse duration t_{pulse} is inversely proportional to the width of laser emission (for Gaussian lines $t_{pulse}\Delta\upsilon_l \geq 0.44$) and laser materials with strong homogeneous or/and inhomogeneous line broadening of are necessary. Homogeneous broadening is determined by strong electron-phonon coupling, such with the spin-allowed transitions of 3-d ions (vibronic lasers), whereas the inhomogeneous broadening is caused by a distribution of crystal field interactions, either by a distribution of the structure of the site (glasses), by presence of several sites offered for substitution or by crystal field perturbations induced by disorder in the neighboring cationic sites. Low threshold for these lasers requires large $\sigma_e \tau_{eff}$ products. The repetition rate in case of ML lasers is very high, tens to hundred MHz, and the pulse energy is very low, several nJ.

The pulse energy can be subsequently increased by chains of amplifiers. In a regenerative amplifier the pulse duration and spectral width are determined by the oscillator, whereas the amplifier controls the energy. A major characteristics of the amplifier is the saturation fluence $E_s = h\nu/\sigma$, related to the stored energy per unit volume by $J = g_0 E_s$, where $g_0 = \sigma n_e$ is the small-gain coefficient. Efficient extraction of the stored energy requires input fluences similar to the saturation fluence of the material. This imposes construction of large aperture amplifiers for

generation of high energy. Amplification factors to 10^{10} can be obtained in such complex amplifier chains. The high peak power of the ultrashort (fs) pulses could make such amplifier systems unpractical and in such case CPA should be used. The fundamental emission of the infrared lasers is multiplied in frequency to obtain visible or UV radiation using large-size non-linear crystals, such the KDP family.

The analysis of the processes for efficient generation and amplification of high energy pulses necessary for the nuclear fusion experiments indicate several requirements for the laser material, pumping scheme and resonator design:

– *the electronic structure of the active ion* should grant suitable energy levels and transition probabilities for pump absorption and laser emission: (i) good absorption of pump radiation; (ii) large radiative lifetime τ_{rad} and high emission quantum efficiency η_{qe} to grant high storage of inversion and low heat generation; a long lifetime τ_{eff} can reduce the pump power necessary to produce the desired inversion; (iii) the emission cross-section should determine saturation fluences below the laser damage threshold of the laser material or of the optical coatings but large enough to enable high energy pulses and avoid de-excitation by pre-lasing or amplified spontaneous emission; this recommends σ_e of 2 to 6×10^{-20} cm^2.

– *the laser host material* should grant suitable conditions for the static and dynamic properties of the active laser center and for high energy regime and efficient dissipation of heat. It should: (a) offer for doping sites of low symmetry, that enable electric dipole transitions of suitable absorption and emission cross-sections; b) enable incorporation of desired concentrations of doping ions, with selected spatial distribution; c) have high thermal conductivity to dissipate efficiently the heat generated in the pumped laser material or to enable efficient external cooling; d) have high thermo-mechanical (thermal shock parameter) to withstand the conditions of generation of short high-energy pulses; e) the laser material should be produced in very large sizes to enable high-aperture amplifiers;

– *the pumping source* should grant efficient laser scheme with minimal losses: high efficiency, high energy, good spectral overlap with the absorption of the laser material, especially in energy levels that enable low quantum defect.

– *the laser design* should grant high superposition of the pumped and laser mode volumes and high extraction of the stored inversion.

2.3.1. Selection of the laser active ion

The laser active ions of highest prospect for fusion lasers are based on the electronic transitions of the rare-earth ions such as Nd^{3+} and Yb^{3+} ions, several ions of the transitional 3d group, which give emission on vibronic transitions.

The Nd^{3+} ion has rich energy level scheme [6, 7], but a unique metastable level, $^4F_{3/2}$, placed in the 11500 cm^{-1} energy range, that could give laser emission in the 900 nm wavelength range (transitions to the $^4I_{9/2}$ ground level in quasi-three-

