

ORIGINAL RESEARCH



Luminescence properties of Tb³⁺, Eu³⁺ ions doped silicon dioxide

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Europium (Eu³⁺) and Terbium (Tb³⁺) singly doped and double doped with Silicon dioxide (SiO₂) is synthesized successfully by the sol-gel process. A xenon lamp light source with wavelength, $\lambda_e = 370$ nm is used for exciting the sample. The excitation spectra of Eu³⁺ ion may show applications in the area of UVB, near UVA and green light regions. Magnetic-dipole transition (⁵D₀→⁷F₁) is the dominant transition in emission spectra of Eu³⁺ ion. The emission spectra of Tb³⁺ ion reveal four strong emission peaks at 488nm, 545nm, 586nm and 623nm due to the transitions of ⁵D₄→⁷F₆, ⁵D₄→⁷F₅, ⁵D₄→⁷F₄ and ⁵D₄→⁷F₃ respectively with the transition ⁵D₄→⁷F₅ (green emission) as the most intense. The excitation of their co-doped spectra also evident that there is transfer of energy from Tb³⁺ ion to Eu³⁺ ion in silicon dioxide.

Received 05 Mar 2026
Accepted 31 Mar 2026*For correspondence:
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mizoacadsci@gmail.com**Keywords** : emission, europium, excitation, silicon dioxide, terbium

Introduction

Rare earth elements have many applications as a result of their unique spectroscopic properties, including long lifetime, numerous narrow and well-separated bands, and significant effective shifts from excitation to emission wavelength.¹⁻⁵ They can serve as ideal candidates for donors and acceptors when studying energy transfer (ET). Since energy migration between RE ions improves the emission intensity, lifetimes, quantum efficiencies. Interaction between luminescent ions that involve f-f transitions only with enhanced emission from one species. Energy transfer process has gained much technological importance in the fields of optoelectronics and photonics owing to its technological application point of view. The ET between Tb and Eu have attracted much attention because the emission bands of Tb³⁺ have overlap with some emission bands of Eu³⁺ and Tb³⁺ can act as a donor for the emission of Eu³⁺. The energy transfer process from Tb³⁺ to Eu³⁺ has been observed in different materials host such as LaPO₄,⁶ Gd₂O₃,⁷ YTaO₄,⁸ A₃Tb(PO₄)₃ (A=Sr, Ba) and Ca₃Tb₂Z₃O₁₂ (Z=Si, Ge),⁹ Sr₃B₂O₆,¹⁰ Ca₂Ga₂SiO₇,¹¹ zinc aluminoborosilicate,¹² lead borate,¹³ metaphosphate,¹⁴ aluminosilicate,¹⁵ zinc phosphate glass¹⁶. However, there is no report on Tb³⁺ ion, Eu³⁺ ion doped pure SiO₂, therefore, we performed such experiment in liquid form. The

results of Eu³⁺ ion:SiO₂ excitation confirmed that this is useful for sterilizing, disinfecting, killing bacteria and viruses and solid-state lighting devices. The excitation spectra of Tb:Eu:SiO₂ also revealed that there is transfer of energy from Tb³⁺ ion to Eu³⁺ ion in SiO₂ host.

Materials and Methods

The sol-gel process has been utilized for preparing 0.50 mol % of Tb³⁺ ion and 0.5 mol% Eu³⁺ ion doped with SiO₂. TEOS is used as main source of SiO₂. Terbium (III) nitrate hexahydrate and Europium (III) nitrate hexahydrate were separately first dissolved in 5 ml of methanol and deionized water. After that, 3 ml of SiO₂ is added in dropwise and stirrer for 1 hour. Next, the sample emission and excitation spectra were recorded in F-4700 Fluorescence Spectrometer.

