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Photoluminescence and EPR studies on gamma irradiated Ce^{3+} doped potassium halide single crystals

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ABSTRACT

Electron Paramagnetic Resonance(EPR), Photoluminescence(PL), Thermoluminescence (TL) and other optical studies of γ -irradiated KBr, KCl: Ce^{3+} single crystals. Cerium when doped into the KBr, KCl is found to enter the host lattice in its trivalent state and act as electron trap during γ -irradiation, thereby partially converting itself to Ce^{2+} . The Photoluminescence(PL) spectra of both KCl and KBr crystals doped with Ce exhibit the strong blue emissions of Ce corresponding to $^5d(^2D) \rightarrow ^2F_{5/2}$ and $^5d(^2D) \rightarrow ^2F_{7/2}$ transitions. The defect centers formed in the Ce^{3+} doped KBr and KCl. Crystals are studied using the technique of EPR. A dominant TL glow peak at 374, 422 K and KCl: Ce^{3+} at 466, 475 K is observed in the crystal. EPR studies indicate the presence at two centers at room temperature. Spectral distribution under the thermoluminescence emission(TLE) and optically stimulated emission(OSL) support the idea that defect annihilation process to be due to thermal release of F electron in KBr, KCl: Ce^{3+} crystals. Both Ce^{3+} and Ce^{2+} emissions were observed in the thermoluminescence emission of the crystals.

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1. Introduction

Thermoluminescence(TL) is now extensively used for dosimetry of ionizing radiations. In recent years, we have developed sensitive TL single crystals, namely KI: CeTb and KBr: Tb^{3+} [1,2]. The electronic energy of the 4f compounds, the rare earth ions have become a subject of enduring interest as they are useful as a spectral probe to study the perturbation effects in a host lattice [3] as they are not much affected by the crystal field due to their inner $4f^n$ shell. Optical properties like absorption and photoluminescence measurements provide an insight into the various electronic transitions of the rare earth ion and their influence on the host lattice.

Substitution of impurities in a crystal lattice modifies the optical and other properties of the host. In most cases, the impurities enhance the performance of the host lattice. If the impurity is luminescent, then several types of luminescent centers can be obtained leading to various arrangements for the impurity in a crystal lattice. cerium is on a major rare earth studied in different host lattices mainly in powder and glass form [4–7]. The spectroscopic properties of cerium have been a subject of numerous investigations in different matrices [8–12]. Cerium doped alkaline earth haloborates [13] is studied for its potential use as storage phosphors for X-ray imaging and thermal neutron detectors [14–16].

Trivalent cerium doped materials could especially show one or two broader emission bands (300–390 nm) that are identified with the 5d–4f transitions. Cerium has been extensively used in

sensitized and charge transfer luminescence with terbium and europium ions [5]. Ce^{3+} energy levels in crystals consist of two ground states 4f levels separated roughly by about 2000 cm^{-1} ($^2F_{5/2}$ and $^2F_{7/2}$) and up to five 5d split excited state energy levels arising from 2D state of free Ce^{3+} ion [6]. Materials doped with Ce^{3+} attracted the interest of solid state researches due to applications of these materials as phosphors, scintillators for elementary particles, detectors for ionizing radiation, UV absorbing filters or UV emitters and activators for energy transfer [7,8]. Other applications are imaging, lithography, optical data recording [9], solid state lasers, displays, etc. The work aims to understand the luminescence properties of Ce ions, their role in the defect production and annihilation process and also to probe the mechanism of the thermoluminescence process in these crystals.

A large amount of work has been carried out in alkali halides doped with different rare earth like europium, gadolinium, samarium, etc [17,18]. Though a good amount of the literature is available on cerium, not much work could be traced to Ce^{3+} in the crystalline lattice especially the alkali halides with NaCl structure. Potassium halides due to their large ionic radius of K^+ is (1.33 Å) permit the substitutional incorporation of a large number of cations. This paper presents and discusses the results obtained on the photoluminescence and thermoluminescence of X/ γ -ray irradiated KBr and KCl crystals doped with Ce^{3+} .

