ELECTRONIC SUPPLEMENTARY INFORMATION

Tuning of bandgaps and emission properties of light-emitting diode materials through homogeneous alloying in molecular crystals

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S1. Experimental details

All the reagents are obtained from Sigma-Aldrich and were used as received without further purification. Gaq₃, Inq₃ and Crq₃ was synthesized by the reaction of M³⁺ salts with hydroxyquinoline which is further neutralized using potassium acetate. The precipitate obtained is vacuum dried.

\[ \text{Ga} \cdot 3 \text{eq. H}_2\text{O} + \text{q}_3 \rightarrow \text{Gaq}_3 (\text{yellow precipitate}) \]

\[ \text{Cr} \cdot (\text{NO}_3)_3 + \text{q}_3 \rightarrow \text{Crq}_3 (\text{brown precipitate}) \]

\[ \text{In} \cdot 3 \text{eq. H}_2\text{O} \rightarrow \text{Inq}_3 (\text{yellow precipitate}) \]

Crystals of pure Mq₃ and their alloys were obtained by two zone sublimation deposition method. In order to obtain single crystals of alloys, the mixtures of Mq₃ compounds of various composition ratios were taken in a glass boat. This mixture was placed in the hot end of a two-zone sublimation condensation apparatus with an Argon gas flow. The zones of the setup were kept at 390 °C and 100 °C and the samples were kept in the setup for ~5 hours before the temperature was lowered to room temperature. Needle like crystals were harvested from the walls of the glass tubes.

![Alq₃, Gaq₃, Crq₃](image)

**Figure S1**: Optical microscopic images of the single crystals of grown by two zone vaporization condensation method in the study.

S2. Crystallographic data collection and structure refinement

The crystals were cooled to 100 K with a liquid nitrogen stream using an Oxford Instruments Cryojet-XL nitrogen gas-stream cooling device. X-ray diffraction data were collected on Agilent Supernova diffractometer using MoKα radiation (wavelength 0.71073 Å, Table S1-S6). The scan width was chosen to be 1° per frame and the crystal to detector distance was fixed at 53 mm (for MoKα data) during the data collection. Cell refinement, crystal face indexing, data integration and reduction were carried out using CrysAlisPro.1 Crystal structures were solved by direct methods and refined using SHELXS v.2013.12 accessed by OLEX2 package [1-3]. The hydrogen atoms were fixed to standard X-ray bond lengths and refined using a riding atom model.
Modeling molecular alloy crystal structures: The occupancies of central metal atoms in the binary alloy crystals were refined using PART command, using an occupancy free variable (FVAR). However, the occupancy values and the convergence of refinements depend on other variables such as atomic displacement parameters (ADPs) and the positions of the metal atoms. Two different refinement strategies were tested to model the binary alloy structures:

(i) The positions of the central metal atoms were refined freely, whereas their ADPs were constrained to be the same using EADP. The occupancies were refined with the help of an occupancy free variable (FVAR) using PART command.

(ii) The positions and atomic displacement parameters (ADPs) of the metal atoms occupying very close positions were constrained to be identical using the constraints EXYZ and EADP. The occupancies were refined with the help of an occupancy free variable (FVAR) using PART command.

After testing these two refinement methods, it was found that allowing the positions to be refined freely resulted in unreasonable ADPs or convergence issues. Hence, we adopted the strategy (ii) to model all the binary alloy structures in this study.

Modeling of ternary and quaternary alloy crystals: For ternary and quaternary alloy crystals, the occupancies of three/ four metal atoms occupying very close positions were found to be difficult to model. Attempts to refine the occupancies using SUMP restraint (the linear restraint that allows more than two atoms to be assigned to a particular site, with the sum of their site occupancy factors restrained to 1 resulted in unreasonable ADPs, in addition to some convergence issues. Hence to model these structures, the occupancies of central metal atoms were fixed to the values from the composition information obtained from energy-dispersive X-ray spectra (EDX) measured on the same single crystals. As in case of the binary alloy structures, the positions and atomic displacement parameters (ADPs) of the metal atoms occupying very close positions were constrained to be identical using the constraints EXYZ and EADP.

The crystallographic information files have been deposited in the CCDC database with CCDC deposition numbers 2073957-2073962, 2074033-2074036, 2074042-2074044, 2074050-2074051, 2074080-2074081.

1. Dolomanov, O.V., Bourhis, L.J., Gildea, R.J, Howard, J.A.K. & Puschmann, H. (2009), *J. Appl. Cryst.* 42, 339-341.
2. Bourhis, L.J., Dolomanov, O.V., Gildea, R.J., Howard, J.A.K., Puschmann, H. (2015). *Acta Cryst. A*, 71, 59-75.
3. Bourhis, L.J., Dolomanov, O.V., Gildea, R.J., Howard, J.A.K., Puschmann, H. (2015). *Acta Cryst. A*, 71, 59-75.
Figure S2: Crystal packing of mer-β Gaq₃ (left) in comparison with the new polymorph mer-ε Gaq₃ (right).
Figure S3: Hirshfeld fingerprint plots for the crystal structures of Mq₃ compounds discussed in this study. Note that mer forms of Mq₃ have three molecules in the asymmetric unit.
Table S1. Percentage contributions of different intermolecular atom…atom contacts estimated by Hirshfeld fingerprint analysis for the crystal structures of Mq₃ compounds discussed in this study.

| Compound     | Molecule 1 | Molecule 2 | Molecule 3 | Molecular volume in Å³ (Hirshfeld surface) |
|--------------|------------|------------|------------|-------------------------------------------|
| Mer-Alq₃     | 6.5        | 34.5       | 10.3       | 535.17                                    |
| molecule 2   | 5.1        | 33.1       | 10.3       | 537.52                                    |
| molecule 3   | 6.9        | 33.0       | 10.3       | 548.70                                    |
| Mer-Gaq₃     | 5.4        | 33.8       | 11.1       | 529.35                                    |
| molecule 2   | 7.1        | 34.0       | 10.9       | 526.76                                    |
| molecule 3   | 5.4        | 35.7       | 11.2       | 540.41                                    |
| Mer-Crq₃     | 7.0        | 33.5       | 10.9       | 530.03                                    |
| molecule 2   | 5.3        | 33.3       | 11.2       | 531.52                                    |
| molecule 3   | 5.4        | 35.0       | 33.3       | 543.54                                    |
| Fac-Alq₃     | 5.1        | 33.2       | 12.6       | 528.80                                    |
| Fac-Inq₃     | 7.4        | 32.7       | 11.6       | 526.23                                    |