level scheme) or in the ~ 1 μm and 1.3 μm ranges, in four-level schemes on transitions to $^4I_{11/2}$ and $^4I_{13/2}$ respectively, the most efficient being the one-micron emission. The emission spectroscopic and dynamic properties depend on the host material: the luminescence lifetime varies between 80 and 400 μsec, and the emission cross-section for one-micron emission is from several 10^{-20} cm^2 to several 10^{-19} cm^2. The rich energy level scheme of Nd^{3+} favors pumping by various sources (lamps, diode lasers); combined with the emission properties, this enables construction of efficient lasers in various regimes, from CW to short (sub-ps) pulses, in various power ranges, from microlasers to hundred kW CW or burst pulse lasers, and to thousands of J pulse energy. The electronic structure of Nd^{3+} could favor parasitic de-excitation of the metastable level by energy transfer selfquenching caused by interaction of the excited Nd^{3+} ion with another Nd^{3+} ion in its ground state (down-conversion by cross-relaxation) or with another excited Nd^{3+} ion (upconversion). The emission selfquenching induces Nd^{3+} concentration dependent reduction of the emission quantum efficiency [8, 9] that increases the laser threshold and enhances the heat generation; this can limit severely the useful Nd concentrations.

The Yb^{3+} ion has a much simpler energy level scheme, with only two manifolds, $^2F_{7/2}$ (ground manifold) and $^2F_{5/2}$, placed in the range of 10000 cm^{-1}. The laser emission lies in the 1 μm range, similar to Nd^{3+}, but in a quasi-three-level scheme, the terminal laser level being a upper Stark component of the ground manifold. This introduces temperature-dependent partial reabsorption of the laser emission that influences the laser parameters. The cooling of Yb-doped materials can modify the Boltzmann thermal population of the Stark levels of the ground manifold, reducing that of the upper levels and reabsorption in these levels. The luminescence lifetime lies in the ~ 1 msec range, 2-4 times larger than for Nd^{3+}, but the room temperature emission cross-section is smaller, several 10^{-20} cm^2; however, it can be increased by cooling the material due to the narrowing of the emission lines. Thus the operation of the Yb^{3+} laser materials at cryogenic temperatures can be very efficient [10-12]. The simple energy level scheme of Yb^{3+} precludes energy transfer selfquenching by down-conversion or upconversion or by excited state absorption but makes the pumping difficult. Compared with Nd^{3+}, Yb^{3+} is more sensitive to the crystal field and electron-phonon interaction: the lines in the optical spectra are larger both by homogeneous and inhomogeneous broadening, the emission spectra show quite strong vibronic satellites and resonant effects [13, 14]. Besides obvious qualities for CW laser emission, the Yb-doped materials could be suitable for Q-switch or ultra-short pulse emission or amplification [15-17].

Several ions of 3d transition elements (Ti^{3+}, Cr^{2+}, Cr^{3+}, Cr^{4+}) in solids show prospect for generation of ultrashort pulses that could be amplified by CPA to reach PW peak power. The Ti^{3+} ion (ground configuration $3d^1$) can substitute for Al^{3+}, the most popular material being the white sapphire (α-Al_2O_3). Ti: sapphire has a very simple electronic structure, with only two energy levels, the ground level $^2T_2$

and the excited level ^2E placed at 16216 cm^{-1} [18-21]. Spin-allowed electric dipole transitions are possible, an intense vibronic absorption band peaking in the green region of spectrum and a strong and broad (180 nm) emission band in red-near infrared, peaking at 780 nm, with σ_e around 3×10^{-19} cm^2. The emission lifetime of 3.2 μsec makes the lamp pumping quite inefficient, whereas the absorption band lies in a region where strong pump diode laser do not exist at the present time. The unique actual possibility for pumping these lasers is with the second harmonic of ~1 μm Nd^{3+} lasers and thus the global efficiency of the Ti: sapphire lasers is quite low. The broad emission band of Ti: sapphire enables tunable laser emission in a quite large range in near IR, as well as generation of ultra-short pulses, as short as 5 fs. CPA using Ti: sapphire enables generation of ultrashort pulses of hundred of TW peak power, the actual limitation being determined by the size of the existing Ti: sapphire crystals. Nevertheless, the low global efficiency of this laser sheds doubts about its utilization as ignition laser in a nuclear fusion power plant.

