**Achievement of high efficiency and thermally stable near-infrared phosphors by designing chromium crystallographic environment for nondestructive testing and night vision**

Guohui Wei, Panlai Li*, Rui Li, Ye Wang, Hao Suo, Yuanbo Yang, Shaoxuan He, Jiehong Li, Yawei Shi, Zhijun Wang*

Hebei Key Laboratory of Optic-electronic Information and Materials, College of Physics Science & Technology, Hebei University, Baoding 071002, China

**Table S1** Refined crystallographic data of $InTaO_4$:$Cr^{3+}$ (x=0- 0.011).

*li_panlai@126.com

*wangzj1998@126.com

| x | 0 | 0.001 | 0.003 | 0.005 | 0.007 | 0.009 | 0.011 |
|---|---|---|---|---|---|---|---|
| Crystal System | monoclinic-type | | | | | | |
| Space group | P2/a | | | | | | |
| a (Å) | 5.1588 | 5.1575 | 5.1578 | 5.1582 | 5.1571 | 5.1575 | 5.1566 |
| b (Å) | 5.7792 | 5.7777 | 5.7779 | 5.7781 | 5.7769 | 5.7772 | 5.7761 |
| C (Å) | 4.8287 | 4.8274 | 4.8274 | 4.8276 | 4.8263 | 4.8265 | 4.8256 |
| Z | 4 | | | | | | |
| Cell Volume (Å$^3$) | 143.920 | 143.823 | 143.803 | 143.744 | 143.727 | 143.671 | 143.651 |
| $R_{\omega p}$ (%) | 8.70 | 9.05 | 8.57 | 8.96 | 8.84 | 10.02 | 8.97 |
| $R_p$ (%) | 6.79 | 7.12 | 6.02 | 6.99 | 6.98 | 7.75 | 7.00 |
| $\chi^2$ | 1.202 | 1.292 | 1.16 | 1.223 | 1.228 | 1.554 | 1.221 |

**Table S2** Refined crystallographic data of $ScTaO_4$:$Cr^{3+}$ (x=0- 0.011).

| x | 0 | 0.001 | 0.003 | 0.005 | 0.007 | 0.009 | 0.011 |
|---|---|---|---|---|---|---|---|
| Crystal System | monoclinic-type | | | | | | |
| Space group | P2/a | | | | | | |
| a (Å) | 5.1876 | 5.1575 | 5.1237 | 5.1145 | 5.1103 | 5.1073 | 5.1052 |
| b (Å) | 5.7781 | 5.7772 | 5.6802 | 5.6674 | 5.6635 | 5.6593 | 5.6572 |
| c (Å) | 4.8277 | 4.8265 | 4.8263 | 4.8072 | 4.8033 | 4.7995 | 4.7971 |
| Z | 4 | | | | | | |
| Cell Volume (Å$^3$) | 143.771 | 143.691 | 143.665 | 143.286 | 142.747 | 142.571 | 142.441 |
| $R_{\omega p}$ (%) | 8.42 | 9.05 | 8.57 | 8.97 | 8.98 | 9.05 | 8.97 |
| $R_p$ (%) | 7.92 | 7.85 | 7.86 | 7.99 | 7.68 | 7.75 | 7.78 |
| $\chi^2$ | 2.245 | 1.992 | 2.216 | 2.41 | 1.998 | 2.134 | 2.129 |

**Table S3** Refined crystallographic data of $GaTaO_4$:$Cr^{3+}$ (x=0- 0.011).

| x | 0 | 0.001 | 0.003 | 0.005 | 0.007 | 0.009 | 0.011 |
|---|---|---|---|---|---|---|---|
| Crystal System | monoclinic-type | | | | | | |
| Space group | P2/a | | | | | | |
| a (Å) | 4.5936 | 4.5934 | 4.5930 | 4.5928 | 4.5863 | 4.5828 | 4.5755 |
| b (Å) | 5.5717 | 5.5711 | 5.5710 | 5.5684 | 5.5664 | 5.5620 | 5.5598 |
| c(Å) | 4.9680 | 4.9666 | 4.9660 | 4.9631 | 4.9615 | 4.9588 | 4.9561 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| Z | | | | 4 | | | |
| Cell Volume (Å$^3$) | 127.150 | 127.099 | 127.067 | 127.007 | 126.990 | 126.702 | 126.607 |
| $R_{op}$ (%) | 9.95 | 9.65 | 8.69 | 8.43 | 8.54 | 8.02 | 8.97 |
| $R_p$ (%) | 7.56 | 7.22 | 6.95 | 6.86 | 6.92 | 6.75 | 7.38 |
| $\chi^2$ | 2.505 | 2.794 | 2.265 | 2.443 | 2.678 | 2.362 | 2 .267 |