Results and Discussions

The PL excitation and emission spectra of ZnS of 0.5 mol % of Eu³⁺ ion is shown in Figure 1(a) and Figure

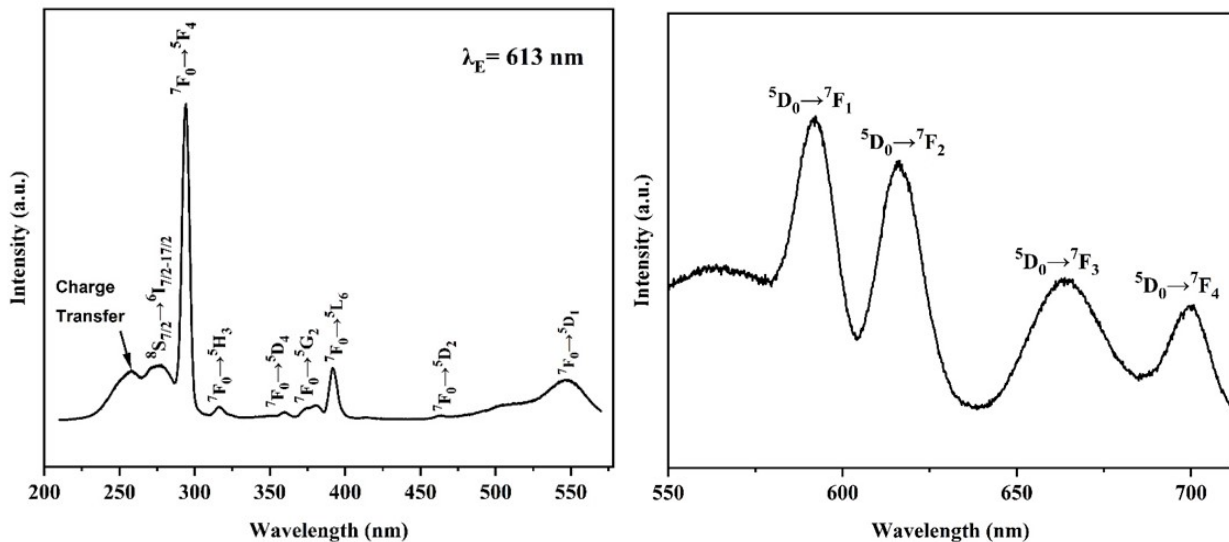


Figure 1. a. Excitation spectra of Eu^{3+} ion (0.5 %) in SiO_2 **b.** Emission spectra of Eu^{3+} ion in SiO_2 .

1(b). Figure 1(a) shows the excitation spectrum monitored at 613 nm which shows a sharp peak from 230- 580 nm due to the transition of $^8\text{S}_{7/2} \rightarrow ^6\text{I}_{7/2-17/2}$ (275 nm), $^5\text{F}_0 \rightarrow ^5\text{F}_4$ (294 nm), $^5\text{F}_0 \rightarrow ^5\text{H}_3$ (316 nm), $^5\text{F}_0 \rightarrow ^5\text{D}_4$ (359 nm), $^5\text{F}_0 \rightarrow ^5\text{G}_2$ (380 nm), $^5\text{F}_0 \rightarrow ^5\text{L}_6$ (391 nm), $^5\text{F}_0 \rightarrow ^5\text{D}_2$ (463 nm), $^5\text{F}_0 \rightarrow ^5\text{D}_1$ (553 nm). Three prominent peaks were observed at 294 nm, 391 nm and 553 nm which indicates that this Eu^{3+} ion: SiO_2 could be excited by the middle UV (UVB), near UV (UVA) light and green light¹⁷ which may find applications sterilizing, disinfecting, killing bacteria and viruses and solid-state lighting devices. Since the position of the $\text{O}^{2-} \rightarrow \text{Eu}^{3+}$ charge transfer band typically locates in the 200–300 nm range, the broad band from 257 nm is primarily caused by the charge transfer transitions of the ligand O^{2-} atom to Eu^{3+} ions. Figure 1(b) represents