Of more prospect for fusion could be the lasers based on Cr^{3+}, which has richer energy level diagram of Cr^{3+}. The Cr^{3+} ion has preference for octahedrally-coordinated crystal sites and function on the crystal field strength the lowest excited level could be the doublet ^2E (strong field) or the triplet $^4T_2$ (weak field). In strong field, specific for substitution of Cr^{3+} in small-size sites, such as Al^{3+} in Al_2O_3 (ruby) pure electronic sharp and quite weak spin-forbidden emission transitions with lifetime in the range of milliseconds to the ground level $^4A_2$ take place. By contrary, in weak crystal field sites, such as the large sites occupied normally by ions such as Ca^{2+}, Sr^{2+}, Sc^{3+}, In^{3+}, strong and broad vibronic bands of the spin-allowed transitions from $^4T_2$ to $^4A_2$ are observed, with lifetime from ~60 to hundreds of μsec. In such sites Cr^{3+} show strong and broad vibronic absorption bands in blue and red that can be pumped with lamps or red diode lasers. Efficient ultra-short laser emission was demonstrated with Cr^{3+} -doped colquirites $LiCaAlF_6$ - LiCAF (emission range 720-840 nm, lifetime 170 μsec) [22] and $LiSrAlF_6$ – LiSAF (emission range 780-920 nm peaking at 830 nm with cross-section 4.8×10^{-20} cm^2 and lifetime 67μsec) [23] or alexandrite [24].

The Cr^{4+} ion in YAG has strong absorption in the ~ 1 micron range, so it can be pumped with Nd lasers: its very broad (200-250 nm) $^3T_2\rightarrow{}^3A_2$ emission in IR peaking in the 1250-1450 nm range is very promising for generation of ultrashort pulses with very high peak power (100 PW range) [25].

The saturation fluence for the various active ions depends on the host, which determines the emission cross-section and on temperature. At room temperature for Nd^{3+} it has the values of 0.62 Jcm^{-2} in YAG, 6 Jcm^{-2} for silicate glasses and 8 Jcm^{-2} for phosphate glasses, for Yb^{3+} it has the value of 9.2 Jcm^{-2} in YAG, 15 Jcm^{-2} in sesquioxides and 35-40 Jcm^{-2} in glasses, for Ti: sapphire it is 0.8 Jcm^{-2} and for Cr: LiSAF 5 Jcm^{-2}. Obviously, the saturation fluence can be modified by changing the temperature. Since the most efficient extraction implies work close to the saturation fluence and the laser damage fluences of the optical coatings are around 10 Jcm^{-2} ,

it is obvious that not all the doped laser materials satisfy the condition for efficient extraction.

2.3.2. The pumping system

Regardless of the regime of laser emission, the laser threshold and the slope efficiency are influenced by the flow of excitation inside the laser material, $P_{th} \propto K^{-1}$ and $\eta_{sl} \propto K$, with $K = \eta_v \eta_a \eta_p \eta_{qd}^{(l)}$. At the same time, the heat loading coefficient that expresses the fraction of absorbed power transformed into heat, $\eta_h = 1-(1-\eta_l)\eta_p \eta_{qe} \eta_{qd}^{(f)} - \eta_p \eta_l \eta_{qd}^{(l)}$, contains the contribution of the excited ions that do not participate to lasing and that of the ions involved in the laser emission process; these two categories of excited ions are delineated by the laser emission efficiency η_l that depends on the volume superposition efficiency η_v and on extraction efficiency of stored inversion η_{se}; for CW lasers $\eta_{se} = 1-(P_{th}/P)$. The quantum defect ratio $\eta_{qd}^{(f)} = \lambda_p / \overline{\lambda}$ corresponds to the average wavelength of the luminescence emission and can be calculated from the emission spectra of each particular laser material. It is then evident that optimization of the laser parameters and reduction of heat generation would recommend optimization of the pumping process: good superposition of the pumped and laser mode volumes, good matching with the absorption spectrum of the laser material and emission as close as possible to the emitting laser level. Additionally, in case of pulse pumping, high-energy pump pulses with optimized duration should be provided.

The major pumping sources for the solid-state laser are the flashlamps and the diode lasers [5]. The energy of the flashlamps could be very high, of the order of kJ; in case of emission of several hundred μs pulses the peak power could reach tens of MW. The broad UV-VIS-NIR emission spectrum of flashlamps can excite various energy levels of the Nd^{3+} ion, from which the excitation decays very fast on the metastable level $^4F_{3/2}$. The absorption lines of Nd^{3+} are sharp and their cross-sections are not very strong, and thus high Nd concentrations and large paths of the pump radiation inside the laser material are desirable. The poor matching of the flashlamp emission with the Nd absorption spectrum determines low global absorption efficiency η_a. Moreover, the effective quantum defect between the average lamp pump quantum and the laser emission quantum of Nd is very large, the average $\eta_{qd}^{(l)}$ lies in the range of 0.5, leading to reduced laser performances and intense heat generation. Such shortcomings can be largely avoided by resonant diode laser pumping. The power of individual diode lasers is low and in order to increase the power, the diode lasers can be assembled in linear bars or in two dimensional stacks. Usually the diode lasers are operated in CW regime, at power levels of up to several kW and in pulsed operation at peak levels by about an order