**Table S4** Content ratio of each element of $GaTaO_4$:$Cr^{3+}$

| Element | Atomic (%) |
|---|---|
| O | 84.8 |
| Ga | 18.8 |
| Ta | 37.2 |
| Cr | 0.7 |

**Table S5** Content ratio of each element of $ScTaO_4$:$Cr^{3+}$

| Element | Atomic (%) |
|---|---|
| O | 64.4 |
| Sc | 23.2 |
| Ta | 51.9 |
| Cr | 1.1 |

**Table S6** Content ratio of each element of $InTaO_4$:$Cr^{3+}$

| Element | Atomic (%) |
|---|---|
| O | 54.1 |

| | |
|---|---|
| **In** | **21.9** |
| **Ta** | **22.3** |
| **Cr** | **1.7** |

**Table S7.** Calculation of the relative formation energy of $GaTaO_4$ and $Ga[Ta_{Cr}]O_4$.

| **Substituting model** | **$GaTaO_4$** | **$GaTa_{Cr}O_4$** |
|---|---|---|
| **Formation energy (eV)** | **-14.3671 eV** | **-8.6370 eV** |

**Measurements of quantum efficiency**

The quantum efficiency (QE) measurement was performed at room temperature by using a HORIBA FLuorolog3 fluorescence spectrometer with a 450 W Xe lamp. The IQE is defined as the percentage of the number of emitted photons to that of absorbed photons, which can be calculated using the following equation:

$$IQE = \frac{\int L}{\int E_R - \int E_S} \quad (1)$$

$L$ is the luminescence spectrum of the sample, $E_S$ is the excitation spectrum of the sample, and $E_R$ is the excitation spectrum referenced by $BaSO_4$. In addition, the EQE is defined as the percentage of the number of emitted photons to the number of exciting photons:

$$EQE = \frac{\int L}{\int E_R} \times 100\% \qquad (2)$$

The absorption efficiency (AE) is defined as the percentage of the number of absorbed photons (by the sample) to that of excitation photons:

$$AE = \frac{\int E_R - \int E_S}{\int E_R} \times 100\% \qquad (3)$$

**Measurements of Huang–Rhys factor (S)**

$$FWHM = 2.36\sqrt{S}\hbar\omega\sqrt{coth}(\frac{\hbar\omega}{2kT}) \qquad (4)$$

where the *FWHM* (eV) is the full width at half maximum of emission spectrum at temperature T(K), and the hω and k are the phonon energy and Boltzmann's constant, respectively.

$$\coth(x) = \frac{e^x + e^{-x}}{e^x - e^{-x}} \qquad (5)$$

$$FWHM^2 = 5.57 \times S \times (\hbar\omega)^2(1 + \frac{1}{e^{\frac{\hbar\omega}{kT}} - 1}) \qquad (6)$$

$$\frac{\hbar\omega}{kT} \approx 10^{-3} \qquad (7)$$

$$FWHM^2 = 5.57 \times S \times (\hbar\omega)^2(1 + \frac{1}{\frac{\hbar\omega}{2kT}}) \qquad (8)$$

$$FWHM = a + b \times 2kT \qquad (9)$$

$$a = 5.57 \times S \times (\hbar\omega)^2 \qquad (10)$$

$$b = 5.57 \times S \times (\hbar\omega)^2 \qquad (11)$$

Straight-line fitting equation:

**$InTaO_4$:** $y = 0.36454x + 0.00454$

**$ScTaO_4$:** $y = 0.57179x + 0.01892$

**$GaTaO_4$:** $y = 0.39389x + 0.01987$

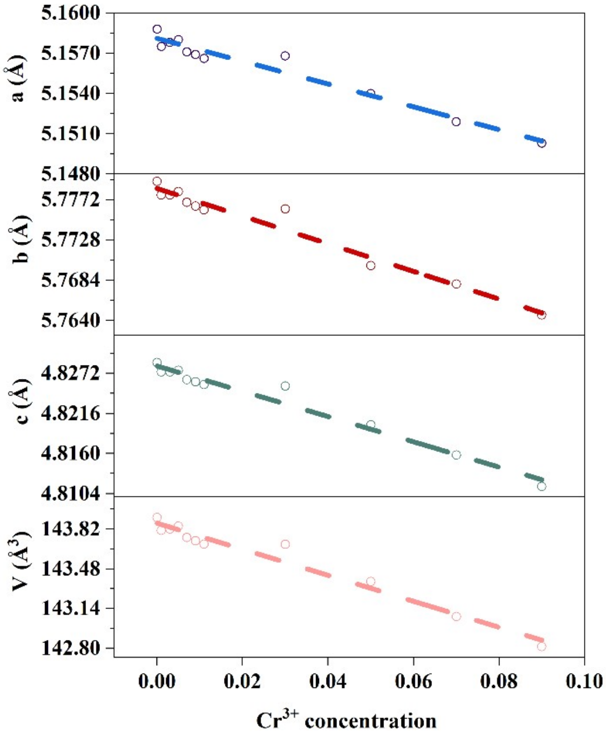


**Figure S1** The trend of volume change of a, b, c, V of $InTaO_4$: $xCr^{3+}$ (x=0-0.1).

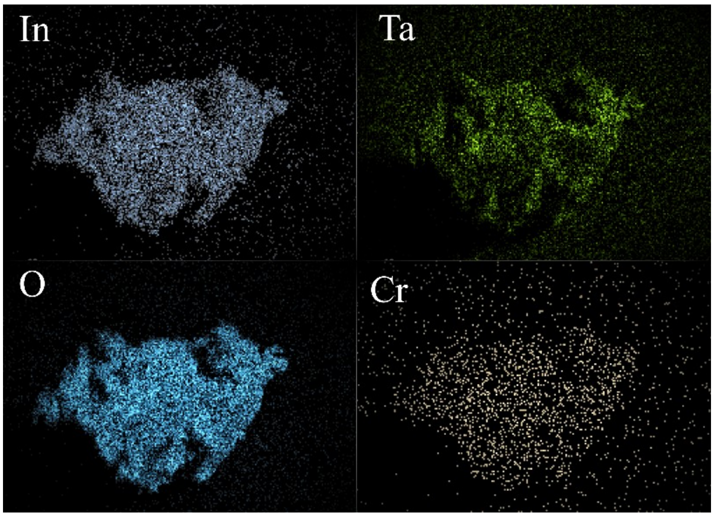


