Luminescence and tunable color properties of uniphase white-emitting $\text{Sr}_3\text{B}_2\text{SiO}_8$:Tm$^{3+}$/Dy$^{3+}$/Eu$^{3+}$ phosphors by energy transfer for UV-excited white LEDs

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Abstract
Multi-color single-phase white emission $\text{Sr}_3\text{B}_2\text{SiO}_8$:Tm$^{3+}$/Dy$^{3+}$/Eu$^{3+}$ fluorescent powders were prepared by solid-state fritting method. The fluorescence performance of Tm$^{3+}$, Dy$^{3+}$ and Eu$^{3+}$ ions single-activated $\text{Sr}_3\text{B}_2\text{SiO}_8$ phosphors were investigated and they exhibit desirable behaviors in their characteristic emissions. Meanwhile, the energy required for migration from Tm$^{3+}$ to Dy$^{3+}$ and from Dy$^{3+}$ to Eu$^{3+}$ in co-doped fluorescent substance was determined by the aids of steady-state and transient-state photoluminescence spectra/decay measurements. The energy transmission process from Tm$^{3+}$ to Dy$^{3+}$ and Dy$^{3+}$ to Eu$^{3+}$ has been explored to be a dipole–quadrupole interaction and a dipole–dipole interaction of the resonant type, respectively. More significantly, by tuning the molarity ratio of doping Dy$^{3+}$/Eu$^{3+}$ ions, multicolor emission from yellow to red shall be achieved in $\text{Sr}_3\text{B}_2\text{SiO}_8$:Dy$^{3+}$, Eu$^{3+}$ phosphors for its potential adhibitions in solid-state lighting applications. The $\text{Sr}_3\text{B}_2\text{SiO}_8$:Tm$^{3+}$, Dy$^{3+}$ samples can implement white light emission by the appropriate changeable doping molar quantity of Tm$^{3+}$ and Dy$^{3+}$, applying potentially in white LEDs. The energy migrations between co-doped rare earth ions play a decisive role in these processes.

Keywords $\text{Sr}_3\text{B}_2\text{SiO}_8$:Tm$^{3+}$/Dy$^{3+}$/Eu$^{3+}$ · Energy transfer · Multi-color emission · WLED

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1 Introduction

Considered as the significant emerging lighting source, white light emitting diodes (WLEDs) are infiltrating the global optics space with a high speed of development due to their unique features of high luminous yield, long service life and small space usage (Chen et al. 2014; Bharat et al. 2016; Shang et al. 2014; Liu et al. 2018). Nowadays, the core technology of commercializing white LED is packaging of broadband yellow phosphor YAG:Ce$^{3+}$ with InGan blue chip to switch blue photons to long-wavelength ones (Wang et al. 2016). Unfortunately, the unsolved challenging issues of yellowing and red component absence leading to the decrease of white light softness and color rendering for the combination restrict its expansion in relevant application (Ye et al. 2010; Smet et al. 2011). A new format of realizing white LED with ultraviolet chip coating red, green, and blue multiphase phosphors was introduced to ameliorate the deficiency of blue chip system. It is a pity that the serious energy reabsorption and proportion imbalance occurs in the mixing of multi-component phosphors, which have a certain impact on the luminous efficiency and color reductivity (Liu et al. 2014; Luwang et al. 2011; Zhu et al. 2007). Under this circumstance, another effective alternate was major concentrated in the single-phase phosphor with white or multi-color emission, which was more stimulated in the ultraviolet region. In generally, white or multi-color emission could be carried out by controlling the luminous color of materials effectively, on account of designing the energy transfer between different sensitizer/activator ions in the single-phase substrate (Patnam et al. 2019; Zhu et al. 2014; Chen et al. 2012). Therefore, seeking for novel single-phase phosphors and high efficient energy transfer has become greatly in demand.

Rare earth ions as the frequently used phosphor activators and sensitizers have been playing a critical part in pc-WLEDs due to the ample emission colors range from near-infrared to ultraviolet (Song et al. 2015; Setlur 2009). It is common knowledge that Tm$^{3+}$ ions possessing of 4f$^{12}$ electron configuration are generally served as blue-light emission active centers, which is mainly attributed to the characteristic level transition $^1D_2 \rightarrow ^3F_4$ (Hu et al. 2014). Comfortable white light is more likely to be emerged in the co-doped system with Tm$^{3+}$ ions as sensitizers. Furthermore, owing to $^4F_{9/2} \rightarrow ^6H_{13/2}$ and $^4F_{9/2} \rightarrow ^6H_{15/2}$ transitions of Dy$^{3+}$ ions, the emission color is centralized in the blue and yellow range of the visible spectrum, respectively (Pavitra et al. 2013). Hence, there is adequate possibility for Tm$^{3+}$ to behave as a sensitizing donor which diverts energy to Dy$^{3+}$ in co-doped system for white light being engendered by the recombination of different light color. Deemed as excellent red emitter for phosphor, the Eu$^{3+}$ ion exhibits efficient typical emissions belonging to the transition from upper states $^5D_{0,1,2}$ to ground states $^7F_{1,2,3,4,5}$ (Huang et al. 2017). Thus, by adding Eu$^{3+}$ ions into the same host lattice for offering intense red composition, the deficiency of red component can be conquered in the Dy$^{3+}$ single-doped phosphor. Remarkably, the Tm$^{3+} \rightarrow$ Dy$^{3+}$ and Dy$^{3+} \rightarrow$ Eu$^{3+}$ energy transfer is studied in very few material families as white or multi-color emitting phosphor for construction of w-LEDs, such as K$_3$Gd(PO$_4$)$_2$:Tm$^{3+}$,Dy$^{3+}$ (Zhao et al. 2017), NaGdTiO$_4$:Tm$^{3+}$,Dy$^{3+}$ (Bharat et al. 2016), NaBi(MoO$_4$)$_2$:Dy$^{3+}$,Eu$^{3+}$ (Li et al. 2019) and Ca$_2$ZnSi$_2$O$_7$:Dy$^{3+}$,Eu$^{3+}$ (Mondal and Manam 2018). For all that, the strict synthesis conditions and not high luminescence efficiency of these materials hamper their further applications.

