A novel optical thermometry based on the energy transfer from charge transfer band to Eu$^{3+}$-Dy$^{3+}$ ions

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Optical thermometry based on the up-conversion intensity ratio of thermally coupled levels of rare earth ions has been widely studied to achieve an inaccessible temperature measurement in submicron scale. In this work, a novel optical temperature sensing strategy based on the energy transfer from charge transfer bands of W-O and Eu-O to Eu$^{3+}$-Dy$^{3+}$ ions is proposed. A series of Eu$^{3+}$/Dy$^{3+}$ co-doped SrWO$_4$ is synthesized by the conventional high-temperature solid-state method. It is found that the emission spectra, emission intensity ratio of Dy$^{3+}$ (572 nm) and Eu$^{3+}$ (615 nm), fluorescence color, lifetime decay curves of Dy$^{3+}$ (572 nm) and Eu$^{3+}$ (615 nm), and relative and absolute sensitivities of Eu$^{3+}$/Dy$^{3+}$ co-doped SrWO$_4$ are temperature dependent under the 266 nm excitation in the temperature range from 11 K to 529 K. The emission intensity ratio of Dy$^{3+}$ (572 nm) and Eu$^{3+}$ (615 nm) ions exhibits exponentially relation to the temperature due to the different energy transfer from the charge transfer bands of W-O and Eu-O to Dy$^{3+}$ and Eu$^{3+}$ ions. In this host, the maximum relative sensitivity S can be reached at 1.71% K$^{-1}$, being higher than those previously reported material. It opens a new route to obtain optical thermometry with high sensitivity through using down-conversion fluorescence under ultraviolet excitation.

Recently, white light emitting diode (LED) technology has attracted much attention in the solid-state lighting industry, due to the advantages of white LEDs including power saving, long lifetime, and environmental benefit. Single-phase luminescent materials that can directly emit white light under UV excitation have been explored in oxyfluoride glass and oxides matrices. The Eu$^{3+}$ and Dy$^{3+}$ ions were chosen as the red, green, and blue emitting activator centers through the transitions $^{5}D_{0} \rightarrow ^{7}F_{2}$ (Eu$^{3+}$) and $^{5}D_{0} \rightarrow ^{4}H_{11/2}$ (Dy$^{3+}$) under ultraviolet excitation. For examples, Das and co-authors reported the controllable white light emission from Dy$^{3+}$-Eu$^{3+}$ co-doped KCaBO$_3$ phosphor. Laguna reported the shape controlled white light emission from Dy$^{3+}$-Eu$^{3+}$ co-doped CaMoO$_4$ microarchitectures. Hirai obtained the white light emission from Dy$^{3+}$-Eu$^{3+}$ co-doped SrCeO$_4$. In these works, the white light emission was controlled by changing doping concentration and host types.

It was reported that the temperature was a key parameter to adjust the emission intensity, the fluorescence intensity ratio, and emission color. Berry found that the lifetime of $^{5}D_{0} \rightarrow ^{7}F_{2}$ (Eu$^{3+}$) ion was temperature dependent in Europium Tris(2,2,6,6-tetramethyl-3,5-heptanedionato). Morgan observed that the homogeneous linewidth of the $^{5}D_{0} \rightarrow ^{5}F_{2}$ transition of Eu$^{3+}$ was dependent on temperature in amorphous hosts. Eckert observed that the phosphorescence decay lifetime of the Dy$^{3+}$-transitions in Dy$^{3+}$: Al$_2$O$_3$ showed strong temperature dependency in a temperature range from 1100 to 1500 K. Zhou reported that the emission intensity ratio of $^{5}D_{0}$ to $^{5}D_{2}$ of Eu$^{3+}$--doped transparent MF$_2$ (M = Ba, Ca, Sr) glass ceramics increased with the temperature increase. However, the temperature dependent optical property of Dy$^{3+}$-Eu$^{3+}$ co-doped materials has not been studied so far. It is necessary to explore the spectra and energy transfer of Dy$^{3+}$-Eu$^{3+}$ co-doped materials at high temperature.