EPR is a powerful method for investigating the detailed physical properties of paramagnetic defects in ionic materials, including cerium. The presence of a multitude of paramagnetic defects in natural cerium complicates the interpretation of the EPR signal. Nevertheless, natural cerium crystals have been studied

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intensively with EPR and many of the paramagnetic point defects have been identified and described. The EPR spectra of the rare earth ions Ce^{3+} and their paramagnetic centers play a key role in the damage formation and TL processes in natural cerium.

2. Materials and methods

$\text{KBr}:\text{Ce}^{3+}$ and $\text{KCl}:\text{Ce}^{3+}$ crystals were grown from 99.98% purity to KBr and KCl with 99.8% purity Ce by the Bridgman Stockbarger technique. The samples of size $4 \times 4 \times 4 \text{ mm}^3$ were used for all the spectroscopic studies except optically stimulated luminescence (OSL) and PL studies. For OSL and PL $5 \times 5 \times 3 \text{ mm}^3$ samples were used. The results due to the three concentrations in both the host lattices were similar except for a higher yield of Ce emissions at higher concentrations and hence only the results pertaining to a cerium concentration of 5% by weight are presented and discussed. Photoluminescence spectra were recorded at room temperature using Perkin Elmer LS 55 Luminescence spectrophotometer, equipped with a red sensitive photomultiplier R928 as light detector, in the region 200–900 nm with a spectral width of 5 nm. TL glow were recorded using a PC based TL 1007 analyzer at a heating rate of $120^\circ\text{C}/\text{min}$ supplied by Necleonic India Pvt. Ltd. The samples were irradiated using Co^{60} gamma ray source at a dose rate of $10.6 \text{ Gy}/\text{min}$. TL emission spectra were recorded using Perkin Elmer LS 55 with the excitation slit being closed. Before every experiment, the crystals were annealed at 400°C for half an hour and then quenched to room temperature to ensure homogeneous distribution of impurity and to remove any storage effects.

3. Results

3.1. Photoluminescence

To have a better understanding of the electronic states of the cerium ions, photoluminescence studies were made. The photoluminescence emission spectrum for excitation at 240 nm (Fig. 1(b)) for $\text{KBr}:\text{Ce}^{3+}$ is shown in Fig. 1. The emission spectrum for excitation at 240 nm (Fig. 1(b)) exhibits broad emission bands at 300 and 390 nm. The excitation spectrum for emission at 390 nm in $\text{KBr}:\text{Ce}^{3+}$ Fig. 1(a) shows a strong band at 330 nm. On excitation at 230 nm, $\text{KCl}:\text{Ce}^{3+}$ crystals exhibit broad emission bands at 300 and 390 nm (Fig. 2(b)). The excitation bands at 240 nm confirm that the absorption bands at 240 and 230 nm in the UV–vis spectra is due to the Ce^{3+} ions. The emission spectrum for excitation at 240 nm (Fig. 3(a and b)) reveals a broad band at 300 nm and a sharp band at

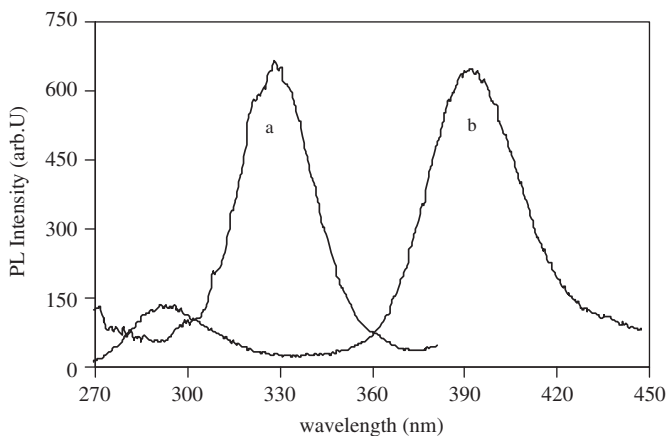


Fig. 1. PL spectrum of $\text{KBr}:\text{Ce}^{3+}$. (a) Excitation spectrum emission at 390 nm. (b) emission spectrum excitation at 240 nm.

