Kinetic characteristics of up-conversion luminescence in fluorophosphate glasses doped with Tm^{3+} and Yb^{3+} ions, excited by CW and pulsed femtosecond infrared radiation

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Abstract

We report kinetics of rise and decay of up-conversion luminescence in processes involving Tm^{3+} ions in fluorophosphate glass upon excitation of Yb^{3+} ions by continuous and pulsed femtosecond radiation. We observe several characteristic times in time-profiles of up-conversion rise and decay fronts. Also, the luminescence decay process with continuous pump behaves in a completely different way in comparison with femtosecond pulsed pump with increasing pump power.

I. Introduction

Up-conversion (UC) luminescence of rare earth ions is a promising area of research in optics and laser physics. Continuous lasers are widely used to excite UC processes, but femtosecond lasers are also of interest for several applications involving UC processes. Femtosecond laser radiation provides a high peak power of excitation pulses [1, 2], which can lead to multi-photon excitation [2, 3] and decay [4] processes. Direct laser excitation of thulium ions due to absorption processes between its excited states, along with the energy transfer from ytterbium ions, makes it possible to control the temporal dynamics of UC processes. One can either enhance or suppress UC processes using coherent control methods [2, 4, 5]. Note that the analysis of the time-hierarchy of UC processes [6], occurring due to both single- and multiphoton laser excitation of thulium ions [4] and energy transfer between ytterbium and thulium ions [7], becomes much more complicated when excited by femtosecond radiation [4, 8]. It is even more complicated in the presence of several resonant frequencies [9]. In our case, the characteristic emission frequencies are determined by the wavelength of the femtosecond laser ($\lambda \approx 1038$ nm) and the wavelength of the energy exchange between ytterbium ions and the thulium ions ($\lambda \approx 975$ nm).

Classical analysis of double logarithmic (log-log) dependences of the intensities of these luminescence bands in the case of continuous pumping showed that the dependences under consideration reflect the mechanism of sequential nonradiative energy transfer from Yb^{3+} ions to Tm^{3+} ions

[7, 10, 12]. At the same time, the dependencies obtained using excitation by femtosecond pulsed radiation suggest that, along with this process, significant contributions can also be made by the processes of both single-photon and multiphoton excitation of Tm^{3+} ions. The latter can be attributed to significantly higher peak power of femtosecond pulses [2]. In addition, an influence of the nonlinear effect of photobleaching [13], associated with the saturation of the population of intermediate levels and leading to a decrease in the slope of the log-log dependence [14], can also take place.

II. Experimental method

In previous studies, we performed a comparative analysis of the UC processes of Tm^{3+} ions in fluorophosphate glass upon excitation of Yb^{3+} ions by continuous [1,7,10] and pulsed femtosecond [1] radiation. The wavelength of CW radiation pump (975 nm) was in the strong absorption band of Yb^{3+} ions, and the wavelength of femtosecond radiation pump (1038 nm) was into the edge of the absorption band of Yb^{3+} ions. In addition, 1038 nm wavelength corresponded to the transitions between some excited states of Tm^{3+} ions (e.g., $^3F_4 \leftrightarrow {}^3F_{2,3}$ и $^3H_4 \leftrightarrow {}^1G_4$ in Fig. 1a, see Fig. 6 in [11]). In both cases, bands in the regions of 440–480 nm, 630–670 nm, and 760–840 nm were observed in the UC luminescence spectra (Fig. 1b). The distribution of their respective intensities obtained by femtosecond and continuous excitation turned out to be different. This indicates different kinetics of the processes of population of the energy levels of ytterbium and thulium and a possible difference in the time-hierarchy of these processes.

