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Spectroscopic Characteristics of Pr-Doped $Na_{0.4}Y_{0.6}F_{2.2}$ Crystals, promising for lasing in near IR and visible range.

A.M. Tkachuk, S.E. Ivanova
ITMO University, Russia
e_mail: am_tkachuk@mail.ru

A.A. Mirzaeva
Vavilov State Optical Institute,
St. Petersburg, Russia
e_mail: amirzaeva@mail.ru

M.-F. Joubert, Y. Guyot
Institut Lumiére Matiére, Université de Lyon, Lion, France

***Abstract*—Spectroscopic characteristics of intracenter transitions in disordered crystals of sodium-yttrium double fluoride Na0.4Y0.6F2.2: Pr3+ with cubic structure, grown by Stockbarger-Bridgeman method, are investigated theoretically and experimentally in order to evaluate their potential for application as active media of solid state lasers emitting in visible spectral range.**

Keywords— cubic fluoride crystals, praseodymium, absorption spectra, oscillator strengths, intensity parameters, branching coefficients, lifetimes, luminescence decay kinetics

I. INTRODUCTION

In the last decade, interest in the spectroscopic properties of crystals doped by ions of rare-earth elements has rekindled. These crystals are especially promising for practical applications as active media for solid-state lasers in the UV, visible, and mid-IR ranges. The upconversion effect is widely used in quantum counters, visualizers, and nanomarkers. In recent years, development of laser diodes emitting in visible range of the spectrum, stimulated interest to Pr^{3+} doped crystals, which have a broad absorption band in the range of 440–480 nm.

II. RESULTS AND CONCLUSION

We studied the series of cubic fluoride crystals $Na_{0.4}Y_{0.6}F_{2.2}$:Pr^{3+} (NYF:Pr^{3+}) with the praseodymium concetration 0.04–9 at % grown by using the Bridgman–Stockbarger technique. All grown single crystals were high optical quality without veils, visible inclusions, bubbles and scattering centers.

The optical spectra of NaYF:Pr^{3+} crystals at room and low (7 K) temperatures in the range of 200–2800 nm were investigated. The low-temperature absorption spectra of NYF:Pr^{3+} crystals have been shown to consist of broad weakly structured bands.

Based on the analysis of low-temperature absorption spectra, the positions of excited multiplets of Pr^{3+} ions were determined. The structure of the Stark splitting of praseodymium levels has been represented in terms of a model of "quasi-centers", which are characterized by an inhomogeneous broadening of Stark components. The energy gaps ΔE_{ji} between praseodymium multiplets were further used to estimate the rates of nonradiative transitions from excited radiative levels.

From experimental absorption spectra at T = 300 K, we have calculated absorption cross-section spectra and the oscillator strengths for transitions from the ground state $^3H_4$ to excited multiplets $^3H_5$, $^3H_6$, ^3F_j (j = 2, 3, 4), $^1G_4$, $^1D_2$, and (^3P_j, $^1I_6$) (j = 0, 1, 2). The maximal values of the peak absorption cross-sections belong to bands that are located in the visible range (λ_{abs} = 441 nm; transitions $^3H_4 \rightarrow {}^3P_{0,1,2}$, $^1I_6$) and in the near-IR range of the spectrum (λ_{abs} = 1526 nm; transition $^3H_4 \rightarrow {}^3F_{3,4}$). Corresponding experimental values of the peak absorption cross sections from the ground state $^3H_4$ are σ_{abs}(441 nm) = 1.01 × 10^{-20} cm^2 and σ_{abs}(1526 nm) = 1.41 × 10^{-20} cm^2.

Using the Judd–Ofelt method, we have determined intensity parameters Ω_t for NYF:Pr^{3+} crystal: Ω_2 = 0, Ω_4 = 4.4 x 10^{-20}, and Ω_6 = 2.28 x 10^{-20} cm^2. With these values, we have calculated the probabilities of radiative transitions, the branching coefficients, and the lifetimes of the radiative levels $^1D_2$ and $^3P_0$. The probabilities of multiphonon nonradiative transitions in NYF:Pr^{3+} crystals have been estimated.

Using the method of kinetic spectroscopy with selective excitation, we have investigated the luminescence decay kinetics of Pr^{3+} from the $^3P_0$ and $^1D_2$ levels upon their selective resonant excitation by nano-second laser pulses. We showed that the decay of the luminescence from the $^1D_2$ and $^3P_0$ levels in NYF:Pr^{3+} crystals with a Pr^{3+} concentration of 0.4 at % has a nonexponential character at the initial stage of the decay. For the radiative $^3P_{0,1}$ level, we obtained good agreement between the calculated and experimental radiative and nonradiative relaxation rates. For the $^1D_2$ level we observed a considerable discrepancy between the experimental and calculated values of the lifetime. This discrepancy can be explained by the fact that the $^1D_2$ level in the specimen with the concentration of 0.4 at % is partially quenched as a result of the self-quenching processes.

Based on the obtained results, we have inferred that NYF:Pr^{3+} crystals are processable; their high incorporation coefficient of Pr^{3+} ions K_{Pr} ~ 0.9 allows to obtain crystals with a high praseodymium concentration, up to 90 at.% (1.3×10^{22}cm^{-3}) without worsening of the optical quality. These crystals can be considered as new materials that are promising for the use as active media for optical converters and solid-state lasers in the visible and IR ranges (~2.3 μm) with a continuous tuning of the radiation frequency.