

PHOTOLUMINESCENCE INVESTIGATION OF LANTHANIDE-DOPED GARNET PHOSPHOR COMPOUNDS

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ABSTRACT:

The research will involve a thorough structural, morphological, and compositional characterization will be conducted utilizing XRD, SEM/TEM, FTIR, and Raman spectroscopy after the synthesis of single- and co-doped garnet compounds using appropriate procedures such solid-state reaction and sol-gel techniques. To assess radiative and non-radiative processes, photoluminescence studies will be conducted, including excitation, emission, decay lifetime, and quantum yield measurements. Energy transfer modelling and Judd–Ofelt (J–O) research will provide further light on sensitizer–activator interactions and transition probabilities. For realistic device integration, temperature-dependent PL tests will evaluate the phosphors' thermal stability and quenching behaviour.

The goal of this methodical study is to clearly identify relationships between luminescence characteristics, dopant site occupancy, and structural aspects in garnet hosts. The discovery of optimal doping concentrations, increased emission intensities, and thermally stable phosphors with adjustable emission throughout the visible spectrum are among the anticipated results. These materials have a great deal of promise for usage in optical sensors, laser gain media, display devices, and white light-emitting diodes. Beyond applications, the research will improve our basic knowledge of site-selective spectroscopy in complicated oxide lattices, energy migration pathways, and host–dopant interactions.

KEYWORDS: Lanthanide-doped Garnets, Photoluminescence (PL), Judd–Ofelt (J–O) Analysis, Energy Transfer, Thermal Quenching, Solid-State Lighting (SSL) (or WLEDs)

1. INTRODUCTION

Luminescent stuff usually has two parts: a base material - often an inorganic compound with a wide band gap - and a glowing ion, like one from transition metals or lanthanides. Instead of just holding things together, the base shapes how the light-emitting ion behaves by controlling its surroundings. Materials like oxide-based crystals, garnets, aluminium-rich compounds, silicon mixes, or nitrogen-containing structures tend to last longer because they resist heat damage, don't break down under light, and stay stable in tough conditions. A major trait of glowing stuff is trapped electron spots where energized electrons can drop back via light-emitting routes. How well they glow hinges on how strongly light-releasing paths stack up against energy-loss ways, vibrations, flaws, or atomic arrangement quirks. Light behaviour gets adjusted using impurity levels, added helpers, shape tweaks, or changing the base structure's layout.

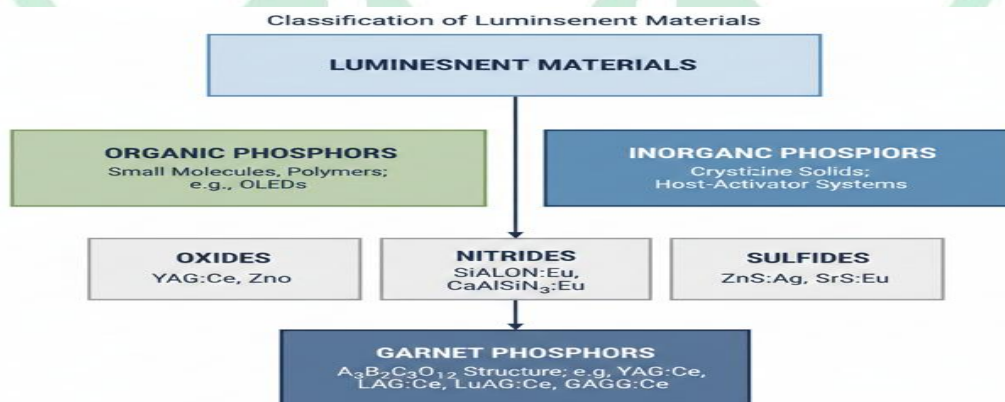


Figure 1: Diagram provides a hierarchical classification of luminescent materials

The figure 1, chart starts wide, covering all glowing stuff, then shifts - narrowing step by step - to focus on your type of material. As solid-state lights and electronic gadgets became common, better glowing materials are needed more than ever. Scientists are hunting fresh crystal structures that hold rare-earth elements well without breaking down or losing light quality. One promising option? Garnets - these $A_3B_2C_3O_{12}$ types stand out because they're tough, adaptable, and work great in bright LEDs or sensors that detect radiation.

A. The Nature of the {4f-4f} Transition

- Intra-configurationally means the shift happens between two energy levels inside the same $4f^n$ setup. Instead, these states are labeled using Russell-Saunders notation ($^{2s+1}L_j$ groups). For example, Eu^{3+} shows terms like 5D_0 or 7F_2 .
- Some jumps between energy levels aren't allowed - when both sides have the same symmetry, light can't easily make that switch. These kinds of shifts go against standard dipole rules, which control how atoms usually release light. So, a move within similar states, say f to f, just doesn't happen under normal conditions.
- Some allowance happens when the crystal's effect on atoms blends inner 4f levels with outer ones - say, from 4f to 5d - that have different symmetry. Because of this blend, the usual light rules don't block the shift completely; it's still faint but possible. That kind of jump gets called a triggered electric dipole move since outside forces tweak how electrons behave.

In short, lanthanide-doped phosphors are notable because of these reasons:

- Crackling, pinpoint bursts of light
- Pure colors plus steady light performance
- Wide energy span - common in 4f to 5d jumps
- Long luminescence lifetimes
- Firm power movement skills
- Stays strong when it gets hot or faces harsh chemicals
- Multicolour light use along with shifting wavelengths back or forth

These special light traits put lanthanide-filled phosphors at the heart of progress in today's photon systems - also seen in opto-electronic devices, screens, LED lights, or sensors that rely on glow effects.

This figure 2, gives a clear picture - helping you see how materials science fits together, while pointing out exactly where your work on Garnet Phosphors lands.

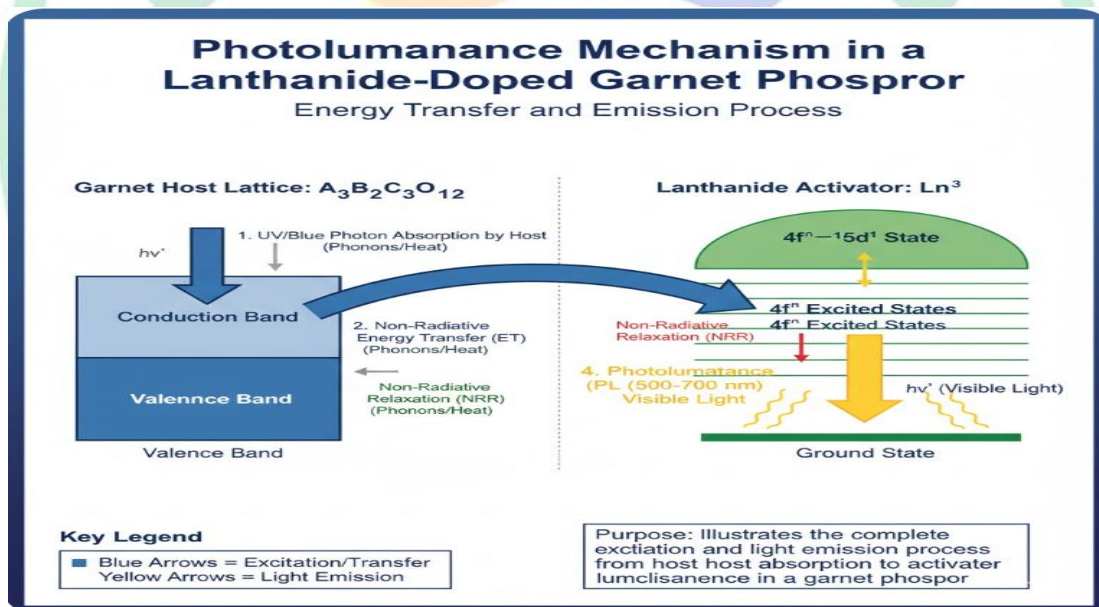


Figure 2: Classification of Luminescent Materials

B. Importance of Garnet-Based Host Matrices ($A_3B_2C_3O_{12}$)

Garnet-like oxides, written as $A_3B_2C_3O_{12}$, are among the most useful crystal frameworks around. What makes them stand out is their adaptable structure, along with solid heat resistance and steady chemistry. These traits go hand in hand with strong light-related and electrical behaviors. Because of that mix, they work well for holding different active additives. Those added elements let them perform in lighting tech, electronics, or radiation detection gear.