the emission spectra under excitation with an UV light source with wavelength 370 nm. The spectra consist of four broad peaks at 578 nm, 593 nm, 613 nm, 653 nm and 703 nm which corresponds to transitions of $^5\text{D}_0 \rightarrow ^7\text{F}_0$, $^5\text{D}_0 \rightarrow ^7\text{F}_1$, $^5\text{D}_0 \rightarrow ^7\text{F}_2$, $^5\text{D}_0 \rightarrow ^7\text{F}_3$, and $^5\text{D}_0 \rightarrow ^7\text{F}_4$ respectively. The emission at around 578 nm and 593 nm may be assigned to magnetic-dipole allowed transition which obeys the selection rule $\Delta J = 1$ ¹⁸ which is independent of the local environment of the host lattice. $^5\text{D}_0 \rightarrow ^7\text{F}_0$ transition is also used for determining the environment in which the Eu^{3+} ions are bonded. The emission bands correspond to $^5\text{D}_0 \rightarrow ^7\text{F}_2$ (613 nm), $^5\text{D}_0 \rightarrow ^7\text{F}_3$ (653 nm) and $^5\text{D}_0 \rightarrow ^7\text{F}_4$ (703 nm) were a transition enabled by an electric dipole, and its intensity is hypersensitive to the variation of the bonding environment of the Eu^{3+} ions. The main causes of

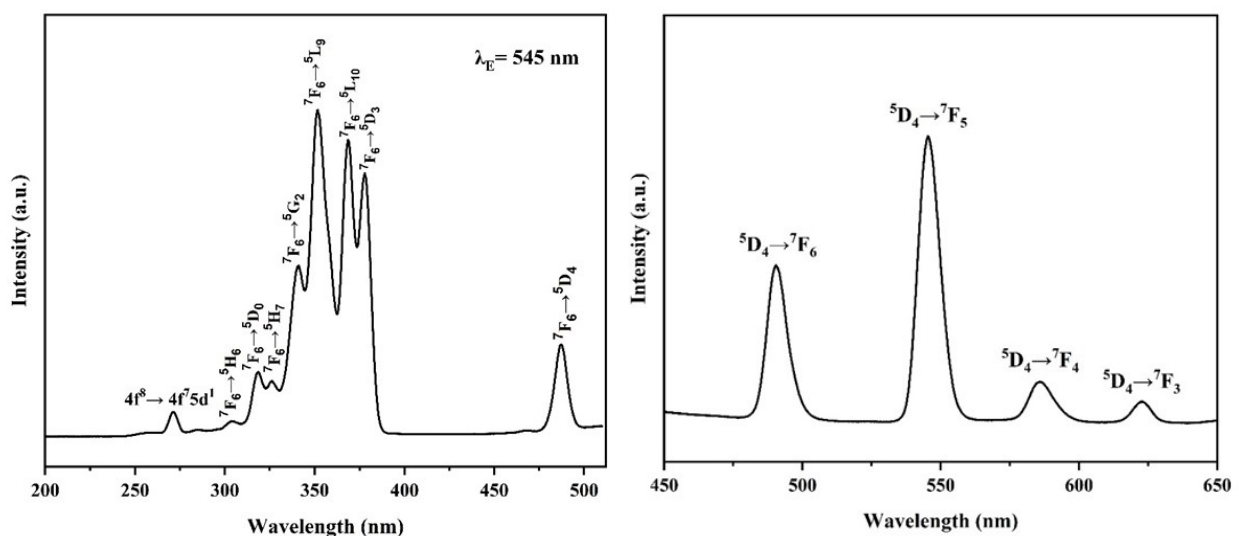


Figure 2. a. Excitation spectra of Tb^{3+} ion in SiO_2 **b.** Emission spectra of Tb^{3+} ion in SiO_2 .

electric dipole transitions are the lack of a centre of symmetry and the mixing of $4f^5$ orbitals with the opposite parity orbital.¹⁹ These transitions appear due to charge imbalance between Zn^{2+} and Eu^{3+} ions. Here, the magnetic-dipole transition $^5D_0 \rightarrow ^7F_1$ (593 nm) is more dominant than the electric-dipole transition $^5D_0 \rightarrow ^7F_1$ (613 nm), indicating that the most of Eu^{3+} locate at the inversion symmetry center in SiO_2 .²⁰