Figure S4: Asymmetric units of Alq₃-Gaq₃ molecular alloys with thermal ellipsoids at the 50% probability level.
**Figure S5:** Asymmetric units of $\text{Alq}_3\text{-Crq}_3$, $\text{Alq}_3\text{-Inq}_3$ and $\text{Gaq}_3\text{-Crq}_3$ molecular alloys with thermal ellipsoids at the 50% probability level.
Table S2. Crystal data and structure refinement for the Mq₃ series of compounds

|                | Alq₃  | Gaq₃    | Crq₃    |
|----------------|-------|---------|---------|
| **Stoichiometric ratio used for alloy crystallization** |       |         |         |
| **Empirical formula** | C₁₂H₁₃N₃Al | C₂₇H₁₅GaN₃O₃ | C₂₇H₁₅CrN₃O₃ |
| **Formula weight** | 306.30 | 502.16  | 484.44  |
| **Temperature/K** | 100(1) | 100.01(10) | 100(1)  |
| **Crystal system**  | triclinic | triclinic | triclinic |
| **Space group**     | P-1   | P-1     | P-1     |
| **a/Å**             | 13.3849(5) | 13.947(4) | 13.4826(9) |
| **b/Å**             | 15.5506(6) | 15.5684(6) | 15.7428(14) |
| **c/Å**             | 18.6315(4) | 18.6801(5) | 18.5238(7) |
| **α/°**             | 95.044(3)  | 94.969(3) | 94.860(5) |
| **β/°**             | 109.792(3) | 109.651(3) | 109.436(5) |
| **γ/°**             | 114.085(4) | 113.851(3) | 114.310(8) |
| **Volume/Å³**       | 3213.8(2)  | 3241.3(2) | 3264.5(4) |
| **Z**               | 9      | 6       | 6       |
| **ρ(calc) g/cm³**   | 1.4242 | 1.544   | 1.478   |
| **μ/mm⁻¹**          | 0.132  | 1.310   | 0.562   |
| **F(000)**          | 1429.0 | 1536.0  | 1494.0  |
| **Crystal size/mm³**| 0.25 × 0.14 × 0.1 | 0.19 × 0.11 × 0.06 | 0.22 × 0.11 × 0.06 |
| **Radiation**       | Mo Kα (λ = 0.71073) | MoKα (λ = 0.71073) | MoKα (λ = 0.71073) |
| **2Θ range for data collection/°** | 2.98 to 52 | 6.606 to 52.998 | 3.436 to 51.996 |
| **Index ranges**    | -16 ≤ h ≤ 15, -18 ≤ k ≤ 18, -23 ≤ l ≤ 22 | -16 ≤ h ≤ 16, -19 ≤ k ≤ 19, -16 ≤ h ≤ 16, -19 ≤ k ≤ 19, -22 ≤ l ≤ 22 |
| **Reflections collected** | 46405 | 53536  | 40042  |
| **Independent reflections** | 12017 [R(int) = 0.0973, R(sigma) = 0.1118] | 13414 [R(int) = 0.0532, R(sigma) = 0.0587] | 12844 [R(int) = 0.1250, R(sigma) = 0.1963] |
| **Data/restraints/parameters** | 12017/0/919 | 13414/0/769 | 12844/0/919 |
| **Goodness-of-fit on F²** | 1.071 | 1.064 | 0.936 |
| **Final R indexes [I>=2σ (I)]** | R₁ = 0.0661, wR₂ = 0.1555 | R₁ = 0.0536, wR₂ = 0.1309 | R₁ = 0.0930, wR₂ = 0.1814 |
| **Final R indexes [all data]** | R₁ = 0.1225, wR₂ = 0.1930 | R₁ = 0.0781, wR₂ = 0.1431 | R₁ = 0.2161, wR₂ = 0.2276 |
| **Largest diff. peak/hole / e Å⁻³** | 0.72/-0.69 | 2.02/-0.68 | 1.35/-0.45 |

Inq₃ single crystals obtained were too weakly diffracting to obtain full data for structure solution. However, the cell parameters were obtained from a SC-XRD dataset, which confirmed that Inq₃ crystallizes in fac-δ form (Z=2) with the following cell parameters:

\[
a = 6.196(7) \text{ Å, } b = 13.44(2) \text{ Å, } c = 14.73(3) \text{ Å, } α = 65.65(17)°, \ β = 88.03(12)°, \ γ = 83.53(11)°, \text{ Volume} = 1110(3) \text{ Å}³
\]
Table S3. Crystal data and structure refinement for the Alq\textsubscript{3}-Ga\textsubscript{3} binary alloys