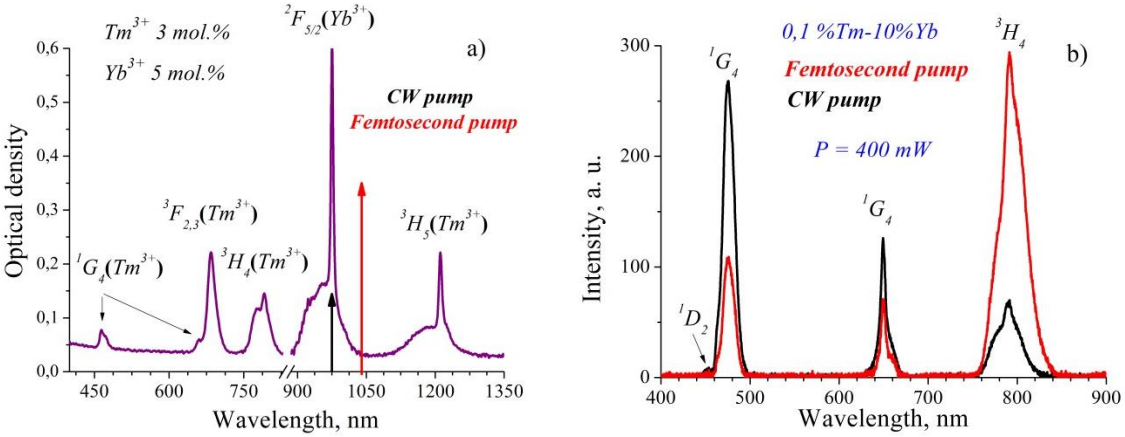

Fig. 1 Absorption spectrum of the sample with excitation wavelengths indicated (a) and up-conversion luminescence spectra of the sample (b) upon excitation by CW radiation (black) and femtosecond pulses (red)

III. Results and Discussions

This short notice presents some preliminary results of a study of the kinetic characteristics of UC luminescence depending on the power of the exciting radiation. In the experiments, the laser sources described in [1] were used to excite UC luminescence. CW pump was modulated using a control unit, and femtosecond-pulsed pump was switched on and off using a mechanical chopper. Photomultiplier tube FEU-118 and a TDS5104 oscilloscope were used to record oscillograms. The oscillograms of the rise and decay fronts of UC luminescence at different powers of the exciting continuous and femtosecond radiation are shown in Fig. 2. This figure shows that in the case of excitation by

continuous radiation, the kinetic dependences are characterized by a sequence of several sections with different characteristic times, τ, corresponding, for example, to slow and fast processes (Fig. 2 (a, c)). The presence of a slow component might be associated with the reverse energy transfer from Tm^{3+} ions to Yb^{3+}. With femtosecond excitation, this process can be characterized by a single exponential (Fig. 2 (b, d)).