The luminescent material host of borosilicate for rare earth ions emission has attracted the interest of numerous researchers because of their economical prepared temperature and prominent corrosion resistance (Chang and Chen 2007). Among the borosilicate materials, Sr$_3$B$_2$SiO$_8$ has been affirmed as an excellent matrix with good application prospects in the
phosphor-converted white LEDs, for example, Sr$_3$B$_2$SiO$_8$:Eu$^{3+}$ (Wang and Wang 2011), Sr$_3$B$_2$SiO$_8$:Eu$^{2+}$, Dy$^{3+}$ (Sabikoglu 2013), Sr$_3$B$_2$SiO$_8$:Sm$^{3+}$ (Sun et al. 2016). However, to our knowledge, white light emitting and tunable color properties on the strength of energy transfer between active centers, Tm$^{3+}$→Dy$^{3+}$, Dy$^{3+}$→Eu$^{3+}$ has not been yet reported in Sr$_3$B$_2$SiO$_8$ system. Herein, we present the design of novel Sr$_3$B$_2$SiO$_8$:Tm$^{3+}$/Dy$^{3+}$/Eu$^{3+}$ phosphor synthesized by solid-state fritting reaction which shows unique white light emitting and tunable characteristics. Additionally, the energy transfer of Tm$^{3+}$→Dy$^{3+}$ and Dy$^{3+}$→Eu$^{3+}$ has been confirmed to exist in the Sr$_3$B$_2$SiO$_8$ matrix and the mechanism were discussed indetail on account of steady-state and transient-state photoluminescence spectra/decay consequences. All results indicated that the tunable color from yellow to red luminescence has been obtained in Sr$_3$B$_2$SiO$_8$:Dy$^{3+}$/Eu$^{3+}$ phosphor by the alteration of Dy$^{3+}$/Eu$^{3+}$molarity ratio and the phosphor Sr$_3$B$_2$SiO$_8$:Tm$^{3+}$/Dy$^{3+}$ is a potential candidate for warm UV or near-UV WLEDs.

2 Experimental

2.1 Synthesis procedure

Phosphors with the chemical constituents of Sr$_3$B$_2$SiO$_8$:Tm$^{3+}$/Dy$^{3+}$/Eu$^{3+}$ were prepared via the traditional solid-state process. The starting materials such as SrCO$_3$, H$_3$BO$_3$, SiO$_2$, Li$_2$CO$_3$ and Tm$_2$O$_3$ (99.99%), Dy$_2$O$_3$ (99.99%), Eu$_2$O$_3$ (99.99%) were blended in accordance with the chemometry proportion, and well-milled evenly for 30 min in an agate mortar and pestle. All the starting chemicals were of the analytical reagent grade. The precursors were diverted into corundum crucibles, then calcined at 600 °C for 1 h in elema stove. Since removed, secondary milled and programmed temperature up to 1080 °C, holding for 3 h in a tube furnace under ambient atmosphere. After the reaction, nature air cooling and milled again into smalls to acquire the final products.

2.2 Characterization

The poly-crystalline phases of Sr$_3$B$_2$SiO$_8$:Tm$^{3+}$/Dy$^{3+}$/Eu$^{3+}$ phosphors were analyzed by X-ray powder diffraction (XRD) on a Rigaku Miniflex 600, with Cu Kα radiation. The diffraction patterns were scanned at 6° min$^{-1}$ from 10° to 60°. The refinements of crystal structure were operated applying GSAS program based on the Rietveld method. The steady-state photoluminescence excitation/emission spectra and the quantum efficiency (QE) were measured (Edinburgh FS5) using a 150 W Xenon lamp as the excitation source and an integrating sphere. The transient-state luminescence decay measurements were obtained (Edinburgh FL5920 spectrophotometer) utilizing 350 W Xenon lamps as the excitation source. The element composition and content of phosphors was inspected by using a scanning electron microscope (SEM, JSM-6490LV) equipped with an energy-dispersive X-ray spectrometer (EDS). The electronic structure of SBSO was evaluated on a personal computer by using the module CASTAP in the software Materials Studio based on the density functional theory (DFT). The particle diameter distribution of phosphors was carried out using particle size and Zeta potential analyzer (NanoBrook Omni).
Fig. 1 XRD pattern for the Rietveld refinement of $\text{Sr}_3\text{B}_2\text{SiO}_8$

Fig. 2 The crystal structure of $\text{Sr}_3\text{B}_2\text{SiO}_8$ host
3 Results and discussion

3.1 Crystalline characteristic and Phase purity analysis

The as recorded, computed and difference XRD patterns obtained by Rietveld refinement corresponding to SBSO are represented in Fig. 1. The weighted and profile R-factors, Rwp, Rp and $\chi^2$ computed after multiple iterations are intended to be 11.29%, 7.48% and 1.09, respectively, well fitted the reflection condition. The raw structure model (Fig. 2) were constructed consulting to previously literature for SBSO (JCPDS 32–1224). The synthesized sample SBSO is pure and has the orthorhombic symmetry structure (space group: Pnma) and the lattice constants are computed as $a = 12.361$ Å, $b = 3.9061$ Å, $c = 5.4068$ Å. The unit cell parameters are recorded as $V = 259.82$ Å$^3$ and $Z = 4$. The refinement crystallographic parameters of lattice structure, the coordinates of the atom as well as the occupancy ratio are presented in Tables 1 and 2. Figure 3 illustrates the representative XRD patterns for SBSO:Tm$^{3+}$/Dy$^{3+}$/Eu$^{3+}$ phosphors with diverse doping concentration and also the powder diffraction standard of SBSO (JCPDS 32–1224). The measured XRD patterns revealed the crystal structure match of different doping samples with standard diffraction pattern corresponding to JCPDS 32–1224. The absence of secondary phases confirm that

| Table 1 Crystal structural data of SBSO as the results of Rietveld refinement |
|---------------------------------|
| **Crystal structural and lattice parameters of SBSO** |
| **Formula** | Sr$_3$B$_2$SiO$_8$ |
| **Crystal system** | Orthorhombic |
| **Space group** | Pnma |
| **a/Å** | 12.3025 |
| **b/Å** | 3.9061 |
| **c/Å** | 5.4068 |
| **α/deg** | 90 |
| **β/deg** | 90 |
| **γ/deg** | 90 |
| **V/Å$^3$** | 259.82 |
| **Rp/%** | 11.29 |
| **Rwp/%** | 14.78 |
| **$\chi^2$** | 1.09 |

| Table 2 Atomic Coordinates and Atomic Occupancy Ratio of SBSO Obtained by the Rietveld Refinement |
|---------------------------------|
| **Atom** | x | y | z | SOF |
|-------|----|----|----|----|
| Sr1   | 0.1470 | 0.75 | 0.9058 | 1.0 |
| B1    | 0.4136 | 0.75 | 0.9342 | 0.67 |
| Si1   | 0.4136 | 0.75 | 0.9342 | 0.33 |
| O1    | 0.4213 | 0.75 | 0.6732 | 0.48 |
| O2    | 0.5215 | 0.9015 | 0.9351 | 0.71 |
| O3    | 0.3440 | 0.75 | 0.1154 | 0.29 |
| O4    | 0.3655 | 0.75 | 0.667 | 0.60 |
| O5    | 0.3121 | 0.75 | 0.0821 | 0.90 |
Fig. 3  X-ray powder diffraction patterns of SBSO:Tm$^{3+}$/Dy$^{3+}$/Eu$^{3+}$ and the standard data (JCPDS 32–1224) for SBSO

![X-ray powder diffraction patterns](image)

Table 3 The EDS quantitative analysis of SBSO:7%Dy$^{3+}$ sample

| Label | Element | Weight % | Atomic % |
|-------|---------|----------|----------|
| 1     | Boron   | 5.826    | 15.765   |
| 2     | Oxygen  | 32.050   | 58.796   |
| 3     | Silicon | 6.060    | 7.362    |
| 4     | Strontium | 53.092   | 17.722   |
| 5     | Dysprosium | 2.973    | 0.535    |
the doping level of Tm$^{3+}$, Dy$^{3+}$ and Eu$^{3+}$ ions is within the solubility limit and the small introduction is stabilized in the crystal structure of the matrix without inducing significant effect. The ionic radii values for 8 coordination number indicate that the Tm$^{3+}$ (1.134 Å), Dy$^{3+}$ (0.908 Å) and Eu$^{3+}$ (0.950 Å) ions should occupy the lattice position of Sr$^{3+}$ (1.18 Å) rather than B$^{3+}$ after doping into the crystal lattice (Bharat et al. 2016).