A big plus of the garnet setup is how it can hold different cations at dodecahedral (A), octahedral (B), or tetrahedral (C) spots while keeping the crystal frame stable. Because this structure bends but doesn't break, you can tweak things like spacing between atoms, vibration energy, and tiny shifts in electric fields - each playing a role in making dopants work better. So instead of one rigid option, garnets give a flexible base for rare-earth or transition metals that power lasers, glow-in-the-dark materials, and LED tech.

From how light moves through them, garnet structures let a wide range of wavelengths pass, bend light strongly, plus have little vibration noise - so glowing shifts happen more easily. Because of these traits, materials like $Y_3Al_5O_{12}$ (YAG) work well in strong lasers, while $(Lu,Y)_3Al_5O_{12}$ helps make white LED lights using phosphors. They can hold lots of added elements without losing glow strength, which makes them better for bright emitters or fast energy sharing.

The $A_3B_2C_3O_{12}$ garnet setup is tough to beat when it comes to flexibility and function. Because it's naturally stable but still easy to tweak, researchers keep using it for new kinds of light-based tech. On top of that, it's helping build better low-energy lights. It also plays a key role in improving tools that catch harmful radiation.

By swapping elements at A, B, or C spots on purpose, we can tweak how charges move, where they get stuck, also what light gets absorbed - so each use works better. Changing one part shifts how the whole system behaves, depending on needs in figure 3.

Dopant Substitution in Garnet Host Lattice: Influence on Photoluminescence

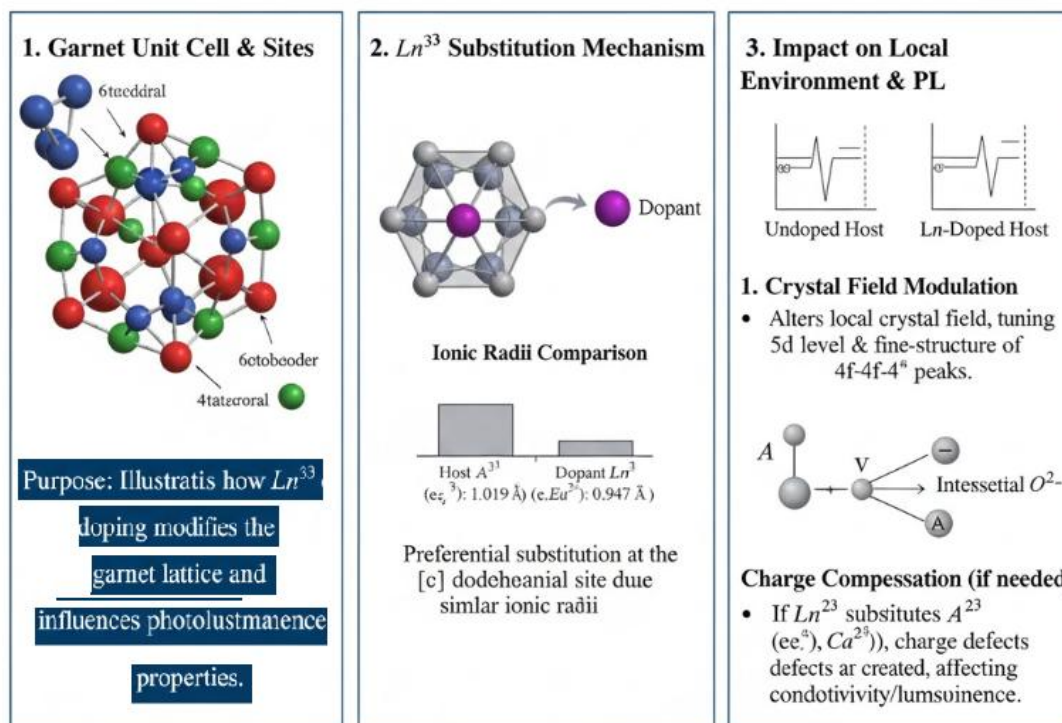


Figure 3: Dopant Substitution in Garnet Lattice

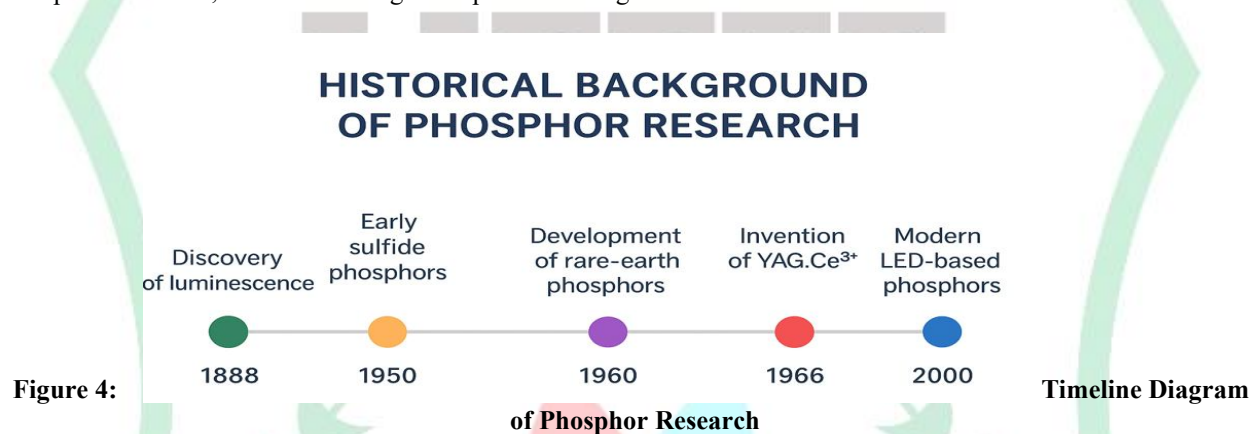
The $A_3B_2C_3O_{12}$ garnet setup is known for being flexible and useful in many ways. Because it's naturally stable, while also easy to adjust, researchers keep using it for new types of light-based tech and low-energy lighting solutions.

The target sketch shows how a lanthanide ion (Ln^{3+}) fits into a base crystal structure $A_3B_2C_3O_{12}$, using $Y_3Al_5O_{12}$ (YAG) as an example material. Instead of general terms, it zooms in on actual atom placement. While keeping clarity, it avoids vague descriptions. Because precision matters here, each step highlights real positioning. So rather than abstract ideas, the focus stays on physical integration within the lattice.

2. LITERATURE REVIEW

A. Historical Background of Phosphor Research

Start with old glow-in-the-dark stuff like sulfides - those powered early screens. Then came silicates, a bit better, lasting longer under electron beams in CRTs. Over time, these dim, unstable picks gave way to sharper, tougher mixes. Now, solid-state lighting runs on advanced crystals that shine bright with less waste heat. Each step dropped the weak links, kept what worked faster and cleaner. The arrival of the blue LED brought a tricky challenge - one only the garnet setup could handle, the timeline diagram represents on figure 4.



- **Benchmark Material:** Back in the late '90s, finding $Y_3Al_5O_{12}:Ce^{3+}$ - called YAG:Ce - and quickly bringing it to market was a game changer. This material's an oxide-based phosphor built on a garnet framework. Instead of matching just one color, its wide yellow-green glow pairs smoothly with blue LEDs. That combo makes bright white light without wasting much energy.
- **Advantage of Garnet Hosts:** Fast rise in use of YAG:Ce and similar materials like LuAG or GdAG happened because
 - The cubic garnet setup holds up really well - its toughness stands out.
 - Its high melting point plus minimal phonon energy cut down heat-related dimming, so brightness stays steady when powerful LEDs run hot.
 - Lanthanide Fit: That big twelve-sided spot works great for holding Ce^{3+} - also other Ln^{3+} ions - which helps bright light come out easily.