| Stoichiometric ratio used for alloy crystallization | Alq\textsubscript{3}-Ga\textsubscript{3}(9:1) | Alq\textsubscript{3}-Ga\textsubscript{3}(1:1) | Alq\textsubscript{3}-Ga\textsubscript{3}(1:3) |
|--------------------------------------------------|---------------------------------|---------------------------------|---------------------------------|
| Empirical formula                                | \( \text{C}_{27}\text{H}_{18}\text{Al}_{0.91}\text{Ga}_{0.09}\text{N}_{3}\text{O}_{3} \) | \( \text{C}_{27}\text{H}_{18}\text{Al}_{0.48}\text{Ga}_{0.52}\text{N}_{3}\text{O}_{3} \) | \( \text{C}_{27}\text{H}_{18}\text{Al}_{0.27}\text{Ga}_{0.73}\text{N}_{3}\text{O}_{3} \) |
| Formula weight                                   | 463.21                          | 481.88                          | 490.70                          |
| Temperature/K                                     | 100.01(10)                      | 100.00(10)                      | 99.94(19)                       |
| Crystal system                                    | triclinic                       | triclinic                       | triclinic                       |
| Space group                                       | P-1                             | P-1                             | P-1                             |
| \(a/Å\)                                           | 13.3967(6)                      | 13.3851(4)                      | 13.3810(4)                      |
| \(b/Å\)                                           | 15.5597(7)                      | 15.5551(5)                      | 15.5698(5)                      |
| \(c/Å\)                                           | 18.6662(5)                      | 18.6583(5)                      | 18.6754(4)                      |
| \(\alpha/°\)                                      | 95.00(3)                        | 94.96(2)                        | 94.98(2)                        |
| \(\beta/°\)                                       | 109.81(3)                       | 109.74(2)                       | 109.72(2)                       |
| \(\gamma/°\)                                      | 113.987(4)                      | 113.903(3)                      | 113.79(3)                       |
| Volume/Å\(^3\)                                    | 3227.9(3)                       | 3228.32(18)                     | 3237.2(2)                       |
| \(Z\)                                             | 6                               | 6                               | 6                               |
| \(\rho_{\text{calc}}\text{g/cm}^3\)               | 1.4296                          | 1.487                           | 1.5101                          |
| \(\mu\text{mm}^{-1}\)                            | 0.236                           | 0.753                           | 0.995                           |
| \(F(000)\)                                        | 1438.6                          | 1485.0                          | 1508.7                          |
| Crystal size/mm\(^3\)                            | 0.25 × 0.1 × 0.06               | 0.28 × 0.12 × 0.1               | 0.25 × 0.14 × 0.11              |
| Radiation                                         | Mo Kα (\(\lambda = 0.71073\))  | Mo Kα (\(\lambda = 0.71073\)) | Mo Kα (\(\lambda = 0.71073\))  |
| 2Θ range for data collection/°                    | 3.3 to 54                       | 6.612 to 62.008                 | 3.3 to 54                       |
| Index ranges                                      | -16 ≤ \(h\) ≤ 16, -19 ≤ \(k\) ≤ 19, -19 ≤ \(h\) ≤ 19, -21 ≤ \(k\) ≤ 22, -16 ≤ \(h\) ≤ 17, -19 ≤ \(k\) ≤ 19, -24 ≤ \(l\) ≤ 23 | -26 ≤ \(l\) ≤ 26 | -24 ≤ \(l\) ≤ 24 |
| Reflections collected                             | 42561                           | 84947                           | 56372                           |
| Independent reflections                           | 13933 [\(R_{\text{int}} = 0.0794, R_{\text{sigma}} = 0.0731, R_{\text{int}} = 0.0639, R_{\text{sigma}} = 0.0703\)] | 18704 | 13945 [\(R_{\text{int}} = 0.0959\)] |
| Data/restraints/parameters                        | 13933/0/920                     | 18704/0/812                     | 13945/0/920                     |
| Goodness-of-fit on \(F^2\)                       | 1.052                           | 1.037                           | 1.053                           |
| Final R indexes [\(I\geq2\sigma\ (I)\)]      | \(R_1 = 0.0596, wR_2 = 0.1358, R_1 = 0.0556, wR_2 = 0.0971, R_1 = 0.0425, wR_2 = 0.0852\) | \(R_1 = 0.0971, wR_2 = 0.1629, R_1 = 0.1098, wR_2 = 0.1193, R_1 = 0.0676, wR_2 = 0.0981\) | \(R_1 = 0.0971, wR_2 = 0.1629, R_1 = 0.1098, wR_2 = 0.1193, R_1 = 0.0676, wR_2 = 0.0981\) |
| Largest diff. peak/hole / e Å\(^{-3}\)           | 0.75/-0.49                      | 0.72/-0.63                      | 0.87/-0.54                      |
| Percentage of Al                                  | 0.9118(12)                      | 0.4751(14)                      | 0.2687(14)                      |
| Percentage of Ga                                  | 0.0882(12)                      | 0.5249(14)                      | 0.7313(14)                      |
| Empirical formula | Alq₃-Cr₂(O₃)₃(95:5) | Alq₃-Cr₂(O₃)₃(9:1) | Alq₃-Cr₂(O₃)₃(3:1) | Alq₃-Cr₂(O₃)₃(1:1) |
|-------------------|---------------------|---------------------|---------------------|---------------------|
| Stoichiometric ratio used for alloy crystallization | | | | |
| Content | Formula weight | Temperature/K | Crystal system | Space group | a/Å | b/Å | c/Å | α/° | β/° | γ/° | Volume/Å³ | Z | ρ(calc) | μ/mm⁻¹ | F(000) | 2Θ range for data collection/° | Index ranges | Reflections collected | Independent reflections | Data/restraints/parameters | Goodness-of-fit on F² | Final R indexes [I>2σ (I)] | Final R indexes [all data] | Largest diff. peak/hole / e Å⁻³ | Percentage of Al | Percentage of Cr |
|-------------------|---------------------|---------------------|---------------------|---------------------|-----|-----|-----|------|------|------|-----------|-----|--------|--------|-------|-----------------------------|-----------------|------------------------|------------------------|------------------------|----------------|----------------------|----------------------|----------------------|---------------|-------------|
| C₂₇H₁₈Al₀.₉₄Cr₀.₀₆N₃O₃ | 461.01 | 100(1) | triclinic | P-1 | 13.4084(6) | 15.5505(9) | 18.6432(5) | 95.029(4) | 109.772(3) | 114.071(5) | 3222.7(3) | 6 | 1.425 | 0.159 | 1432.0 | 3.296 to 53.994 | -16 ≤ h ≤ 16, -19 ≤ k ≤ 19, -23 l ≤ 23 | 53059 | 13808 [R(int) = 0.2480, R(sigma) = 0.2384] | 13808/0/920 | 0.911 | 0.0862, wR₂ = 0.2194 | 0.2207, wR₂ = 0.2876 | 0.67/-0.52 | 0.935(3) | 0.065(3) |
| C₂₇H₁₈Al₀.₈₁Cr₀.₁₉N₃O₃ | 464.21 | 100.00(11) | triclinic | P-1 | 13.4069(6) | 15.5667(9) | 18.6269(6) | 95.032(4) | 109.659(4) | 114.181(5) | 3222.5(3) | 6 | 1.4351 | 0.215 | 1441.9 | 3.3 to 54 | -17 ≤ h ≤ 16, -19 ≤ k ≤ 19, -24 ≤ l ≤ 24 | 56151 | 13898 [R(int) = 0.0946, R(sigma) = 0.1292] | 13898/0/920 | 1.052 | 0.0925, wR₂ = 0.1346 | 0.1705, wR₂ = 0.1698 | 1.70/-0.75 | 0.809(4) | 0.191(4) |
| C₂₇H₁₈Al₀.₇₇Cr₀.₂₃N₃O₃ | 465.22 | 100(1) | triclinic | P-1 | 13.5617(12) | 15.5617(12) | 18.6118(7) | 95.032(4) | 109.711(5) | 114.128(7) | 3219.4(4) | 6 | 1.440 | 0.233 | 1443.0 | 3.452 to 51.998 | -10 ≤ h ≤ 16, -18 ≤ k ≤ 12, -21 ≤ l ≤ 23 | 20022 | 10587 [R(int) = 0.0847, R(sigma) = 0.1717] | 10587/0/920 | 0.2384 | 0.3957, wR₂ = 0.2194 | 0.1595, wR₂ = 0.1698 | 0.61/-0.33 | 0.768(3) | 0.232(3) |
| C₂₇H₁₈Al₀.₄4Cr₀.₆₆N₃O₃ | 474.56 | 100.00(10) | triclinic | P-1 | 15.6445(19) | 15.6445(19) | 18.5794(6) | 95.032(4) | 109.452(6) | 114.370(10) | 3246.3(6) | 6 | 1.456 | 0.394 | 1468.0 | 6.896 to 53.996 | -16 ≤ h ≤ 14, -18 ≤ k ≤ 19, -22 ≤ l ≤ 23 | 20588 | 11360 [R(int) = 0.0538, R(sigma) = 0.1054] | 11360/0/920 | 1.0946 | 0.1167, wR₂ = 0.2746 | 0.1599, wR₂ = 0.3000 | 0.14 × 0.08 × 0.06 | 0.935(3) | 0.396(7) |

