Supporting Information

Data-Driven Photoluminescence Tuning in Eu^{2+} Doped Phosphors

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EXPERIMENTAL SECTION

Reagents. Potassium carbonate (K_2CO_3, A.R.), rubidium carbonate (K_2CO_3, A.R.), lutecia (Lu_2O_3, 99.99%), silicon dioxide (SiO_2, A.R.), and europia (Eu_2O_3, 99.99%). All chemicals were used as received.

Synthesis. $R_{1-x}K_xLSO:0.01Eu^{2+}$ ($0 \leq x \leq 1$) powders were synthesized via high-temperature solid-state reaction. The stoichiometric mixtures of K_2CO_3, Rb_2CO_3, Lu_2O_3, SiO_2, and Eu_2O_3 were thoroughly grounded and transferred into an alumina crucible. After that, the mixed starting materials were sintered at 1000 - 1200 ^oC for 6 h under a reducing atmosphere (N_2 : H_2 = 80% : 20%) in a tube furnace. After the calcinations, the samples were slowly cooled to room temperature and crushed into powders for further characterization.

Characterization. The phase and purity of the as-prepared powder samples were examined by X-ray diffraction (XRD) analysis using an Aeris powder diffractometer (PANalytical Corporation, Netherlands) operating at 40 kV and 15 mA with monochromatized Cu Kα radiation (λ = 1.5406 Å). Rietveld refinement was conducted using TOPAS 4.2 software. Photoluminescence (PL) and Photoluminescence excitation (PLE) spectra were measured by an Edinburgh FLS920 fluorescence spectrometer equipped with continuous xenon (Xe) lamps (450 W). The fluorescent decay curves were also measured by the FLS920 instrument with an nF900 flash lamp as the excitation source.

Computations via the regression model. To construct the general prediction model, one should have information on two numeric characteristics for each member of a group, which are related to each other. In such a situation, it is able to plot the data related to the characteristics of a scatter diagram. If the dots fall roughly along a straight line, sloping either upwards or downwards, it will conclude that a relationship exists. As a next step, linear regression analysis is used to make the relationship computable, which means that knowing the value of one variable can predict the value of the other variable in the group. The known value variable refers to the independent variable, and the predicted variable refers to the dependent variable. The independent and dependent variables, by convention, refer to "x" and "y", plotting as horizontal and vertical axes, respectively. Namely, linear regression analysis on two variables (x and y) in a sample can be looked upon as plotting the data to a best-fit line as is shown in Figure S1 in the supporting information (SI). This best fit line is chosen in the minimum sum of squares of all the residuals (the vertical distance of each point from the line), as least-squares line. This line can be mathematically defined by an equation of the form:

$$Y = a + bx \qquad (1)$$

where "x" is the known value of the independent variable, "Y" is the predicted value of "y" for the given value of "x", "a" is called as the "intercept" of the estimated line, representing the value of Y when $x = 0$, and "b" is called as the "slope" of the estimated line representing the amount by which Y changes on average as "x" increases by one unit. It is also referred to "coefficient", "regression coefficient", or "gradient".[1,2]