B. Overview of Garnet Structure and Crystal Chemistry

The sturdy framework along with adjustable chemistry of garnet-style oxides - formula $A_3B_2C_3O_{12}$ packing 120 oxygens per cell - makes them key in tech uses due to their role as base materials [1].

Crystallographic Features

Garnets form in a cube-like pattern, part of the $Ia\bar{3}d$ setup - space group 230. Their chunky building block holds 160 atoms, held together through a tight web of oxygen shapes wrapping around positive ions. Three separate spots where metal ions sit, each unique and not interchangeable.

1. Dodecahedral {c} Site (24c positions):

- **Arrangement:** eight-sided shapes (MO_8 units).
- **Cation:** taken up by an A ion - often a big, three-times charged rare-earth type such as Y^{3+} or Gd^{3+} , also Lu^{3+} [2].
- **Importance:** It's the biggest spot - main place where big Ln^{3+} activator particles fit best, mainly because space allows it more easily than elsewhere.

2. Octahedral [a] Site (16a positions):

- **Arrangement:** six-part (MO_6 shapes).
- **Cation:** taken up by element B - think tiny triple-charged particles such as Al^{3+} or Ga^{3+} .
- These closely connected octahedra make up the core framework, boosting strength and heat resistance in a major way.

3. Tetrahedral (d) Site (24d positions):

- Arrangement: four parts linked together (MO_4 shape).
- Cation: filled with C-type ions - commonly Al^{3+} or Ga^{3+} , sometimes Si^{4+} instead.
- Importance: These shapes link at corners, which helps pack the structure tightly.

The setup's like $A_3^{3+} B_2^{3+} C_3^{4+} O_{12}^{2-}$, or take YAG - there it's $Y_3Al_2Al_3O_{12}$, just rolled into $Y_3Al_5O_{12}$.

C. Isomorphous Substitution and Host Modification

The garnet structure can hold different-sized ions without falling apart - this staying power is called isomorphous substitution. That shift happens naturally, making it useful for crafting new materials [4]:

- Lanthanide ions fit well into the host material because they carry the same +3 charge and are fairly big. So, Ln^{3+} tends to replace A^{3+} at the c-site with twelve sides. When there's a difference in ion sizes, it bends the nearby structure just a bit. That small distortion changes how atoms interact with light. As a result, the color of emitted light can be adjusted precisely.
- Swapping out the main metal ions at A, B, or C spots changes how the garnet behaves - each shift tweaks performance a bit differently [5]:
 - Swapping bigger Y^{3+} ions with smaller Lu^{3+} ones - like in $Lu_3Al_5O_{12}$ or LuAG - shrinks the lattice spacing while boosting the local crystal field around doped ions. That shift typically pushes Ce^{3+} 's light emission toward shorter wavelengths; it also tends to improve heat resistance [6].
 - Swapping Al^{3+} with bigger Ga^{3+} ions - like in $Gd_3Al_2Ga_3O_{12}$, also called GAGG - makes the crystal structure expand. Because of this expansion, the electric field around dopant ions gets weaker. That shift causes Ce^{3+} light emission to move toward longer wavelengths. Changes in atomic vibrations happen at the same time, altering how quickly excited states lose energy without emitting light [7].

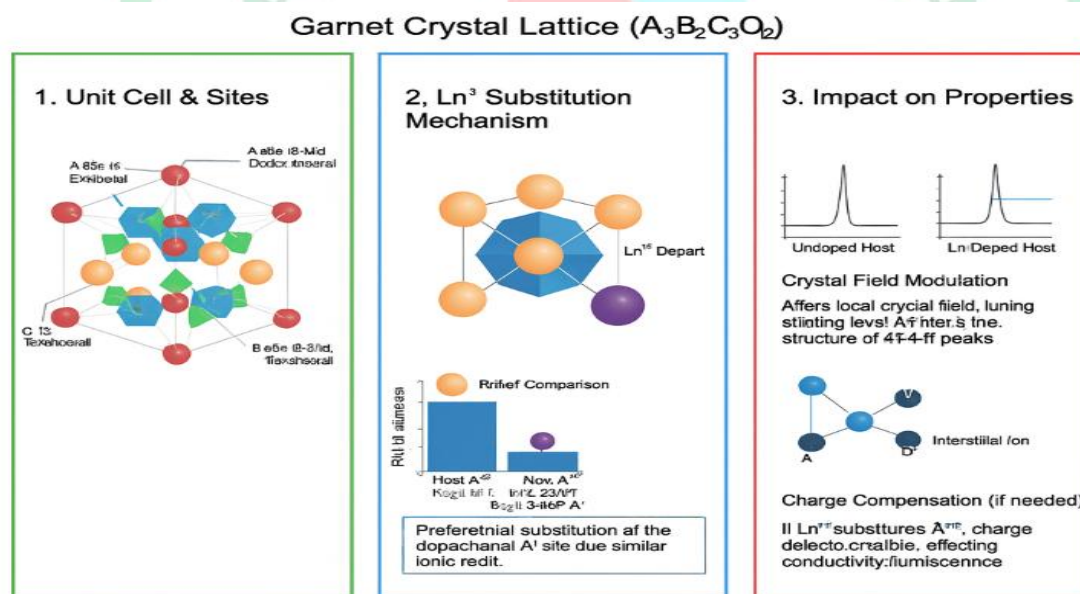


Figure 5: Garnet Crystal Lattice

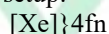
The strong $A_3B_2C_3O_{12}$ structure works like a flexible base, so scientists can carefully adjust how Ln^{3+} ions are surrounded - this helps fine-tune light output for particular uses in photonics represented on figure 5.

D. Lanthanide Ions: Electronic Configuration and Transitions

Lanthanide ions (Ln^{3+}) have special light-related traits thanks to their half-filled 4f shells. Because of this, they're widely used in glow materials, medical scans, lasers, or LED tech. Knowing how electrons are arranged - and how they shift - helps explain how these ions emit light when placed in garnet crystals [8].

Electronic Configuration of Ln^{3+} Ions

Trivalent lanthanide ions (Ln^{3+}) follow this electron setup:



with n going from 0 (in the case of La^{3+}) up to 14 (when it's Lu^{3+}), based on which element you're looking at [21].

The key trait shaping how they interact with light is the protection around their 4f electrons. Deep within the atom's

center, those 4f regions are tucked away - shielded by full outer layers of 5s² and 5p⁶ [9].

Consequences of 4f Electron Shielding:

- Crystals shape a faint force around atoms - this barely touches hidden 4f parts inside, so energy states stay clear. That's why light shots out in thin, unique bands when these bits react [10].
- Slight color changes: light output stays almost the same no matter the base - like YAG or LuAG [11].
- Shielding cuts down how much electrons interact with atomic jiggling, which means less heat messing things up. This also slashes energy leaks through vibrations - giving a solid edge without fluff or hype [12].

This setup explains why particles such as Eu³⁺ and Tb³⁺ show sharp color bands.

E. Types of Electronic Transitions in Lanthanide Ions

Ln³⁺ ions show two main types of changes shaping how they're used - either one involves shifting energy states while the other relies on magnetic interactions instead [13].

1. . 4fⁿ → 4fⁿ Transitions (Intra-configurational)

- These shifts happen inside the 4f orbit - like when 5D0 jumps to 7FJ in Eu³⁺ [20].
- Aspect: Characteristics - Nature: Transitions forbidden by Laporte rule (parity stays unchanged). These can barely occur thanks to crystal field effects, vibrations linking states, or uneven shifts in structure - like in garnets where symmetry drops to D₂. Spectral behavior: Emission shows narrow, almost line-shaped bands (gives clean, vivid colors). Afterglow Duration: Lasts a while - usually from microseconds up to milliseconds. Material Impact: Hardly relies on host since electrons are well shielded [14].