Figure 2(a) shows the excitation spectrum of Tb^{3+} ion (0.5%) when monitored at 545 nm which reveals several prominent peaks between 200-500 nm due to the transitions of ground state 7F_6 to the different excited states of Tb^{3+} i.e., at 303 nm (5H_6), 319 nm (5D_0), 326 nm (5H_7), 341 nm (5G_2), 352 nm (5L_9), 368 nm ($^5L_{10}$), 378 nm (5D_3) and 486 nm (5D_4) respectively.¹⁷ The sharp band observed at 275 nm is due to the spin allowed ($4f^8 \rightarrow 4f^7 5d^1$) and spin forbidden ($4f^8 \rightarrow 4f^7 5d^1$) transitions of Tb^{3+} ion. Figure 2(b) represents the emission spectra Tb^{3+} ion by exciting an UV light source wavelength 370 nm. The spectra reveal four strong emission peaks at 488nm, 545nm, 586nm and 623nm due to the transitions of $^5D_4 \rightarrow ^7F_6$, $^5D_4 \rightarrow ^7F_5$, $^5D_4 \rightarrow ^7F_4$ and $^5D_4 \rightarrow ^7F_3$ respectively. As expected, the transition from $^5D_4 \rightarrow ^7F_5$ (green emission) is the most intense and has the highest affinity for both electric-dipole and magnetic-dipole-induced transitions.²¹ It is also sensitive to the nature of the surrounding atoms. No emission was observed from higher level of 5D_3 due to cross-relaxation effect.

The inset of Figure 3(a) depicts the excitation spectrum of $Tb(0.5\%) : Eu(0.5\%)$ co-doped by monitoring of Tb^{3+} emission wavelength at 545

nm. The spectrum shows similar spectra observed for Tb^{3+} ion alone, no extra peaks of Eu^{3+} excitation were observed suggesting that there is no transfer of energy from Eu^{3+} to Tb^{3+} ion. The excitation spectrum of Tb (0.5%): Eu (0.5%) co-doped by monitoring of Eu^{3+} emission at 613 nm is shown in Figure 3(a). The spectrum reveals new prominent peaks different from Eu^{3+} (see Figure 1(a)) because of the combination of spin allowed/ spin forbidden ($4f^8 \rightarrow 4f^7 5d^1$) of Tb^{3+} and $O^{2-} \rightarrow Eu^{3+}$ charge transfer band of the Eu^{3+} ion. Because of this several peaks were observed at 303, 317, 351 nm due to the $^7F_6 \rightarrow ^5H_6$ and $^7F_6 \rightarrow ^5D_0$ transitions of Tb^{3+} and at 375, 393, 414, 463 and 553 nm due to $^5F_0 \rightarrow ^5D_3$, $^5F_0 \rightarrow ^5D_2$, $^5F_0 \rightarrow ^5D_1$, $^5F_0 \rightarrow ^5G_2$ and $^5F_0 \rightarrow ^5L_6$ transitions of Eu^{3+} respectively. Therefore, it is evident that the co-doped sample excitation spectrum contains every characteristic of the single-doped Tb^{3+} ion and Eu^{3+} ion. When the co-doped excitation spectrum is compared to the single doped glasses, the two match precisely in terms of spectrum summation (see Figure 1(a) and 2(a)). Consequently, energy transfer from the Tb^{3+} to Eu^{3+} ions is clearly demonstrated by the presence of the Tb^{3+} excitation peaks among the various photo-stimulation paths for the activation of the Eu^{3+} (613 nm) $^5D_0 \rightarrow ^7F_2$ transition. The Eu^{3+} excitation spectra and Tb^{3+} emission is also highlighted for direct comparison (see Figure 3(b)). Figure 3(b) reveal that there is strong overlap between Eu^{3+} excitation peaks $^5F_0 \rightarrow ^5L_6$ (391 nm), $^5F_0 \rightarrow ^5D_2$ (463 nm), $^5F_0 \rightarrow ^5D_1$ (553 nm) and Tb^{3+} emissions peaks $^5D_4 \rightarrow ^7F_6$ (488 nm), $^5D_4 \rightarrow ^7F_5$ (545 nm) and respectively clearly manifest the confirmation of possible transfer of energy from Tb^{3+} to Eu^{3+} in SiO_2 host.