Table S4. Crystal data and structure refinement for the Alq₃-Cr₂(O₃)₃ binary alloys
Table S5. Crystal data and structure refinement for the Alq₃-Inq₃ binary alloys

| Stoichiometric ratio used for alloy crystallization | Alq₃-Inq₃(9:1)                           | Alq₃-Inq₃(9:1)                           |
|---------------------------------------------------|----------------------------------------|----------------------------------------|
| Empirical formula                                 | C₂₇H₁₈Al₀.₈₉In₀.₁₂N₃O₃                  | C27H18Al0.87In0.13N3O3                 |
| Formula weight                                    | 470.09                                 | 470.51                                 |
| Temperature/K                                     | 100(1)                                 | 99.98(12)                              |
| Crystal system                                    | triclinic                              | triclinic                              |
| Space group                                       | P-1                                    | P-1                                    |
| a/Å                                               | 13.4101(13)                            | 13.3999(10)                            |
| b/Å                                               | 15.6147(17)                            | 15.6033(13)                            |
| c/Å                                               | 18.6245(10)                            | 18.6507(8)                             |
| α°                                                | 95.104(7)                              | 95.062(5)                              |
| β°                                                | 109.621(7)                             | 109.708(5)                             |
| γ°                                                | 114.045(10)                            | 114.012(8)                             |
| Volume/Å³                                         | 3237.0(6)                              | 3236.1(5)                              |
| Z                                                 | 6                                      | 6                                      |
| ρcalc/g/cm³                                       | 1.447                                  | 1.4485                                 |
| μ/mm⁻¹                                            | 0.250                                  | 0.257                                  |
| F(000)                                            | 1454.0                                 | 1455.6                                 |
| Crystal size/mm³                                  | 0.2 × 0.15 × 0.11                      | 0.21 × 0.14 × 0.1                      |
| Radiation                                         | MoKa (λ = 0.71073)                     | Mo Ka (λ = 0.71073)                    |
| 2Θ range for data collection/°                   | 3.28 to 54                             | 3.28 to 54                             |
| Index ranges                                      | -16 ≤ h ≤ 16, -19 ≤ k ≤ 19, -23 ≤ l ≤ | -16 ≤ h ≤ 16, -19 ≤ k ≤ 19, -25 ≤ l ≤ |
|                                                   | 23                                      | 24                                     |
| Reflections collected                             | 40517                                  | 56377                                  |
| Independent reflections                           | 13918 [R_int = 0.1923, R_sigma =     | 13930 [Rint = 0.2247, Rsigma =       |
|                                                   | 0.3085]                                | 0.3270]                                |
| Data/restraints/parameters                        | 13918/0/884                            | 13930/0/920                            |
| Goodness-of-fit on F²                              | 0.899                                  | 0.987                                  |
| Final R indexes [I>=2σ (I)]                       | R₁ = 0.1108, wR₂ = 0.2364             | R₁ = 0.1092, wR2 = 0.2323              |
|                                                   | R₁ = 0.2792, wR₂ = 0.3033             | R₁ = 0.2731, wR2 = 0.3376              |
| Largest diff. peak/hole / e Å⁻³                   | 1.38/-0.66                             | 2.20/-1.40                             |
| Percentage of Al                                  | 0.8922(13)                             | 0.8740(17)                             |
| Percentage of In                                  | 0.1185(15)                             | 0.1260(17)                             |
| Table S6. Crystal data and structure refinement for the Ga$_3$-Cr$_3$ binary alloys |
|-------------------------------------------------|-------------------------------------------------|
| Stoichiometric ratio used for alloy crystallization | Ga$_3$-Cr$_3$(1:1) | Ga$_3$-Cr$_3$(9:1) |
| **Empirical formula** | C$_{27}$H$_{18}$Cr$_{0.74}$Ga$_{0.26}$N$_3$O$_3$ | C$_{27}$H$_{18}$Cr$_{0.23}$Ga$_{0.78}$N$_3$O$_3$ |
| **Formula weight** | 489.08 | 498.11 |
| **Temperature/K** | 100.01(10) | 100.01(10) |
| **Crystal system** | triclinic | triclinic |
| **Space group** | P-1 | P-1 |
| **a/Å** | 13.4304(3) | 13.4017(7) |
| **b/Å** | 15.5926(5) | 15.5902(9) |
| **c/Å** | 18.5705(3) | 18.6416(7) |
| **α/°** | 94.833(2) | 94.981(4) |
| **β/°** | 109.506(2) | 109.585(4) |
| **γ/°** | 114.045(2) | 114.021(5) |
| **Volume/Å$^3$** | 3236.16(15) | 3237.0(3) |
| **Z** | 6 | 6 |
| **ρ$_{calc}$/g/cm$^3$** | 1.506 | 1.5330 |
| **μ/mm$^{-1}$** | 0.762 | 1.141 |
| **F(000)** | 1505.0 | 1528.5 |
| **Crystal size/mm$^3$** | 0.19 $\times$ 0.13 $\times$ 0.1 | 0.17 $\times$ 0.12 $\times$ 0.08 |
| **Radiation** | MoKα ($λ = 0.71073$) | MoKα ($λ = 0.71073$) |
| **2θ range for data collection/°** | 6.616 to 55.998 | 3.3 to 52 |
| **Index ranges** | -17 $\leq$ h $\leq$ 17, -20 $\leq$ k $\leq$ 20, -24 $\leq$ l $\leq$ 24 | -17 $\leq$ h $\leq$ 17, -20 $\leq$ k $\leq$ 19, -24 $\leq$ l $\leq$ 24 |
| **Reflections collected** | 76466 | 42649 |
| **Independent reflections** | 15576 [R$_{int}$ = 0.0451, R$_{sigma}$ = 0.0345] | 12724 [R$_{int}$ = 0.0968, R$_{sigma}$ = 0.1452] |
| **Data/restraints/parameters** | 15576/0/920 | 12724/0/920 |
| **Goodness-of-fit on F$^2$** | 1.139 | 1.013 |
| **Final R indexes [I>=2σ (I)]** | R$_1$ = 0.0661, wR$_2$ = 0.1870 | R$_1$ = 0.0711, wR$_2$ = 0.1684 |
| **Final R indexes [all data]** | R$_1$ = 0.0777, wR$_2$ = 0.1924 | R$_1$ = 0.1183, wR$_2$ = 0.2012 |
| **Largest diff. peak/hole / e Å$^{-3}$** | 2.80/-1.28 | 2.69/-1.55 |
| **Percentage of Ga** | 0.262(7) | 0.770(8) |
| **Percentage of Cr** | 0.738(7) | 0.230(8) |
Table S7. Crystal data and structure refinement for the Alq$_3$-Ga$_q$$_3$-Crq$_3$ ternary alloys and Alq$_3$-Ga$_q$$_3$-Crq$_3$-Inq$_3$ quaternary alloy crystals.*