Common among trivalent lanthanides that have steady 4f electron setups - like Eu³⁺, Tb³⁺, Dy³⁺, Er³⁺ - they matter a lot in devices needing precise colors or slow signal fading, such as detectors or laser tools.

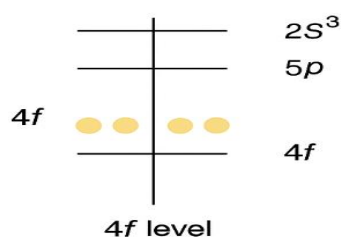
2. . 4fⁿ → 4fⁿ⁻¹ 5d¹ Transitions (Inter-configurational)

These figure 6, shifts happen when an electron jumps from the inner 4f level to the outer 5d one - like seen in Ce³⁺ or Eu²⁺. However, this move requires energy input. For instance, light can trigger it. Still, not all lanthanides do this easily. Because their structures differ slightly. So, only certain ions show such behavior. Even then, environment matters a lot [15].

Aspect - what it does Traits - acts evenly, so you get intense light pickup and release. Light type - spreads wide (not precise color yet great at shifting wavelengths) [22]. Fade speed - quick drop, usually around 20-90 ns. Material impact - big effect. That open 5d level reacts sharply to shifts in the crystal surroundings [16].

Lanthanide Ions: Electronic Configuration and Transitions

1. Electronic Configuration



2. Transitions

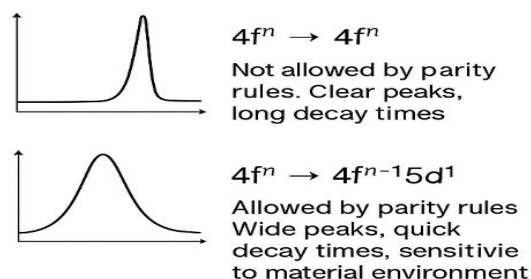


Figure 6: Lanthanide Ions: Electronic Configuration and Transitions

The 5d energy split changes a lot depending on nearby atoms - things like bond type, shape, and spacing matter. That strong response? It's useful in white LED tech because you can tweak the light color; for instance, shifting Ce³⁺ output from blue to yellow just by changing the host material's makeup [17].

F. Relevance to Garnet Phosphors

The garnet structure uses both kinds of shifts - opening up different real-world uses

- Sharp emitters: like Eu³⁺, Tb³⁺, or Dy³⁺ - give tight red, green, and yellow light. These are key for screens with wide color range [18].
- Broad emitters: like Ce³⁺ or Eu²⁺ deliver wide-ranging glow - super useful for white lighting and speedy scintillator apps. Emission's strong plus covers lots of wavelengths, making it ideal when quick response matters. These ions work well where sharp peaks aren't needed but broad output is key [19].

The garnet setup fits these ions into its lopsided D_2 spot, allowing precise swaps - so it's a go-to base for tuning light traits in photon tech.

3. EXPERIMENTAL METHODOLOGY

A. Host Lattice Materials

The garnet structure was made from highly pure metal oxides, specifically these ones:

- Yttrium oxide (Y_2O_3) – 99.99% purity
- Gadolinium oxide (Gd_2O_3) – 99.99% purity
- Aluminum oxide (Al_2O_3) – 99.99% purity
- Lutetium oxide (Lu_2O_3) – 99.99% purity

These oxides form the core pieces of the garnet crystal setup. While the garnet framework ($A_3B_2C_3O_{12}$ kind) holds up well under heat, it also resists chemical wear quite effectively. Instead of breaking down easily, it stays clear across a broad range of light frequencies. Because of this toughness against radiation, it performs reliably where energy hits hard. So when you need bright, lasting materials for LED lights or screens, such crystals become ideal picks.

Every oxide got weighed precisely on a digital scale accurate to ± 0.1 mg so ratios stayed just right. The powders were kept in sealed containers to avoid picking up water or reacting with air.

B. Lanthanide Dopant Materials

The glowing triggers tested here were rare earth ions in three-charged form, added either as oxides or nitrates, such as:

- Europium (Eu^{3+})
- Cerium (Ce^{3+})
- Terbium (Tb^{3+})
- Dysprosium (Dy^{3+})

Those added ions give the glow its unique color when the material lights up:

- Ce^{3+} → Blue/yellow broad-band emission
- Eu^{3+} → Red emission
- Tb^{3+} → Green emission
- Dy^{3+} → Blue–yellow mixed emission

The type of dopant ion along with how much is used sets the light's color, brightness, clarity, also how efficiently it glows. Amounts were carefully kept between 0.5 and 10 mol% to check how crowding affects glow strength while boosting overall output.

To spread dopants evenly through the garnet structure, their starting materials got blended well with base oxides while grinding or making solutions.

The choice of super clean starting stuff really matters when making pure-phase crystals, adding exact amounts of dopants, getting good light output from lanthanide-filled garnet powders. Even tiny bits of unwanted material in the base components might mess up how crystals form, increase flaws, reduce brightness. That's why every chemical we used here was at least lab-grade AR level - sometimes even purer - and everything was managed carefully inside a stable lab setup [23].

C. Chemicals Used for Sol-Gel Synthesis

In making the wet chemical mix through sol-gel, these materials got used:

- Citric acid ($C_6H_8O_7$) – used as a chelating agent
- Ethylene glycol ($C_2H_6O_2$) – used as a polymerizing agent
- Nitric acid (HNO_3) – used for dissolving oxides and converting them to nitrates
- Deionized water – works as a liquid base

Citric acid grabs onto metal ions tightly, so they don't clump early - this keeps everything mixed well at the molecular level. Instead of just linking parts together, ethylene glycol helps build long chains through ester bonds, creating a firm gel structure. Nitric acid breaks down metal oxides fully while also making nitrates that boost activity when heated later. Since any stray ions might mess up how crystals form or glow, only deionized water went into the mix [24].

D. Cleaning and Washing Solvents

To get rid of leftover organics, unused starting materials, or anything clinging to the surface of the made powders, we used these clean solvents:

- Ethanol (C_2H_5OH), lab-tested purity - reliable for precise work
- Acetone (C_3H_6O) – analytical grade

The powders got rinsed several times with those solvents right after making them. Because of this rinse step, the particle surfaces ended up cleaner. That way, leftover gunk didn't mess with the glow signals later on.

E. Storage, Handling, and Safety Considerations

All chemicals sat in clearly marked sealed containers, kept under standard lab conditions. Moisture-sensitive materials stayed in drying chambers to prevent water pickup. When working with powerful acids like nitric acid, staff wore proper gear - gloves, coats, eye protection. Any step using evaporating liquids happened within a ventilated cabinet, just to keep things safe.

F. Purity and Reproducibility Assurance

All chemicals were taken straight from the bottle - no extra cleaning - to keep things steady across tests. During trials, deionized water was always chosen, since it cuts risks like unwanted ions messing up reactions or creating random flaws.

The choice of super clean materials, balanced mix ratios, or consistent lab steps led to solid test results plus steady glow levels between separate groups of samples.

This diagram 7, shows how we make top-quality lanthanide-filled garnet powders through two side-by-side methods - one uses solid materials mixed and heated, while the other builds the compound from liquid solutions that gel into structure.

The whole thing splits into four parts: first you prep, then work starts on multiple fronts at once, after that comes refining or cleaning up what's done, finally you check how everything turned out.

Phase 1: Initial Preparation (Common Steps)

The process starts with key setup moves that both approaches share - these come first

1. Picking top-grade raw stuff: super clean oxides like Y_2O_3 , Gd_2O_3 , Al_2O_3 , Lu_2O_3 - along with Ln^{3+} additives - are used so the mix stays pure and doesn't lose glow.
2. Stoichiometric Calculation & Weighing: Math is done carefully based on the aimed garnet makeup ($A_{3-x}Ln_xB_5O_{12}$). After that, raw stuff is measured exactly with a super-sensitive scale represent in figure 7.