The energy-dispersive spectrometer (EDS) of SBSO:7%Dy$^{3+}$ samples were studied to further analyze the element composition and content. As shown in Fig. 4, the EDX spectrum results confirm the presence of boron (B), oxygen (O), silicon (Si), strontium (Sr) and dysprosium (Dy) elements in Sr$_3$B$_2$SiO$_8$:0.07Dy$^{3+}$ sample, no other peaks of impurity elements were detected. In addition, from the results of quantitative analysis (Table 3), it can be seen that the values of Weight% and Atomic% lists are in agreement with the stoichiometric ratio of chemical formula Sr$_3$B$_2$SiO$_8$:0.07Dy$^{3+}$ basically. Therefore, the information of EDS gives further support for the XRD analysis.

### 3.2 Luminescence and energy transfer characteristics of SBSO:Tm$^{3+}$, Dy$^{3+}$

The normalized photoluminescence excitation and emission spectra of SBSO:3%Tm$^{3+}$ sample is displayed in Fig. 5. It can be seen that a sharp absorption peak at 357 nm corroborated with the f-f parity-forbidden Tm$^{3+}$:3H$_6^-$→1D$_2$ appears in the excitation spectrum when monitored at 456 nm, which confirms a perfect match with the UV-LED chip bands. Upon the stimulation of 357 nm irradiation, an intense blue emission locating at about 456 nm is observed assigned to the shift of Tm$^{3+}$ from 1D$_2$ to 3F$_4$. The special characteristic luminescence indicates that Tm$^{3+}$ ions are appropriate as sensitizers to transfer energy while comfortable white light is more likely to be obtained in co-doping system.

On the side, the comparison result of the emission curves of SBSO:x%Tm$^{3+}$ (x = 0.5–5) were presented in the upper inset Fig. 5. It can be surveyed that the emission intensities of Tm$^{3+}$ heighten sustainably with the doping content raising until the concentration is reaching 3 mol% (the emission intensity is at the maximum position), then showed a conspicuous monotonous decrement for dopant level increase, ascribed to the attainment of solubility limit. The concentration quenching of Tm$^{3+}$ ions is a physical phenomenon of
nonradiative relaxation, that is, the luminescence center returns to the ground state from the excited state in nonradiative pattern. In this case, the energy emitted by the system will be lost in the lattice in form of vibrational kinetic energy. The theoretical formula of nonradiative transition probability is as follows:

\[
W = Ce^{-\alpha \Delta E} (1 + m)^{\Delta E/h\nu}
\]

where \( \Delta E \) is energy difference between the excited state and the ground state, \( h\nu \) represents photon energy (\( \nu \) is the ambient vibration frequency), \( C \) and \( \alpha \) are proportional constants depending on matrix, and \( m \) is a temperature dependent parameter (Blass 1979). It can be seen from the formula that the nonradiative transition probability (\( W \)) is only related to \( \Delta E \) and \( h\nu \) when the environment and matrix material are determined. When the concentration of luminescent centers increases, the distance between ions decreases, so that the electron-lattice coupling and interaction strength increases. Under the circumstances, energy transfer occurs among the neighboring centers (\( h\nu \)), and \( W \) will raise with the enhancing of \( h\nu \) according to the equation. It means, the excitation energy is transferred from the absorption site to the nonradiative depletion site, and then lost in the lattice of system in form of vibrational energy, the concentration quenching occurs. Thus, the energy transfer from one activator to another will strongly depend on the distance (\( R \)) between \( \text{Tm}^{3+} \) ions, when the value reaches the critical transfer distance (\( R_c \)), the concentration quenching will take place. Blasse has pointed out that the critical transfer distance (\( R_c \)) is approximately equal to twice the radius of a sphere with this volume (Blass 1969):

\[
R_c = 2 \left[ \frac{3V}{4\pi X_c N} \right]^{1/3}
\]

where \( X_c \) is the critical concentration, \( N \) is the number of cations in the unit cell and \( V \) is the volume of the unit cell. By taking the experimental and analytic values of \( X_c \), \( N \) and \( V \) (0.03, 4, 263.05, respectively), the critical distance \( R_c \) is estimated by Eq. (2) to be about 15.79 Å.

Furthermore, based on the previous refined structure parameters, the band gap energy and orbital density of pure SBSO are calculated by using CASTAP module of Materials Studio software. As shown in Fig. 6a, b, it is clear that the computed energy gap is approximately 1.7 eV, implying that SBSO belongs to semiconductor materials with no large bandgaps. It is expected that SBSO is an appropriate luminescent host because the characteristic structure with electron–hole carriers can increase the transition probability of luminescent ions.

The optical band gap (\( E_g \)) of SBSO:3%Tm\(^{3+} \) can be obtained from the diffuse reflectance spectrum (DRS), estimated according to the equation:

\[
(\alpha h\nu)^n = A(h\nu - E_g)
\]

where \( \alpha \) is absorption coefficient, \( h\nu \) represents photon energy, \( A \) is a proportional constant and the exponent \( n = 1/2 \) and 2 is the allowed indirect transition and allowed direct transitions, respectively (Zhao et al. 2018). The value of \( \alpha \) can be obtained from the reflection coefficient \( R \):

\[
\alpha = \frac{(1 - R^2)}{2R}
\]
The graphs plotting for $(\alpha h \nu)^2$ and $(\alpha h \nu)^{1/2}$ verses $h \nu$ give the straight line with intercept indicating the optical energy band gap. As shown in Fig. 7, the optical band gap energy is determined to be 3.6, 3.1 eV for direct and indirect allowed transitions of the SBSO:3%Tm$^{3+}$ phosphor, respectively. Measuring the band gap of experimental sample SBSO:3%Tm$^{3+}$ using the DRS is to verify the theoretical calculation results. Because the GGA exchange–correlation function that was adopted usually underestimates the size of the energy band gap, simultaneously together with the influence of Tm$^{3+}$ ion doping, the experimental values of 3.6, 3.1 eV for direct and indirect allowed transitions is greater than the calculated value (1.73 eV), but within the reasonable range.

