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Growth and Polarized Spectroscopy of Red-Emitting Monoclinic Eu:CsGd(MoO₄)₂ Crystal with a Layered Structure

Anna Volokitina^{1,2,*}, Pavel Loiko³, Anatoly Pavlyuk⁴, Sami Slimi¹, Rosa Maria Solé¹, Magdalena Aguiló¹, Francesc Díaz¹, and Xavier Mateos¹

1. Universitat Rovira i Virgili (URV), Marcel·lí Domingo 1, 43007 Tarragona, Spain

2. ITMO University, 49 Kronverkskiy Pr., 197101 St. Petersburg, Russia. *E-mail: anna.itmo@gmail.com

3. Centre de Recherche sur les Ions, les Matériaux et la Photonique (CIMAP), UMR 6252 CEA-CNRS-ENSICAEN, Université de Caen Normandie, 6 Boulevard du Maréchal Juin, 14050 Caen Cedex 4, France

4. A.V. Nikolaev Institute of Inorganic Chemistry, Siberian Branch of Russian Academy of Sciences, 3 Lavrentyev Ave., 630090 Novosibirsk, Russia

Trivalent europium ions (Eu³⁺) are known as luminescent centers leading to red emission of high color purity; the property utilized in red phosphors. Eu³⁺ ions are also recognized as structural probes (their luminescence is sensitive to the local site symmetry and its distortion). Recently, CW laser action was also demonstrated in certain Eu³⁺-doped crystals such as double tungstates KRE(WO₄)₂ (RE = Gd or Y) [1]. The laser operation was relying on the ⁵D₀ → ⁷F₄ transition corresponding to the deep-red spectral range (~700 nm). We report on the growth, structural study and polarized spectroscopy of another promising crystal, CsGd(MoO₄)₂, suitable for Eu³⁺ doping in high concentrations.

Single-crystals of cesium gadolinium double molybdate, 17 at.% Eu:CsGd(MoO₄)₂ (Eu:CsGdMo) were grown from the flux under low thermal gradients using cesium trimolybdate (Cs₂Mo₃O₁₀) as a solvent by A. Pavlyuk [2]. The as-grown crystals were transparent and had rose coloration, Fig. 1(a). Their structure (monoclinic, sp. gr. *P2*/*c*) was determined by powder XRD (lattice parameters: *a* = 9.5180(8) Å, *b* = 5.0775(3) Å, *c* = 8.0791(4) Å, β = 90.8998(0)°, obtained by the Le Bail method), Fig. 1(b). Eu:CsGdMo possesses a layered structure: the stacking of alternating layers – [Gd|EuO₈] – [MoO₄] – Cs – ... – occurs along the *a*-axis, Fig. 1(c). This determines the perfect natural cleavage of this material along the (100) plane, Fig. 1(d).

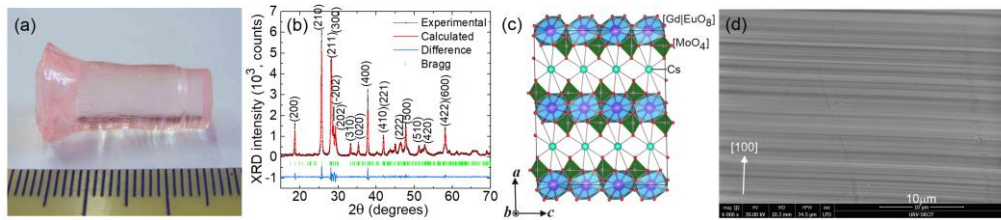


Fig. 1 17 at.% Eu:CsGdMo crystal: (a) Photograph of the as-grown crystal, the growth direction is along the [100] axis; (b) X-ray powder diffraction (XRD) pattern showing the Le Bail refinement, (*hkl*) – Miller's indices; (c) fragment of structure: a supercell (2*a* × 2*b* × 2*c*) in projection to the *a-c* plane; (d) ESEM image of a cleaved crystal plate (side view).

Under excitation by an UV lamp, Eu:CsGdMo provides intense red emission, see Fig. 2(a). Its color coordinates (CIE 1931) are determined. Eu³⁺ ions in CsGdMo exhibit extremely strong polarization-anisotropy of emission properties, Fig. 2(b), which is due to the low symmetry of the rare-earth site (C₂) and the layered crystal structure. The observed emissions originate from the ⁵D₀ → ⁷F_{*J*} (*J* = 1...6) Eu³⁺ transitions. No spectroscopic evidence of Eu²⁺ species is found. The most intense emission at ~614 nm is due to the ED allowed transition to the ⁷F₂ multiplet. The stimulated-emission (SE) cross-sections, σ_{SE}, for the potential laser transition, ⁵D₀ → ⁷F₄, are determined using the experimental branching ratio β_{*JJ'*} of 15.9%, see Fig. 2(c). The maximum σ_{SE} is 1.81 × 10⁻²⁰ cm² at 703.3 nm for light polarization *E* ∥ *b*. The luminescence decay from the metastable ⁵D₀ state is single exponential with a luminescence lifetime τ_{lum} = 693 μs. Highly-doped Eu:CsGdMo crystals are promising for solid-state lighting and laser applications.

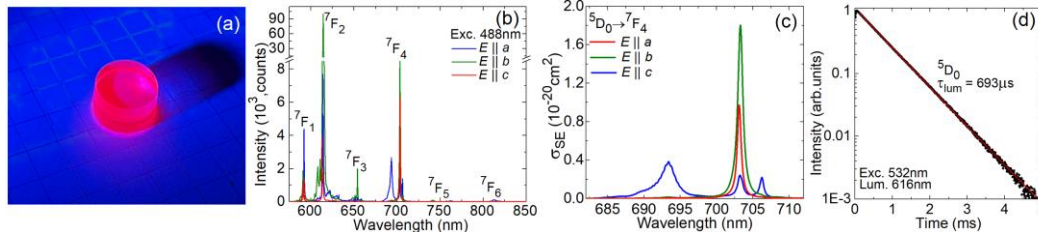


Fig. 2 Emission properties of Eu³⁺ ions in monoclinic CsGdMo: (a) photograph of red luminescence under excitation by an UV lamp; (b) polarized luminescence spectra, λ_{exc} = 488 nm; (c) SE cross-sections, σ_{SE}, for the ⁵D₀ → ⁷F₄ transition; (d) luminescence decay curve, λ_{exc} = 532 nm, λ_{lum} = 616 nm.

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