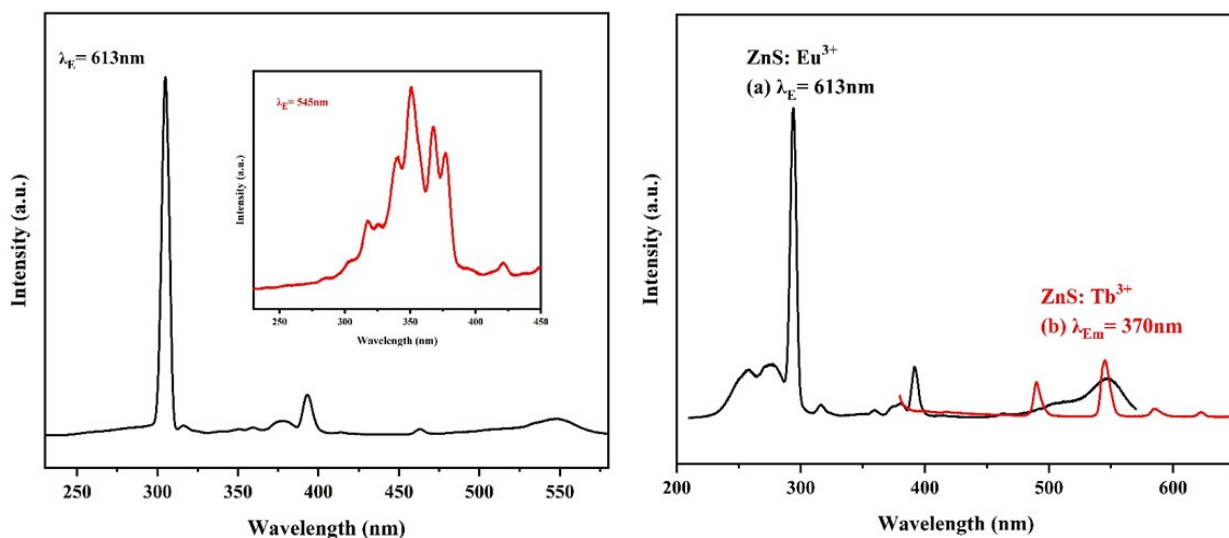


Figure 3. a. Excitation spectra of $Tb:Eu:SiO_2$ at $\lambda_E = 613$ nm and $\lambda_E = 545$ nm **b.** Spectral overlap between Tb emission and Eu excitation in SiO_2 .

Conclusion

The excitation and emission spectra of a single phase of Eu^{3+} ion and Tb^{3+} ion doped and co-doped with silicon dioxide have been studied. Eu^{3+} ion excitation spectrum three prominent peaks were observed at 294 nm, 391 nm and 553 nm which indicates this may find applications sterilizing, disinfecting, killing bacteria and viruses and solid-state lighting devices. Tb^{3+} ion emission spectra confirmed that green emission as the most intense and this transition has the highest affinity for both electric-dipole and magnetic-dipole-induced transitions. It is also evident that the co-doped sample excitation spectrum contains every characteristic of the single-doped Tb^{3+} ion and Eu^{3+} ion. The co-doped sample has characteristics peaks due to the combination of spin allowed/ spin forbidden of Tb^{3+} and $\text{O}^{2-} \rightarrow \text{Eu}^{3+}$ charge transfer band of the Eu^{3+} ion which clearly confirmed the transfer of energy from Tb^{3+} ion to Eu^{3+} ion.

Acknowledgement

The author is thankful to Ministry of Tribal Affairs, Govt. of India for their financial support through NFST with Award letter 202021-NFST-MIZ-0247.

Conflict of Interest

The author declares there is no conflict of interest.

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