We compared the band gap structure of SBSO matrix with some other reported (Jiang et al. 2009; Wang et al. 2017; An et al. 2020; Geng et al. 2019), as shown in Table 4. It can be seen that the band gap value of SBSO is lower than that of other host. In general, if the band gap is about 1–3.6 eV, it belongs to semiconductor; if the band gap is greater than 3.6 eV, it belongs to nonconductor, they have their own advantage to the luminescent behavior. The nonconductor materials have a wide band gap to
accommodate both the ground and excited states of luminescent ions within its wide band structure, which is more favorable to the broad band emission for rare earth ions with 4f–5d transition (all the insulator materials listed in the table are doped with Ce³⁺ ions). The semiconductor materials have no wide band gap, but its characteristic structure with electron–hole carriers promotes more electron-transition, thus increasing the transition probability of luminescent ions, which is beneficial to luminescent behavior. In this study, the doped rare earth ion: Tm³⁺ belongs to sharp line emission with the parity forbidden 4f–4f transition, semiconductor substrate is more appropriate for it. It is determined that the matrix belongs to semiconductor through the calculation of the band gap structure, moreover, with such a band gap, it is predicted that SBSO is an appropriate luminescent host. While, the appropriate luminescent semiconductor band structure of pure SBSO and SBSO:3%Tm³⁺ confirms the above luminescent properties of Tm³⁺ ions (the obvious sharp line peak in the emission spectrum).

As 7 mol% Dy³⁺ singly doped SBSO host, the PLE and PL spectra are shown in Fig. 8a. It is observed that monitored with 576 nm emission of Dy³⁺, the excitation spectrum within the section of 275–500 nm is composed of several narrow bands centered
The comparison of band gap structure between the SBSO matrix and other reported materials

| Label | Material Composition | Bandgap (eV) | Structure type | Advantage | References            |
|-------|----------------------|--------------|----------------|-----------|-----------------------|
| 1     | Sr$_3$B$_2$SiO$_8$   | 1.73         | Semiconductor  | Electron–hole carriers increase the transition probability of luminescentions | Present work |
| 2     | Ca$_2$GeO$_4$       | 4.0          | Nonconductor   | Accommodating both the ground and excited states of luminescent ions within its wide band gap | Jiang et al. (2009) |
| 3     | Ca$_2$YZr$_2$Al$_3$O$_{12}$ | 4.56      | Nonconductor   |           | Wang et al. (2017)    |
| 4     | CaMg$_{0.8}$Al$_{0.4}$Si$_{1.8}$O$_6$ | 5.28      | Nonconductor   |           | An et al. (2020)      |
| 5     | NaBaSc(BO$_3$)$_2$  | 5.81         | Nonconductor   |           | Geng et al. (2019)    |
at 296, 324, 349, 387, 425 and 452 nm, which corresponds to the parity-forbidden f-f transitions \(^6H_{15/2} \rightarrow ^4H_{13/2}, ^6H_{15/2} \rightarrow ^6P_{3/2}, ^6H_{15/2} \rightarrow ^6P_{7/2}, ^6H_{15/2} \rightarrow ^4P_{7/2}, ^6H_{15/2} \rightarrow ^4G_{11/2}\) and \(^6H_{15/2} \rightarrow ^4I_{15/2}\). The emission spectrum excited by 349 nm arises two dominating incisive bands at around 487 and 576 nm, ascribed to the typical energy level for \(\text{Dy}^{3+}\) ions, the \(^4F_{9/2} \rightarrow ^6H_{15/2}\) of magnetic dipole (blue) and the \(^4F_{9/2} \rightarrow ^6H_{13/2}\) of electric dipole (yellow), respectively. The stronger yellow luminescence than blue demonstrates that the crystal field of \(\text{Dy}^{3+}\) centers is no inversion symmetry in SBSO host (Li et al. 2019; Mondal 2018). A series of diverse emission spectra for SBSO: \(x\%\text{Dy}^{3+}\) (\(x = 1–9\)) phosphors under 349 nm excitation were depicted in Fig. 8b. The results of this section can be discussed with reference to Fig. 5 and the maximum luminous molality is 7 mol% for \(\text{Dy}^{3+}\) ions in SBSO host.

As described in Fig. 9a, amidst the incisive emission of \(\text{Tm}^{3+}\) and the \(\text{Dy}^{3+}\) absorption transitions, a manifest spectral overlap emerges ranging from 440 to 470 nm. Moreover,
the excitation spectrum for SBSO:Tm$^{3+}$, Dy$^{3+}$ (Fig. 9b) reveals some absorption bands belonging to the feature f-f parity-forbidden of Tm$^{3+}$ and Dy$^{3+}$, observed by the relevant characteristic emission (456 nm of Tm$^{3+}$ and 576 nm of Dy$^{3+}$). All the above discussion spredict convincingly that the process of energy transfer from Tm$^{3+}$ to Dy$^{3+}$ is likely to occur in SBSO host. Therefore, the synergy of yellow (Dy$^{3+}$) and blue (Tm$^{3+}$) emissions via the energy transfer could be considered to achieve white light emission in a single substance upon UV irradiation stimulation.

The speciality of multi-color white light in single-phase substrate is of consequence for promoting the application of w-LEDs. Diverting energy from donor ions to acceptor ions is critical in this process (Li et al. 2014a; Wang et al. 2009). A series of emission spectra for SBSO:3%Tm$^{3+}$, y%Dy$^{3+}$ (y = 0, 1, 3, 5, 7, 9) were illustrated in Fig. 10, to verify the energy migration in co-doping single-phase system. It can be revealed that the blue proper emission for Tm$^{3+}$ and the yellow sharp peak for Dy$^{3+}$ ions emerge simultaneously in the PL spectrum of SBSO:Tm$^{3+}$, Dy$^{3+}$ under the 357 nm excitation. With
the invariant of Tm$^{3+}$ doping density (3 mol%) while the content for Dy$^{3+}$ increasing gradually, the Tm$^{3+}$ emission strength decreased uniformly, to the contrary that of Dy$^{3+}$ enhanced by degrees and the maximal intensity was at y = 7. Then the intensity of Dy$^{3+}$ emission shows a reduction caused by the self-quenching effect which were the interactions between Dy$^{3+}$ ions. In terms of the appearance above, it could reflect the fact that the energy transfer from Tm$^{3+}$ to Dy$^{3+}$ occurred in SBSO:Tm$^{3+}$, Dy$^{3+}$ system, while the emission of white light shall be attributed to the luminous color composition of Tm$^{3+}$ and Dy$^{3+}$ on account of the energy migration. This can be verified by the CIE chromaticity coordinates.