**Figure S2** Mapping of $InTaO_4:Cr^{3+}$.

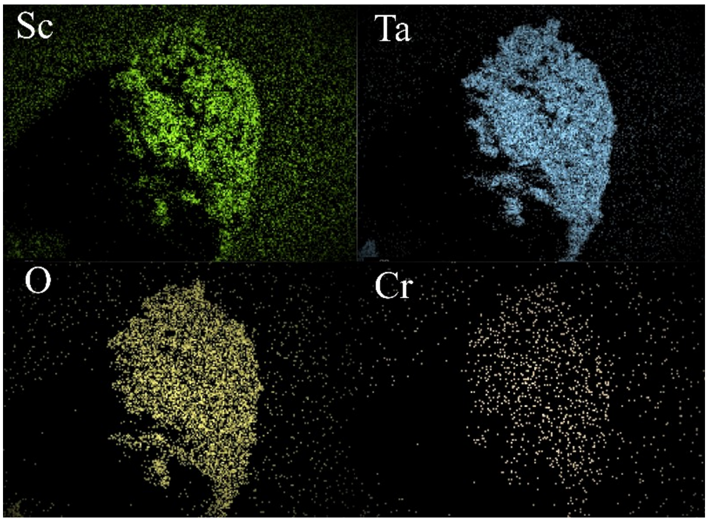


**Figure S3** Mapping of $ScTaO_4:Cr^{3+}$.

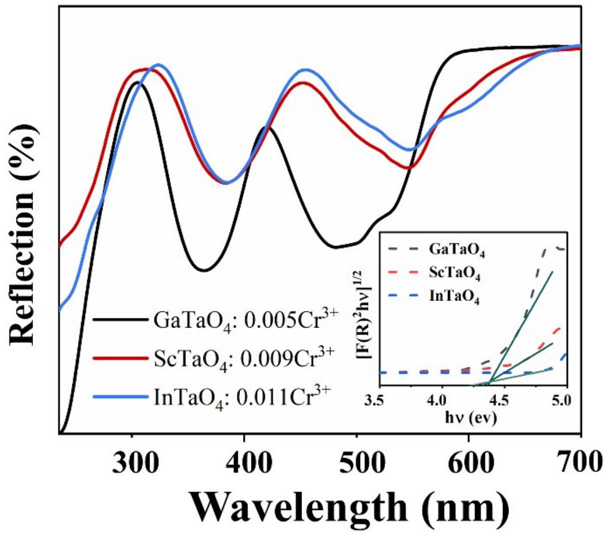


**Figure S4** The diffuse reflectance spectra (DRs) of $XTaO_4:Cr^{3+}$ (X=Ga, Sc, In). The inset shows the $XTaO_4$ (X=Ga, Sc, In) host of linear relationship with $[F(R)^2hv]^{1/2}$ and *hv*.