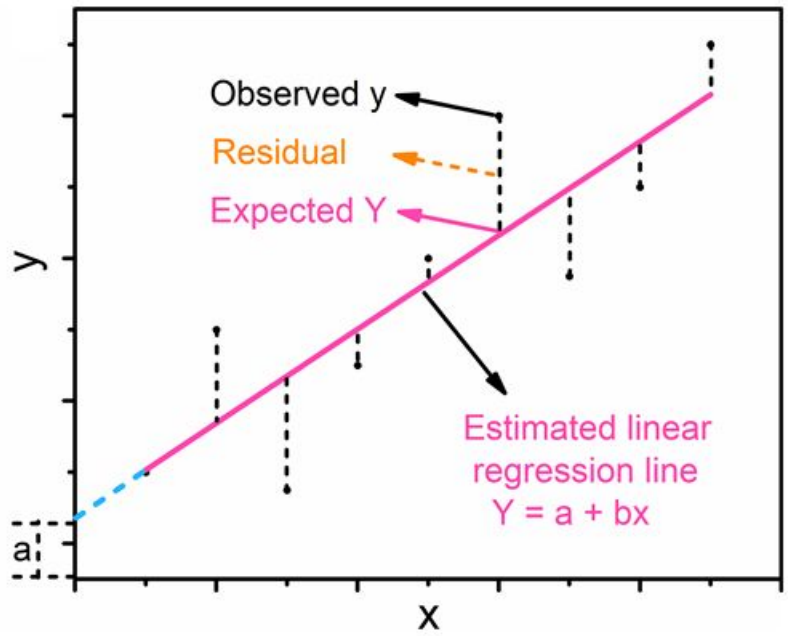

Figure S1. Data from a sample and the estimated linear regression line for these data. Each dot corresponds to a data point, i.e., an individual pair of values for *x* and *y*, and the vertical dashed lines from each dot represent residuals. The capital letters (*Y*) are used to indicate predicted values and lowercase letters (*x* and *y*) for known values. Intercept is shown as "*a*" and slope or regression coefficient as "*b*".

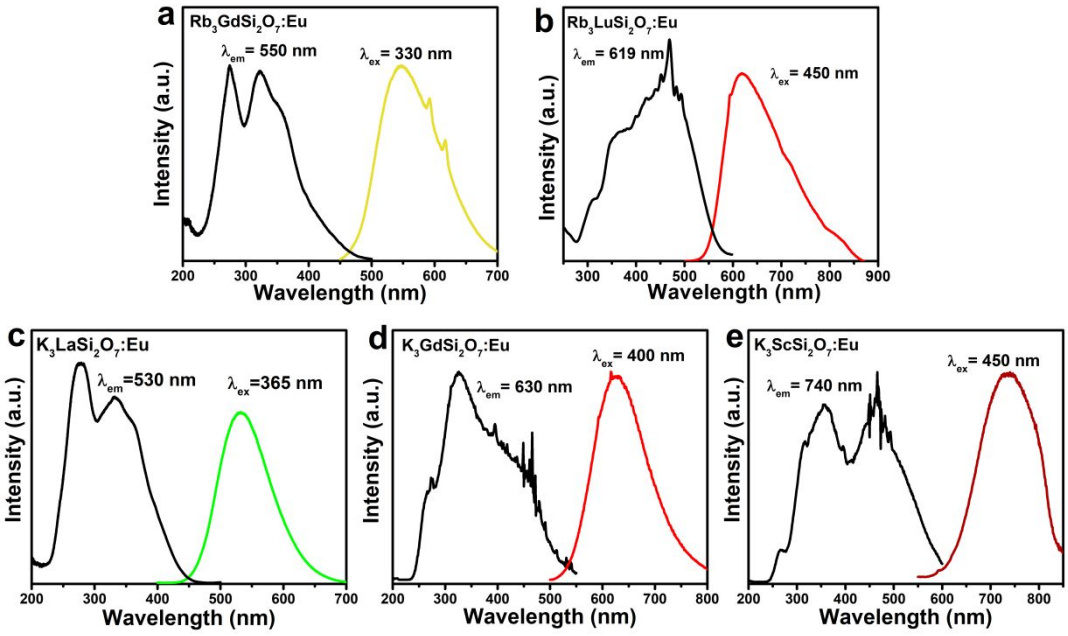

Figure S2. PLE and PL spectra of as-prepared $Rb_3GdSi_2O_7:Eu^{2+}$, $Rb_3LuSi_2O_7:Eu^{2+}$, $K_3LaSi_2O_7:Eu^{2+}$, $K_3GdSi_2O_7:Eu^{2+}$ and $K_3ScSi_2O_7:Eu^{2+}$, and the corresponding monitoring and excitation wavelengths are marked.