| Stoichiometric ratio used for alloy crystallization | Alq$_3$-Ga$_q$$_3$-Crq$_3$(1:1:1) | Alq$_3$-Ga$_q$$_3$-Crq$_3$(1:1:1) | Alq$_3$-Ga$_q$$_3$-Crq$_3$-Inq$_3$(1:1:1:1) |
|--------------------------------------------------|---------------------------------|---------------------------------|------------------------------------------|
| Empirical formula                                | C$_{27}$H$_{18}$N$_3$Al$_{0.23}$Cr$_{0.51}$Ga$_{0.26}$O$_3$ | C$_{20}$H$_{20}$N$_3$AlCrGa     | C$_{27}$H$_{18}$N$_3$Al$_{0.14}$Cr$_{0.54}$Ga$_{0.14}$In$_{0.18}$O$_3$ |
| Formula weight                                   | 483.25                          | 481.28                          | 494.73                                   |
| Temperature/K                                     | 100.0(4)                        | 99.99(10)                       | 100.00(10)                               |
| Crystal system                                    | triclinic                       | triclinic                       | triclinic                                |
| Space group                                       | P-1                             | P-1                             | P-1                                      |
| a/Å                                               | 13.4222(7)                      | 13.4296(15)                     | 13.4055(7)                               |
| b/Å                                               | 15.6206(8)                      | 15.589(3)                       | 15.6401(10)                              |
| c/Å                                               | 18.5990(4)                      | 18.5731(10)                     | 18.6009(6)                               |
| α/°                                               | 95.021(3)                       | 94.902(9)                       | 94.842(4)                                |
| β/°                                               | 109.472(4)                      | 109.511(8)                      | 109.500(4)                               |
| γ/°                                               | 114.159(5)                      | 114.214(14)                     | 113.676(5)                               |
| Volume/Å$^3$                                      | 3238.8(3)                       | 3228.8(9)                       | 3257.2(3)                                |
| Z                                                 | 6                               | 6                               | 6                                        |
| ρ__$\text{calc}$/g/cm$^3$                         | 1.487                           | 1.4850                          | 1.513                                    |
| μ/mm$^{-1}$                                       | 0.659                           | 0.613                           | 0.707                                    |
| F(000)                                           | 1490.0                          | 1486.5                          | 1518.0                                   |
| Crystal size/mm$^3$                               | 0.16 x 0.1 x 0.07               | 0.2 x 0.12 x 0.08               | 0.15 x 0.1 x 0.08                        |
| Radiation                                        | MoKα(λ = 0.71073)               | Mo Kα(λ = 0.71073)              | MoKα(λ = 0.71073)                        |
| 2Θ range for data collection/°                   | 3.292 to 53.998                 | 3.3 to 52                       | 6.584 to 58.83                           |
| Index ranges                                      | -16 ≤ h ≤ 16, -19 ≤ k ≤ 19, -16 ≤ l ≤ 17, -19 ≤ k ≤ 18, -17 ≤ h ≤ 16, -19 ≤ k ≤ 20, -23 ≤ l ≤ 23 | -24 ≤ l ≤ 23                     | -25 ≤ l ≤ 23                             |
| Reflections collected                             | 41246                           | 36489                           | 42679                                    |
| Independent reflections                           | 13935 [R$_{int}$ = 0.0583, R$_{sigma}$ = 0.0695] | 12696 [R$_{int}$ = 0.2736, R$_{sigma}$ = 0.4993] | 15344 [R$_{int}$ = 0.1195, R$_{sigma}$ = 0.2095] |
| Data/restraints/parameters                        | 13935/0/919                     | 12696/0/853                     | 15344/0/919                              |
| Goodness-of-fit on F$^2$                          | 1.099                           | 0.920                           | 0.986                                    |
| Final R indexes [I>2σ (I)]                       | R$_1$ = 0.0880, wR$_2$ = 0.2280 R$_1$ = 0.1245, wR$_2$ = 0.2854 R$_1$ = 0.0756, wR$_2$ = 0.1181 | R$_1$ = 0.3058, wR$_2$ = 0.4091 R$_1$ = 0.1931, wR$_2$ = 0.1620 |
| Final R indexes [all data]                       | R$_1$ = 0.1222, wR$_2$ = 0.2499 R$_1$ = 0.3058, wR$_2$ = 0.4091 R$_1$ = 0.1931, wR$_2$ = 0.1620 |
| Largest diff. peak/hole / e Å$^{-3}$              | 3.02/-0.80                      | 4.04/-1.79                      | 0.89/-0.53                               |
| Percentage of Al                                 | 0.24                            | 0.29                            | 0.14                                     |
| Percentage of Cr                                 | 0.5                             | 0.48                            | 0.54                                     |
| Percentage of Ga                                 | 0.26                            | 0.23                            | 0.14                                     |
| Percentage of In                                 | ---                             | ---                             | 0.18                                     |

* The occupancy values of metal atoms are fixed to the compositions obtained from EDX spectra
Table S8. Ga$_3$In$_3$ binary alloys: single crystals of Ga$_3$In$_3$ binary alloys obtained were too weakly diffracting to obtain full data for structure solution. However, the cell parameters were obtained from SC-XRD datasets showed mer-$\varepsilon$ form ($Z=6$) with the following cell parameters (for the different crystals tested):

| Crystal | Ga-Inq$_3$ (50:50) | Ga-Inq$_3$ (50:50) | Ga-Inq$_3$ (90:10) | Ga-Inq$_3$ (90:10) | Ga-Inq$_3$ (90:10) |
|---------|---------------------|---------------------|---------------------|---------------------|---------------------|
| Crystal system | triclinic | triclinic | triclinic | triclinic | triclinic |
| a/Å | 13.419 | 13.437 | 13.400 | 13.483 | 13.510 |
| b/Å | 15.706 | 15.710 | 15.574 | 15.656 | 15.780 |
| c/Å | 18.664 | 18.642 | 18.689 | 18.632 | 18.612 |
| $\alpha/^\circ$ | 94.987 | 94.709 | 95.043 | 95.171 | 94.836 |
| $\beta/^\circ$ | 109.445 | 109.491 | 109.708 | 109.297 | 109.461 |
| $\gamma/^\circ$ | 113.450 | 113.800 | 113.831 | 114.727 | 114.970 |
| Volume/Å$^3$ | 3291.68 | 3285.20 | 3243.09 | 3250.13 | 3272.01 |
| Presumed crystal form | mer-$\varepsilon$ | mer-$\varepsilon$ | mer-$\varepsilon$ | mer-$\varepsilon$ | mer-$\varepsilon$ |
| Z | 6 | 6 | 6 | 6 | 6 |
| Presumed space group | $P-1$ | $P-1$ | $P-1$ | $P-1$ | $P-1$ |