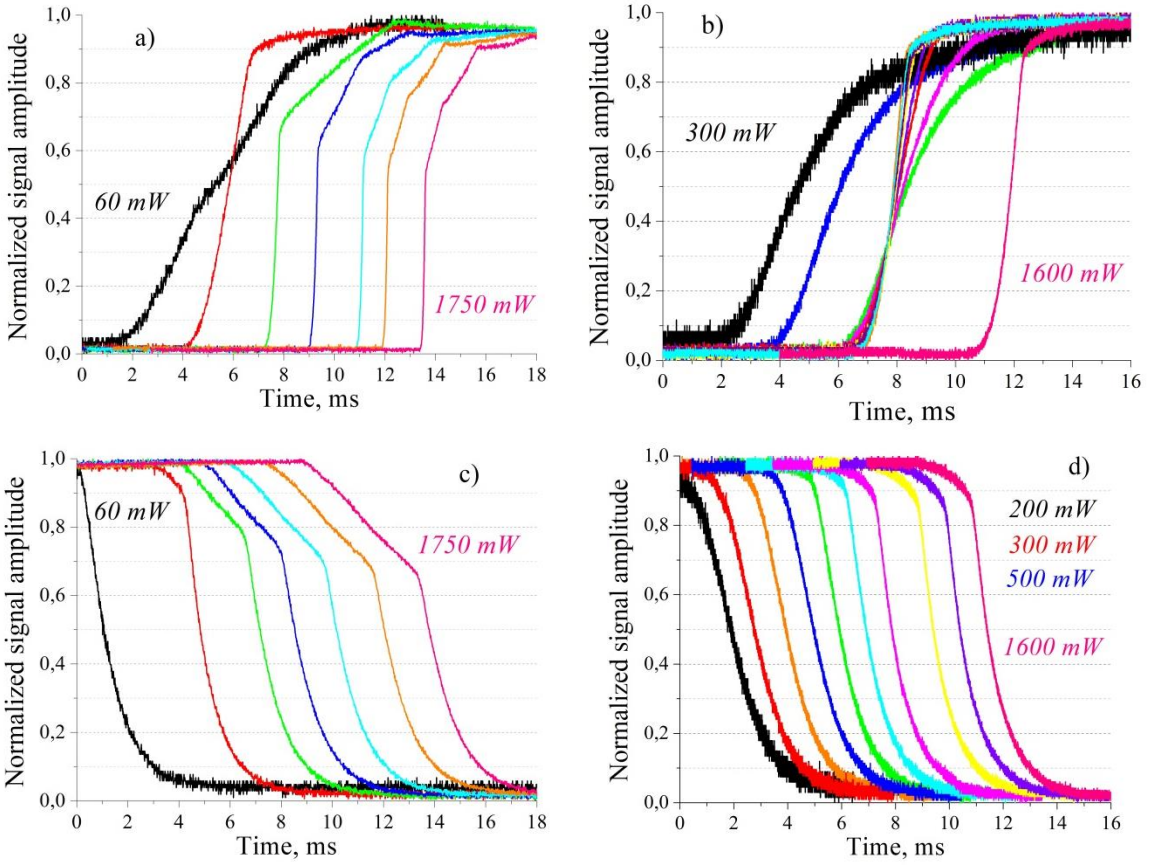

Fig. 2 Oscillograms of the rise (a, b) and decay (c, d) fronts of the UC luminescence (475 nm) excited by modulated CW (a, c) and femtosecond pulsed (b, d) pump. For better clarity, the oscillograms of the fronts are spaced apart, and the corresponding powers of the exciting radiation are indicated by numbers.

The rise time decreases with increasing power of both continuous and femtosecond excitation radiation (Fig. 3a).

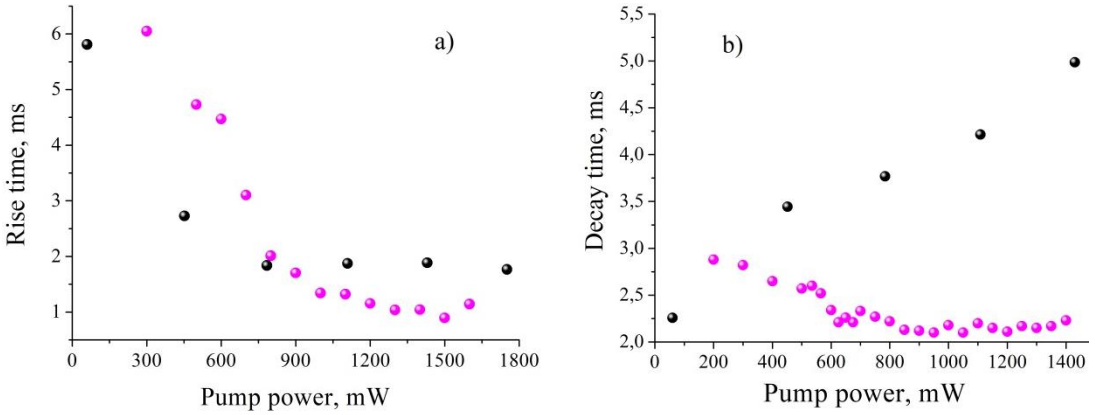

Fig. 3 Dependences of the rise time (a) and decay time (b) on the CW pump power of the exciting continuous (black balls) and femtosecond pulsed (magenta balls) pump.

With CW excitation, along with the energy transfer from Yb^{3+} to Tm^{3+}, the population of the $^1G_4$ level can also increase due to cooperative processes by transferring energy from excited pairs of ytterbium ions Yb*-Yb* [7]. This is possible at a high pump power and a sufficiently high concentration of ytterbium ions. With femtosecond pulsed pump, a sharp jump-like decrease of the rising-front formation time is observed. The parameters of the obtained dependencies of the rise and decay kinetics of the UC luminescence for the boundary values of the studied ranges of the exciting radiation powers are given in the table.

Characteristic decay and rise times of UC luminescence at different pump powers

CW pump		
P, mW	Rise time, ms	Decay time, ms
60	3	1.23
1750	2.4	1.21
Femtosecond pulsed pump		
P, mW	Rise time, ms	Decay time, ms
300	2.9	3.06
1600	2.2	1.18

It should be noted that the luminescence rise time decreases with increasing femtosecond pulsed pump, like how it occurs with continuous excitation (Fig. 3a). However, the luminescence decay process with CW pump behaves in a completely different way in comparison with femtosecond pulsed pump with increasing pump power (Fig. 3b). In the latter case, the change in the decay rate of the process from the $^1G_4$ level can be associated with both the influence of relaxation processes from higher excited states ($^1D_2$, $^1I_6$ и $^3P_{0,1,2}$) and exchange processes involving other excited states, depending on the pump power and the concentration of Tm^{3+} and Yb^{3+} ions [15]. Thus, the kinetic characteristics of the UC luminescence of thulium ions from the $^1G_4$ multiples confirm the difference in the mechanisms of its population and interaction between Tm^{3+} and Yb^{3+} ions, both depending on the excitation mode and on the pump power.

