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Stark level structure and oscillator strengths of Nd³⁺ ion in different fluoride single crystals

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Abstract

Optical properties of three fluoride single crystals (YF₃, LiLuF₄ and KY₃F₁₀) doped with Nd³⁺ ions have been studied. Low temperature absorption spectra lead to the Stark energy level scheme up to 29 000 cm⁻¹. From room temperature absorption measurements and using the Judd–Ofelt theory, the Ω_2 , Ω_4 and Ω_6 parameters have been obtained in the LiLuF₄:Nd³⁺ and KY₃F₁₀:Nd³⁺ crystals and radiative lifetimes have been calculated. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Recently there has been considerable progress in the development of up-conversion lasers based on rare earth doped materials [1]. It has been shown for the Nd³⁺ ions that the operation of up-conversion laser in the blue to ultraviolet (UV) spectral regions depends strongly on the crystal host [2,3]. It is therefore important to understand the factors determining the lifetimes of the ²P_{3/2} and ⁴D_{3/2} upper states of the 4fⁿ configuration of Nd³⁺ incorporated in different surroundings. These emitting levels of the Nd³⁺ ions have rather small energy gaps to the levels below them and the low phonon frequencies of the fluorides result in good quantum efficiencies for blue or near UV luminescence from these levels. ⁴F_{3/2} is also an important energy level to investigate as it can serve as population reservoir for different scheme of up-conversion pumping and because it is a well known starting level for infrared (IR) stimulated emissions.

In this work the optical properties of the Nd³⁺ ions have been studied in three fluoride single crystals (YF₃, LiLuF₄ and KY₃F₁₀). Raman spectra have been done to estimate the highest vibrational frequency in each host. Judd–Ofelt parameters were derived from the absorption spectra and used to calculate the radiative lifetimes of the ⁴F_{3/2}, ⁴D_{3/2}

and ²P_{3/2} levels in the different crystals. Moreover, the absorption spectra obtained at liquid helium temperature permit to obtain the energy level schemes for the Stark components of the Nd³⁺ manifolds in these crystals.

2. Experimental

The samples studied (YF₃:Nd³⁺, LiLuF₄:Nd³⁺ and KY₃F₁₀:Nd³⁺) were grown from carbon crucibles using the Stockbarger method. As determined by electron beam micro-probe analysis, the Nd³⁺ concentration is 4.13 10¹⁹ at cm⁻³ (0.29 at.%) in Nd:LiLuF₄ (refractive index 1.468, thickness 0.304 cm with the crystallographic c axis perpendicular to this thickness) and 2.37 10²⁰ at cm⁻³ (1.51 at.%) in Nd:KY₃F₁₀ (refractive index 1.4712 and thickness 0.359 cm). The Nd:YF₃ crystal is too small for the concentration to be measured but 1% of Nd was introduced during the growth; it is also too small to be oriented and cut along the crystallographic directions, so we did not record polarized spectra with it.

Absorption spectra in the range from 200 to 900 nm were recorded using a Cary 2300 Varian Spectrophotometer equipped with a continuous flow helium refrigerator. Experimental fluorescence decays were measured under pulsed laser excitation at 355 nm (for the ⁴D_{3/2} and ²P_{3/2} decays) or at 733 nm (for the ⁴F_{3/2} decays). Raman spectra

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