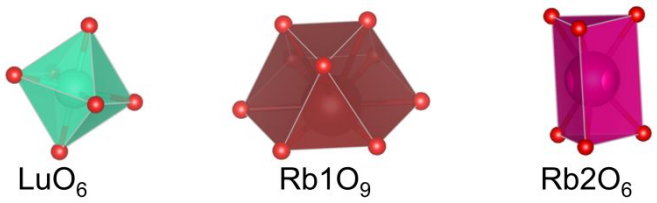

Figure S3. The coordination of metal cations in $Rb_3LuSi_2O_7$.

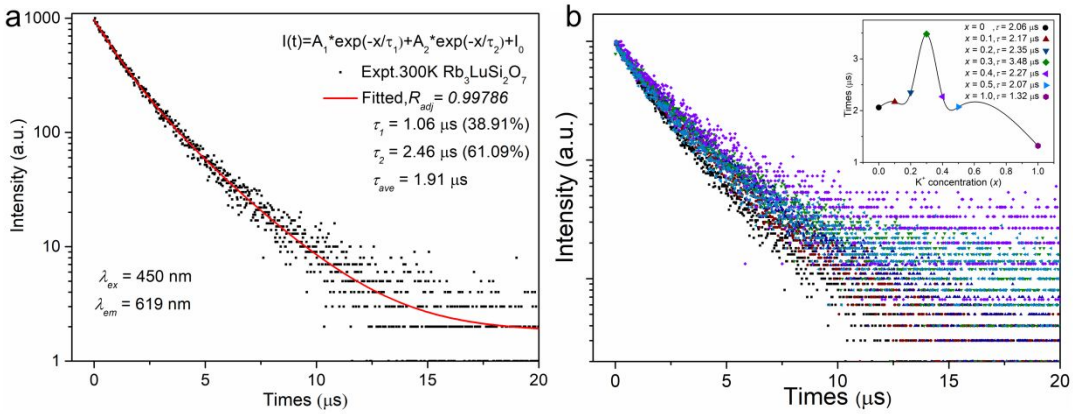

Figure S4. (a) Photoluminescence decay curve of $Rb_3LuSi_2O_7$:0.01Eu^{2+}. (b) Photoluminescence decay curves of $R_{1-x}K_xLSO$:0.01Eu^{2+} phosphors under excitation at 450 nm monitored at each maximum emission wavelengths. The inset shows the lifetime variation of $R_{1-x}K_xLSO$:0.01Eu^{2+} phosphors.

Table S1. Main parameters of processing and refinement of the $R_{1-x}K_xLSO$ samples.

x (K^+)	Space Group	Cell parameters (Å), Cell Volume (Å^3)	R_{wp}, R_p, R_B, χ^2
0	$P6_3/mmc$	a = 5.81351 (8), c = 13.9448 (2), V = 408.151 (13)	6.09, 4.75, 2.04, 1.34
0.1	$P6_3/mmc$	a = 5.79870 (4), c = 13.92373 (13), V = 405.459 (6)	6.29, 4.89, 1.57, 1.75
0.2	$P6_3/mmc$	a = 5.78072 (7), c = 13.9046 (2), V = 402.396 (12)	7.82, 5.78, 1.64, 2.13
0.3	$P6_3/mmc$	a = 5.76999 (2), c = 13.90978 (7), V = 401.053 (3)	5.25, 4.04, 3.62, 1.09
0.4	$P6_3/mmc$	a = 5.75900 (2), c = 13.91350 (7), V = 399.633 (3)	5.38, 4.10, 1.00, 1.11
0.5	$P6_3/mmc$	a = 5.74994 (3), c = 13.91202 (10), V = 398.334 (5)	6.02, 4.74, 1.02, 1.51
1	$P6_3/mmc$	a = 5.72585 (7), c = 13.9246 (2), V = 395.361 (12)	6.53, 5.12, 1.13, 1.71

Table S2. Fractional atomic coordinates and isotropic displacement parameters (Å^2) of $R_{1-x}K_xLSO$.

