Supplementary Information

$K_5Yb_{1-x}Eu_x(MoO_4)_4$ phosphors: aperiodic structures and luminescence properties.

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Table S1. Element analysis results of α-$K_5Yb_{1-x}Eu_x(MoO_4)_4$ ($0 \leq x \leq 1$).

x	K, at.%	Mo, at.%	Yb, at.%	Eu, at.%	Yb:Eu ratio	Method
0.1	47.80±0.18	41.50±0.40	9.71±0.53	0.99±0.07	0.91:0.09	SEM-EDX
0.3					0.73:0.27	ICP-MS
0.4	48.60±0.37	41.55±0.26	6.12±0.40	3.73±0.10	0.62:0.38	SEM-EDX
			60.05±1.27	39.95±1.27	0.60:0.40	TEM-EDX
0.5	48.33±0.18	41.76±0.16	5.39±0.20	4.53±0.14	0.54:0.46	SEM-EDX
0.7					0.33:0.67	ICP-MS
0.9	48.97±0.18	41.53±0.10	1.36±0.05	8.14±0.07	0.14:0.86	SEM-EDX

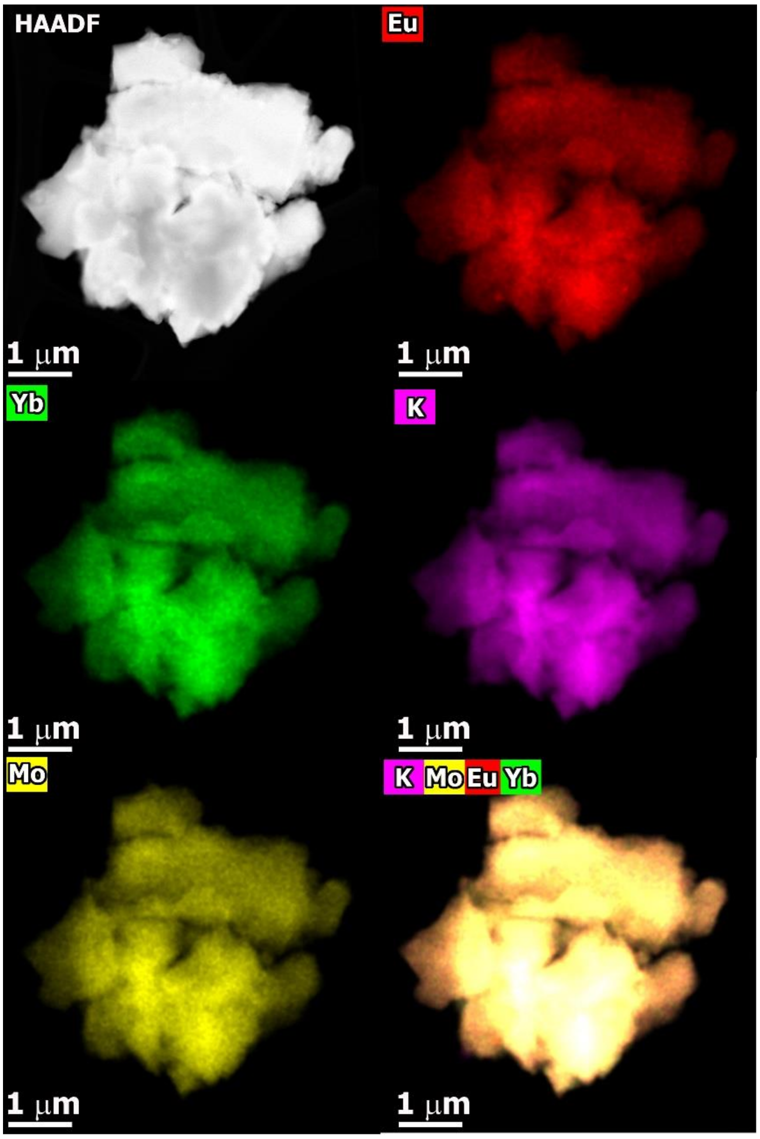

Figure S1. HAADF-STEM images of the particles in the $K_5Yb_{0.6}Eu_{0.4}(MoO_4)_4$ sample along with the corresponding color-coded elemental maps and the mixed EDX map for K, Mo, Yb, Eu.