of magnitude larger. The state of the art of diode lasers enables generation of laser radiation at desired wavelength in a quite narrow bandwidth (1-4 nm) enabling efficient resonant pumping of the energy levels of the laser active ions, although sometimes the absorption lines of the rare earth ions could be very narrow. Unfortunately, the emission wavelength of the diode lasers shifts with temperature so in many cases thermal stabilization could be necessary. The emission wavelength ranges of the diode lasers are determined by composition: the Nd^{3+} lasers can be pumped with AlGaAs or InGaAsP diodes (790-890 nm range), Yb^{3+} by InGaAs (940–970 nm) and Cr^{3+} by AlGaInP (670 nm).

In order to grant low quantum defect for the laser emission a pump level as close as possible to the metastable level must be choosen: in case of Nd laser materials the diode laser pumping is made traditionally into the strongly absorbing level $^4F_{5/2}$, placed in the 12,500 cm^{-1} (800 nm) range, i.e. 850–1,000 cm^{-1} above the emitting level $^4F_{3/2}$. This grants good pump absorption and quite low quantum defect, with Stokes ratio $\eta_{qd}^{(l)} \approx 0.76$ in case of the one-micron laser. In case of very efficient laser emission $(\eta_l \approx 1)$ this determines a heat loading parameter of 0.24, more than twice lower than for lamp pumping. It was recently shown [26-31] that pumping the Nd lasers directly into the emitting level $^4F_{3/2}$, in the range of 880 nm, reduces further the quantum defect and increases the Stokes ratio to 0.84, leading to a similar improvement of the laser parameters and to reduction of the heat generated by non-radiative processes by about 30% compared with the 808 nm pumping. This approach was also used for improving the performances of the Nd lasers in other wavelength ranges [32-34]. With this approach and optimizing the laser resonator, slope efficiencies of 80% in CW one-micron [35, 36] and 84% in the 900 nm laser emission [37] were demonstrated. By taking into account the reduction of the quantum defect in 880 nm pumping it was calculated that the power scaling of the one-micron Nd lasers, i.e. the laser power for given heat generation, can be extended by more than 50% compared with the 800 nm pump [38]. The direct pumping into the emitting level increases dramatically the global efficiency of the frequency multiplication devices based on the Nd lasers [39].

The Yb-doped materials do not show absorption in the visible range, so they cannot be pumped by lamps. These materials have a quite strong absorption line in region of 970 nm, corresponding to the first Stark component of the $^2F_{5/2}$ manifold, but this line is usually very narrow [12-14, 40, 41], especially at low temperatures, so diode pumping in the region of broad but weaker absorption band in region of 940 nm is preferred. There is a large variety of Yb-doped crystals that could generate ultrashort pulses [42]; most of these crystals are of low symmetry and their size is limited. Despite of the success in pumping laser crystals with diode lasers, very little attempt to use this approach in case of Nd or Yb doped glasses was reported, most probably because of the quite low absorption.

2.3.3. Selection of the host material

Laser emission was investigated in a large variety of active solid materials. These materials can be single crystals, glasses and, more recently, transparent polycrystalline materials produced by ceramic techniques. The crystals have usually ordered structure, with well defined crystallographic sites, occupied by specific ions; partially disordered crystals that preserve the basic crystallographic structure but where a given crystallographic site could be occupied by several types of ions of different species or valence can be used because they provide controlled inhomogeneous broadening of the spectral lines. The ordered crystals have usually high thermal conductivity k, for instance in case of YAG at the room temperature this is around 10.3 $Wm^{-1}K^{-1}$, for cubic sesquioxides it could go to 17 $Wm^{-1}K^{-1}$ and so on, and it increases strongly by reducing the temperature. The doping with other ions could reduce the thermal conductivity, especially when the atomic mass of the doping ion differs markedly from that of the substituted cation; a similar reduction is observed for the partially disordered crystals.