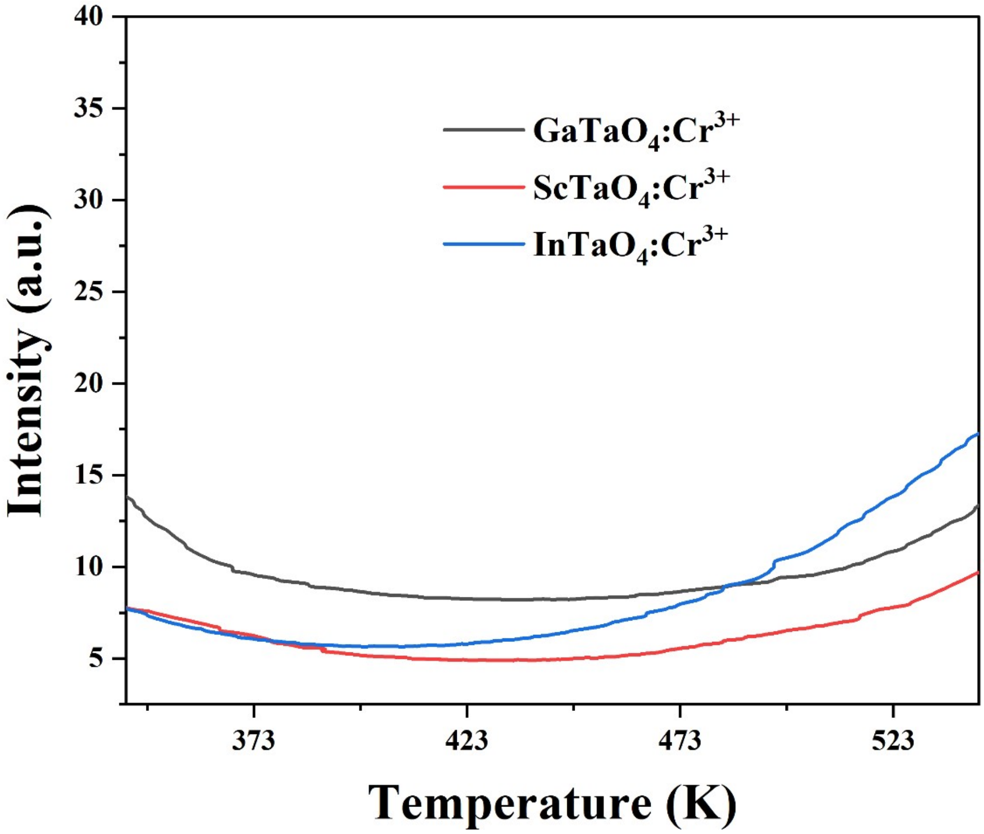


**Figure S5** The thermoluminescence spectra of $XTaO_4:Cr^{3+}$ (X=Ga, Sc, and In).

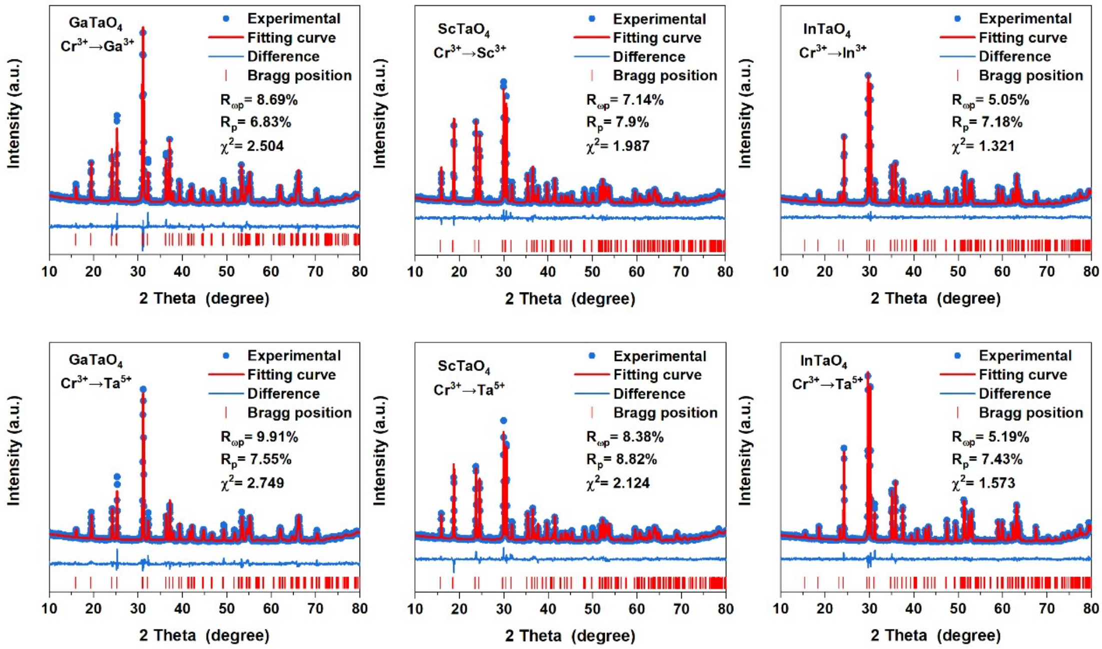


**Figure S6** Rietveld refinement of XRD results of $X^{3+}$ and $Ta^{5+}$ ions are substituted by $Cr^{3+}$ ions, respectively.