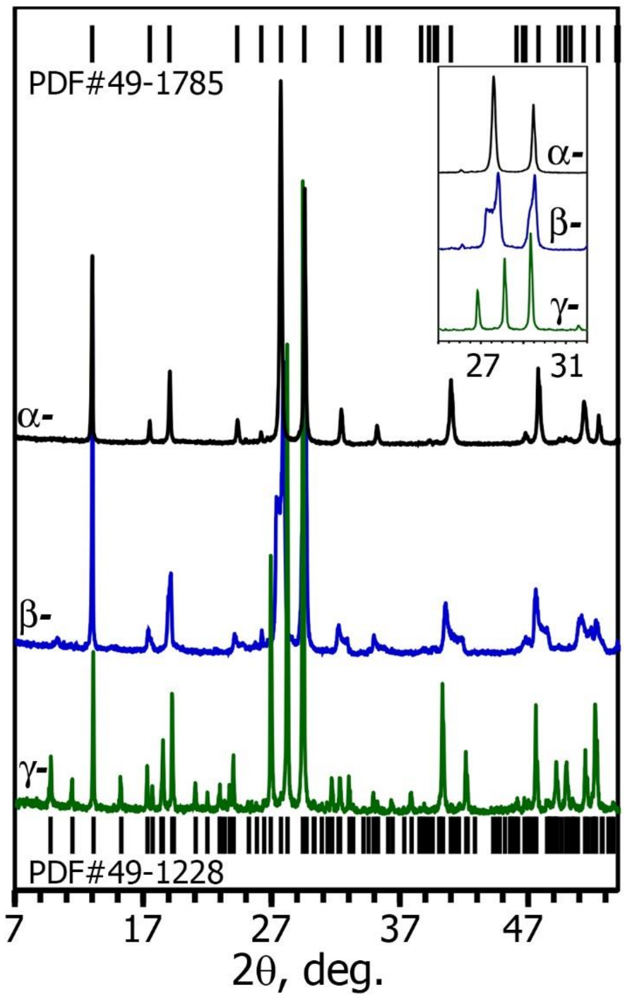

Figure S2. Parts of PXRD patterns for γ-, β- and α-modification of $K_5Yb(MoO_4)_4$. The inset shows fragments in 2θ ranges of 25-32°. Tick marks denote the peak positions of possible Bragg reflections for γ- (PDF#49-1228) and α-$K_5Yb(MoO_4)_4$ (PDF#49-1785).

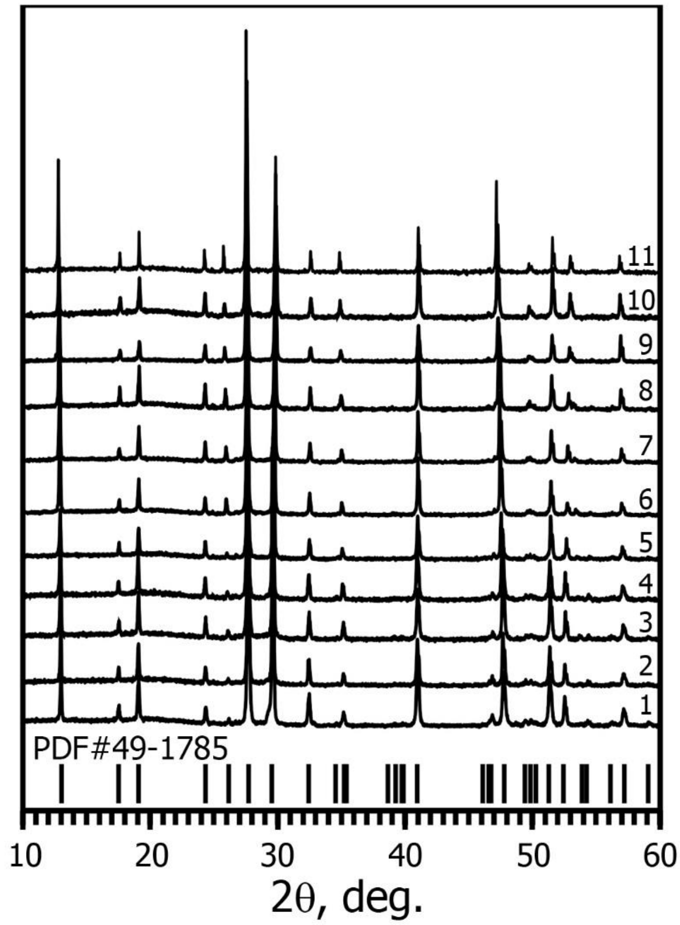

Figure S3. Parts of PXRD patterns for α-$K_5Yb_{1-x}Eu_x(MoO_4)_4$ (*x*: 0(1)-1(11), step 0.1) annealing at 1123±10 K followed by quenching to the liquid nitrogen temperature. Tick marks denote the peak positions of possible Bragg reflections for α-$K_5Yb(MoO_4)_4$ (PDF#49-1785).