A major criterion of selection for the lasers used in nuclear fusion is the possibility to produce large-size laser materials. At the present time no crystal can met such requirement; however, large active components of very high optical quality can be produced from glasses: in these materials the site offered to the doping ion has not a well defined structure (anionic coordination number, anion-cation distances and bond angles) and this determines inhomogeneous broadening of the lines. The most popular for lasers are the silicate and phosphate glasses, which can be doped with high concentrations of Nd or Yb. The thermal conductivity of glass is quite low (below 1 $Wm^{-1}K^{-1}$) and the thermo-mechanical properties are quite poor, precluding good dissipation of the heat generated in the laser material and limiting severely the repetition rate for the high energy lasers.

These limitations could be eliminated to large extent by the polycrystalline laser materials produced by ceramic techniques. These materials are composed of closely-packed crystalline grains with very shallow grain boundaries and very low density (order of ppm) of intergrain pores. The production technologies can be grouped in two classes according to the method of synthesis of the materials: solid-state synthesis [43-46], and wet (soft) synthesis [47-49], where a solution containing the desired species of cations is precipitated to form a precursor that is subsequently reduced to the final compound and transformed in nanocrystalline powder by thermal treatment. The solid-state-synthesized compound is submitted to isostatic compression to reduce the density of pores and is finally sintered in vacuum, whereas the nanocrystalline powder is collected to desired size and shape by slip casting and submitted to final sintering. Since the orientation of the ceramic grains produced by these methods is random, they have been limited to optically isotropic materials, i.e. to cubic compounds such as garnets or sesquioxides. The average grain sizes of ceramics is 30 to 50 μm in case of the solid-state synthesis and of several μm for the wet synthesis. Recently, attempts to orient the grains of

non-cubic materials in strong magnetic field before sintering produced nearly transparent bodies, but the optical quality proved too low to grant laser emission.

The transparent ceramic materials join structural and thermo-mechanical properties specific to crystals with the technologic capabilities specific to glasses:

– Characteristics of fabrication techniques: temperatures by 400 to 700°C lower than for the melt-growth of crystals; high technological yield; good control; high reproducibility; possibility to produce bodies of large (meter) size; possibility to produce composite materials;

– Structural properties: uniform grain sizes; low size and volume density of residual intergrain pores (order of ppm); very shallow grain boundaries;

– Functional properties: high compositional versatility; large doping concentrations; controlled profile of composition or doping (homogeneous, gradient, step change); optical transmission similar to the single crystals; similar or improved thermal and mechanical properties

– Economical aspects: Reduced production cost; good use of the raw material; low energy consumption.

Investigation of the doped garnet (YAG, GSGG) or cubic sesquioxide (Sc_2O_3, Y_2O_3) ceramics indicates that the spectroscopic and microstructural properties are similar or improved with respect to the crystals [9, 28, 29, 50-52]:

– the variety, nature and structure of the centers formed by the doping ions in ceramics are similar to those of the single crystals: although in case of ceramics defective structural centers could exist at the grain boundaries, their concentration is low compared with the regular sites; this was connected with the low ratio surface/volume and with the very shallow grain boundaries. Moreover, it was found that specific defects connected with departure from the ideal stoichiometry in the melt-grown garnet crystals have much lower concentrations in ceramics.

– the quantum states of the doping ions (energy levels, transition probabilities, crystal field effects, electron-phonon interactions) are similar to the corresponding single crystals;

– the distribution of the doping ions at the available lattice sites is random in systems that do not need charge compensation, although increased concentration could occur at grain boundaries;

– the interactions between the doping ions and the energy transfer processes at low doping concentrations are similar to single crystals. Moreover, investigation of such processes in ceramics with high doping concentrations, unavailable in crystals, evidenced new types of energy transfer processes, including cooperative down-conversion or upconversion of excitation.

The capabilities of the ceramics in construction of lasers have been demonstrated for various materials with garnet and cubic sesquioxide structure. The similarity of the laser emission in the low concentrated Nd:YAG ceramics and single crystals as well as the laser emission at high doping concentrations in

ceramics (to 7 % Nd) [28-30, 43, 46, 47] was demonstrated and these studies were soon extended to other Nd or Yb doped garnet and sesquioxide ceramics. The Nd-doped YAG ceramic plates of 10cm×10cm size proved already crucial in extension of the heat capacity burst mode lasers to unprecedented 67 kW emission [53] and Yb-doped ceramic are envisaged for construction of cryogenic lasers for nuclear fusion [54]. It became then obvious that the polycrystalline ceramic material could be not only substitutes for the existing crystal lasers but they can extend considerably their performances and scale: moreover, it was demonstrated that the ceramic techniques can tailor new laser materials adapted to the requirements of specific applications, such as large size, good thermo-mechanical Based on these aspects, these materials cam offer breakthroughs in construction of the lasers for inertial nuclear fusion by offering optimal spectroscopic properties for pump absorption and laser emission, high thermo-mechanical properties to enable highly efficient, ten of Hz laser emission and large size to enable very high energy pulses.