From the published work on the spectra of Dy$^{3+}$-Eu$^{3+}$ co-doped materials, one can find that it had a little overlap of the excitation spectrum between $^{5}D_{0} \rightarrow ^{5}F_{2}$ (Eu$^{3+}$) and $^{5}F_{2} \rightarrow ^{5}I_{15/2}$ (Dy$^{3+}$) overlaps. It is necessary to find another ion to sensitize the Dy$^{3+}$ and Eu$^{3+}$ simultaneously. Notably, the charge transfer band of W-O was reported to have the wide absorption band in the ultraviolet range from 200 nm to 300 nm. It may be a promise sensitizer to excite Dy$^{3+}$ and Eu$^{3+}$ simultaneously. Thus, in this work, the optical temperature property of Eu$^{3+}$/Dy$^{3+}$...
co-doped SrWO₄ are studied under 266 nm excitation. It is observed that the fluorescence intensity ratio between Eu³⁺ and Dy³⁺ emissions are strongly dependent on the temperature in the temperature range from 11 K to 529 K. The Eu³⁺/Dy³⁺ co-doped SrWO₄ phosphors are proved as an excellent materials used for optical thermometry, due to its maximum value of Sr as high as 1.71% K⁻¹.

**Results**

The X-ray diffraction (XRD) patterns of the SrWO₄, SrWO₄: 0.4 mol% Eu³⁺, and SrWO₄: x Eu³⁺, 4 mol% Dy³⁺ (x = 0, 0.2 mol%, 0.4 mol%, 0.6 mol%, 0.8 mol%, 1 mol%) samples synthesized by high-temperature solid-state reaction method are shown in Fig. 1. The peaks of all the products can be easily indexed to tetragonal system of SrWO₄, which has a I41/a space group (PDF# 08-0490, unit cell parameters: a = b = 5.416 Å, c = 11.95 Å). No trace of impurity peaks can be found when Dy³⁺ and Eu³⁺ ions are introduced into the system. Compared with the pure SrWO₄, the diffraction peaks of the Eu³⁺ single-doped and Eu³⁺/Dy³⁺ co-doped SrWO₄ exhibit a slight shift toward high-angle side, due to substitution of Sr²⁺ ions by smaller size Dy³⁺ (1.03 Å, CN = 8) and Eu³⁺ (1.07 Å, CN = 8) ions, which revealing that Dy³⁺ and Eu³⁺ ions have been successfully doped into the system. Figure 2 shows the unit cell parameters of a (Å) and c (Å) as well as unit cell volume (Å³) at different Eu³⁺ concentration in tetragonal SrWO₄: x Eu³⁺, 4 mol% Dy³⁺ (x = 0, 0.2 mol%, 0.4 mol%, 0.6 mol%, 0.8 mol%, 1 mol%).

**Figure 1.** (a) XRD patterns of the as-synthesized SrWO₄, SrWO₄: 0.4 mol% Eu³⁺, and SrWO₄: x Eu³⁺, 4 mol% Dy³⁺ (x = 0, 0.2 mol%, 0.4 mol%, 0.6 mol%, 0.8 mol%, 1 mol%) phosphors. The standard data of tetragonal SrWO₄ (PDF# 08-0490) is given as a reference; (b) Partially enlarged XRD patterns of the corresponding phosphors (2θ = 27–29°).

**Figure 2.** (a) Unit cell parameters of a (Å) and (b) c (Å) and (c) unit cell volume (Å³) at different Eu³⁺ concentration in tetragonal SrWO₄: x Eu³⁺, 4 mol% Dy³⁺ (x = 0, 0.2 mol%, 0.4 mol%, 0.6 mol%, 0.8 mol%, 1 mol%).

It can be observed that the fluorescence intensity ratio between Eu³⁺ and Dy³⁺ emissions are strongly dependent on the temperature at the temperature range from 11 K to 529 K. The Eu³⁺/Dy³⁺ co-doped SrWO₄ phosphors are proved as an excellent materials used for optical thermometry, due to its maximum value of Sr as high as 1.71% K⁻¹.
differences between the different valence state cations. The same tendency can be observed in the values of parameter $c$ (Å) and volume (Å$^3$). It reveals that Eu$^{3+}$ and Dy$^{3+}$ ions can be easily doped into SrWO$_4$ lattice, and the lattice can be distorted by the doping ions.

The scanning electron microscopy (SEM) image of a representative SrWO$_4$: 0.4 mol% Eu$^{3+}$, 4 mol% Dy$^{3+}$ sample is shown in Fig. 3a, exhibiting sphere-like morphology with a particle size of about 1 µm. The energy dispersive spectrometer (EDS) spectrum (Fig. 3b) confirms the presence of Sr, W, O, Eu, and Dy elements, and further providing the evidence that Dy$^{3+}$ and Eu$^{3+}$ ions have been successfully doped into the SrWO$_4$ host lattice.