The decay curves for Tm$^{3+}$ emission with variations of Dy$^{3+}$ molarity in SBSO system were measured by the stimulation of 357 nm and monitoring at 456 nm, to explore and certify the energy transfer thoroughly. All the decay behaviors of luminescence as displayed in Fig. 11 shall be well-explained by a double-exponential relation:
where $I_0$ denotes the initial luminous intensity, $I$ denotes the emission intensity at a certain time, $A_1$, $A_2$ are constant coefficients, $t$ is time, $\tau_1$ and $\tau_2$ express the decay lifetimes for different luminous components, respectively. Furthermore, the following expression can determine the average values of decay lifetime:

$$
\tau_{avg} = \frac{A_1 \tau_1^2 + A_2 \tau_2^2}{A_1 \tau_1 + A_2 \tau_2}
$$

The lifetime values of Tm$^{3+}$ ions in SBSO:3%Tm$^{3+}$, y%Dy$^{3+}$ system were figured out, 6.18, 4.05, 3.14, 2.53 and 2.16 μs, respectively, when y = 0, 3, 5, 7 and 9. The energy transfer of Tm$^{3+} \rightarrow$ Dy$^{3+}$ was affirmed dramatically via the tendency of lifetime for Tm$^{3+}$ ions shortening monotonously with the rise in the level of Dy$^{3+}$ ions.
Normally, the energy transfer efficiency ($\eta_T$) from energy donor (sensitizer) to energy acceptor (activator) is worked out in terms of formula as follows (Dexter 1954):

$$\eta_T = 1 - \frac{I_S}{I_{SO}}$$  \hspace{1cm} (7)

where $I_S$ and $I_{SO}$ express the luminous intensities of the energy donor (Tm$^{3+}$) co-doping and no co-doping the energy acceptor (Dy$^{3+}$), respectively. The relation curve of $\eta_T$ as the function of Dy$^{3+}$ doping content is presented in Fig. 12. The $\eta_T$ value increases persistently with the rising in Dy$^{3+}$ concentrations nevertheless the growth rate slackens inch by inch, ultimately the energy transfer verges to saturation caused by the identical Tm$^{3+}$ concentration. The peak efficiency can be up to 80% at $y = 9$ which reveals that it is efficient.

Through the above discussion, it can be found that there are both spectral overlap and energy level matching between Tm$^{3+}$ and Dy$^{3+}$ ions, indicating that the energy migration for Tm$^{3+} \rightarrow$ Dy$^{3+}$ contains two kinds of manner: radiative transfer and resonant non-radiative transfer. According to Dexter’s theory, the energy transfer rate for Tm$^{3+} \rightarrow$ Dy$^{3+}$ can be defined by the following formula:

$$H_{DA} = \frac{2\pi}{\hbar} \langle D, A^* | H_{DA} | D^*, A \rangle \cdot \int gD(E) \cdot gA(E) dE$$  \hspace{1cm} (8)

where D represents energy donor, A represents energy acceptor, $H_{DA}$ is the Hamiltonian operator of the interaction, the integral item denotes as spectral overlap (radiative transfer) and the matrix element expresses the interaction between the initial state $|D^*, A\rangle$ and the final state $|D, A^*\rangle$ (resonant transfer interaction, related to the distance between ions) (Dexter 1954). In accordance with the above expression, it can been seen that when the concentration of two centers is very low and the distance between them is large, the energy transfer depends on the spectral overlap (radiation transfer is dominant); when the center concentration increases and the distance between centers shortens to a certain range, the nonradiative transfer of electric multipole interaction occurs; if the concentration of the centers is further increased, nonradiative transfer of exchange interaction may appear. The probability of energy transfer for Tm$^{3+} \rightarrow$ Dy$^{3+}$ in SBSO via electronic exchange interaction is excluded, since this mechanism is only effective at a shorter $R_c$ of $< 5$ Å (it can be estimated that $R_{\text{Tm-Dy}}$ is greater than 5 Å by Eq. 2). To analyze the occurrence of electrostatic interaction for resonant energy transfer, the following expression is given with respect to the Dexter’s energy transfer theory for multipolar interaction and Reisfeld’s approximation (Dexter 1954; Reisfeld and Lieblich-Soffer 1979):

$$\frac{I_{SO}}{I_S} \propto C^n$$  \hspace{1cm} (9)

where C is the sum of molarity for all doping ions (Tm$^{3+}$ and Dy$^{3+}$); Eq. (9) for n taking values such as 6, 8 and 10 can categorize as dipole–dipole, dipole–quadrupole and quadrupole–quadrupole interactions, respectively. Equation 9 is manifested in Fig. 13, and the best linear relation with better fitting at $n = 8$ demonstrates the mechanism of dipole–quadrupole interactions for energy transfer.

For a dipole–quadrupole interaction, the energy transfer probability ($P_{SA}$) from a donor to an acceptor is given by the following formula:
where $\lambda_S$ is the emission wavelength of the donor ions, $\tau_S$ is the radiative decay time of the donor, $R$ is the donor–acceptor average distance. The critical distance ($R_c$) of the energy transfer from the donor to acceptor is defined as the distance for which the probability of transfer equals the probability of radiative emission of the donor, i.e., the distance for which $P_{SA}\tau_S = 1$. Further, the donor–acceptor critical distance for energy transfer can be reckoned by the following expression in terms of the electric dipole-quadrupole mechanism (Blass 1969; Dexter 1954):

$$R_c = \left(\frac{3 \times 10^{12}\lambda_S^2 f_d}{R^8 \tau_S}\right)^{1/2} \int \frac{F_S(E)F_A(E)}{E^4} \, dE$$  \hspace{1cm} (11)

where $f_d \approx 0.02$ denotes the oscillator strength for the activator Dy$^{3+}$ ions electric dipole. $\int F_S(E)F_A(E)\, dE/E^4$ represents the spectral overlap amongst the normalized shapes of Tm$^{3+}$ emission $F_S(E)$ and Dy$^{3+}$ excitation $F_A(E)$, and the value is determined to be 0.000123 eV$^{-1}$ in our condition. Ultimately, the critical distance ($R_c$) of 7.22 Å is confirmed through Eq. (11).

Quantum efficiency (QE) is a very important physical parameter for phosphor materials in the practical application, defined as the ratio of the number of photons emitted to the number of photons absorbed. It can be measured by the integrated sphere (Liu et al. 2011), in accordance to the following equation:

$$P_{SA}(dq) = \frac{3 \times 10^{12}\lambda_S^2 f_q}{R^8 \tau_S} \int \frac{F_S(E)F_A(E)}{E^4} \, dE$$  \hspace{1cm} (10)
were $E_i(\lambda)$ is the integrated luminescence upon direct excitation, and $E_0(\lambda)$ is the integrated luminescence excited by indirect illumination from the integrated sphere. The value $L_e(\lambda)$ is the integrated excitation profile acquired from the empty integrated sphere (absence of sample). The optical absorbance $A$ can be gained via using the following formula:

$$A = \frac{L_0(\lambda) - L_i(\lambda)}{L_0(\lambda)}$$

where $L_0(\lambda)$ is the integrated excitation profile when the sample is diffusely illuminated by the integrated sphere’s surface, and $L_i(\lambda)$ is the integrated excitation profile when the sample is directly excited by the incident beam (Li 2014b). The corresponding value for SBSO:3%Tm$^{3+}$, y%Dy$^{3+}$ ($y = 0$, 3–9) phosphors excited at 357 nm are determined to be about 25.69%, 26.77%, 28.53%, 31.05% and 30.26%, respectively.