Atom	*x*	*y*	*z*	B_{iso}	*Occ.*
			x = 0		
Rb1	1/3	2/3	0.10189 (14)	1.17 (7)	1
Rb2	0	0	1/4	1.46 (8)	1
Lu	0	0	0	0.50 (5)	1
Si	2/3	1/3	0.1322 (4)	0.52 (12)	1
O1	0.3546 (10)	0.1773 (5)	0.0958 (5)	0.71 (14)	1
O2	2/3	1/3	1/4	0.8 (4)	1
			x = 0.1		
Rb1	1/3	2/3	0.10163 (12)	0.47 (6)	0.855 (7)
K1	1/3	2/3	0.10163 (12)	0.47 (6)	0.145 (7)
Rb2	0	0	1/4	0.27 (8)	0.680 (7)
K2	0	0	1/4	0.27 (8)	0.320 (7)
Lu	0	0	0	0.20 (5)	1
Si	2/3	1/3	0.1288 (4)	0.50 (10)	1
O1	0.3621 (8)	0.1810 (4)	0.0951 (4)	0.50 (11)	1
O2	2/3	1/3	1/4	0.5 (3)	1
			x = 0.2		
Rb1	1/3	2/3	0.10230 (18)	0.53 (9)	0.725 (10)
K1	1/3	2/3	0.10230 (18)	0.53 (9)	0.275 (10)
Rb2	0	0	1/4	0.74 (12)	0.546 (9)
K2	0	0	1/4	0.74 (12)	0.454 (9)
Lu	0	0	0	0.20 (7)	1
Si	2/3	1/3	0.1298 (5)	0.53 (15)	1
O1	0.3627 (10)	0.1814 (5)	0.0932 (5)	0.77 (15)	1
O2	2/3	1/3	1/4	0.6 (4)	1
			x = 0.3		
Rb1	1/3	2/3	0.10070 (9)	1.11 (4)	0.551 (4)
K1	1/3	2/3	0.10070 (9)	1.11 (4)	0.449 (4)
Rb2	0	0	1/4	0.99 (5)	0.350 (4)
K2	0	0	1/4	0.99 (5)	0.650 (4)
Lu	0	0	0	0.41 (3)	1
Si	2/3	1/3	0.1314 (2)	0.50 (6)	1
O1	0.3592 (5)	0.1796 (2)	0.0952 (2)	0.61 (7)	1
O2	2/3	1/3	1/4	0.74 (16)	1
			x = 0.4		
Rb1	1/3	2/3	0.09952 (10)	1.13 (5)	0.426 (4)
K1	1/3	2/3	0.09952 (10)	1.13 (5)	0.574 (4)
Rb2	0	0	1/4	0.88 (6)	0.248 (4)
K2	0	0	1/4	0.88 (6)	0.752 (4)
Lu	0	0	0	0.28 (3)	1
Si	2/3	1/3	0.1316 (2)	0.50 (6)	1

O1	0.3593 (5)	0.1796 (2)	0.0951 (2)	0.66 (7)	1
O2	2/3	1/3	1/4	1.06 (17)	1
			x = 0.5		
Rb1	1/3	2/3	0.09862 (11)	0.84 (5)	0.297 (5)
K1	1/3	2/3	0.09862 (11)	0.84 (5)	0.703 (5)
Rb2	0	0	1/4	0.89 (6)	0.170 (5)
K2	0	0	1/4	0.89 (6)	0.830 (5)
Lu	0	0	0	0.20 (3)	1
Si	2/3	1/3	0.1326 (2)	0.50 (7)	1
O1	0.3579 (6)	0.1789 (3)	0.0953 (2)	0.78 (8)	1
O2	2/3	1/3	1/4	1.24 (19)	1
			x = 1		
K1	1/3	2/3	0.0931 (3)	0.63 (8)	1
K2	0	0	1/4	1.42 (8)	1
Lu	0	0	0	0.24 (4)	1
Si	2/3	1/3	0.1324 (4)	0.50 (10)	1
O1	0.3594 (9)	0.1797 (4)	0.0956 (4)	0.70 (12)	1
O2	2/3	1/3	1/4	0.9 (3)	1