2.4. PRESENT AND PLANED INERTIAL FUSION EXPERIMENTS

The first projects for demonstration of the break-even point are based on central confinement, the most advanced being the National Ignition Facility (NIF) at the National Lawrence Livermore Laboratory in USA [55-57], whose aim is to demonstrate ignition of a ~10 mg D-T contained in a Be, plastic or diamond capsule of 2 mm diameter, centrally compressed under the action of the X-rays produced by laser irradiation of a heavy metal (uranium-gold alloy) hohlraum containing the fuel capsule. Since the only available type of laser material of large aperture that could be used was the Nd-doped glass, the NIF project is based on Q-swithed Nd-glass lasers, pumped by high-energy flashlamps. The Nd-doped glass has of quite large fluorescence lifetime and moderate cross-section for 1.06 μm emission, favorable to storage of population inversion and generation of high energy pulses by Q-swithching. Two bunches of 96 laser beams each are introduced through two holes in the ends of the hohlraum and the angles are thus chosen as to enable generation of a highly symmetrical field of X-Rays that act on the surface of the fuel pellet. The total energy of the 192 laser beams is 4.2 MJ at the fundamental emission wavelength of 1. 053 μm and duration of 5 to 10 ns. In order to reduce the strong coupling with the hot electrons this radiation is tripled in frequency using large size DKDP plates resulting a total of 1.82 MJ at 351 nm, with total peak power of 400-500 TW. The aperture of the laser is 40×40 cm and is obtained by using glass plates of 46 cm × 81 cm and 4.11 cm thick. A nuclear reaction gain of 10-20, i.e. generation of 20-40 MJ nuclear power for each laser pulse is expected. The very high generation of heat, coupled with the low thermal conductivity of the glass and with the large thickness of plates, that preclude the efficient removal of the heat and with the reduced thermo-mechanical parameters limits the repetition rate of these lasers to a pulse each two hours. The construction

of the laser was completed and laser pulses in excess of 1 MJ have been already demonstrated. Although the efficiency of the laser system is very low, under 1%, and the cadence is not practical for production of energy, the NIF plays a crucial role in the laser inertial nuclear fusion, the demonstration of possibility to control the nuclear reaction beyond the break-even point.

A high-energy laser fusion facility now in construction is the Megajoule laser at Bordeaux-France. The equipment will use 240 beams of third harmonic Nd:glass lasers to compress and ignite indirectly a D-T capsule placed in a gold hohlraum. The total energy supplied by this equipment will be around 1.8 MJ (around 550 TW peak power) and is envisaged to get a nuclear reaction gain slightly larger than 10 (20 MJ nuclear power for each laser pulse) [58].

Experiments for characterization of the central ignition with direct drive of the laser radiation on the D-T capsule are performed with OMEGA facility at Univ. of Rochester using 60 beams of a tripled Nd: glass laser of 30 kJ UV radiation [59]. This was completed with PW lasers of 2.6 kJ at 1053 nm, duration 1-100 ps using CPA (project OMEGA-EP) [60-62].

In order to increase the repetition rate of the lasers for fusion, an experimental laser amplifier based on the diode laser pumped Yb-doped $Sr_5(PO_4)_3F$ –SFAP crystal [63, 64] is in construction. This will be a single-beam laser delivering 100 J at 1047 nm with duration 3 to 15 nsec at 10 Hz repetition rate in a 3×5 cm aperture. The laser is built in a scalable concept in frame of the project Mercury [65].

The requirement on the laser energy is somewhat relaxed in case of the fast ignition lasers. The calculation indicates that the necessary energy for the ns compression laser is of 300 kJ, almost an order of magnitude lower than for central ignition, whereas that of the 10 ps ignition laser is of the order of 10 kJ. This prompted search for alternative lasers, which would enable the increase of laser efficiency and cadence. Some of these new solutions are envisaged for the fast ignition laser facilities in construction.