Alq$_3$In$_3$ binary alloys: good quality single crystals of Alq$_3$In$_3$ binary alloys could be only for very low composition of In (as given in Table 4). Alloy crystal phases of Alq$_3$In$_3$ 50:50 and 30:0 binary phases were characterized by powder XRD Lebail fitting as given below.
Figure S6. Le Bail fitting PXRD profile for the Alq₃-Inq₃ (30:70) binary alloy sample. Cell parameters: \(a=13.458\ \text{Å}, \ b=15.534\ \text{Å}, \ c=18.606\ \text{Å}, \ \alpha=95.104^\circ, \ \beta=110.973^\circ, \ \gamma=114.223^\circ\).

Figure S7. Le Bail fitting PXRD profile for the Alq₃-Inq₃ (50:50) binary alloy sample. Cell parameters: \(a=13.480\ \text{Å}, \ b=15.534\ \text{Å}, \ c=18.671\ \text{Å}, \ \alpha=95.349^\circ, \ \beta=110.507^\circ, \ \gamma=114.219^\circ\).
Figure S8. Le Bail fitting PXRD profile for the Inq$_3$ sample (fitted to cell parameters corresponding to fac-$\delta$ form).

Table S9: Average metal-oxygen and metal nitrogen bond distances observed in the Mq$_3$ crystal structures and their alloys.

| Composition | Metal-N | e.s.d. | Metal-O | e.s.d. |
|-------------|---------|--------|---------|--------|
| Alq$_3$     | ---     | 2.027  | 0.013   | 1.861  | 0.0185 |
| Gaq$_3$     | ---     | 2.075  | 0.016   | 1.948  | 0.014  |
| Crq$_3$     | ---     | 2.041  | 0.009   | 1.956  | 0.0115 |
| Alq$_3$-Gaq$_3$ | 91:09 (Al:Ga) | 2.041  | 0.016   | 1.873  | 0.014  |
| Alq$_3$-Gaq$_3$ | 48:52 (Al:Ga) | 2.058  | 0.015   | 1.910  | 0.015  |
| Alq$_3$-Gaq$_3$ | 27:73 (Al:Ga) | 2.066  | 0.015   | 1.929  | 0.014  |
| Alq$_3$-Crq$_3$ | 40:60 (Al:Cr) | 2.046  | 0.011   | 1.931  | 0.007  |
| Alq$_3$-Crq$_3$ | 77:23 (Al:Cr) | 2.030  | 0.013   | 1.889  | 0.010  |
| Alq$_3$-Crq$_3$ | 81:19 (Al:Cr) | 2.032  | 0.014   | 1.887  | 0.011  |
| Alq$_3$-Crq$_3$ | 94:06 (Al:Cr) | 2.029  | 0.012   | 1.875  | 0.014  |
| Gaq$_3$-Crq$_3$ | 77:23 (Ga:Cr) | 2.068  | 0.015   | 1.949  | 0.012  |
| Gaq$_3$-Crq$_3$ | 26:74 (Ga:Cr) | 2.061  | 0.010   | 1.955  | 0.011  |
S3: Raman spectroscopic studies of solid solutions

Raman spectra were recorded on a Renishaw inVia Reflex Micro Raman spectrometer using a 785 nm laser operated at 10% power. The spectra of the single crystal samples were recorded with an exposure time of 1 s using a 50× objective. The instrument was calibrated using a silicon standard.

Figure S9. Raman spectra collected from crystals of (a) Gaq₃-Crq₃ (b) Gaq₃-Inq₃ alloy crystals

Table S10. Raman peak positions and their corresponding vibrational assignments of mer-Alq₃.¹

| Mer Alq₃ | Vibration Assignments                                      |
|-----------------|-------------------------------------------------------------|
| 118 cm⁻¹        | Al-HQ deformation + Al-N stretching                          |
| 153 cm⁻¹        | 8-HQ wagging                                                |
| 166 cm⁻¹        | Butterfly mode                                             |
| 195 cm⁻¹        | 8-HQ wagging                                                |
| 238 cm⁻¹        | Ring torsion                                               |
| 314 cm⁻¹        | Al-N stretching + C-C-O bending                             |
| 503 cm⁻¹        | Ring deformation                                           |
| 525 cm⁻¹        | Ring deformation + Al-O stretching                          |
| 540 cm⁻¹        | Al-O str.+ Al-N str.+ ring deformation                       |
| 576 cm⁻¹        | Ring deformation + Al-O-C bending                           |
| 645 cm⁻¹        | Al-O stretching+ ring deformation                           |
| 756 cm⁻¹        | CH wagging                                                  |
Table S11. Raman peak positions and their corresponding vibrational assignments of fac-Alq$_3$.

| Fac-Alq$_3$ | Vibration Assignments                          |
|-------------|-----------------------------------------------|
| 118 cm$^{-1}$ | Al-HQ deformation + Al-N stretching          |
| 166 cm$^{-1}$ | Butterfly mode                               |
| 183 cm$^{-1}$ | Ring torsion                                 |
| 195 cm$^{-1}$ | 8-HQ wagging                                 |
| 224 cm$^{-1}$ | 8-HQ wagging                                 |
| 292 cm$^{-1}$ | Ring torsion                                 |
| 503 cm$^{-1}$ | Ring deformation                             |
| 535 cm$^{-1}$ | Ring deformation + Al-O stretching           |
| 548 cm$^{-1}$ | Al-O str.+ Al-N str.+ ring deformation        |
| 576 cm$^{-1}$ | Ring deformation + Al-O-C bending            |

1) Halls, M. and R. Aroca *Can. J. Chem.* **1998**, **76**, 1730-1736.
2) Hung, Shang-Yu, Kao, Ruei-Lin, Lin, Ku-Yen, Yang, Chun-Chuen, Lin, Kuen-Song, Chao, Yu-Chiang, Chiu, Kuan-Cheng. *Materials Chemistry and Physics*, **2015**, **154**, 100-106.