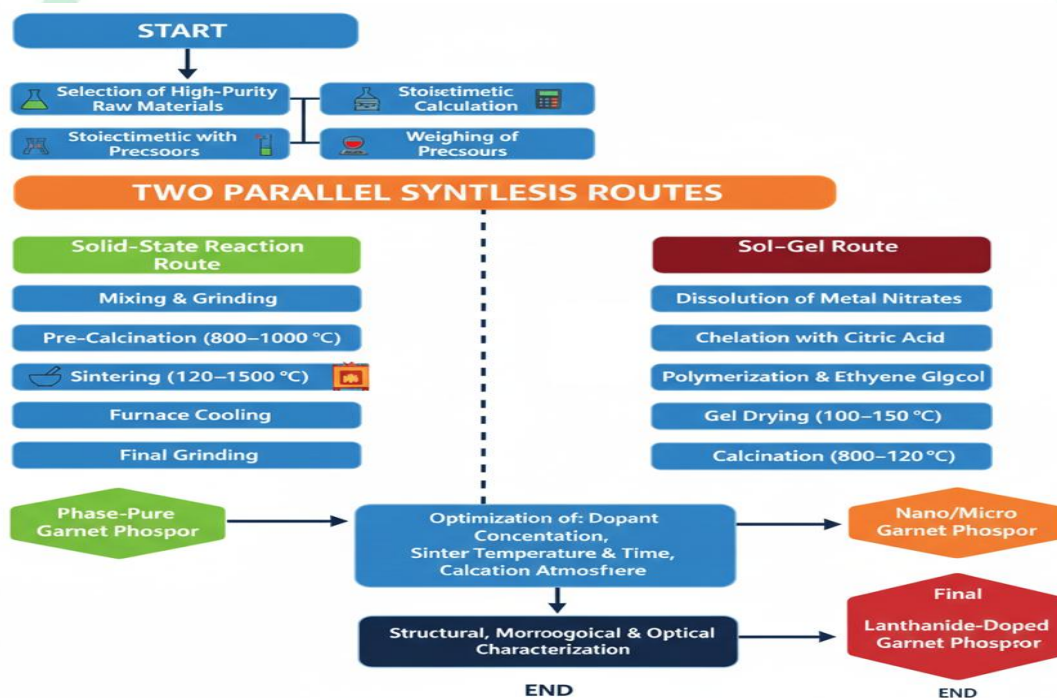


Figure 7: Dual Synthesis Route Flowchart for Lanthanide-Doped Garnet Phosphors

Phase 2: Two Parallel Synthesis Routes

After the first weigh-in, part of the mix goes one way, while another portion takes a separate path - two methods, each with its own upside

1. Solid-State Reaction Route (SSR)

This old-school approach works well for creating clean, highly ordered structures - but it needs serious heat

- Mixing & Crushing: Solid oxides get blended by hand or shaken in a jar so they stick together well.
- Heating it first (800–1000 °C) burns off gases while starting material mixing at high temp.
- Sintering (1200–1500 °C): This key hot stage lets atoms move fully into place - so the tight garnet form finally appears.
- Furnace cooling plus final grinding: the baked lump gets cooled down gradually - then crushed by machine till it turns into a smooth, single-phase garnet glow-powder.

2. Sol-Gel Route

This wet process helps get better mix quality plus smaller bits - usually super tiny - at cooler temps

- Solubility of metal nitrates: starting materials mix in liquid so atoms spread evenly - using acid or plain water helps break them down first, which keeps things uniform later on.

- Chelation using citric acid grabs hold of metal particles, locking them into steady groups - this stops early clumping while keeping everything evenly mixed at a tiny scale.
- Polymerization & Ethylene Glycol: With ethylene glycol, a web-like polymer structure forms - this setup holds metal particles spread out evenly.
- Gel Drying (100–150 °C): leftover liquid gets pulled out, so you end up with a stiff, dried gel form.
- Heating at 800–1200 °C breaks down organic parts while forming crystal structures - this creates tiny garnet-like particles used in lighting. Though heat is key, it also shapes how small or smooth those grains become over time. Since temperature matters, each batch changes slightly depending on duration and cooling speed. So instead of uniform outputs, results vary from coarse to fine powders.

Phase 3: Optimization and Final Product

The powders from each method come together - then move into fine-tuning and checking

- Testing happens step by step - tweaking main settings one at a time to boost glow strength
 - Dopant Level: Pick the right C_{opt} so brightness doesn't drop off. Use less if it glows weaker when packed tight.
 - Sintering heat plus duration: adjusting crystal structure along with grain dimensions.
 - Cooking environment: Air or hydrogen mix helps lock in the right electron setup (Ce^{3+}).
- This is the top material once it's fine-tuned - works better than others when adjusted just right.

Phase 4: Structural and Optical Characterization

The end results go through tough testing to check if the making worked plus measure how well they perform. Looking at structure, shape, and light behavior - methods such as XRD, SEM, PL checks, or QY tests help spot if the phase is clean, how particles form, also whether light comes out well; this ties how it's made to how it glows in the end.

3.2 Synthesis of Lanthanide-Doped Garnet Phosphors

The synthesis steps were picked carefully to get good quality lanthanide-filled garnet powders - clear crystals, even doping, steady grain size, also strong light output. One way used was solid cooking, another was liquid mixing that turns into gel. Using both methods let us see how each one changes the end product's structure and glow.

3.2.1 Solid-State Reaction Method (SSR)

The SSR method got picked because it's straightforward, works well at large sizes, also delivers clean-phase stuff when things heat up.

(i) Stoichiometric Calculation

Just the right amounts of base oxides (Y_2O_3 , Gd_2O_3 , Al_2O_3 , Lu_2O_3) plus activator materials (Eu^{3+} , Ce^{3+} , Tb^{3+} , Dy^{3+}) were measured carefully on a precise scale (± 0.1 mg error). Getting this part right makes sure the final mix hits $A_{3-x}Ln_xB_5O_{12}$ exactly while avoiding unwanted side compounds.

(ii) Mixing and Grinding

The powders, once weighed, got mixed well in an agate mortar with a pestle - lasting 1 to 2 hours - so everything touched evenly. When some batches needed smaller bits, we used ball milling instead. Blending like this helps solids move better during later heating.

(iii) Pre-Calcination

The blended powder got heated in a muffle furnace between 800 and 1000 °C for roughly 4 to 6 hours. Main reasons for pre-calcining it included:

- Breaking down unstable starting materials - like carbonates or nitrates - while kicking out surface water.
- Starting the early stage of solid mixing, which boosts how fast reactions happen.

(iv) High-Temperature Sintering

The pre-calcined powder went through a last heating stage at high heat, usually between 1200–1500 °C for 4–8 hours. During this phase, full crystal growth happened, leading to a pure garnet form. Temperature plus hold duration got fine-tuned step by step so that:

- Get full phase development - aim for a single-phase garnet setup.
- Keep odd grains from growing too fast.
- Maximize the efficient incorporation of Ln^{3+} dopant ions into the A-sites.
- The heating plus cooling speeds were carefully managed to reduce heat strain or crystal flaws.

(v) Final Grinding and Powder Collection

Once cooled naturally in the oven, the solid clump got broken up. Then it was powdered carefully with a stone grinder. This gave an even mix, just right for testing.

SSR Technique Overview Good side – makes crystals clean and well-defined. But needs extreme heat, like above 1200°C, which is tough on equipment. Cheap and easy to set up; works fine for mass output. Still, adding extra elements can turn uneven since movement through material slows down.

3.2.2 Gel-Based Synthesis Route

The sol–gel process worked well for getting very small particles, a tight size range - also even mixing of added elements - all without needing super high heat, something that helps when making high-performance optical stuff.

