Optical Materials 35 (2013) 499–503

Contents lists available at SciVerse ScienceDirect

Optical Materials

journal homepage: www.elsevier.com/locate/optmat

Laser ceramics $Tm:Lu_2O_3$. Thermal, thermo-optical, and spectroscopic properties

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ARTICLE INFO

Article history:
Received 30 July 2012
Received in revised form 2 October 2012
Accepted 3 October 2012
Available online 30 October 2012

Keywords:
Laser ceramics
Transient grating
Thermal diffusivity
Refractive index
Linear expansion
Lifetime

ABSTRACT

In this paper we report the measurements of in-plane thermal diffusivity, lifetime of metastable level $^3F_4$, thermal coefficients of linear expansion and refractive index change of laser ceramics $Tm(2\ at.\%):Lu_2O_3$ by the transient grating technique. The wavelengths 213, 266, 355 and 796 nm from pulsed lasers were chosen for sample excitation in accordance with the desire to measure particular parameters. The experimentally determined thermal diffusivity 0.05 cm^2/s was attributed to subsurface layer, which has thickness of order of grating period. The applied approach ensured more reliable measurement of lifetime of metastable $^3F_4$ level as compared to the conventional luminescence detection. The dynamic value of linear expansion coefficient was measured under sample excitation at 213 nm and was found to be close to that as measured elsewhere in static regime. Coefficient of refraction index change was found to be as high as $2 \times 10^{-5}\ K^{-1}$.

The value of absorption coefficient of ceramics at 213 nm was estimated by excitation of acoustic grating in a thin layer of air adjacent to the sample surface. After the estimation, it exceeded $10^4\ cm^{-1}$.

1. Introduction

Optical ceramics based on rear earth doped sesquioxides Lu_2O_3, Sc_2O_3, Y_2O_3 currently attracts attention as a promising alternative to single crystalline laser materials [1–3]. The favorable spectral and luminescent peculiarities, together with relatively high and nearly insensitive to dopant concentration heat conductivity, make the Lu_2O_3 ceramics prospective for production of high-power solid-state laser systems [4–9]. As the internal structure and parameters of ceramics are substantially dependent on features of numerous technologies of novel gain materials fabrication, the intrinsic thermal and optical properties of laser ceramics need to be thoroughly controlled.

In this paper, we report an optical characterization of laser ceramics $Tm(2\ at.\%):Lu_2O_3$, fabricated by Konoshima Chemical Co. (Japan). Thermal and thermo-optical parameters, as well as laser upper-state lifetime, have been measured by transient gratings (TG), excited at different wavelengths. Phase gratings (bulk and surface-relief types) were recorded in a sample by two-beam interference from pulsed laser. The intensity of diffracted probe beam from a CW laser was monitored as a function of time. Homodyne mixing and phase sensitive detection were applied for the diffracted signal enhancement [10–12]. The initial and time-resolved values of TG efficiency were controlled with the aim of determination of in-plane thermal diffusivity, lifetime of $^3F_4$ level, as well as for calculation of temperature coefficient of refractive index dn/dT and dynamic coefficient of linear expansion β_{eff}.

Optical glass K8 (an analog to glass BK7) was used as a reference material for the metrological verification of the measurements.

2. Preface: Spectral and luminescent studies

The sample under study was cut to form a 1 mm thick slab with 4 mm aperture in diameter. The image of the parent sample can be found in [8]. Both sides were mechanically polished up to optical grade. The absorption spectrum was measured by a spectrophotometer Cary 500 (Varian) with spectral resolution 2 nm. According to the spectrum, the edge of intensive absorption was near 266 nm, where α was found to be about $7\ cm^{-1}$. The much larger value of α at 213 nm was estimated indirectly via excitation of acoustic wave in air (see Section 4.2).

The absorption spectrum in near IR and the luminescence of ceramics $Tm:Lu_2O_3$ under pumping at the wavelength 796 nm one can find in [8]. We observed an additional band of fluorescence with maximum at 810 nm under sample pumping at 460 nm.

Time-resolved emission at the most intensive luminescence line (1950 nm, corresponding to the depletion of the level $^3F_4$, see Fig. 1) was studied. The luminescence lifetime τ_{lum} was found to be 4 ± 0.1 ms under pumping at the wavelengths of 796 and 460 nm. The studies of single crystalline $Tm(2\ at.\%):Lu_2O_3$ conducted by the pinhole method gave $\tau_{lum} = 3.8$ ms [13]. According to [8], the lifetime

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http://dx.doi.org/10.1016/j.optmat.2012.10.002

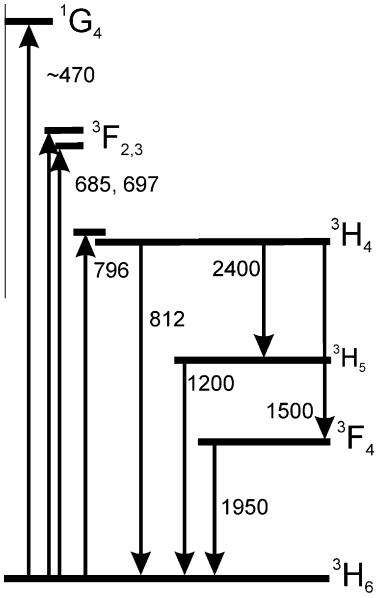

Fig. 1. Energy levels of Tm^{3+} ion in Lu_2O_3 matrix. Figures denote the wavelengths in nm.

of $^3F_4$ level is 3.7 ± 0.2 ms. Our measurements via luminescence appeared to give higher values, which could be attributed to the well known effect of reabsorption.