It is pointed out by Ronda that if the transition from the excited state to the ground state contains both radiative and nonradiative transitions, the transition rate ($k$, in $s^{-1}$) can be written as (Ronda 2008)

$$k = k_r + k_{nr}$$

where $k_r$ is the transition rate of only the emission process, and $k_{nr}$ is the rate of the nonradiative process. The existence of the nonradiative process will increase the transition rate, namely, a shorter decay time ($\tau$, equals $1/k$). If the decay time ($\tau_0$) without nonradiative transitions is known, the QE of the phosphor can be expressed as

$$QE = \tau / \tau_0$$

from the equation, it can be known that the change of QE should be consistent with the decay time, and the above results (decay time and QE) for our phosphors also confirm the conclusion. Therefore, the decrease of QE values for phosphors may be due to the increase of the nonradiation rate.

### 3.3 Luminescence and energy transfer characteristics of SBSO:Dy$^{3+}$, Eu$^{3+}$

As we all know, the high efficiency red emission corresponding to the typical 4f-4f parity-forbidden is the characteristic luminescence for Eu$^{3+}$ ions. It is considerable that Eu$^{3+}$ ions are applied in the Dy$^{3+}$ single-doped phosphor for supplying plentiful red constituent to implement the multicolor emission in accordance with the energy transfer of Dy$^{3+} \rightarrow$ Eu$^{3+}$. The photoluminescence excitation and emission spectra of Eu$^{3+}$-doped SBSO samples are exhibited in Fig. 14a. The excitation spectrum through the typical emission (611 nm) monitoring comprises a sharp line at 392 nm deriving from the transition $^7F_0 \rightarrow ^5L_6$ as well as a broad absorption band from 250 to 315 nm contributed principally by the Eu$^{3+}$–O$^{2-}$ and matrix charge transfer. In addition, there is a sharp absorption line appearing at 465 nm, ascribed to the energy level for $^7F_0 \rightarrow ^5D_2$, that is the hypersensitive transition of Eu$^{3+}$ ions. According to the Judd–Ofelt (JO) theory (Shanmuga Sundari et al. 2010), the hypersensitive transition follows the selection rule $|\Delta J| \leq 6, |\Delta L| \leq 2, \Delta S = 0$ and is related to parameter $\Omega_2$. In the present work, parameter $\Omega_2$ indicates the covalence of Eu–O bond and the asymmetry around Eu$^{3+}$ site. The parameter $\Omega_2$ of hypersensitive transition is of special
sensitivity to surrounding environment and related to the linear term of crystal field action. That is, the hypersensitive transition can occur only when the local symmetry makes the crystal field interaction having a linear term. Thus, the hypersensitive transition absorption (465 nm) in SBSO:Eu$^{3+}$ indicates that Eu$^{3+}$ is in the symmetry of C$_{2v}$ point group with linear term crystal field in the orthorhombic symmetry structure.

In the section of 500–750 nm (Fig. 14a), the emission spectrum upon stimulation of 392 nm exhibits several emission bands at 577, 590, 611 and 649 nm corresponding to the typical $^5D_0 \rightarrow ^7F_{0,1,2}$ transitions for Eu$^{3+}$ ions. The special emission transition of $^5D_0 \rightarrow ^7F_{0}$ at 577 nm disconfirms the selection rule (Judd–Ofelt theory), belonging to the parity-forbidden transition. But the crystal field interaction having a linear term of C$_{4v}$, C$_{2v}$ or C$_{3v}$ point group symmetry site can prompt the emission for $^5D_0 \rightarrow ^7F_{0}$. The appearance of $^5D_0 \rightarrow ^7F_{2}$ in SBSO:Eu$^{3+}$ confirms the conclusion for the hypersensitive transition absorption again. It is observed that the emission transition for $^5D_0 \rightarrow ^7F_{2}$ follows

![Excitation and Emission Spectra](image-url)
the selection rule $|\Delta J| \leq 6$ which comes under electric dipole induced in nature and remaining transition of $^5D_0 \rightarrow ^7F_1$ which obey the selection rule $|\Delta J|=0, \pm 1$ comes under magnetic dipole in nature. Generally, the parity of all states in $4f^n$ configuration of rare earth ions is the same. According to the parity selection rule of rare earth ions transition, it is known that the electric dipole is parity-forbidden and the magnetic dipole is parity-allowed in $4f^n$ configuration. When the sites of Eu$^{3+}$ ions are of inversion symmetry, only magnetic dipole allowed transition for $^5D_0 \rightarrow ^7F_1$ appears due to the crystal field without odd term. If the sites are lack of inversion symmetry, the opposite parity is mixed into $4f^n$ configuration, which makes the rule of parity selection relaxed because of the odd term of crystal field. At this time, the electric dipole transition of $^5D_0 \rightarrow ^7F_2$ dominates. In the present work, it can be found that the featured transition of $^5D_0 \rightarrow ^7F_2$ for electric dipole at 611 nm is superior to other transitions prominently (the magnetic dipole transition of $^5D_0 \rightarrow ^7F_1$ at 590 nm), intimating that the sites of Eu$^{3+}$ ions is lack of inversion symmetry. This also confirms the conclusion obtained from the emission spectrum of Dy$^{3+}$ ion. The emission spectra of SBSO:x\%Eu$^{3+}$ (x = 1–13) with diverse molarity of Eu$^{3+}$ are revealed in Fig. 14b. The optimal emission concentration for Eu$^{3+}$ ions is confirmed to be 9 mol% in SBSO host.

As contrasted in Fig. 15, the excitation spectrum for SBSO:7\%Dy$^{3+}$, 9\%Eu$^{3+}$ via the 611 nm emission (Eu$^{3+}$) monitoring is constituted by the composition of manifest Eu$^{3+}$ and Dy$^{3+}$ excitation bands, which substantiates the energy migration for Dy$^{3+} \rightarrow$ Eu$^{3+}$ preliminarily.

