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Scintillation studies of CdS(In): effects of various semiconductor doping strategies

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Abstract

We present room-temperature photoluminescence and pulsed X-ray measurements of powder samples of CdS(In) codoped with three hole traps (Te, Ag, and Na). Te is an isoelectronic hole trap and Ag and Na are acceptor hole traps. The emission of CdS(In) excited at 430 nm is centered at \approx 520 nm (near the band edge) with \approx 20 nm FWHM. The emissions from CdS(Te) and the three codoped samples are shifted to longer wavelengths and are characterized by broad emission bands peaking near 630 nm. Whereas the decay of the CdS(Te) emission is nonexponential with times >10 ns, the decay of CdS(In,Te) is 3.3 ns and exponential over three decades. The decay of the acceptor-doped samples CdS(In,Ag) and CdS(In,Na) are also fast, 2.5 and 2.8 ns, respectively. These results show the potential for developing fast inorganic scintillators based on direct-gap semiconductors that can be codoped to provide fast radiative recombination. Additional work is needed to increase the luminosity.

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1. Introduction

Cadmium sulfide is a n-type direct-gap semiconductor characterized by excitonic and selfactivated luminescence. It has a density of 4.82 g/cm^3 and a band gap of 2.485 eV at 300 K. Many years ago Lehmann reported ultrafast (<1 ns) near-band-edge emission from In-doped CdS [1]. The intensity was moderate at low temperatures but low at room temperature. Red excitonic emission from CdS doped with the isoelectronic impurity Te was studied by Cuthbert and Thomas [2]. Subsequently Schotanus et al. investigated the scintillation properties of CdS:Te where

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moderately bright and fast emission was observed [3]. Recently, in the quest for a fast, bright scintillator, we obtained fast scintillation from CdS(In) codoped with Te [4]. Here, we report further investigations of CdS(In) codoped with the isoelectronic hole trap Te and also with the acceptor impurities Ag and Na.

2. Background

The luminescence of CdS has been well studied [5-7]. At low temperatures the emission consists of intrinsic and bound exciton emission in the range 486–508 nm and emission in the range 510–520 nm due to shallow ($\approx 30 \text{ meV}$) donors and deep ($\sim 150 \text{ meV}$) acceptors arising from various impurities. At room temperature an exciton band peaks at 505 nm. The binding energy of intrinsic excitons is 29.4 meV, hence at room temperature the band is weak because of thermal disassociation and hole trapping on nonradiative centers.

A donor band of electrons can be provided by cation dopants such as Al, Ga, In or anion dopants such as Cl. Indium is a shallow ($\approx 34 \text{ meV}$) donor in CdS. At high doping levels (>10¹⁹/cm³), the conduction band of CdS:In is degenerate and partly filled with donor electrons [8,9]. We seek to show that codopants can be added to CdS(In) to provide additional radiative centers with fast decay times.

3. Experimental

3.1. Material preparation

The samples were prepared from CdS powder (Strem Chemicals, 99.999% Puratrem), In powder, 325 mesh (Alfa Aesar, 99.999% Puratronic), CdTe powder (Strem Chemicals, 99.999%), Ag₂S powder (Alfa Aesar, 99.998% Premion), and NaCl powder (Alfa Aesar, 99.998% Premion), and NaCl powder (Alfa Aesar, 99.999% Puratronic). CdS(In) was prepared by incorporating 1% In into CdS via the following steps: (1) mix the dry powders in a vortex mixer, (2) seal the result a quartz ampoule under vacuum with a residual pressure of 5×10^{-5} Torr, (3) place the ampoule in

a tube furnace at 900 °C for 10 h, (4) cool the ampoule at a rate of ~200 °C/h, (5) crush the resulting material into a fine powder using a mortar and pestle and (6) repeat steps (2) to (5). CdS(Te), CdS(Ag) and CdS(Na) were prepared by incorporating 0.01% CdTe, or 0.05% Ag₂S, or 0.05% NaCl, respectively into CdS using steps (1) to (5) above. CdS(In,Te), CdS(In,Ag), and CdS(In,Na) were prepared by combining equal amounts of CdS(Te), CdS(Ag) and CdS(Na), respectively, and CdS(In) and performing steps (1) to (5). All dopant percentages relate to mole fractions.

3.2. Luminescence measurements

Powdered samples were contained in fused silica cuvettes for measurements. Decay times were measured using a pulsed X-ray system that produces 60 ps (FWHM) pulses of X-rays with a mean energy of 18 keV [10,11]. The attenuation length of 18 keV X-rays in CdS is approximately 100 μ m. Fluorescent photons from the sample are detected by a microchannel phototube with 35 ps (FWHM) response. Optical excitation and fluorescence spectra were recorded using a Fluorolog spectrophotometer. All measurements were made at room temperature.

4. Results

4.1. CdS(In)

Figs. 1 and 2 show the near-band-edge emission in CdS(In) prepared with 1% In as described above. The spectrum peaks at 520 nm. The decay time is approximately 0.2 ns, and the luminosity is approximately 1.5% of $Bi_4Ge_3O_{12}$ (BGO) powder [12].

4.2. CdS(Cd), CdS(Ag) and Cds(Na)

When a small amount (e.g. 0.01%) of excess Cd is incorporated into CdS, an intense near-infrared band peaking at 735 nm with a long (>1 µs) decay time is observed. This is similar to the intense green emission of ZnO(Zn) which is attributed to

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