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Determination of the thermo-optic coefficient *dn/dT* of ytterbium doped ceramics (Sc_2O_3, Y_2O_3, Lu_2O_3, YAG), crystals (YAG, CaF_2) and neodymium doped phosphate glass at cryogenic temperature

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ABSTRACT

In this paper, we report the measurements of the thermal expansion coefficient and the thermo-optic coefficient *dn/dT* for the ytterbium doped cubic sesquioxides (Sc_2O_3, Y_2O_3, Lu_2O_3) at cryogenic temperature. These materials appear to have very interesting properties for setting up high average power laser chains useful for plasma physics and for inertial fusion energy drivers. Measurements have also been done on YAG ceramic and crystal, CaF_2 crystal, and neodymium phosphate glass (Hoya, LHG-8).

1. Introduction

Since a few tenth years, important developments of laser chains for inertial fusion confinement have been investigated. These include fusion applications like National Inertial Fusion (NIF) in USA, MegaJoule Laser (LMJ) in France, Gekko II in Japan or Sheng Guang III in China. These chains require high energy but very low repetition rate and are generally based on neodymium phosphate glass which is of poor thermal conductivity material but available in large scales. Beside these investigations, some works are devoted on the development of high average power laser chains capable to deliver comparable energy at high repetition rate (MERCURY in USA [1], LUCIA in France [2] and HALNA in Japan [3]). In these systems, crystalline amplifier medium must be used in order to reach high thermal conductivity of the amplifier medium for evacuating the heat generated by the high repetition rate pumping and stored in the medium. With this goal, we have investigated the potentiality of the Yb doped cubic sesquioxides. Thermo-optical values, such as thermal conductivity, thermo-optical coefficient *dn/dT*, coefficient of thermal expansion, absorption and emission cross sections are actually unknown in the literature, especially at low temperatures. Therefore, measuring these parameters is very important for the development of high-power lasers.

2. The ytterbium ion and the host choice

Ytterbium ion brought a lot of advantages compared to neodymium ion for the amplifier mediums. First, its energy level diagram is made of only two electronic levels spitted in sublevels by Stark effect generated by the crystal field making it free of up-conversion processes, excited states absorption and concentration quenching. Its upper level life time is long enough making it possible for diode pumping and, the absorption bandwidths are large avoiding accurate control of the pumping wavelength even in pulsed pump mode [4].

The low quantum defect is a result of the quasi-three level nature of the Yb ion. In this system, the fundamental level and the terminal level of the pumping transition are closed to the upper level and the terminal level of the laser transition respectively making then possible to achieve low quantum defect favourable to low thermal loading. Unfortunately, the terminal level of the laser transition is closed to the fundamental and it is thermally populated. As a result, pumping requires minimum pump energy for insuring the population inversion. This amount of pump energy does not participate to the laser process and reduces the efficiency.

High conductivity crystals are essentially the garnets and the sesquioxides. YAG is the most used laser host but higher thermal conductivity at low temperature can be achieved using sesquioxides [5]. These materials are now available as sintered ceramics making then possible obtaining large pieces with the same properties of single crystals [6].

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3. Cryogenic cooling

Cryogenic cooling largely improves the properties of laser materials [7,8]. First, with Yb doped materials, the terminal level of the laser transition is unpopulated and the system acts as a four-level system. Therefore, the absorption and emission cross sections are larger [9–11] and, as a consequence, the saturation fluences are lower. Then, both pump energy storage and laser energy extraction are larger and more efficient. Last, the thermal conductivity increases and the thermal elongation coefficient decreases when decreasing temperature [12,13]. The cooling limit seems to be the narrowing of the absorption line width [14]. Then, for designing a laser chain, the variation of these parameters versus temperature must be accurately measured [15], which is the purpose of our paper.

4. Samples

We have measured both the thermal expansion and the thermo-optic *dn/dT* coefficients at cryogenic temperature of different laser materials. We have tested Y_2O_3 and YAG both undoped and ytterbium doped ceramics, together with 1 at.% Yb doped Lu_2O_3 and Sc_2O_3 ceramics. Measurements are also done on Yb:YAG crystals with different doping levels, Yb:CaF_2 both undoped and doped crystals and 4 at.% neodymium doped phosphate glass (Hoya LHG-8). All the samples are cylinders with 4 mm diameter and 6 mm length.

All the ceramics used in our experiments have been supplied by Konoshima Chemical Co., Japan. YAG crystals come from Laserayin Co., Armenia and CaF_2 is from Hellma, Germany. LHG-8 glass comes from NIF/LMJ glass production [16].