The ultraviolet-visible diffuse reflectance spectrum of the SrWO$_4$: 0.4 mol% Eu$^{3+}$, 4 mol% Dy$^{3+}$ phosphor at room temperature is shown in Fig. 4. A broad band and several absorption peaks corresponding to the doped ions can be observed. The broad band is located from 200 to 350 nm, corresponding to the O-W ligand-to-metal charge transfer in the WO$_4^{2-}$ group. Four absorption peaks located at 365, 384, 426 and 454 nm can be assigned to the intra 4f electronic transitions of $7F_0 \rightarrow 5D_4$ (Eu$^{3+}$), $7F_0 \rightarrow 5G_2$ (Eu$^{3+}$), $6H_{15/2} \rightarrow 4G_{11/2}$ (Dy$^{3+}$), and $6H_{15/2} \rightarrow 4I_{15/2}$ (Dy$^{3+}$), respectively.

The PLE spectra of SrWO$_4$: 4 mol% Dy$^{3+}$, SrWO$_4$: 0.4 mol% Eu$^{3+}$, and SrWO$_4$: 0.4 mol% Eu$^{3+}$, 4 mol% Dy$^{3+}$ samples are shown in Fig. 5. The PLE spectrum of SrWO$_4$: 4 mol% Dy$^{3+}$ (Fig. 5a) illustrates a broad charge transfer band centered at 247 nm from 200 nm to 280 nm and a series of sharp lines extended to visible
The broad band can be ascribed to the charge transfer from WO$_4^{2−}$ group to Dy$_3^{2+}$, and the seven sharp lines can be ascribed to f-f transitions of Dy$_3^{2+}$, $^{4}f$ configuration, which are $^{6}H_{15/2} \rightarrow ^{4}K_{15/2}$ (296 nm), $^{6}H_{15/2} \rightarrow ^{4}M_{15/2}$ (350 nm), $^{6}H_{15/2} \rightarrow ^{4}P_{3/2}$ (365 nm), $^{6}H_{15/2} \rightarrow ^{4}M_{21/2}$ (387 nm), $^{6}H_{15/2} \rightarrow ^{4}G_{11/2}$ (426 nm), and $^{6}H_{15/2} \rightarrow ^{4}I_{15/2}$ (450 nm), respectively. The excitation spectrum of SrWO$_4$: 0.4 mol% Eu$^{3+}$ is shown in Fig. 5b. Monitored at 615 nm, an intense broad band can be found in the range of 250–320 nm, which is due to the Eu-O charge transfer transition. While in the range of 200–250 nm, no obvious band can be found, indicating the energy transfer from WO$_4^{2−}$ group to Eu$^{3+}$ is negligible. Additionally, a series of sharp lines corresponding to the intra 4f electron transitions of Eu$^{3+}$ ion can also be observed, which are 360 nm ($^{7}F_{0} \rightarrow ^{5}D_{4}$), 380 nm ($^{7}F_{0} \rightarrow ^{5}L_{6}$), 393 nm ($^{7}F_{0} \rightarrow ^{5}L_{7}$), 414 nm ($^{7}F_{0} \rightarrow ^{5}D_{3}$), and 463 nm ($^{7}F_{0} \rightarrow ^{5}D_{2}$), respectively. The excitation spectrum of SrWO$_4$: 0.4 mol% Eu$^{3+}$ is shown in Fig. 5c.

Figure 5. Excitation spectra of (a) SrWO$_4$: 4 mol% Dy$^{3+}$, (b) SrWO$_4$: 0.4 mol% Eu$^{3+}$, and (c) SrWO$_4$: 0.4 mol% Eu$^{3+}$, 4 mol% Dy$^{3+}$ phosphors at room temperature.
total emissions intensity, when the doping concentration of Eu$^{3+}$ reaches to 0.8 mol%, the strongest total emission intensity is obtained. Thus, the sample co-doped with 0.4 mol% Eu$^{3+}$ and 4 mol% Dy$^{3+}$ should be selected as the optimum doping concentration to study optical properties at different temperature.