A series of SBSO:7\%Dy$^{3+}$, y\%Eu$^{3+}$ (y = 0, 1, 3, 5, 7, 9, 11, 13) were prepared and Fig. 16 displays the PL spectra corresponding to the as-synthesized samples with a 349 nm excitation source. The emission spectra exhibit not only the characteristic yellow emissions of Dy$^{3+}$ at 487 and 576 nm but also the red emitting peaks of Eu$^{3+}$ around 611 nm. Although the Dy$^{3+}$ ion concentration remains constant at 0.07, the feature emission (Dy$^{3+}$) weakens along with increasing Eu$^{3+}$ content, in the meantime the red luminescence (Eu$^{3+}$) rises until the concentration quenching at y = 9. The above exploration in the luminescence for Dy$^{3+}$ and Eu$^{3+}$ ions forcefully demonstrate the behavior in the transfer of energy from Dy$^{3+}$ to Eu$^{3+}$.
Fig. 16  a PL spectra for SBSO:7% Dy<sup>3+</sup>, γ% Eu<sup>3+</sup> phosphors with different Eu<sup>3+</sup> doped concentrations  b Dependence of emission intensity on Eu<sup>3+</sup> concentrations

Fig. 17  Decay lifetime tests of SBSO:7% Dy<sup>3+</sup>, γEu<sup>3+</sup> detected at 576 nm for Eu<sup>3+</sup> emission excited at 349 nm
The decay luminescence for Dy$^{3+}$ ion in SBSO:7%Dy$^{3+}$, y%Eu$^{3+}$ (y = 0, 1–13) upon the stimulation at 349 nm with 576 nm emission monitoring were recorded and presented in Fig. 17. All the decay behaviors show a precise fit with Eq. (5). According to Eq. (5) and (6), the characteristic lifetimes of Dy$^{3+}$ emission were reckoned to be 0.67, 0.60, 0.56, 0.53, 0.49 and 0.42 ms for Eu$^{3+}$ concentrations as 0, 3%, 5%, 7%, 9% and 11%, respectively. It reflects the phenomenon that the decay time of Dy$^{3+}$ monotonically decreases with the promoting amount of Eu$^{3+}$ ions, which provides convincing evidence for the energy transfer of Dy$^{3+}$ → Eu$^{3+}$. The ET efficiencies were estimated employing...
Fig. 20  Schematic energy-level diagram of energy transfer mechanism in the SBSO:Tm$^{3+}$/Dy$^3$/Eu$^{3+}$ system

Eq. (7) and the relation curve versus Eu$^{3+}$ molarity is plotted in Fig. 18. The $\eta_I$ value enlarges constantly in the wake of Eu$^{3+}$ concentrations rising and approximately is 82% at y = 13. can be estimated that $R_{\text{Dy-Eu}}$ is greater than 5 Å by Eq. (2), therefore the mechanism of energy transfer for Dy$^{3+} \rightarrow$ Eu$^{3+}$ in SBSO via electronic exchange interaction is excluded. Similarly, based on the relevant theory of multipolar interaction, the most plausible energy migration process for Dy$^{3+} \rightarrow$ Eu$^{3+}$ was analyzed by Eq. (9). The linear relationship of $(I_o/I) \propto C_n^{1/2}$ are represented in Fig. 19, and the best fitting factor $R^2 = 0.8832$ at n = 6 implied a dipole–dipole interaction for the energy transfer mechanism. For a dipole–dipole interaction, the donor–acceptor critical distance for energy transfer can be reckoned by the following expression (Blass 1969; Dexter 1954):

$$R_c^6 = (3 \times 10^{12})f_d \int \frac{F_s(E)F_A(E)}{E^4}dE$$  (16)
Thereby, the critical distance \((R_{\text{Dy-Eu}})\) is computed as 10.79 Å using Eq. (16).

The energy level transition of ET mechanism in \(\text{Tm}^{3+}/\text{Dy}^{3+}/\text{Eu}^{3+}\) co-doped SBSO system was investigated according to the schematic model as shown in Fig. 20. Under the irradiation at 357 nm the electrons located at the ground state \(3\text{H}_6\) are promoted to the emission level \(1\text{D}_2\) and then part of the stimulated electrons return to the lower state \((1\text{D}_2\rightarrow 3\text{F}_4)\) in \(\text{Tm}^{3+}\) ions, and emit a certain wavelength of light. While, the difference between the \(1\text{D}_2\) level of \(\text{Tm}^{3+}\) and the \(4\text{I}_{11/2}\) level of \(\text{Dy}^{3+}\) is not significant, another part of the stimulated electrons can transfer energy to \(\text{Dy}^{3+}\) in a non-radiative form, namely ET1. Meanwhile, a portion of \(\text{Tm}^{3+}\) emission is also received by \(\text{Dy}^{3+}\) ion, namely ET2, which arouses the electrons transition for \(6\text{H}_{15/2}\rightarrow 4\text{P}_{7/2}\) (\(\text{Dy}^{3+}\)), since the manifest overlap emerging in the spectra of \(\text{Dy}^{3+}\) and \(\text{Tm}^{3+}\). The activated electrons in the \(6\text{P}_{7/2}\) and \(4\text{I}_{11/2}\) level of \(\text{Dy}^{3+}\) undergo non-radiative relaxation, liberate to the \(4\text{F}_{9/2}\) level and then display a radiative release to the \(6\text{H}_{15/2}\) and \(6\text{H}_{13/2}\) levels under the combined actions of ET1 and ET2, giving rise to the typical yellow emission of \(\text{Dy}^{3+}\) at 488 and 576 nm. The energy level transition mechanism between \(\text{Dy}^{3+}\) and \(\text{Eu}^{3+}\) only belongs to ET1. Some energy for \(4\text{F}_{9/2}\) level (\(\text{Dy}^{3+}\)) is taken in the \(5\text{L}_6\) level (\(\text{Eu}^{3+}\)) through the modality of ET1, and the activated electrons at the emission level \(5\text{L}_6\) liberate to the lower energetic state \(5\text{D}_0\) by non-radiative relaxation. Further, a radiative release to the ground state \(7\text{F}_{0,1,2,3}\) induces the feature emissions at 579, 591, 611 and 652 nm for \(\text{Eu}^{3+}\) ion, respectively. It can be known from the above analysis that the cross relaxation occurs in the process of energy transfer for \(\text{Tm}^{3+}\rightarrow \text{Dy}^{3+}\) and \(\text{Dy}^{3+}\rightarrow \text{Eu}^{3+}\), that is, the transition of \(1\text{D}_2\) (\(\text{Tm}^{3+}\)) \(\rightarrow 4\text{F}_{9/2}\) (\(\text{Dy}^{3+}\)) \(\rightarrow 6\text{H}_{15/2}\) (\(\text{Dy}^{3+}\)) and \(9\text{P}_{7/2}\) (\(\text{Dy}^{3+}\)) \(\rightarrow 4\text{F}_{9/2}\) (\(\text{Dy}^{3+}\)) \(\rightarrow 6\text{D}_0\) (\(\text{Eu}^{3+}\)) \(\rightarrow 7\text{F}_2\) (\(\text{Eu}^{3+}\)). With the increasing of luminescent center concentration, the distance decreases and the interaction increases, which eventually leads to the raising of lattice vibration frequency. In this case, the energy donor relaxes from the higher excited energy level to the lower excited

![Fig. 21 The CIE chromaticity coordinates for SBSO:Tm\textsuperscript{3+}/Dy\textsuperscript{3+}/Eu\textsuperscript{3+} phosphors under 357 and 349 nm excitation](image-url)
level and then returns to the ground state in form of radiation to make the emission of light. Although this process leads to the emission quenching of high energy level, it also increases the aggregation number at low excited energy level. This is beneficial to the emission of the main energy level, so as to enhance the luminous intensity. Thus, the cross relaxation process in the energy transfer for $\text{Tm}^{3+} \rightarrow \text{Dy}^{3+}$ and $\text{Dy}^{3+} \rightarrow \text{Eu}^{3+}$ improves the luminous efficiency of energy acceptor $\text{Dy}^{3+}$ and $\text{Eu}^{3+}$ ions. However, the concentration quenching will occur when constant is further enhanced (beyond the critical distance), as discussed in the eighth paragraph.