IV. Conclusions

The observed differences in the fronts' kinetics for CW and pulsed excitations points to the conclusion that under the femtosecond and CW excitations the UC dynamics might by caused by the different physical mechanisms. Possibly, in the case of femtosecond excitation occurring on the time-scale less than the typical decoherence time, coherent cooperative processes might influence population exchange. That is the subject of future work.

The authors gratefully acknowledge financial support from the BRFFR grant F23UZB-064 and MFST grant BLR-2024/4

References

[1] Up-conversion in fluorophosphate glasses with ytterbium and thulium ions excited by a sequence of ultrashort pulses at a wavelength of 1038 nm or by continuous radiation in the region of 975 nm / I.A. Khodasevich [et al.] // VII Congress of Physicists of Belarus: collection of scientific papers, Minsk, Belarus, April 26-28, 2023 / OOO "Kovcheg", 2023. - P. 222 - 223.

[2] Femtosecond Laser-Induced Upconversion Luminescence in Rare-Earth Ions by Nonresonant Multiphoton

Absorption / Yu.Yao et al. // J. Phys. Chem. A. – 2016. – Vol. 120. – P. 5522 – 5526.

[3] Upconversion luminescence from aluminoborate glasses doped with Tb^{3+}, Eu^{3+} and Dy^{3+} under the excitation of 2.6-μm femtosecond laser pulses /M. H. Yuan [et al.] // Opt. Express. – 2015. – Vol.23. – P. 21909.

[4] Controlling multiphoton excited energy transfer from Tm^{3+} to Yb^{3+} ions by a phase-shaped femtosecond laser field / Ye. Zheng [et al.] // Photonics Research. – 2019. - Vol. 7. – P. 486.

[5] Optimizing femtosecond laser-induced upconversion luminescence enhancement and suppression of Dy^{3+}-doped glass / Y. Huang [et al.] // Laser Phys. Lett. – 2020. – Vol. 17. – P. 016001.

[6] Mikheeva A.V., Kazakov B.N. Rise kinetics of up-conversion luminescence under pulsed excitation. Probabilistic model and experiment / J. Lumin. – 2019. – Vol. 205. – P. 167.

[7] Kolobkova E., Grabtchikov A. S., Khodasevich I. A. Upconversion Luminescence and Energy Transfer in the Fluorophosphate Glasses with low Phosphate Content doped with Tm^{3+} and Yb^{3+} ions / J. of Non-Crystalline Solids: X. – 2021. – Vol. 11-12. – P. 100065.

[8] Quantitatively identifying the upconversion luminescence mechanisms of Er^{3+}:$NaYF_4$ nanocrystals in aluminosilicate glass ceramics under femtosecond laser field / X. Xie [et al.] // J. Lumin. – 2023. – Vol. 255. – P. 119567.

[9] Realizing up-conversion fluorescence tuning in lanthanide-doped nanocrystals by femtosecond pulse shaping method / S. Zhang, et al. // Scientific Reports. – 2017. – Vol. 7. – P. 45650.

[10] Line Verification in Luminescence Spectra of Fluorophosphate Glass Doped with Ytterbium and Thulium Ions by the Degree of Nonlinearity of Up-Conversion Processes / M. V. Korolkov [et al.] // Journal of Applied Spectroscopy. – 2023. – Vol. 90. – P. 982–987.

[11] Visible and near infra-red up-conversion in Tm^{3+}/Yb^{3+} co-doped silica fibers under 980 nm excitation / D. A. Simpson et al. // Optics Express. – 2008. – Vol.16. – P. 13781.

[12] Power dependence of upconversion luminescence in lanthanide and transition-metal-ion systems / M. Pollnau [et al.] // Phys. Rev. B. – 2000. – Vol. 61. – P. 3337–3346.

[13] Nyk M., Wawrzynczyk D., Parjaszewski K., Samoc M. Spectrally Resolved Nonlinear Optical Response of Upconversion Lanthanide-Doped $NaYF_4$ nanoparticles / J. Phys. Chem. C. – 2011. – Vol. 115. – P. 16849 – 16855.

[14] Riedener T., Guedel H., McFariane R. A. Infrared to visible upconversion in $Cs_3Yb_2C_{19}$:Tm^{3+} / J. Lumin. – 1995. – Vol. 63. – P. 327 – 337.

[15] Excitation scheme for the upper energy levels in a Tm:Yb:BaY_2F_8 laser crystal / M. A. Noginov [et al.] // J. Opt. Soc. Am. B. – 1997. – Vol. 14. – P. 2126 – 2136.