The effective lifetimes of 4F9/2 and 5D0 energy levels can be expressed as

$$\tau_{\text{eff}} = \frac{\int I(t)dt}{\int I(t)dt}$$

where $I(t)$ represents the emission intensity at time $t$. The decay curves of Dy$^{3+}$ ($^{4}F_{9/2}$) and Eu$^{3+}$ ($^{5}D_{0}$) ions at different Eu$^{3+}$ concentration were recorded by monitoring at 572 nm and 615 nm, respectively. The decay curves support the existence of energy transfer progress for doped and co-doped samples. The values of lifetimes of SrWO$_4$: $x$ Eu$^{3+}$, 4 mol% Dy$^{3+}$ ($x = 0, 0.2$ mol%, 0.4 mol%, 0.6 mol%, 0.8 mol%, 1 mol%) phosphors were calculated by using equation (1), in Fig. 6c. The decreasing tendency of lifetimes of both $^{4}F_{9/2}$ and $^{5}D_{0}$ energy levels can be found with the rise of Eu$^{3+}$ concentration. The Fig. 6c shows the inhomogeneous change of lifetimes of the 572 nm (Dy$^{3+}$) and 615 nm (Eu$^{3+}$) emissions. It means that the energy transfer from charge transfer band of W-O to Dy$^{3+}$ and the energy transfer from charge transfer band of Eu-O to Eu$^{3+}$ as well as energy transfer from Eu$^{3+}$ to Dy$^{3+}$ are different at different Eu$^{3+}$ concentrations.

To further study the temperature-dependent photoluminescence performance, the emission spectra of the SrWO$_4$: 0.4 mol% Eu$^{3+}$, 4 mol% Dy$^{3+}$ samples are investigated in the temperature range from 11 K to 592 K, as shown in Fig. 7a. One can see that the emission intensity of Dy$^{3+}$ ions increases with the rise of temperature, while the emission intensity of Eu$^{3+}$ ions decreases. The emission bands of Dy$^{3+}$ ions at 572 nm ($^{4}F_{9/2} \rightarrow ^{4}H_{3/2}$) and Eu$^{3+}$ ions at 615 nm ($^{5}D_{0} \rightarrow ^{7}F_{2}$) were enlarged and shown in Fig. 7b. One can find that the intensity of 572 nm (Dy$^{3+}$) increases with the temperature increase, while the intensity of 615 nm (Eu$^{3+}$) decreases with the temperature increase. It means that the energy transfer from charge transfer bands to Eu$^{3+}$ and Dy$^{3+}$ ions is temperature dependent. The Commission International de L'Eclairage (CIE) diagram (Fig. 7c) shows that the emission color of the SrWO$_4$: 0.4 mol% Eu$^{3+}$, 4 mol% Dy$^{3+}$ sample can be turned from the orange-red to the yellow region with the increase of temperature from 11 K to 529 K.

In order to study the energy transfer among charge transfer bands, Eu$^{3+}$ and Dy$^{3+}$, the decay curves of $^{4}F_{9/2}$ and $^{5}D_{0}$ energy levels at different temperature were measured by monitoring at 572 nm and 615 nm, respectively, and calculated by using equation (1). The values of the effective lifetimes are shown in Fig. 7d. It can be found that...
the lifetimes of $^4\text{F}_{9/2}$ energy level of Dy$^{3+}$ ion increase with the increase of temperature, while the lifetimes of $^5\text{D}_0$ energy level of Eu$^{3+}$ ion decrease, demonstrating the different energy transfer rates from charge transfer bands to Dy$^{3+}$ and Eu$^{3+}$ ions.

To study the temperature dependence of energy transfer from charge transfer bands to Eu$^{3+}$-Dy$^{3+}$ ions, the dynamic balance rate-equation model for the energy transfer between charge transfer bands and Eu$^{3+}$-Dy$^{3+}$ ions are established in Fig. 8. We supposed $^7\text{F}_J$ ($J = 0, 1, 2, 3, 4, 5, 6$), $^4\text{H}_{12/2}$ ($J = 15, 13, 11, 9, 7$), or $^1\text{B}(^1\text{T}_2)/^1\text{E}(^1\text{T}_2)/^1\text{E}(^1\text{T}_1)$ energy levels as a same level in the case of the fixed temperature. The energy transfer between Eu$^{3+}$ and WO$_4^{2-}$ is neglected. The corresponding rate equations are as follows:

$$\frac{dN_i}{dt} = N_i W_{j1} - N_i A_{10}$$

$$\frac{dN_2}{dt} = N_2 W_{32} - \beta_2 N_2 N_4 - N_2 W_{31}$$

$$\frac{dN_3}{dt} = \sigma_1 \rho_1 N_0 - N_3 W_{32}$$

$$\frac{dN_4}{dt} = N_4 W_{45} - N_4 A_{56}$$

$$\frac{dN_5}{dt} = \sigma_2 \rho_2 N_7 - N_5 A_{87} - \beta_2 N_5 N_4$$

where $\sigma$, and $\rho$, are the cross-section of the ground state absorption of $^7\text{F}_J$ and $^1\text{A}_1$, $\rho_1$ and $\rho_3$ are the incident pumping power density, $N_0$, $N_1$, $N_2$, $N_3$, $N_4$, $N_5$, $N_6$, $N_7$, and $N_8$ are the population densities of the levels of Eu$^{3+}$, (WO$_4$)$^{2-}$, and Dy$^{3+}$ respectively. $\beta_1$ and $\beta_2$ correspond to the energy transfer rates from $^5\text{D}_3$ and $^1\text{B}(^1\text{T}_2)/^1\text{E}(^1\text{T}_2)/^1\text{E}(^1\text{T}_1)$ to $^4\text{I}_{15/2}$, respectively. The terms of $W_{ij}$ represent the nonradiative decay rates between the levels $i$ and $j$. $A_{ij}$ is the radiative transition rates between the levels $i$ and $j$.

By solving the above equations, we have...
The nonradiative relaxation possibility is proportional to ref. 34

\[ w_{ij} \propto e^{-h\omega/kT} \]  

The luminescence intensity of an emission band can be expressed as

\[ I_i = h\nu A_{ij} N_j \]  

where \( h\nu \) is transition energy per photon, \( A_{ij} \) is spontaneous radiative emission probability from an \( i \) state to a \( j \) state, and \( N_j \) is the state population of the \( i \) state 35.

The emission intensity ratio of Dy\(^{3+}\) (572 nm) and Eu\(^{3+}\) (615 nm) ions, defined as \( \text{FIR}(I_{Dy}/I_{Eu}) \), is adopted to study the temperature-dependent photoluminescence property. Combining with above equations, the \( \text{FIR}(I_{Dy}/I_{Eu}) \) can be fitted as

\[ \text{FIR} = \frac{I_{Dy}}{I_{Eu}} = A \exp\left(-\frac{2h\omega/kT}{1 - \exp(-h\omega/kT)}\right) + B \]  

where \( A \) is the fitting constant that depends on the experimental system and intrinsic spectroscopic parameter; \( h\omega \) is the phonon energy; and \( k \) is a Boltzmann constant 36. The absolute sensitivity and relative sensitivity can be defined as

\[ S_a = \frac{dR}{dT} = \frac{A h\omega \exp(-3h\omega/kT)}{k(1 - \exp(-h\omega/kT))^2 T^2} + \frac{2A h\omega \exp(-2h\omega/kT)}{k(1 - \exp(-h\omega/kT))^2 T^2} \]  

\[ S_r = \frac{1}{R} \frac{dR}{dT} = \frac{A h\omega \exp(-3h\omega/kT)}{k(1 - \exp(-h\omega/kT))^2 T^2} + \frac{2A h\omega \exp(-2h\omega/kT)}{k(1 - \exp(-h\omega/kT))^2 T^2} \frac{1}{B + \frac{A h\omega \exp(-h\omega/kT)}{1 - \exp(-h\omega/kT)}} \]  