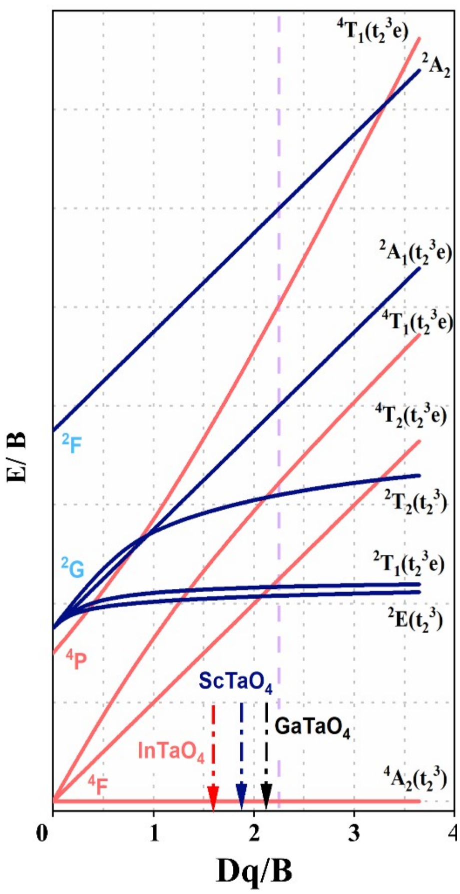


**Figure S7** T–S diagram of $Cr^{3+}$ at an octahedron crystal environment.

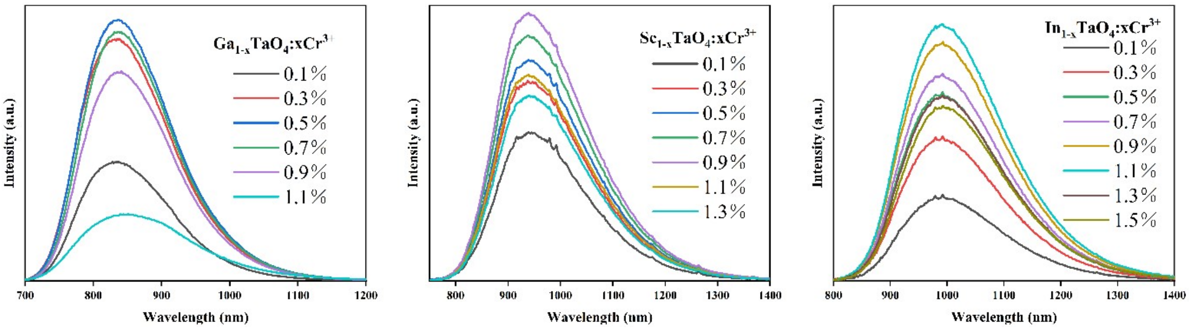


**Figure S8** The PL spectrum of $XTaO_4$:$Cr^{3+}$ (X=Ga, Sc, In).

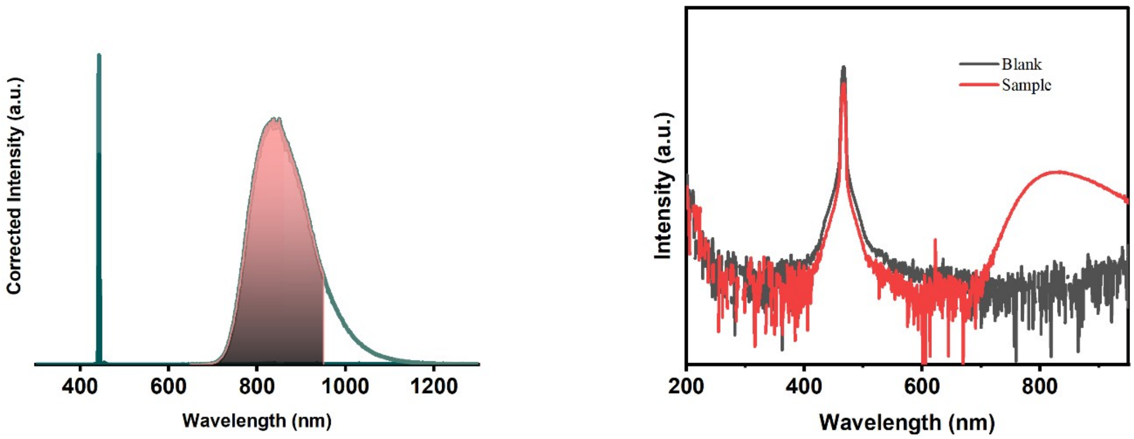


**Figure S9** The luminescent spectra for the IQE of $GaTaO_4$:0.005$Cr^{3+}$.

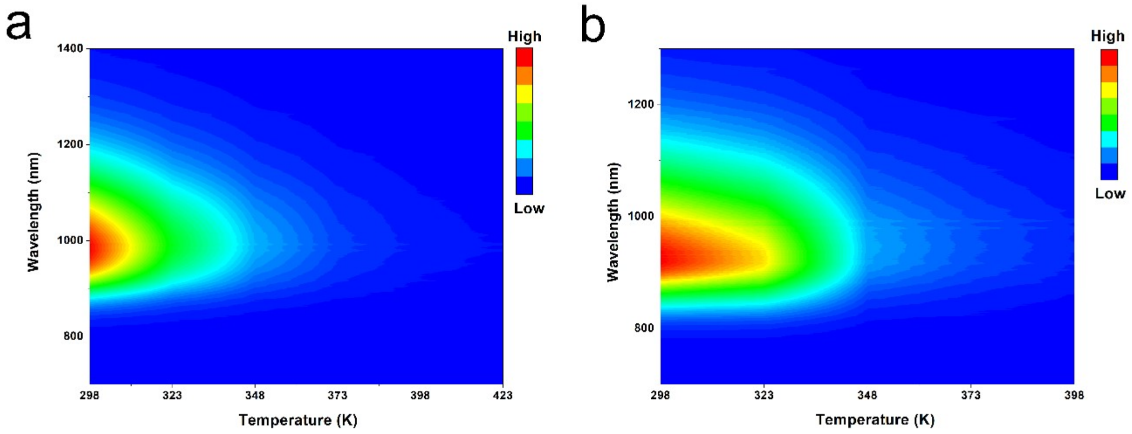