The laser KOYO will use laser radiation of 350 kJ for implosion and 50 kJ for ignition [66]. The facility KOYO-F will use a16 Hz 1.1 MJ frequency tripled (343 nm) cryogenic Yb:YAG laser with active mirror for compression and 100 kJ at 1030 nm for fast heating. FALCON-D (Fast Ignition Advanced Laser Fusion Reactor Concept with a Dry Wall Chamber) will use implosion lasers of 350 kJ and ignition laser of 50 kJ at 30 Hz repetition rate. A fusion yield of 40 MJ is envisaged, which will correspond to a multiplication factor of 100 [67]. The European facility HiPER will use third harmonic Nd:glass 250 kJ, 10 ns for compression and 2^{nd} or 3^{rd} harmonic 70 kJ in 10 ps for ignition [68-70].

A very advanced concept is the hybrid fusion-fission facility LIFE which will use the neutrons released from the fusion reaction to burn a blanket comosed by any tipe of fissile material, natural Uranium, burnt fuel from fision nuclear plants and so on. This will be a sub-critical reactor, with safe control of the fission process, which will act only when the fusion equipment is in function [56, 57].

2.5. DIRECTIONS OF FURTHER DEVELOPMENT OF LASERS FOR INERTIAL NUCLEAR FUSION

The actual state of the art of the high energy lasers for inertial nuclear fusion shows that intensified effort is necessary to identify new laser materials able to generate with high efficiency high energy pulses with repetition rates of 10-15 Hz. Research for enhancement of performances of the nuclear fusion lasers develops along several simultaneous lines of action, such as new or improved laser materials and pumping schemes. The search for new laser materials for generation of high-energy pulses in Q-switch or ultrashort regimes concentrates on materials that could be produced by ceramics techniques and can be pumped efficiently with laser diodes or lamps. These investigations refer mainly to Yb^{3+} or 3d-doped materials in materials of cubic symmetry that will enable utilization of polycrystalline ceramics.

The crystals with disordered structure determined by the multiple occupancy of specific crystallographic cationic sites with ions of different species or with different valence states could enable strong inhomogeneous broadening of the spectral lines. Among these materials, the rare earth (RE^{3+}) doped calcium niobium gallium garnets (CNGG) or calcium lithium niobium gallium garnets (CLNGG) [71] gained interest in recent years. Spectroscopic investigation of Nd^{3+} in these crystals revealed multisite structure and inhomogeneous broadening caused by local disorder in the octahedral coordination site occupied by Li^{+}, Nb^{5+} and Ga^{3+} around the doping ion [72]. This broadening made possible sub-picosecond laser emission of Nd^{3+} in these materials [73]. Recent investigation of Yb-doped CNGG crystal revealed multisite structure of the spectra and broadening of the lines [74] and the possibility of ultrashort (73 fs) pulse generation was demonstrated [75]. Our recent investigation revealed that CLNGG can be produced by ceramic techniques and that the spectroscopic properties of Yb^{3+} in CLNGG ceramics and crystals are similar and show pronounced inhomogeneous broadening that determines strong and fairly broad absorption band at 972 nm and the very broad emission band around 1027 nm of Yb: CLNGG in a very large temperature range. Moreover, it was shown that further disordering by replacing half of the Nb ions by Ta increases the width of the 1027 nm emission line to 24 nm at 300 K and 17 nm at 10 K, making this material suitable for short-pulse generation. The use of ceramic materials could enable scaling to very high energy laser emission, making these materials of interest for high energy lasers for nuclear fusion.

The second direction of action is the improvement of pumping schemes. A promising direction of investigation is the sensitization of Yb emission by co-doping with Nd that would enable the pumping of Yb with flashlamps using the absorption in Nd. Our investigation revealed that energy transfer efficiencies larger than 98% could be obtained in case of (Nd, Yb)-doped Sc_2O_3 [76] and Y_2O_3 ceramics [41,77]. Such sensitization process was considered [78] as a possibility to excite the Yb^{3+} ions with high energy pulses from flashlamps *via* the Nd^{3+} ions in order to

generate high power laser pulses. Direct pumping into the level $^4F_{3/2}$ of Nd^{3+} could optimize the quantum defect in such lasers. This approach might prove useful in reducing the quantum defect for the Nd:glass lasers.

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