S4. Diffuse reflectance spectroscopic studies of solid solutions

For the diffuse reflectance spectroscopy, a Shimadzu UV-3600 spectrophotometer was used. The finely powdered samples were spread onto a layer of BaSO$_4$ powder and the spectra are measured in the wavelength range of 200 nm to 900 nm. The band gaps of the samples were estimated using Tauc plots$^{1,2}$ assuming direct allowed transition, i.e. plot of $[F(R)h\nu]^2$ against $h\nu$ where $F(R)$ is the Kubelka-Munk function and $h\nu$ is incident photon energy. In case of Crq$_3$ and its alloy crystals, there is a fair amount of absorption below the band gap energy and therefore a modified Tauc plot is used in these cases.$^3$
Figure S10. Tauc’s plots obtained from the diffuse reflectance spectra of pure Alq$_3$ and Gaq$_3$ and their alloys of varying compositions.

Figure S11. Tauc’s plots obtained from the diffuse reflectance spectra of pure Alq$_3$ and Inq$_3$ and their alloys of varying compositions.
Figure S12. Tauc’s plots obtained from the diffuse reflectance spectra of pure Alq₃ and Crq₃ and their alloys of varying compositions.

Figure S13. Tauc’s plots obtained from the diffuse reflectance spectra of pure Gaq₃ and Inq₃ and their alloys of varying compositions.
Figure S14. Tauc’s plots obtained from the diffuse reflectance spectra of pure Gaq$_3$ and Crq$_3$ and their alloys of varying compositions.

1) J. Tauc, R. Grigorovici, A. Vancu, *physica status solidi (b)* **1966**, *15*, 627-637
2) E. A. Davis, N. F. Mott, *The Philosophical Magazine: A Journal of Theoretical Experimental and Applied Physics* **1970**, *22*, 0903-0922.
3) Patrycja Makula, Michal Pacia, and Wojciech Macyk, *J. Phys. Chem. Lett.* **2018**, *23*, 6814–6817.

S5. Photoluminescence studies

Photoluminescence (PL) measurements on the samples were performed in solid state using FluoroMax-3 fluorometer, Jobin Yvon, Horiba. The single crystals of Mq$_3$ and their alloy samples were placed on a quartz plate with minimum amount of paratone oil to hold the crystal. The emission was collected with an excitation wavelength of 370 nm.

Figure S15: Optical microscopic images of single crystals of Alq$_3$-Gaq$_3$ alloy crystals of varying compositions, under UV light (395 nm).
Figure S16. Optical microscopic images of single crystals of Alq₃-Inq₃ alloy crystals of varying compositions, under UV light (395 nm).

Figure S17. Optical microscopic images of single crystals of Gaq₃-Inq₃ alloy crystals of varying compositions, under UV light (395 nm).

Figure S18. Photoluminescence spectra of (a) Gaq₃-Crq₃ 95:5 alloy crystal (b) ternary and quaternary alloy crystals (background corrected spectra). The sample is excited at $\lambda = 370$ nm.
S6. Energy-dispersive X-ray spectroscopy (EDX) measurements

Elemental mapping was obtained on a TALOS F200A with a TWIN lens system, X-FEG electron source. Spatially resolved elemental analysis, with a spatial resolution better than 2 nm, was obtained using the same TALOS microscope in STEM mode. Exposure times of 5 minutes were used to create elemental distribution maps with satisfactory counting statistics. STEM pictures were obtained using a High Angle Annular Dark Field detector (HAADF). RG overlays of the STEM EDX elemental maps were made using the FIJI (v.1.49q) software.¹

Figure S19. EDX maps of the a) Ga₀.₉₂In₀.₈ b) Al₀.₅₂Ga₀.₄₈ c) Al₀.₇₂In₀.₂₈ alloy crystal highlighting regions of different elemental contents and overlapped map showing nearly homogenous distribution of the individual components in the alloy crystal.
Elemental analysis

EDX measurements were performed on the Nova NANO SEM 600 using a high-resolution scanning electron microscope. The microscope was operated in high vacuum. It is equipped with an EDAX detector for energy dispersive X-Ray spectroscopy (EDX) measuring of samples.

Table S12: Elemental analysis of Al\(_{0.91}\)Ga\(_{0.09}\) alloy crystal from the EDX measurement.

| Sample       | Element | Point 1 |     |     |     |     |     |     |     |     |     |
|--------------|---------|---------|-----|-----|-----|-----|-----|-----|-----|-----|-----|
|              |         | Wt %    | At %| Wt %| At %| Wt %| At %| At %|     |     |     |
| Al\(_{0.91}\)Ga\(_{0.09}\) | Al      | 78.87   | 90.61| 80.93| 91.64| 79.89| 91.12|     | 91.1(4) |     |     |
|              | Ga      | 21.13   | 9.39 | 19.07| 8.34 | 20.11| 8.88 |     | 8.9(4)  |     |     |
| Al\(_{0.89}\)Ga\(_{0.11}\) | Al      | 74.78   | 88.46| 78.37| 90.35| 76.73| 89.5 |     | 89.4(8) |     |     |
|              | Ga      | 25.22   | 11.54| 21.63| 9.65 | 23.27| 10.5 |     | 10.6(8) |     |     |
| Al\(_{0.78}\)Ga\(_{0.22}\) | Al      | 56.59   | 77.11| 60.35| 79.73|     |     |     | 78(1)   |     |     |
|              | Ga      | 43.41   | 22.89| 39.65| 20.27|     |     |     | 22(1)   |     |     |
| Al\(_{0.59}\)Ga\(_{0.41}\) | Al      | 29.35   | 51.78| 37.19| 60.47| 42.89| 66   |     | 59 (5)  |     |     |
|              | Ga      | 70.65   | 48.22| 62.81| 39.53| 57.11| 34   |     | 41 (5)  |     |     |
| Al\(_{0.49}\)Ga\(_{0.51}\) | Al      | 22.17   | 42.39| 28.53| 50.78| 28.22| 50.39|     | 48(4)   |     |     |
|              | Ga      | 77.83   | 57.61| 71.47| 49.22| 71.78| 49.61|     | 52(4)   |     |     |
Table S13: Elemental analysis of Alq$_3$-Inq$_3$ alloy crystal from the EDX measurement.