**Figure S10** Temperature-dependent PL spectra of samples $ScTaO_4$:0.009$Cr^{3+}$ and $InTaO_4$:0.011$Cr^{3+}$.

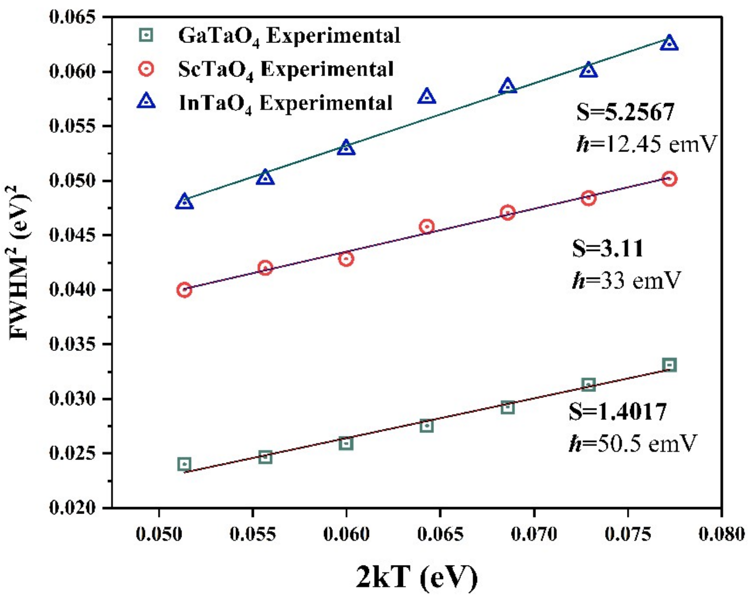


**Figure S11** Fitting results of $FWHM^2$ as a function 2 kT.

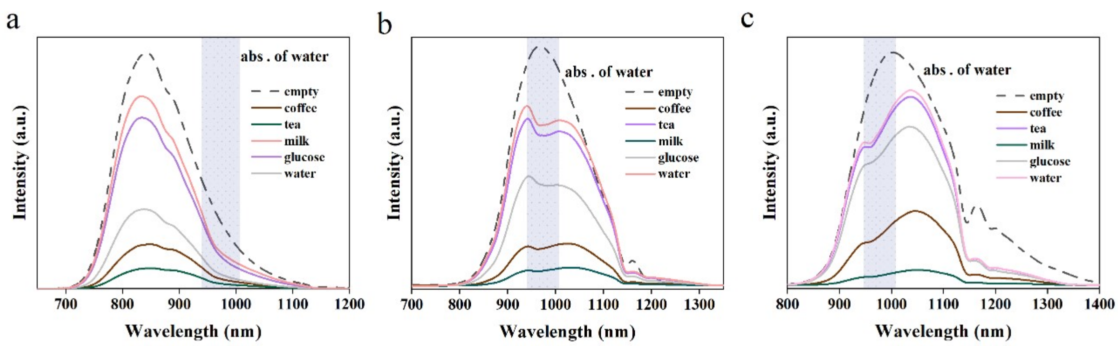

**Figure S12** Spectra of LEDs coupled with $XTaO_4$:(X= Ga, Sc, and In) penetrating five different solutions respectively