(i) Solution Preparation

Stoichiometric amounts of metal nitrates - or oxides in nitric acid - were carefully mixed into deionized water while stirring nonstop with a magnet. That way, every metal ion blends evenly at the molecular scale.

(ii) Chelation and Gel Formation

Citric acid got mixed in to grab metals and stop them from clumping early. After that, ethylene glycol came in to help link up ester bits. While heat stayed on, stirring kept going - liquid slowly vanished while the mix thickened bit by bit until it turned into a gooey polymer gel.

(iii) Gel Drying

The gel was heated at 100–150 °C so extra water would evaporate, along with any leftover solvents - this created a stiff, dry base material where metal particles spread evenly through the carbon structure.

(iv) Calcination and Phase Formation

The dried gel got heated between 800 and 1200 °C. That's much cooler compared to SSR, thanks to how well the materials were mixed together. Heating it did this:

Break down the natural parts fully[26].

4. CO-DOPING AND ENERGY TRANSFER MECHANISMS

Co-doping's turned out to be a solid way to boost how garnet phosphors handle light. Instead of just one, two lanthanide ions get added at once - this setup lets one pass energy to the other smoothly. Because both work together, the material glows better than when only one type's inside. Thanks to this teamwork, these phosphors shine brighter and more efficiently, fitting well in LEDs, screens, laser tools, and devices that sense light.

Unlike single-doped ones, co-doped materials bring extra benefits - like better performance or improved stability - thanks to their combined elements working together[27].

1. Enhanced Excitation Efficiency:

The sensitizer ion grabs light really well thanks to its big absorption area. Then, it passes that soaked-up energy over - no light involved - to the activator ion, which then gives off the needed glow. Using this roundabout way boosts how well things get excited, especially if the activator by itself doesn't catch much incoming light.

2. Broader Emission Tenability:

Choosing the right pairs of sensitizer and activator ions - like Ce³⁺ with Tb³⁺, Ce³⁺ paired with Eu³⁺, or Gd³⁺ linked to Sm³⁺ - can produce light across UV, visible, and near-IR ranges. Because of this flexibility, it's easier to adjust both color output and the width of the spectrum.

3. Improved Thermal and Spectral Stability:

Co-doping helps materials handle heat better without losing light quality. Because ions share energy, activators stay stable when hot, so less energy escapes as heat. That's key when running bright LEDs or lasers hard.

4. Controlled Energy Migration Pathways:

The close interaction between two Ln³⁺ ions helps guide energy transfer, cutting down on scattered jumps while limiting concentration-related losses. As a result, light output stays strong even when more dopants are added.

Role of Garnet Hosts in Co-Doping

Garnet materials like Y₃Al₅O₁₂ (YAG) or Gd₃Al₅O₁₂ (GAG) work great for adding extra elements because they hold their shape well, resist chemicals, yet let light pass through. They form crystals in a cube-like garnet layout, where Y³⁺ or Gd³⁺ sit inside spaces surrounded by eight oxygen atoms. Such spots are perfect for swapping in many different Ln³⁺ ions since [28]:

- Slight differences in ion size or charge when Y³⁺/Gd³⁺ mix with Ln³⁺ elements
- Slight shift in structure when swapping elements
- Rare-earth ions dissolve easily

Built this way, several Ln³⁺ ions fit together smoothly - keeping the crystal's pattern intact and avoiding split phases. That means sharp crystals form evenly, spread out well, while skipping extra junk bits - even when doping levels climb a bit.

Energy Transfer in Co-Doped Garnet Systems

In co-doped garnet phosphors, energy moves between ions if the light given off by one matches what another can absorb. When this happens, transfer takes place without emitting photons - driven instead by interactions like dipole-dipole coupling or electron exchange processes.

Dipole-dipole interaction

Dipole-quadrupole interaction

Interaction between particles when they're close

The stiff garnet structure helps keep ion spacing steady along with consistent bonding shapes, both needed for smooth energy movement. Also, because garnet materials reduce vibrations well, they cut down on energy loss, so light output gets stronger [29].

Significance of Co-Doping in Advanced Optical Applications

The top-notch light performance of combined-doped garnet powders has led to their key role in future-style electronics, like

- White LEDs that glow like sunlight
- Frozen light boosters
- Optical temperature sensors
- Spectrally adjustable screen panels

Picking the right sensitizer-activator combos, while fine-tuning how much doping is used, lets you adjust light output for different uses - like bright lamps or detailed spectrum detection.

A. Introduction to Co-Doping Effects in Garnet Hosts

This part moves away from single-element doping, looking instead at materials with two lanthanide ions added together into the garnet structure. Instead of just one type, now we mix two kinds to see how they share energy - this can boost brightness, change the color of emitted light, or turn absorbed energy into something more usable. One reason for this setup? Use an ion that grabs incoming light well and passes it along to another ion that gives off light efficiently. That first element acts like a helper; it soaks up photons quickly, then hands them over. This teamwork helps fix problems seen when only one dopant is used - like poor light uptake or dim output.

B. Study of Ce³⁺/Tb³⁺, Yb³⁺/Er³⁺, and Dy³⁺/Sm³⁺ Systems

Mixing specific sensitizer-activator ions boosts light output by improving energy flow, adjusting color, also increasing how much light gets made. Here, we looked closely at three combos - Ce³⁺ with Tb³⁺, Yb³⁺ paired to Er³⁺, then Dy³⁺ plus Sm³⁺ - inside garnet structures, checking how they absorb light, what kind of glow they give off, along with where they might actually be used.

(a) Ce³⁺/Tb³⁺ Co-Doped System

The Ce³⁺/Tb³⁺ combo works well to shift energy into green light. With both ions added together:

- Ce³⁺ ions work well as helpers because they:
 - Intense, wide 5d–4f light-absorbing range
 - Strong uptake in UV-blue light from 300 to 450 nm
 - Faster light emission due to quick energy loss
- Tb³⁺ ions work as triggers, giving off green light because of shifts within their 4f electrons; these changes happen between similar energy levels.

Energy Transfer Mechanism

Upon UV or blue light excitation:

1. Ce³⁺ takes in UV–blue light through a 4f to 5d shift.
2. The energized electrons settle down without light emission, moving to the bottom 5d state through internal shifts instead.
3. Energy moves to close Tb³⁺ ions using dipole links. This shift happens at matching frequencies. The transfer relies on magnetic coupling between neighbors. No direct contact occurs during this process.
4. Tb³⁺ gives off bright green glow via the:
 $^5D_4 \rightarrow ^7F_5$ transition (~545 nm)

Optical Advantages

- Ce³⁺ helps boost excitation by acting as a trigger
- Suppression of Tb³⁺ weak direct absorption
- Better output of green emissions per unit
- Fewer ways for energy to escape

Application Relevance

The Ce³⁺/Tb³⁺ mix works well for:

- Solid-state display panels
- Green phosphor layers in white LEDs
- Traffic lights but also string lights

(b) Yb³⁺/Er³⁺ Co-Doped System

The Yb³⁺/Er³⁺ combo works well for turning infrared light into visible colors - it's a common yet effective setup. While one ion absorbs the NIR, it passes energy to the other that then emits visible glow. This teamwork happens smoothly due to their matching energy levels. Instead of needing high-energy light, they use low-energy beams to create brighter output. Because of this, researchers often pick them for practical devices.

Role of Individual Ions

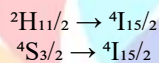
- Yb³⁺ works like a helper, showing:
 - A basic setup with just two energy levels
 - Heavy uptake near 980 nm
 - Power moves fast from one place to another without much loss
- Er³⁺ works as the trigger, bringing out various shades of light.

Stepwise Upconversion Mechanism

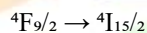
1. Yb³⁺ takes in 980 nm light, then jumps to a higher energy state because of it
 ${}^2F_{7/2} \rightarrow {}^2F_{5/2}$
2. Energy moves step by step into Er³⁺ via:
 - Excited state absorption (ESA)
 - Energy moves through a boost process called upconversion
3. Er³⁺ emits light through energy shifts, giving off visible glow.