As displayed in Fig. 9a, the \( \text{FIR} \) data could be exponentially fitted by the equation (10) from 11 K to 529 K. The parameters \( A, B \) and \( h\omega \) can be determined to be 3250.7, 0.55 and 903.8 cm\(^{-1}\) for the SrWO\(_4\): 0.4 mol% Eu\(^{3+}\), 4 mol% Dy\(^{3+}\) sample by using the fitting equation. The fitted phonon energy of 903.8 cm\(^{-1}\) is close to the literature reported of 917.7 cm\(^{-1}\) 38. The error of the fitted phonon energy is about 1.5%. On the basis of the equations (11) and (12), the absolute sensitivity \( S_a \) and relative sensitivity \( S_r \) are calculated and shown in Fig. 9b,c. One can
see that the absolute sensitivity is as high as 0.27 K\(^{-1}\) at 529 K. It is much higher than the literature reported\(^{39,40}\). For example, the absolute sensitivity in Eu\(^{3+}\) doped Gd\(_2\)Ti\(_2\)O\(_7\) phosphor was 0.015 K\(^{-1}\)\(^{41}\), and in Dy\(^{3+}\) doped Gd\(_2\)O\(_3\) phosphor was 0.01 K\(^{-1}\)\(^{42}\). The maximum relative sensitivity of 1.71% K\(^{-1}\) is obtained at 335 K. It is higher than the reported phosphors, 0.014 K\(^{-1}\) in Eu\(^{3+}\) doped CaGd\(_2\)(WO\(_4\))\(_4\) scheelite\(^{43}\) and 0.003 °C\(^{-1}\) in Dy\(^{3+}\) doped Y\(_2\)Al\(_2\)O\(_3\) phosphor\(^{44}\). The improvement of both the relative sensitivity and absolute sensitivity of this material may be owing to different energy transfer ratio from charge transfer bands to Eu\(^{3+}\)-Dy\(^{3+}\) ions at different temperatures, leading to a significant change in the emission intensity ratio.

The error analysis of measured and calculated FIR\((I_{572}/I_{615})\) is shown in Fig. 10a. One can see that the measured and the calculated FIR match well at low temperature, while the error appears at high temperature more than 400 K. The error may originated from the active nonradiative relaxation and energy transfer between Eu\(^{3+}\)/Dy\(^{3+}\) ions and host\(^{39,45}\). Notably, this error affects little on the values of S\(_a\) and S\(_r\), as shown in Fig. 10b,c.

Conclusions
In this work, a series of Eu\(^{3+}\)/Dy\(^{3+}\) co-doped SrWO\(_4\) phosphors were prepared by the high-temperature solid-state method. The structural property was studied by the X-Ray diffraction. The emission intensity, fluorescence color, and lifetimes of Dy\(^{3+}\) (572 nm) and Eu\(^{3+}\) (615 nm) of the SrWO\(_4\): 0.4 mol% Eu\(^{3+}\), 4 mol% Dy\(^{3+}\) are investigated in the temperature range from 11 K to 529 K under the 266 nm excitation. The emission intensity ratio of Dy\(^{3+}\) and Eu\(^{3+}\) ions was found to be temperature dependent. The maximum value of S\(_a\) can be reached 1.71% K\(^{-1}\) at 335 K, being higher than those previously reported material. This work opens a new route to obtain optical thermometry with high sensitivity through using down-conversion fluorescence under ultraviolet excitation.

Methods
A series of Eu\(^{3+}\)/Dy\(^{3+}\) single doped and co-doped SrWO\(_4\) phosphors were prepared by the high-temperature solid-state method. According to the appropriate stoichiometric ratio, the starting materials, SrCO\(_3\) (Aldrich, 99.9%), WO\(_3\) (Aldrich, 99.9%), Eu\(_2\)O\(_3\) (Aladdin, 99.99%), and Dy\(_2\)O\(_3\) (Aldrich, 99.99%) were weighted and ground thoroughly in an agate mortar for 30 minutes with ethanol. Then the homogenous mixture was collected into a crucible and sintered at 1000 °C to 529 K under the 266 nm excitation. The emission intensity ratio of Dy\(^{3+}\) and Eu\(^{3+}\) ions was found to be temperature dependent. The maximum value of S\(_a\) can be reached 1.71% K\(^{-1}\) at 335 K, being higher than those previously reported material. This work opens a new route to obtain optical thermometry with high sensitivity through using down-conversion fluorescence under ultraviolet excitation.
The photoluminescence excitation (PLE) spectra are recorded by a Pjoton Technology International (PTI, USA) fluorimeter with a 60 W Xe-arc lamp as the excitation light source at room temperature. The photoluminescence (PL) spectra and decay lifetimes are collected by a 266 nm-pulsed laser with a pulse width of 5 ns and a repetition rate of 10 Hz (Spectron Laser Sys. SL802G).

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Author Contributions

X.W. and H.S. developed the idea and supervised the project. J.W. did all the synthetic experiments and performed measurements. Y.B. analyzed the structure and spectra properties. All authors discussed the results and contributed to writing the manuscript.

Additional Information

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