Kinetics of luminescence at 1518 nm (transitions from the level $^3H_4$) can be approximately considered as consisting of two exponents with decay times of about 7 and 40 μs. Nearly the same values were obtained in [8] for a similar sample, where time-resolved luminescence at spectral band shorter than 1200 nm was studied, and where the process of $^3F_4$ level populating was assigned to the effect of cross-relaxation, as well as to radiative and non-radiative transitions.

The energy-level diagram of the Tm-doped ceramics is shown in Fig. 1, where the results obtained elsewhere and our spectral studies are taken into account.

3. Experimental method description

The detailed description of the TG method, used in this study, can be found in [14,15], for example. In our experiment (see Fig. 2), two interfering laser beams were formed by a phase-grating beam-splitter of period 4Λ with deep rectangular surface profile. The beam-splitter was imaged with magnification 0.5^x into the sample by the two-lens telescope through the spatial filter in common focal plane, so that the high quality and about 100% modulated interference pattern of a spatial period Λ was obtained in a plane of a sample. In such a way the sample was symmetrically one-side illuminated by two intersecting beams, so that the interference planes were parallel to the plane *xz*. Orthogonal axes *z* and *x* were normal to the sample's surface and to the interference planes inside the sample respectively.

The Lu_2O_3 matrix was pumped by the radiation of the third, fourth, and fifth harmonics from a Nd:YAG laser LS-2151 (Lotis TII). Pulse width and pulse repetition rate were 60 ps and 10 Hz respectively. 10 ns pulses at wavelength 796 nm from a Ti:Sapphire laser, model CF123 M (Solar TII) were used for excitation of Tm^{3+} ions. Pulse duration in both cases of pumping was much shorter than any temporal process studied. As the pumping energy *E* was distributed non-homogeneously over the beam profile, only the central part (2 mm in diameter) of the beam – where energy density could be considered approximately uniform – was used

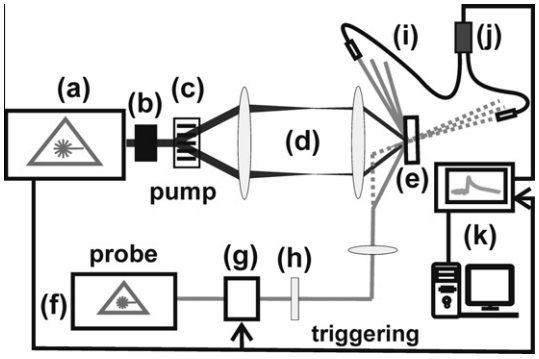

Fig. 2. Experimental setup: (a) pulsed laser; (b) attenuator; (c) beam-splitter; (d) telescope; (e) sample; (f) CW laser; (g) optoacoustic modulator; (h) half-wave plate; (i) fiber; (j) photomultiplier tube; and (k) digital oscilloscope.

for sample excitation. This point was of significance during the measurements of TG diffraction efficiency (see Section 4.4).

TG were probed by a CW He–Ne laser (λ = 633 nm) in reflection and transmission modes. The readout beam impinged the sample in the direction close to normal. This enabled us to test the gratings recorded both due to surface relief formation and the refractive index change. The probe beam was synchronously modulated by acousto-optical gate in order to diminish the parasitic light influence. The first order diffraction beam was detected by a photomultiplier tube (PMT), model Hamamatsu H6780-20. The time-resolved signal was recorded and stored by a digital oscilloscope Tektronix TDS 3032B with 300 MHz frequency band width. The net FWHM of pulse response function of the detecting system was either 4 or 70 ns, depending on load resistance.

For enhancement of the diffracted signal, the procedure of homodyne mixing was used by adding of some portion of coherent light, scattered by the sample imperfections, to photo-sensitive aperture of PMT. The phase difference between these two interfering fields of probe light was controlled, so the phase-sensitive detection and storage were conducted [11] for further signal processing.

4. TG experiments and calculations

Investigations of ceramic sample were verified and controlled by comparison with the results obtained for optical glass K8 as a reference material with well-known parameters.

4.1. In-plane thermal diffusivity measurement

In case of pumping at 213 nm, two processes basically contributed to TG recording: surface corrugation, caused by thermal expansion of the material, and temperature change of the refractive index. The diffraction signal in reflection mode of probing was mostly formed by the relief grating, whereas the signal observed in the transmission mode contained the contributions of both gratings. Typical decay curves of the diffracted signal are shown in Fig. 3. In accordance with a theory for absorbing samples, the intensity of a beam, diffracted by thermal surface relief grating, decays as the complementary error function $I_R(t) = I_R(0)\mathrm{erfc}(t/\tau_{th})^{0.5}$ [16,17], where τ_{th} is the thermal grating lifetime. The best fit to the experimental data (Fig. 3a) gave τ_{th} = 12.7 ± 0.3 μs.

In contrast, the exponential decay $I_n(t) = I_n(0)\exp(-t/\tau_{th})$ appeared to be more adequate to the experimental data (Fig. 3b) in case of TG probing in transmission mode. This is a consequence of much higher efficiency of a bulk refractive index grating in comparison with surface relief TG. The fitting procedure yielded τ_{th} = 12.9 ± 0.3 μs. Quadratic dependence of τ_{th} on Λ was observed for both modes of probing, confirming the diffusive nature of grating