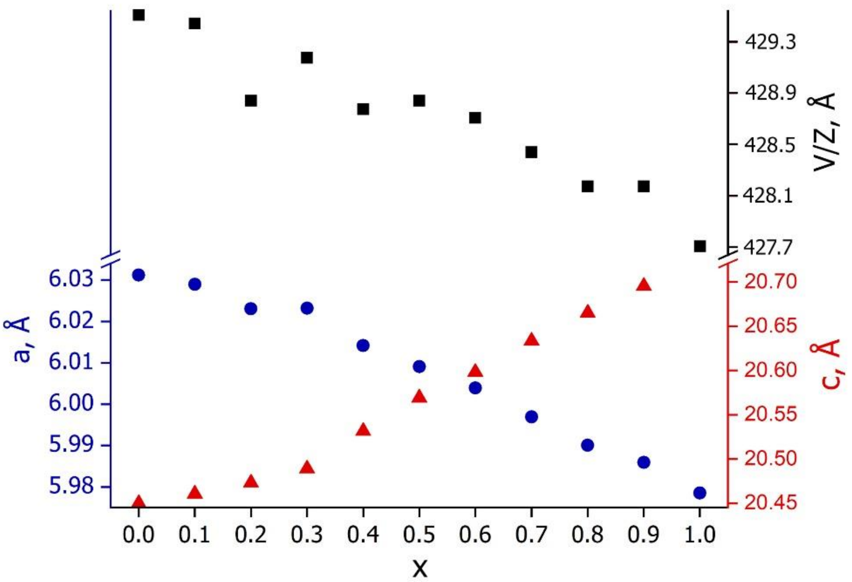

Figure S4. The lattice cell parameters of α-$K_5Yb_{1-x}Eu_x(MoO_4)_4$ (*x*=0.1–1).

Table S2. Unit cell parameters for α-$K_5Yb_{1-x}Eu_x(MoO_4)_4$ (0≤x≤1, SG $R\bar{3}m$, Z=1.5 (α-phase)) annealed at 1123±10 K samples followed by quenching from the high to the liquid nitrogen temperature.

x	*a*, Å	*c*, Å	*V*, Å^3	*V/Z*, Å^3	*Ref.*
0	6.0372(1)	20.4045(2)	644.06(1)	429.37	[14]
0	6.0312(2)	20.4499(8)	644.21(4)	429.47	
0.1	6.0290(2)	20.4606(7)	644.07(4)	429.38	
0.2	6.0231(1)	20.4731(4)	643.21(1)	428.81	
0.3	6.0232(1)	20.4890(5)	643.74(1)	429.16	
0.4	6.0142(2)	20.5316(7)	643.14(4)	428.76	
0.5	6.0090(1)	20.5692(6)	643.22(3)	428.81	
0.6	6.0039(1)	20.5983(5)	643.03(2)	428.69	
0.7	5.9969(1)	20.6335(5)	642.63(3)	428.42	
0.8	5.9901(2)	20.6654(7)	642.15(4)	428.10	
0.9	5.9859(2)	20.6954(8)	642.20(4)	428.13	
1	5.9785(1)	20.7253(5)	641.53(3)	427.69	
1	5.9785(1)	20.7254(5)	641.54(3)	427.69	[12]
1	5.980	20.74	642.53	428.35	PDF-2, №45–0340