Table S3. Main bond lengths (Å) of $R_{1-x}K_xLSO$.

		x = 0	
Rb1—O1	2.910 (4)	Lu—O1	2.230 (5)
Rb1—O1^{i}	3.173 (7)	Si—O1	1.651 (5)
Rb2—O1	2.795 (6)	Si—O2	1.643 (6)
		x = 0.1	
Rb1—O1	2.905 (4)	Rb2—O1	2.821 (5)
Rb1—O1^{i}	2.904 (4)	K2—O1	2.821 (5)
Rb1—O1^{ii}	3.137 (5)	Lu—O1	2.249 (4)
K1—O1	2.905 (4)	Si—O1	1.600 (4)
K1—O1^{i}	2.904 (4)	Si—O2	1.688 (6)
K1—O1^{ii}	3.137 (5)		
		x = 0.2	
Rb1—O1	2.897 (4)	Rb2—O1	2.837 (6)
Rb1—O1^{i}	2.897 (4)	K2—O1	2.837 (6)
Rb1—O1^{ii}	3.115 (7)	Lu—O1	2.231 (5)
K1—O1	2.897 (4)	Si—O1	1.605 (5)
K1—O1^{i}	2.897 (4)	Si—O2	1.671 (7)
K1—O1^{ii}	3.115 (7)		
		x = 0.3	
Rb1—O1	2.889 (2)	K2—O1	2.803 (3)
Rb1—O1^{i}	3.128 (3)	Lu—O1	2.231 (2)
K1—O1	2.889 (2)	Si—O1	1.617 (2)
K1—O1^{i}	3.128 (3)	Si—O2	1.650 (3)

Rb2—O1	2.803 (3)		
		x = 0.4	
Rb1—O1	2.883 (2)	Rb2—O1	2.803 (3)
Rb1—O1^{i}	2.883 (2)	K2—O1	2.803 (3)
Rb1—O1^{ii}	3.112 (3)	Lu—O1	2.228 (2)
K1—O1	2.883 (2)	Si—O1	1.615 (2)
K1—O1^{i}	2.883 (2)	Si—O2	1.647 (3)
K1—O1^{ii}	3.112 (3)		
		x = 0.5	
Rb1—O1	2.878 (3)	Rb2—O1	2.794 (3)
Rb1—O1^{i}	2.878 (3)	K2—O1	2.794 (3)
Rb1—O1^{ii}	3.105 (3)	Lu—O1	2.221 (3)
K1—O1	2.878 (3)	Si—O1	1.623 (3)
K1—O1^{i}	2.878 (3)	Si—O2	1.633 (3)
K1—O1^{ii}	3.105 (3)		
		x = 1	
K1—O1	2.866 (4)	Lu—O1	2.225 (4)
K1—O1^{i}	3.037 (6)	Si—O1	1.608 (4)
K2—O1	2.793 (5)	Si—O2	1.638 (6)

Symmetry codes: (i) x, y+1, z; (ii) -x+1, -y+1, -z

Table S4. Emission wavelengths of $R_{1-x}K_xLSO:Eu^{2+}$ phosphors.

x	Emission wavelength (nm)
0	619
0.1	643
0.2	662
0.3	691
0.4	703
0.5	717
1	737

References

(1) Wei, Y.; Gao, Z.; Liu, S.; Chen, S.; Xing, G.; Wang, W.;Dang, P.; Li, G., Highly Efficient Green-to-Yellowish-Orange Emitting Eu^{2+}-Doped Pyrophosphate Phosphors with Superior Thermal Quenching Resistance for w-LEDs. *Adv. Opt. Mater.* **2020,** *8*, 1901859-1901872.
(2) Aggarwal, R.; Ranganathan, P., Common Pitfalls in Statistical Analysis: Linear Regression Analysis. *Perspect Clin. Res.* **2017,** *8*, 100-102.