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Excitation and temperature tuned photoluminescence in $HgGa_2S_4$ single crystals

V.V. Ursaki[a], P.C. Ricci[b,*], I.M. Tiginyanu[c], A. Anedda[b], N.N. Syrbu[d], V.E. Tezlevan[d]

[a]*Institute of Applied Physics, Academy of Sciences of Moldova, 2028 Chisinau, Moldova*
[b]*INFM, Dipartimento di Fisica, Universitá di Cagliari, Cittadella Universitaria S.P. Monserrato-Sestu Km 0-700-09042 Monserrato, Cagliari, Italy*
[c]*Laboratory of Low-Dimensional Semiconductor Structures, Technical University of Moldova, 2004 Chisinau, Moldova*
[d]*Center of Nanoelectronics, Technical University of Moldova, 2004 Chisinau, Moldova*

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Abstract

Mercury thiogallate single crystals are shown to exhibit bright photoluminescence (PL) tunable in a wide spectral range from green to red when changing the excitation power density, wavelength and sample temperature. Taking into account the excitation and temperature dependencies of the PL characteristics, a model accounting for the radiative electron transitions in $HgGa_2S_4$ is proposed. The values of the energy gap extrapolated to 0 K and of the linear and quadratic temperature coefficients of the energy gap [$Eg(0) = 2.997$ eV; $a = -1.34 \times 10^{-4}$ eV K^{-1}; $b = -1.29 \times 10^{-6}$ eV K^{-2}] were deduced from PL excitation experiments.

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1. Introduction

Mercury thiogallate, $HgGa_2S_4$, is a defect chalcopyrite semiconductor with the space group S_4^2 (I4) [1,2], which offers a combination of attractive properties for applications. It was first prepared as powder by Hahn et al. [1]. Beun et al. [3] and Krausbauer et al. [4] obtained yellow needles of $HgGa_2S_4$ by vapor transport with iodine. Single crystals of $HgGa_2S_4$ were grown from the melt [5–9]. The growth of optical-quality ingots with the dimensions of $10 \times 15 \times 200$ mm^3 by the Lockheed Sanders Company using directed crystallization was recently reported [9]. $HgGa_2S_4$ crystals are characterized by high nonlinear susceptibility coefficients, wide transparency range (from 0.5 to 13 μm) and considerable birefringence [5]. The large birefringence ensures phase matching in a wide frequency range [6,7]. This material can be used for up-conversion of the frequency of the CO_2-laser radiation with an efficiency of up to 60%, for detecting radiation in the atmospheric transmission window (10–12 μm), and for visualizing the spectra of fast processes in the range from 8 to 12 μm [10]. Recently, a novel traveling-wave-type optical parametric generator based on mercury thiogallate was demonstrated [11]. A wave pumping of 1.25 μm $HgGa_2S_4$ produces tunable, high-power, transform-limited proportional to 200-fs pulses in the mid-IR from 5 to 9 μm. The conversion efficiency is more than two times better than the results obtained with an analogous sample of the widely studied $AgGaS_2$ compound. The advantages of $HgGa_2S_4$ in comparison with $AgGaS_2$ for efficient difference-frequency generation in mid-IR spectral range were demonstrated as well [12].

Nevertheless, little attention has been paid to the investigation of the basic physical properties of this material [4,9,13,14]. In this paper we study the radiative properties of $HgGa_2S_4$ single crystals and show that this material is promising for applications in light emitting devices.

2. Experimental

$HgGa_2S_4$ single crystals in the form of yellow needles

* Corresponding author.
E-mail address: ricci@alfis.dsf.unica.it (P.C. Ricci).

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