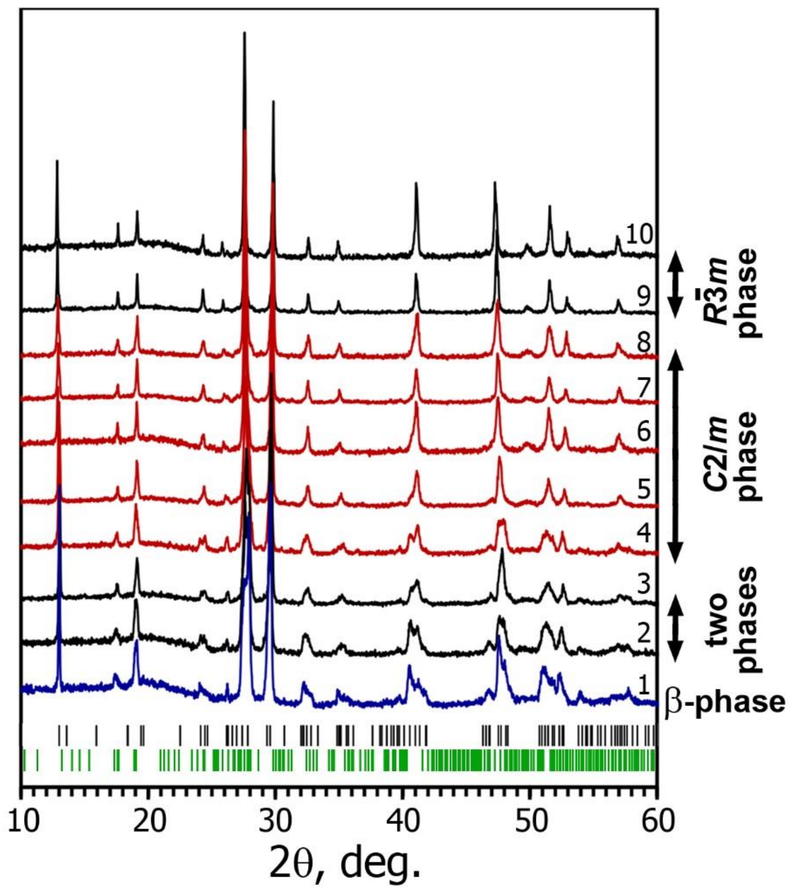

Figure S5. Parts of PXRD patterns for IM-$K_5Yb_{1-x}Eu_x(MoO_4)_4$ (*x*: 0(1)-0.9(10), step 0.1) annealing at 1123±10 K then slow cooling. Positions of main (black) and satellite (green) reflections for incommensurately modulated β-$K_5Yb(MoO_4)_4$ also are presented by bars.

Table S3. Unit cell parameters for LT-$K_5Yb_{1-x}Eu_x(MoO_4)_4$ ($0 \le x \le 1$) samples synthesized by a solid state reaction at 893±10 K followed by slow cooling to T_R.

x	*SG*	*a*, Å	*b*, Å	*c*, Å	*β*	*V*, Å^3	*Ref.*
0	*C*2/*c*	14.8236(1)	12.1293(1)	10.5151(1)	114.559(1)	1719.58(3)	[14]
0	*C*2/*c*	14.8550(1)	12.1449(4)	10.5270(3)	114.576(1)	1727.17(4)	
0.1	*C*2/*c*	14.8777(9)	12.1309(3)	10.5141(2)	114.645(1)	1724.73(5)	
	*C*2/*m*	10.4950(2)	6.0504(1)	7.7753(6)	118.309(7)	434.67(2)	
0.2	*C*2/*c*	14.9154(5)	12.0864(5)	10.5586(1)	115.021(4)	1724.8(1)	
	*C*2/*m*	10.6573(3)	6.0394(3)	7.8377(5)	119.455(9)	439.30(6)	
0.3	*C*2/*m*	10.4554(8)	6.0303(4)	7.7138(4)	117.906(2)	430.06(1)	
0.4	*C*2/*m*	10.4400(2)	6.0223(8)	7.7180(9)	117.610(1)	430.00(1)	
0.5	*C*2/*m*	10.4312(6)	6.0154(1)	7.7257(3)	117.547(1)	429.81(1)	
0.6	*C*2/*m*	10.4217(1)	6.0074(7)	7.7238(7)	117.335(1)	429.50(9)	
0.7	*C*2/*m*	10.3985(4)	6.0014(4)	7.7173(2)	117.042(1)	429.00(2)	
0.8	$R\bar{3}m$	5.9906(1)		20.6622(5)		642.15(1)	
0.9	$R\bar{3}m$	5.9918(1)		20.7176(8)		643.96(1)	
1	$R\bar{3}m$	5.9795(3)		20.7348(5)		642.00(1)	
1	$R\bar{3}m$	5.9780(2)		20.7257(7)		641.44(3)	[12]

Table S4. Unit cell parameters for IM-$K_5Yb_{1-x}Eu_x(MoO_4)_4$ ($0 \le x \le 1$) samples synthesized by annealing at 1123±10 K for 3 h in air followed by slow cooling to T_R.