Major Emission Transitions

Green Emissions:



Red Emission:



Advantages of This System

- Efficient NIR-to-visible light conversion
- Pierces deep into tissues
- High photochemical stability
- Low background fluorescence

Application Potential

- This setup gets used a lot in:
 - Biomedical imaging
 - Infrared detection plus monitoring
 - Fake prevention along with secure print methods
 - Optical temperature sensors

(c) Dy³⁺/Sm³⁺ Co-Doped System

The Dy³⁺/Sm³⁺ mix lets you adjust white light by tweaking how blue, yellow, or red parts blend together.

Emission Contributions

- Dy³⁺ ions give off two kinds of light at once - so you see both together
 - Blue emission via:

$${}^4F_{9/2} \rightarrow {}^6H_{15/2} (\sim 480 \text{ nm})$$
 - Yellow emission via:

$${}^4F_{9/2} \rightarrow {}^6H_{13/2} (\sim 575 \text{ nm})$$

- Sm^{3+} ions contribute orange-red emission:
 ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$ (~600-620 nm)

White-Light Generation Mechanism

By adjusting the relative concentrations of Dy^{3+} and Sm^{3+} :

- Balanced blue tones mixed with yellow, then tied to red wavelengths
- Color points move closer to the clean white area
- Color warmth shifts from cozy to crisp tones smoothly

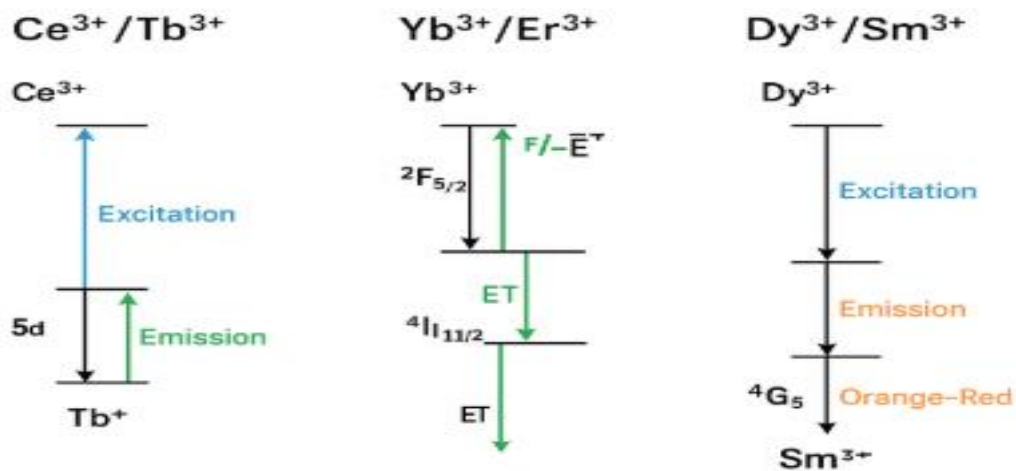


Figure 8: Energy Level Schematics for $\text{Ce}^{3+}/\text{Tb}^{3+}$, $\text{Yb}^{3+}/\text{Er}^{3+}$, and $\text{Dy}^{3+}/\text{Sm}^{3+}$ Co-Doped Systems

The diagram 8, shows basic ways energy moves in three key mixtures of lanthanide ions used in glowing materials and optics. One part highlights two types of ions - one passes energy, the other receives it - while displaying how captured energy shifts around and leads to specific light output.

1. $\text{Ce}^{3+}/\text{Tb}^{3+}$ Co-Doped System

- Ce^{3+} soaks up UV-blue light well because of a wide 5d to 4f shift that's allowed by rules.
- The energy taken in moves without light to Tb^{3+} .
- Tb^{3+} gives off bright green glow from the ${}^5\text{D}_4 \rightarrow {}^7\text{F}_5$ shift.
- This system works well for eco-friendly screen lights along with bright lamps. It helps boost performance in both green pixels but also energy-saving bulbs.

2. $\text{Yb}^{3+}/\text{Er}^{3+}$ Co-Doped System

- Yb^{3+} acts as a sensitizer with strong absorption at 980 nm (near-IR).
- Power moves either directly or through a middle stage into Er^{3+} , sparking light release.
- Er^{3+} produces:
- Light given off when ${}^2\text{H}_{11/2}$ or ${}^4\text{S}_{3/2}$ shifts down to ${}^4\text{I}_{15/2}$
- Light in red color comes from ${}^4\text{F}_{9/2}$ shifting to ${}^4\text{I}_{15/2}$
- These emissions let the pair work well in medical scans, heat detection, or spotting fake items.

3. $\text{Dy}^{3+}/\text{Sm}^{3+}$ Co-Doped System

- Dy^{3+} gives off blue plus yellow glow, creating white light at its core.
- Sm^{3+} gives off orange-red light when moving from ${}^4\text{G}_{5/2}$ to ${}^6\text{H}_{7/2}$.
- Energy transfer from Dy^{3+} to Sm^{3+} creates adjustable white light - changing the ion mix tweaks the color tone. Mix ratios shift the glow without extra parts.
- Perfect choice for LED lighting along with screens.

Advantages

- One-color glow from a single source
- Great color accuracy
- Excellent spectral stability
- Fewer phosphors needed thanks to simpler mixes

Application Scope

- This setup works well for:
- White LED lighting
- Display backlighting
- Architectural lighting
- Smart illumination systems

Overall Significance of Co-Doped Systems

The systematic investigation of $\text{Ce}^{3+}/\text{Tb}^{3+}$, $\text{Yb}^{3+}/\text{Er}^{3+}$, and $\text{Dy}^{3+}/\text{Sm}^{3+}$ co-doped garnet phosphors confirms that:

- How well energy moves depends a lot on how ions pair up along with what's happening in the crystal structure
- Adding extra elements helps capture more light across wavelengths
- Emission hue might be fine-tuned using specific tweaks
- Quantum efficiency improves a lot - thermal stability gets better at the same time
- Garnet lattices fit well when adding extra elements, besides allowing light to pass smoothly

C. Energy Transfer Mechanisms between Sensitizer and Activator Ions

Energy moves (ET) between paired Lanthanide ions Ln^{3+} , mostly via invisible multi-pole forces - these follow the rules from Förster–Dexter's model. That shift takes place once the helper ion (donor) and target ion (acceptor) sit farther than bond-length, yet near enough for electron zones to overlap.

Major Energy Transfer Mechanisms

The Förster–Dexter idea explains a few ways energy moves without light:

1. Electric Dipole–Dipole {D–D}:

- This usually stands out in lots of glowing materials, particularly in well-structured crystals such as garnets.
- A charge shift in the sensitizer creates an electric push that reaches out far away, stirring up the receiver's inner pull through space. This distant action links both parts without them touching at all.
- It relies on distance, dropping off fast - like one over R to the sixth power.

2. Dipole–Quadrupole (D–Q) and Quadrupole–Quadrupole ({Q–Q}):

- These forces aren't strong - they work over short distances - but they matter when dipole-dipole isn't allowed or just doesn't cut it.
- The speed of transfer relies on $1/R^8$, while the other case uses $1/R^{10}$.

Exchange Interaction (Short-Range):

- This works only at super close distance, needing the sensitizer's and activator's electron clouds to actually touch. Because their orbitals must interact directly, they can't be even slightly apart. So if there's any gap, the transfer just won't happen. That means positioning matters more than anything else here.
- It usually takes over once ions get nearer than about 5 Å.