The Commission International de L’Eclairage (CIE) chromaticity coordinates of SBSO:$\text{Tm}^{3+} / \text{Dy}^{3+} / \text{Eu}^{3+}$ specimen subjected to ultraviolet light excitation are disclosed in Fig. 21 and Table 5. For SBSO:7$\%$ $\text{Dy}^{3+}$, $y$%$\text{Eu}^{3+}$ system, as aggrandizing the value of $y$ (0–0.13), the color coordinates are converted piecemeal from (0.372, 0.401) to (0.553, 0.395) in accordance with the color tones from yellow to red. Based on the results, the SBSO:7$\%$ $\text{Dy}^{3+}$, $y$%$\text{Eu}^{3+}$ phosphors can achieve multicolor emission from yellow to red by modulating the molarity ratio of doping $\text{Dy}^{3+} / \text{Eu}^{3+}$ ions for its potential applications in solid-state lighting. In addition, the SBSO:3$\%$ $\text{Tm}^{3+}$ phosphor excited by 357 nm can exhibit bright blue emission, and then the emitting color transferred from blue to white region after introducing $\text{Dy}^{3+}$ ions. Furthermore, complete emission of white light is obtained when the doping levels of $\text{Tm}^{3+}$ and $\text{Dy}^{3+}$ in the samples are appropriately changed, which shall be potentially applied in white LEDs. Generally, particle size of phosphors is considered as an important parameter in the application of white LEDs. The particle size distributions of SBSO:3$\%$ $\text{Tm}^{3+}$, 7$\%$ $\text{Dy}^{3+}$ and SBSO:7$\%$ $\text{Dy}^{3+}$, 9$\%$ $\text{Eu}^{3+}$ phosphors were measured and exhibited in Fig. 22. The average diameters are calculated to be 7.89 and 7.43 μm, respectively, conforming to the requirement of application.

| Label | sample | CIE (x, y) | Color |
|-------|--------|------------|-------|
| 1     | SBSO:3$\%$ $\text{Tm}^{3+}$ | (0.163, 0.052) | Blue |
| 2     | SBSO:3$\%$ $\text{Tm}^{3+}$, 1$\%$ $\text{Dy}^{3+}$ | (0.226, 0.158) | Blue |
| 3     | SBSO:3$\%$ $\text{Tm}^{3+}$, 3$\%$ $\text{Dy}^{3+}$ | (0.253, 0.200) | Blue |
| 4     | SBSO:3$\%$ $\text{Tm}^{3+}$, 5$\%$ $\text{Dy}^{3+}$ | (0.264, 0.253) | White |
| 5     | SBSO:3$\%$ $\text{Tm}^{3+}$, 7$\%$ $\text{Dy}^{3+}$ | (0.287, 0.294) | White |
| 6     | SBSO:3$\%$ $\text{Tm}^{3+}$, 9$\%$ $\text{Dy}^{3+}$ | (0.305, 0.310) | White |
| 7     | SBSO:7$\%$ $\text{Dy}^{3+}$ | (0.372, 0.401) | Yellow |
| 8     | SBSO:7$\%$ $\text{Dy}^{3+}$, 1$\%$ $\text{Eu}^{3+}$ | (0.401, 0.378) | Yellow |
| 9     | SBSO:7$\%$ $\text{Dy}^{3+}$, 3$\%$ $\text{Eu}^{3+}$ | (0.423, 0.365) | Yellow |
| 10    | SBSO:7$\%$ $\text{Dy}^{3+}$, 5$\%$ $\text{Eu}^{3+}$ | (0.432, 0.344) | Yellow |
| 11    | SBSO:7$\%$ $\text{Dy}^{3+}$, 7$\%$ $\text{Eu}^{3+}$ | (0.487, 0.323) | Orange-red |
| 12    | SBSO:7$\%$ $\text{Dy}^{3+}$, 9$\%$ $\text{Eu}^{3+}$ | (0.512, 0.311) | Orange-red |
| 13    | SBSO:7$\%$ $\text{Dy}^{3+}$, 13$\%$ $\text{Eu}^{3+}$ | (0.553, 0.395) | Orange-red |
| 14    | SBSO:9$\%$ $\text{Eu}^{3+}$ | (0.633, 0.352) | Red |
4 Conclusion

On the whole, a sort of original emission-tunable single-component SBSO: Tm$^{3+}$/Dy$^{3+}$/Eu$^{3+}$ phosphor is prepared by high temperature solid state route and their luminescence characteristics were minutely studied. The steady-state and transient-state photoluminescence spectra/decay behaviors indicated the existence of the energy migration for Tm$^{3+}$→Dy$^{3+}$ and Dy$^{3+}$→Eu$^{3+}$ in the SBSO host and the ET efficiency can reach about 80%. While the efficient energy transfer emerges through the dipole–quadrupole mechanism for Tm$^{3+}$→Dy$^{3+}$ and the dipole–dipole interaction for Dy$^{3+}$→Eu$^{3+}$, and the critical distance (R_{Tm-Dy} and R_{Dy-Eu}) is estimated to be 7.22 Å and 10.79 Å, respectively. Furthermore, the tunable yellow-to-red emission can be achieved in SBSO: Dy$^{3+}$, Eu$^{3+}$ systems and Tm$^{3+}$, Dy$^{3+}$ co-doping samples can emit light entirely in the white region by the appropriate changeable doping molar quantity of Dy$^{3+}$/Eu$^{3+}$ and Tm$^{3+}$/Dy$^{3+}$ according to the energy migration in SBSO. These outstanding features make SBSO: Tm$^{3+}$/Dy$^{3+}$/Eu$^{3+}$ promising w-LED phosphors for its potential adhibitions in solid-state optical applications.
Authors' contributions All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Bo Yuan, Chaochao Qi and Xiangting Zhang. The first draft of the manuscript was written by Bo Yuan and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Code availability The authors declare that all data and materials as well as software application or custom code support our published claims and comply with field standards.

Declarations

Conflict of interest The authors declare that we have no known competing financial interests or personal relationships that could have appeared to influence the work reported in the manuscript entitled “Luminescence and tunable color properties of uniphase white-emitting Sr₃B₂SiO₈:Tm³⁺/Dy³⁺/Eu³⁺ phosphors by energy transfer for UV-excited white LEDs”.

Consent to participate We confirm that the manuscript has been read and approved by all named authors and that there are no other persons who satisfied the criteria for authorship but are not listed. We further confirm that the order of authors listed in the manuscript has been approved by all of us.

Consent for publication We confirm that we have given due consideration to the protection of intellectual property associated with this work and that there are no impediments to publication, including the timing of publication, with respect to intellectual property. In so doing we confirm that we have followed the regulations of our institutions concerning intellectual property.

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