x	*SG*	*a*, Å	*b*, Å	*c*, Å	*β*	*V*, Å^3	*Ref.*
0	*X*2/*m*(0β0)00	10.405(2)	6.118(1)	19.775(2)	136.63(1)	864.2(3)	[15]
0	*X*2/*m*(0β0)00	10.428(2)	6.0819(4)	19.693(3)	136.292(5)	863.0(2)	
0.1	*X*2/*m*(0β0)00	10.266(3)	6.0680(7)	19.558(3)	135.772(7)	849.8(3)	
	*C*2/*m*	10.456(1)	6.0246(7)	7.6820(7)	117.799(8)	428.1(2)	
0.2	*X*2/*m*(0β0)00	10.106(1)	6.0554(4)	19.422(3)	135.339(5)	835.4(2)	
	*C*2/*m*	10.4191(9)	6.0202(6)	7.6453(5)	117.418(6)	425.7(2)	
0.3	*C*2/*m*	10.444(1)	6.0252(7)	7.6866(8)	117.799(7)	427.86(9)	
0.4	*C*2/*m*	10.419(1)	6.0185(8)	7.6794(6)	117.35(1)	427.72(8)	
0.5	*C*2/*m*	10.419(2)	6.0040(9)	7.7070(7)	117.097(9)	429.18(4)	
0.6	*C*2/*m*	10.409(1)	6.0066(6)	7.6922(3)	117.077(8)	428.21(3)	
0.7	*C*2/*m*	10.399(1)	5.9976(5)	7.7107(6)	117.101(5)	428.11(7)	
0.8	$R\bar{3}m$	5.9929(2)		20.6360(8)		641.84(4)	
0.9	$R\bar{3}m$	5.9877(1)		20.6825(6)		642.17(2)	

Table S5. Final coordinates, amplitudes of Fourier components for the occupational[1] and displacive[2] modulation functions and isotropic atomic displacement parameters for β-$K_5Yb_{0.3}Eu_{0.7}(MoO_4)_4$. Only waves with non-zero amplitudes are shown. The *s* and *c* symbols stand for the sine and cosine components, respectively.

Atom	Occupation; parameters of atomic domains	Wave	*x/a*	*y/b*	*z/c*	U_{iso}, Å^2
*M*1	0.5K+0.5(Eu1+Yb1= $\delta_{K1}+\delta_{(Yb1+Eu1)}$; δ_{K1} = 0.5, x_4^0(K1) = 0, $\delta_{(Yb1+Eu1)}$=0.5, x_4^0(Yb1+Eu1)= 0.5		0	0	0	0.0304(9)
		s	0	0.030(2)	0	
K2	1 K		0.8038(4)	0	0.4159(5)	0.0343(9)
Mo	1 Mo		0.3996(2)	0	0.1972(2)	0.0186(8)
		s	0	0.0070(15)	0	
		c	-0.0040(7)	0	0	
O1	1 O		0.3106(12)	0	0.9556(11)	0.023(5)
		s	0	-0.075(3)	0	
		c	-0.054(2)	0	0	
O2	1 O		0.0056(5)	0.2606(7)	0.2673(6)	0.015(3)
		s	-0.0103(17)	-0.027(2)	-0.0646(17)	
		c	0.010(2)	0.032(5)	0	
O3	1 O		0.2671(10)	0	0.2732(11)	0.031(6)
		s	0	-0.035(6)	0	
		c	-0.048(2)	0	-0.074(3)	

[1] δ_{M1} and x_4^0(*M*1) parameters define the length and the center coordinate x_4, respectively, for the atomic domain of *M*1-cation.

[2] The displacive modulation function for the atom λ is defined as
$U^{\lambda}(x_4) = A_s^{\lambda}\sin(2\pi x_4) + B_c^{\lambda}\cos(2\pi x_4)$, $x_4 = \mathbf{qr}^{\lambda} + t$.

Table S6. Anisotropic atomic displacement parameters in β-$K_5Yb_{0.3}Eu_{0.7}(MoO_4)_4$.