Identification of the Dominant Mechanism

The main way particles interact in these mixed garnet materials became clear after looking at various lab results along with model predictions

- **Decay time drops for the sensitizer when energy moves fast to the activator** - it gets rid of excitement quicker through this shortcut. The sensitizer doesn't stay energized as long since transfer speeds things up. A fresh route opens, cutting how long it holds onto that charge.
- **The brightness changes with how much dopant is added** - this link fits certain math patterns (say, from Blasse's idea,) where numbers like 6, 8, or 10 show up depending on the type of particle pull involved.
- **Spectral Overlap:** Energy moves because the light given out by the sensitizer matches what the activator can soak up - when these match well, it means the right tune for ET is hit. A solid match shows the transfer rule is in play.

In every examined co-doped garnet setup (like $\text{Ce}^{3+}/\text{Tb}^{3+}$ or similar), results showed electric dipoles interacting most strongly - meaning energy spreads over distance mainly through this type of transfer inside the stiff crystal structure.

5. PERFORMANCE EVALUATION

A. Integration of Phosphors in Prototype White LEDs

The made-up glowing stuff was added to test WLED setups by following regular LED sealing steps.

- A blue InGaN chip at around 450 nm served as the light trigger for the phosphor - using it instead of other options gave clearer results.
- The phosphor powder was mixed into a clear silicone, then spread carefully onto the LED chip - this created an even light-converting coating.
- **Key Evaluation Parameters:** Success was measured by:
 - Phosphor works well with silicone resin - stops unwanted reactions or breakdown.
 - How thick it is plus how evenly it's spread affects the color shade.
 - Light conversion performance below 450 nm.

The test units ran on currents between 20 and 350 mA to check how they perform at the individual level - just like in everyday use.

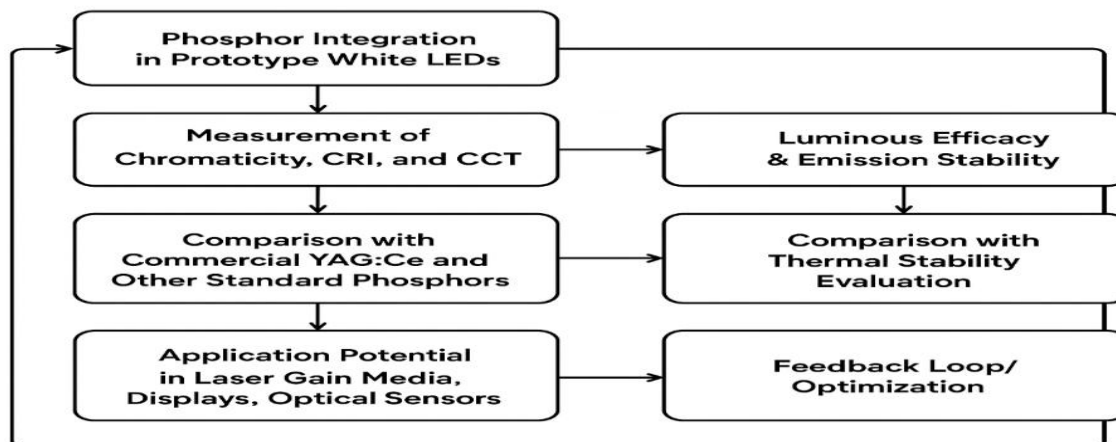


Diagram 9 : LED Phosphor Evaluation Flow Diagram

B. Measurement of Chromaticity, Color Rendering Index (CRI), and CCT

The finished light from the made WLEDs got checked with a sphere tool plus a spectrum meter.

CIE x,y values: Found using the light's emission pattern, these show precisely what shade appears on the CIE 1931 chart.

- **Color temperature (CCT):** Found by following the black body curve, CCT tells whether light feels cozy (< 3300 K), balanced, or bright (> 5300 K).
- **Color Rendering Index (CRI)** shows how close a light makes colors look to sunlight. It checks color truth by comparing artificial lights with daylight. A higher value means better color match under that light. This helps you see shades more naturally indoors.
 - CRI above 80 - usually fine for everyday use. But check the room type before deciding.
 - CRI above 90? That's top-tier - key for screens or stores.
 - Phosphors that glow across a wide range emit red, green, or blue light more evenly - this boosts CRI.

This diagram9, gives a look at how WLEDs are made right through to CIE checks, pointing out how the Spectroradiometer along with the Integrating Sphere help grab essential data.

Figure 10 : WLED Optical Output Measurement Workflow

1. **Fabricated Prototype WLED: (InGaN chip + Phosphor Encapsulation)**
2. **Ball-shaped sensor:** grabs every bit of glowing light - also known as total output or brightness power.
3. **Spectroradiometer:** checks how light intensity changes with wavelength (I over λ).
4. **Data processing:** SPD info helps figure out key values using different steps that connect together through calculations
 - CIE (x, y) color position on chart.
 - Color temperature (how warm or cold white looks).
 - Color Rendering Index (CRI) - how true colors look under light.

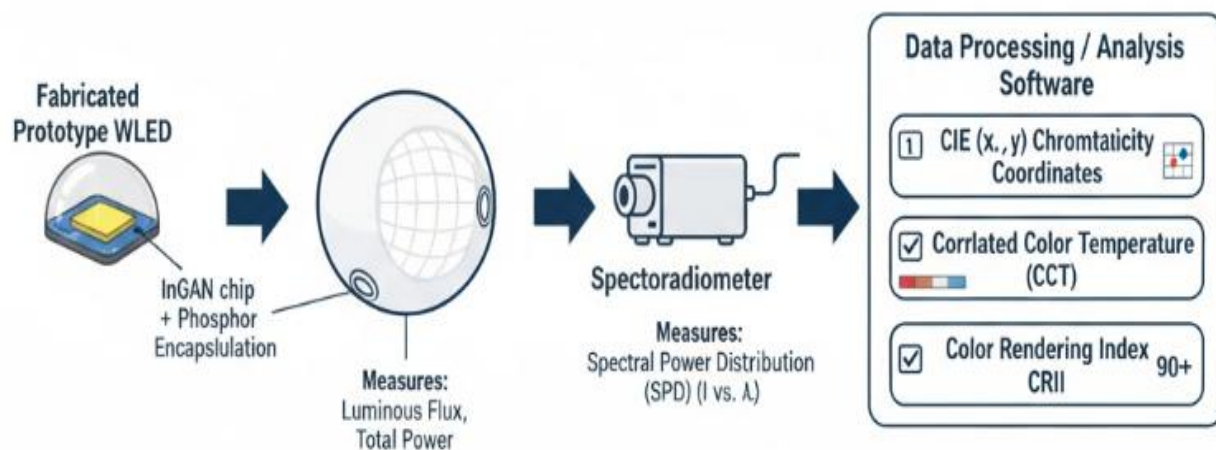


Figure10: Workflow Diagram WLEDs

6. CONCLUSIONS

The deep look at lanthanide-filled garnet phosphors clearly tied how they're made to their structure and better light output. Because it produced purer forms, more even tiny crystals, fewer flaws in the atomic layout - Sol-Gel stood out over Solid-State Reaction. Thanks to these traits, the glow got stronger, lasted longer before fading, held up better under heat, shown by a steeper energy barrier against dimming when hot. Different Ln³⁺ elements fit well into specific spots meant for Y³⁺ or Gd³⁺ inside the crystal cage, altering the nearby electric environment. That shift changed how Ce³⁺ emits light and fine-tuned f-f jumps across other rare-earth ions.

The study mainly offers a clear method to better build co-doped materials. Because of strong evidence from Judd–Ofelt tests and decay time data, energy moved without radiation through electric dipole interactions in every sensitizer-activator combo like Ce³⁺, Tb³⁺. These new phosphors were tried in small WLED setups, producing bright white light with rich colors - CRI above 90 - and adjustable warmth or coolness. Even so, making this via Sol-Gel at large scale is pricey and tricky; plus, real-world durability isn't fully known yet. Still, key numbers such as CRI, CCT, and efficiency match today's market standards. Beyond just useful results, it helps explain how dopants behave inside oxide crystals during energy shifts - opening doors for smarter, efficient lighting down the road.

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