

## Optical spectroscopy of molybdates with composition $\text{Ln}_2\text{Zr}_3(\text{MoO}_4)_9$ (Ln: Eu, Tb)

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### ABSTRACT

This study examined the luminescent properties of systems of double molybdates doped with Eu and Tb — this includes emission, excitation and absorption spectra. The compounds exhibited bright luminescence in the visible region due to the  $[\text{Xe}]4f^n \rightarrow [\text{Xe}]4f^{n-1}$  transitions of rare-earth ions. The luminescence is excited both in the  $[\text{Xe}]4f^n \rightarrow [\text{Xe}]4f^{n-1}$  bands of the rare-earth ions and in the wide band in the ultraviolet region related to the transitions within the Mo-O complex. Because of the energy transfer, the luminescence of  $\text{Tb}^{3+}$  and  $\text{Eu}^{3+}$  in doubly doped molybdates has had common excitation energies.

### 1. Introduction

Sheelite-like compounds show promise as phosphors, scintillators and laser media which host lattice, are capable of storing a large amount of dopant due to highly distorted arrangement of anionic groups [1–5]. Molybdates doped with trivalent europium are excellent red phosphors and as such can be applied in three-color LEDs with effective ultraviolet pumping with near ultraviolet (NUV)-InGaN chips to the  $[\text{Xe}]4f^6 \rightarrow [\text{Xe}]4f^6 \text{Eu}^{3+}$  band  ${}^7\text{F}_0\text{--}{}^5\text{L}_6$  (395 nm) [6,7]. Studies have also shown that terbium molybdates are good luminophores; the luminescence  $[\text{Xe}]4f^8 \rightarrow [\text{Xe}]4f^8$  bands are distributed in a wide spectral region with their maximum in the green region. For practical application of these materials, it is important to know the color of the luminescence of a rare-earth ion (REI) for a particular matrix, or one would need to be able to change the color of the phosphor. Two types of molybdate powders with terbium and europium ions can be used to obtain a different color of luminescence, which is similar to how RGB (tri-color) LEDs work. Otherwise, we can synthesize a molybdate powder, doped twice with terbium and europium. In the luminescence spectrum of such a material, bands characteristic of both ions will be present, but in this case we do not know exactly how the ions will interact with each other. In this paper we discuss the results of spectroscopic studies of trivalent rare-earth ions in a matrix of double zirconium molybdate. The REI-REI and REI-host lattice interactions are described, and some parameters important for practical application are calculated.

### 2. Material and methods

The molybdate powders with composition  $(\text{Eu}_x\text{Tb}_{1-x})_2\text{Zr}_3(\text{MoO}_4)_9$  ( $x = 1, 0.9, 0.5, 0.1, 0$ ) were synthesized according to the ceramic technology by stepwise annealing a mixture of the stoichiometric amounts of  $\text{Eu}_2\text{O}_3$ ,  $\text{Tb}_4\text{O}_7$ ,  $\text{MoO}_3$  and  $\text{ZrO}_2$  for 150 h to a maximum temperature of 700 °C, according to the procedure described in Ref. [8]. The measured X-ray diffraction pattern for  $\text{Tb}_2\text{Zr}_3(\text{MoO}_4)_9$  is shown in Fig. 1.

The crystal structure of the host lattice is a three-dimensional lattice in which the polyhedra  $\text{LnO}_9$  (Ln = Eu, Tb) and  $\text{ZrO}_6$  octahedra are arranged in a rhombohedral order, connected through common oxygen vertices of two types to bridge the Mo tetrahedrons.

The luminescence spectra were recorded with a double monochromator SDL-1 (LOMO) with 600 lines/mm grating and a photoelectron multiplier FEU-106; excitation was carried out using a high-pressure 150 W xenon arc lamp DKSH-150 through a MDR-2 monochromator with a diffraction grating of 1200 lines/mm. The powder was applied to a transparent, undoped LiF crystal. The PLE spectra were corrected due to varying intensity of exciting light. Measurements of the decay times of the luminescence were carried out with excitation of a pulsed argon lamp with a pulse duration of 1.5  $\mu\text{s}$ . Photoluminescence decay curves were recorded on Rigol DS1102E oscilloscope connected to the FEU-106 photomultiplier.

The photoluminescence excitation (PLE) spectra were measured with grating monochromators MDR2 and 150 W xenon arc lamp for