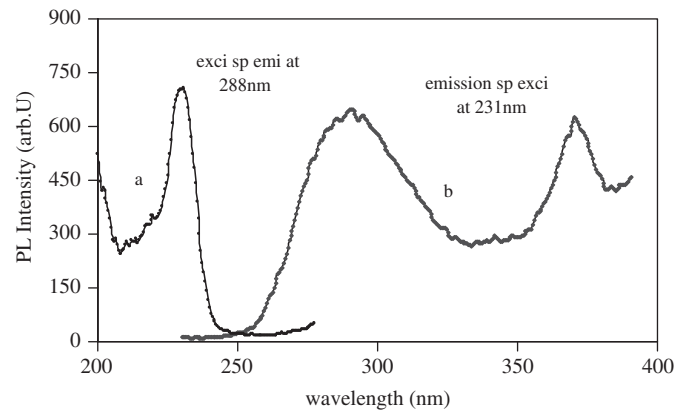


Fig. 2. PL spectrum of $\text{KCl}:\text{Ce}^{3+}$. (a) Excitation spectrum emission at 288 nm. (b) Emission spectrum excitation at 230 nm.

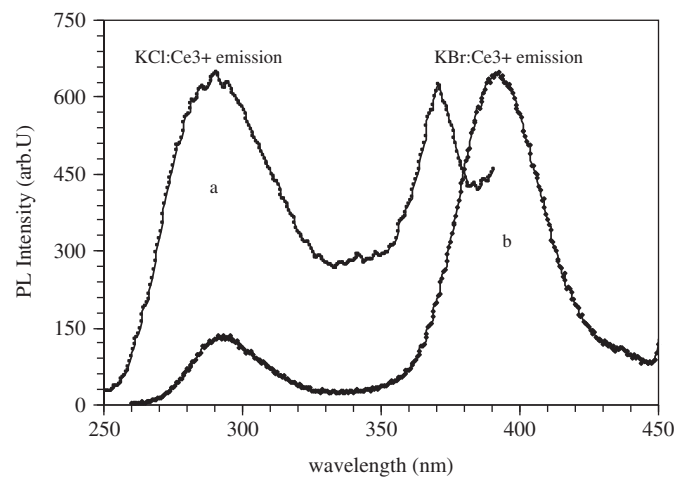


Fig. 3. PL spectrum (a) Ce^{3+} doped KCl emission spectrum excitation 230 nm. (b) Co-doped KBr emission spectrum excitation at 240 nm.

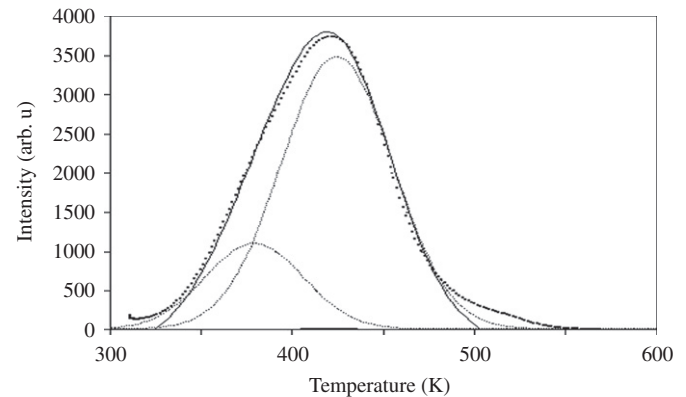


Fig. 4. TL glow curve of a $\text{Ce}^{3+}:\text{KBr}$ X-ray irradiated 5 min (deconvoluted).

390 nm for both Ce^{3+} doped KBr, KCl and this confirms the Ce^{3+} emission.

3.2. Thermoluminescence (TL) glow

The TL glow curves of $\text{KBr}:\text{Ce}^{3+}$ (Fig. 4) exhibit peaks at 374 and 422 K. On F bleaching the low temperature peak grows in intensity at the cost of the high temperature peak. The TL glow curve for $\text{KCl}:\text{Ce}^{3+}$ contains two peaks at 466 and 475 K (Fig. 5) thereby

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