5. Thermal expansion measurement

Thermal expansion measurements were made at LNE [17]. The linear thermal expansion coefficient is determined by a differential method with a push rod dilatometer (Perkin Elmer dilatometer, TMA7 model). This method consists in measuring the changes in length between a reference material and the studied material. These materials are placed into a furnace (working from 130 K to 293 K). The dimensional changes of the specimens are transmitted to an inductive displacement transducer via a fused silica push-rod. Temperature is measured by the means of a type *K* thermocouple in the vicinity of the specimen.

The mean linear thermal expansion coefficient α_L between two temperatures T and T_0 is given by the following formula:

$$\alpha_L = \frac{1}{T - T_0} \frac{\Delta L|_{T_0}^{T}}{L|_{T_0}} \quad (1)$$

where $\Delta L|_{T_0}^{T}$ is the expansion measured from T_0 to T and $L|_{T_0}$ is the length of the material at room temperature (T_0 = 293 K).

Remark . Before the test, the dilatometer is calibrated under identical test conditions (temperature range, heating rate, atmosphere ...) with reference materials of known thermal expansion.

The accuracy of these measurements is ±5% for temperatures below 273 K and ±10% for temperatures between 273 and 293 K mainly because are samples are (only) 6-mm long.

Fig. 1 shows the thermal expansion coefficient results that we have obtained with an undoped Y_2O_3 ceramic. The thermal expansion coefficient tends toward substantially lower values when the temperature decreases below 300 K. Comparison of the thermal expansion coefficient of undoped Y_2O_3 ceramic can be made with Ref. [14]. It is in good agreement with our experimental values, i.e. better than 5% for temperatures higher than 200 K and better than 20% for temperatures below 200 K.

Our results are shown in the following tables. Table 1 presents the thermal expansion coefficient values that we have obtained with sesquioxides Y_2O_3, Sc_2O_3 and Lu_2O_3 ceramics for temperatures between 123 and 300 K. Table 2 shows the thermal expansion coefficients of different YAG materials (ceramic/crystal with different doping levels). In Table 3, the results obtained with neodymium doped phosphate glass and CaF_2 crystals are presented. All the values given in these tables are in ppm/K.

The thermal expansion coefficients decrease when the laser material temperature decreases. From the values of Tables 1–3, we can notice that there is no significant difference between a ceramic and a crystal. Although Xu et al. note that the thermal expansion coefficient varies in function of the Yb^{3+} concentration [10], we did not see a difference for different doping levels.

Besides, the experimental thermal expansion coefficients that we have measured are in reasonable agreement with those given in the literature. Indeed, for undoped YAG crystal, Aggarwal et al. give thermal expansion coefficients of 1.95 ± 0.1 ppm/K and 6.14 ppm/K at 100 and 300 K respectively [18]. In the work of Wynne et al. [19], thermal expansion coefficients have been measured at 125 and 266 K, and they give a value of 2.7 ± 0.2 ppm/K and 6.01 ± 0.2 ppm/K respectively. In our work, we find values of 2.8 ± 0.14 ppm/K and 6.6 ± 0.6 ppm/K, respectively at 123 and 296 K. Our experimental values are close to those of Wynne et al., better than 4% at 123 K, and close to those of Aggarwal et al. better than 10% at 296 K.

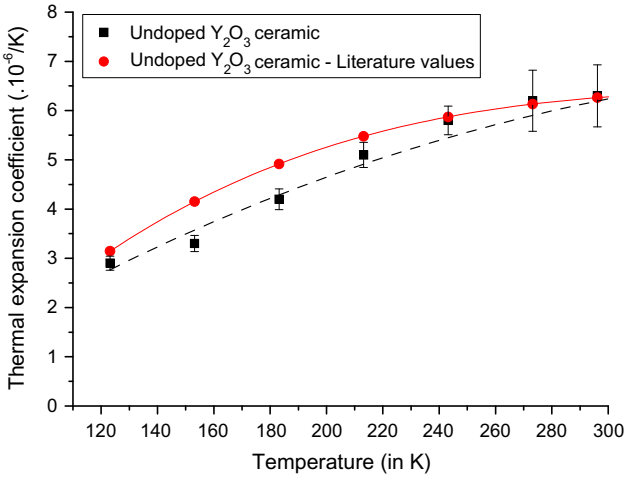

Fig. 1. Thermal expansion coefficient of an undoped Y_2O_3 ceramics. Comparison with literature values is also shown [8].

Table 1
Thermal expansion coefficients of yttria, scandia and lutetia ceramics.

Temperature (in K)	10 at.% Yb:Y_2O_3 ceramic	Undoped Y_2O_3 ceramic	1 at.% Yb:Sc_2O_3 ceramic	1 at.% Yb:Lu_2O_3 ceramic
123	2.8	2.9	2.3	2.9
153	3.5	3.3	3.1	3.4
183	4.3	4.2	4.2	4.4
213	5.1	5.1	5.2	5.3
243	5.8	5.8	5.8	5.9
273	6.4	6.2	6.2	6.1
296	6.7	6.3	6.3	6.1