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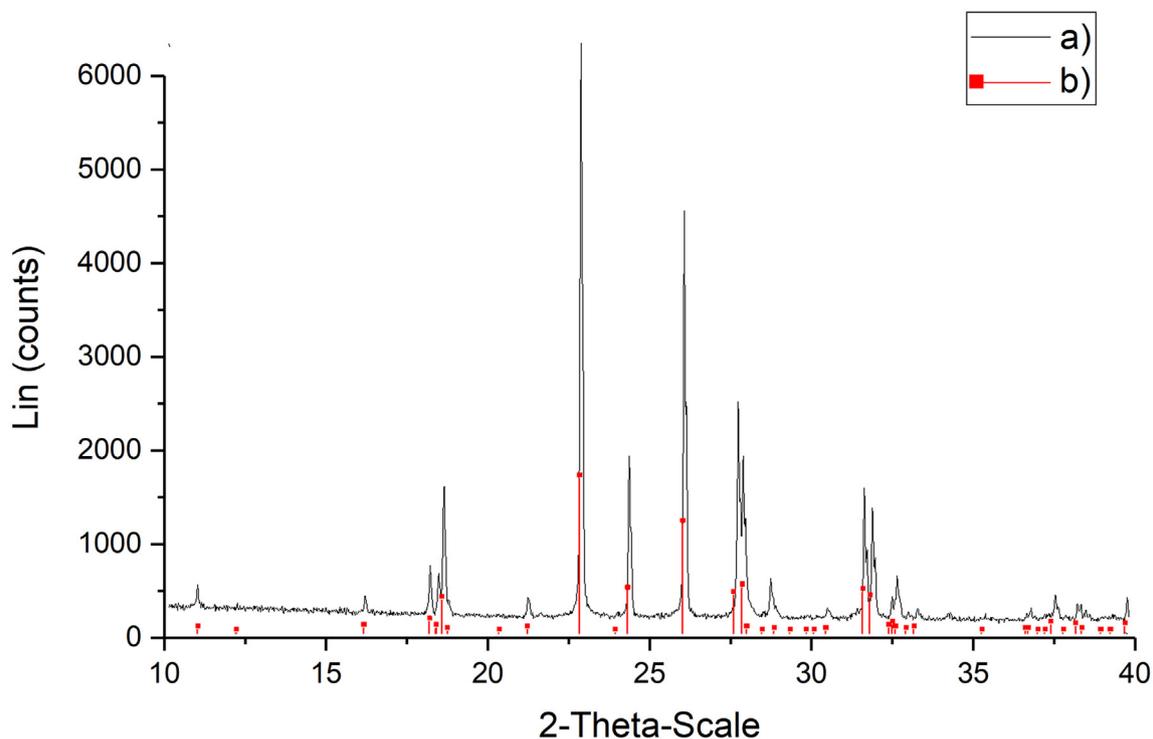


Fig. 1. Measured and calculated X-ray diffraction patterns of a)  $\text{Tb}_2\text{Zr}_3(\text{MoO}_4)_9$  b)  $\text{Eu}_2\text{Zr}_3(\text{MoO}_4)_9$ .

300–500 nm spectral region, a VM-2 (LOMO) vacuum monochromator, and a Hamamatsu deuterium lamp L7292 for measurements in the vacuum ultraviolet (VUV) spectral region. The optical absorption spectra were obtained by a Perkin-Elmer Lambda 950 UV/VIS/NIR spectrophotometer equipped with an integrating sphere at 300 K.

### 3. Results and discussion

The absorption spectra for the samples of  $\text{Tb}_2\text{Zr}_3(\text{MoO}_4)_9$  and  $\text{Eu}_2\text{Zr}_3(\text{MoO}_4)_9$  are given in Fig. 2.

Spectra show broad absorption bands at about 320 nm, identical for both samples, and the narrow weak bands are assigned to  $[\text{Xe}]4f^n \rightarrow [\text{Xe}]4f^{n-1}5d$  electronic transitions of the rare earth ions. The shortest wavelength line peaked at 395 nm corresponds to  $[\text{Xe}]4f^6 \rightarrow [\text{Xe}]4f^6$

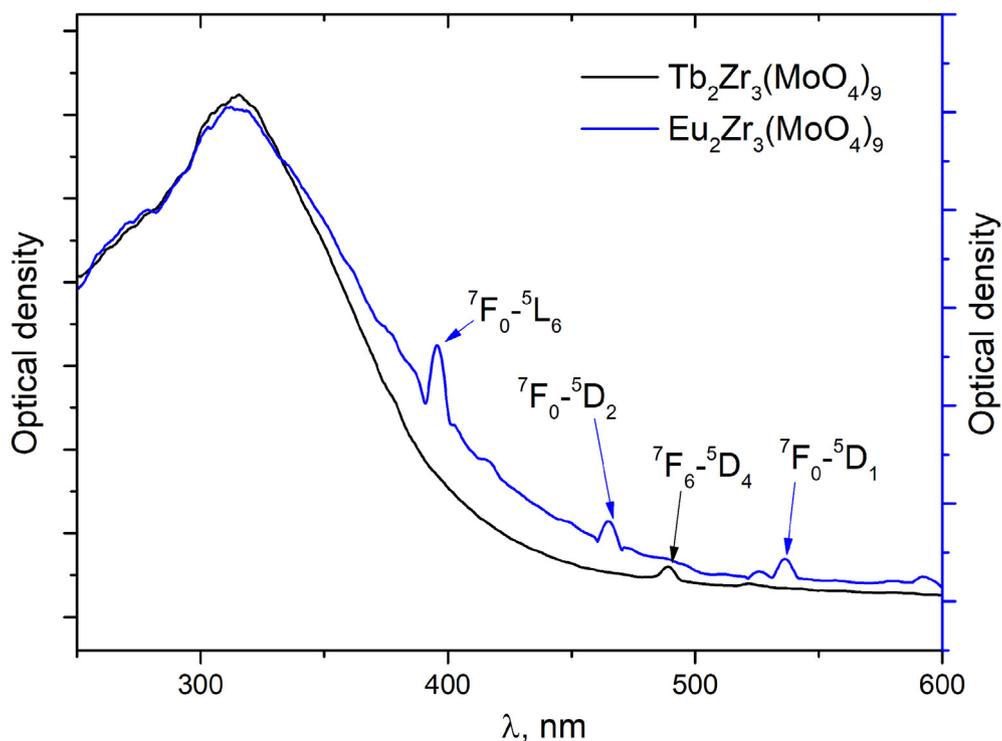


Fig. 2. The absorption spectra of  $\text{Tb}_2\text{Zr}_3(\text{MoO}_4)_9$  and  $\text{Eu}_2\text{Zr}_3(\text{MoO}_4)_9$ .

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