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
*M*1	0.043(2)	0.025(3)	0.020(2)	0	0.0116(17)	0
Mo	0.0168(11)	0.0202(12)	0.0190(11)	0	0.0084(9)	0

Table S7. Main interatomic distances for β-$K_5Yb_{0.3}Eu_{0.7}(MoO_4)_4$ (Å).

distances	average	min.	max.
K1-O1×2	3.08(2)	2.85(2)	3.46(2)
K1-O1×4	3.232(14)	3.02(2)	3.71(2)
K1-O2×4	2.34(2)	2.04(2)	2.96(2)
K1-O3×2	2.297(14)	2.09(2)	2.64(2)
Eu1/Yb1-O1×2	3.775(17)	3.46(2)	3.96(2)
Eu1/Yb1- O1×4	3.830(15)	3.38(2)	4.03(2)
Eu1/Yb1-O2×4	2.820(17)	2.21(2)	3.11(2)
Eu1/Yb1-O3×2	2.957(13)	2.64(2)	3.15(2)
K2-O1	2.607(9)	2.588(11)	2.643(11)
K2-O2×2	2.840(15)	2.62(2)	3.07(2)
K2-O2×2	3.14(2)	2.92(2)	3.35(2)
K2-O2×2	3.24(2)	2.99(3)	3.47(3)
K2-O3×2	3.18(2)	2.92(4)	3.42(4)
K2-O3	2.846(19)	2.63(3)	3.10(3)
Mo-O1	1.737(9)	1.719(10)	1.762(10)
Mo-O2×2	1.78(2)	1.67(3)	1.89(3)
Mo-O3	1.78(2)	1.74(3)	1.84(3)

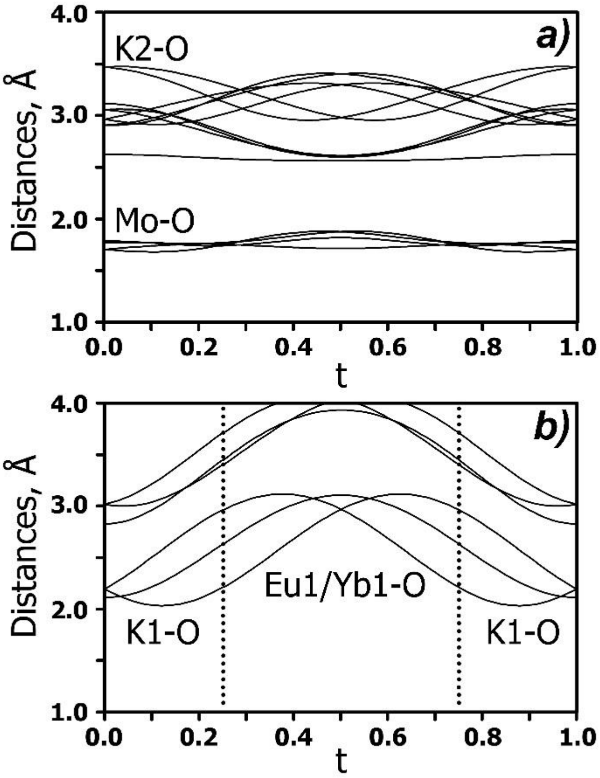

Figure S6. *t*-plots of Mo−O and K2−O (*a*), K1−O (*a*) and Eu1/Yb1−O (*b*) bond lengths for the β-$K_5Yb_{0.3}Eu_{0.7}(MoO_4)_4$ structure.

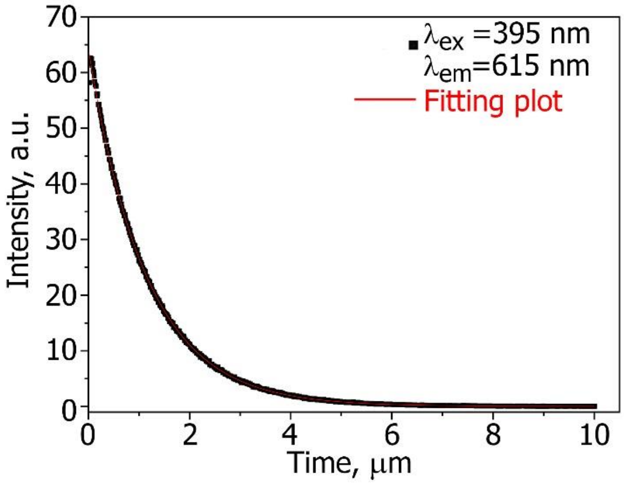

Figure S7. Decay curve of the Eu^{3+} emission of α-$K_5Yb_{0.3}Eu_{0.7}(MoO_4)_4$ at T_R.