| Sample     | Element | Point 1       | Point 2       | Point 3       | Average Atomic % with esd |
|------------|---------|---------------|---------------|---------------|---------------------------|
|            |         | Wt %  | At %  | Wt %  | At %  | Wt %  | At %  | Wt %  | At %  |
| Al$_{0.86}$In$_{0.14}$ | Al      | 61.07 | 86.97 | 58.98 | 85.95 | 86.5(5) |
|            | In      | 38.93 | 13.03 | 41.02 | 14.05 | 13.5(5) |
| Al$_{0.86}$In$_{0.14}$ | Al      | 57.10 | 84.99 | 55.70 | 84.25 | 64.07 | 88.35 | 86(2)  |
|            | In      | 42.90 | 15.01 | 44.30 | 15.75 | 35.93 | 11.65 | 14(2)  |
| Al$_{0.49}$In$_{0.51}$  | Al      | 18.63 | 49.35 | 17.99 | 48.28 | 48.8(5) |
|            | In      | 81.37 | 50.65 | 82.01 | 51.72 | 51.2(5) |
| Al$_{0.27}$In$_{0.73}$  | Al      | 8.44  | 28.19 | 7.67  | 26.13 | 27(1)  |
|            | In      | 91.56 | 71.81 | 92.33 | 73.87 | 73(1)  |

Table S14: Elemental analysis of Alq$_3$-Crq$_3$ alloy crystal from the EDX measurement.

| Sample     | Element | Point 1       | Point 2       | Point 3       | Average Atomic % with esd |
|------------|---------|---------------|---------------|---------------|---------------------------|
|            |         | Wt %  | At %  | Wt %  | At %  | Wt %  | At %  | Wt %  | At %  | Wt %  | At %  |
| Al$_{0.22}$Cr$_{0.78}$ | Al      | 15.28 | 25.79 | 10.13 | 17.85 | 13.20 | 22.66 | 22(3) |
|            | Cr      | 84.72 | 74.21 | 89.87 | 82.15 | 86.80 | 77.34 | 78(3) |
| Al$_{0.85}$Cr$_{0.15}$ | Al      | 73.93 | 84.53 | 74.85 | 85.15 | 84.8(3) |
|            | Cr      | 26.07 | 15.47 | 25.15 | 14.85 | 15.2(3) |
| Al$_{0.89}$Cr$_{0.11}$ | Al      | 80.46 | 88.81 | 75.94 | 85.88 | 83.69 | 90.82 | 89(2) |
|            | Cr      | 19.54 | 11.19 | 24.06 | 14.12 | 16.31 | 9.18  | 11(2) |
### Table S15: Elemental analysis of Gaq\textsubscript{3}-Inq\textsubscript{3} alloy crystal from the EDX measurement.

| Sample      | Element | Point 1 | Point 2 | Point 3 | Average Atomic % with esd |
|-------------|---------|---------|---------|---------|---------------------------|
|             |         | Wt %    | At %    | Wt %    | At %                      |                           |
| Ga\textsubscript{0.88}In\textsubscript{0.12} | Ga      | 81.37   | 87.79   | 82.88   | 88.85                     | 81.85                     | 88.13                     | 88.3(4)                   |
|             | In      | 18.63   | 12.21   | 17.12   | 11.15                     | 18.15                     | 11.87                     | 11.7(4)                   |
| Ga\textsubscript{0.51}In\textsubscript{0.49} | Ga      | 40.69   | 53.05   | 38.57   | 50.84                     | 36.25                     | 48.36                     | 51(2)                     |
|             | In      | 59.31   | 46.95   | 61.43   | 49.16                     | 63.75                     | 51.64                     | 49(2)                     |

### Table S16: Elemental analysis of Gaq\textsubscript{3}-Crq\textsubscript{3} alloy crystal from the EDX measurement.

| Sample      | Element | Point 1 | Point 2 | Point 3 | Average Atomic % with esd |
|-------------|---------|---------|---------|---------|---------------------------|
|             |         | Wt %    | At %    | Wt %    | At %                      |                           |
| Ga\textsubscript{0.62}Cr\textsubscript{0.38} | Ga      | 66.73   | 59.93   | 70.08   | 63.59                     | 67.88                     | 61.18                     | 62(2)                     |
|             | Cr      | 33.27   | 40.07   | 29.92   | 36.41                     | 32.12                     | 38.82                     | 38(2)                     |
| Ga\textsubscript{0.51}Cr\textsubscript{0.49} | Ga      | 55.75   | 48.45   | 59.09   | 51.86                     | 60.55                     | 53.37                     | 51(2)                     |
|             | Cr      | 44.25   | 51.55   | 40.91   | 48.14                     | 39.45                     | 46.63                     | 49(2)                     |

### Table S17: Elemental analysis of Alq\textsubscript{3}-Gaq\textsubscript{3}-Crq\textsubscript{3} ternary alloy crystal from the EDX measurement.

| Sample      | Element | Point 1 | Point 2 | Point 3 | Point 4 | Average Atomic % with esd |
|-------------|---------|---------|---------|---------|---------|---------------------------|
|             |         | Wt %    | At %    | Wt %    | At %    | Wt % | At % | Wt % | At % |                           |                           |
| Al\textsubscript{0.29}Ga\textsubscript{0.23}Cr\textsubscript{0.48} | Al     | 20.64   | 35.52   | 13.07   | 24.36   | 15.56 | 28.48 | 29(5) |                           |                           |
|             | Ga      | 28.17   | 18.76   | 34.49   | 24.89   | 35.90 | 25.43 | 23(2) |                           |                           |
|             | Cr      | 51.20   | 45.72   | 52.44   | 50.75   | 48.54 | 46.09 | 48(3) |                           |                           |
| Al\textsubscript{0.23}Ga\textsubscript{0.26}Cr\textsubscript{0.50} | Al     | 12.02   | 22.88   | 11.46   | 21.79   | 13.19 | 24.4  | 13.09 | 24.54 | 24(1)                     |
|             | Ga      | 38.86   | 28.62   | 36.55   | 26.9    | 31.63 | 22.64 | 36.85 | 26.75 | 26(2)                     |
|             | Cr      | 49.12   | 48.5    | 51.99   | 51.31   | 55.18 | 52.96 | 50.06 | 48.71 | 50(2)                     |
Table S18: Elemental analysis of Alq₃-Gaq₃-Crq₃-Inq₃ quaternary alloy crystal from the EDX measurement.

| Sample | Element | Point 1  | Point 2  | Average Atomic % with esd |
|--------|---------|----------|----------|---------------------------|
|        |         | Wt %     | At %     | Wt % | At % |                   |
| AlGaCrIn | Al      | 4.77     | 11.09    | 7.62  | 17.3 | 14(3)             |
|         | Ga      | 17.01    | 15.32    | 14.30 | 12.56| 14(1)             |
|         | Cr      | 46.64    | 56.32    | 44.21 | 52.07| 54(2)             |
|         | In      | 31.58    | 17.27    | 33.86 | 18.06| 17.7(4)           |

1) C. A. Schneider, W. S. Rasband, K. W. Eliceiri, *Nature Methods* 2012, 9, 671-675.