Supplementary information for

Synthesis of atomically layered and chemically ordered Rare-Earth (RE) *i*-MAX phases; $(Mo_{2/3}RE_{1/3})_2GaC$ with RE = Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu

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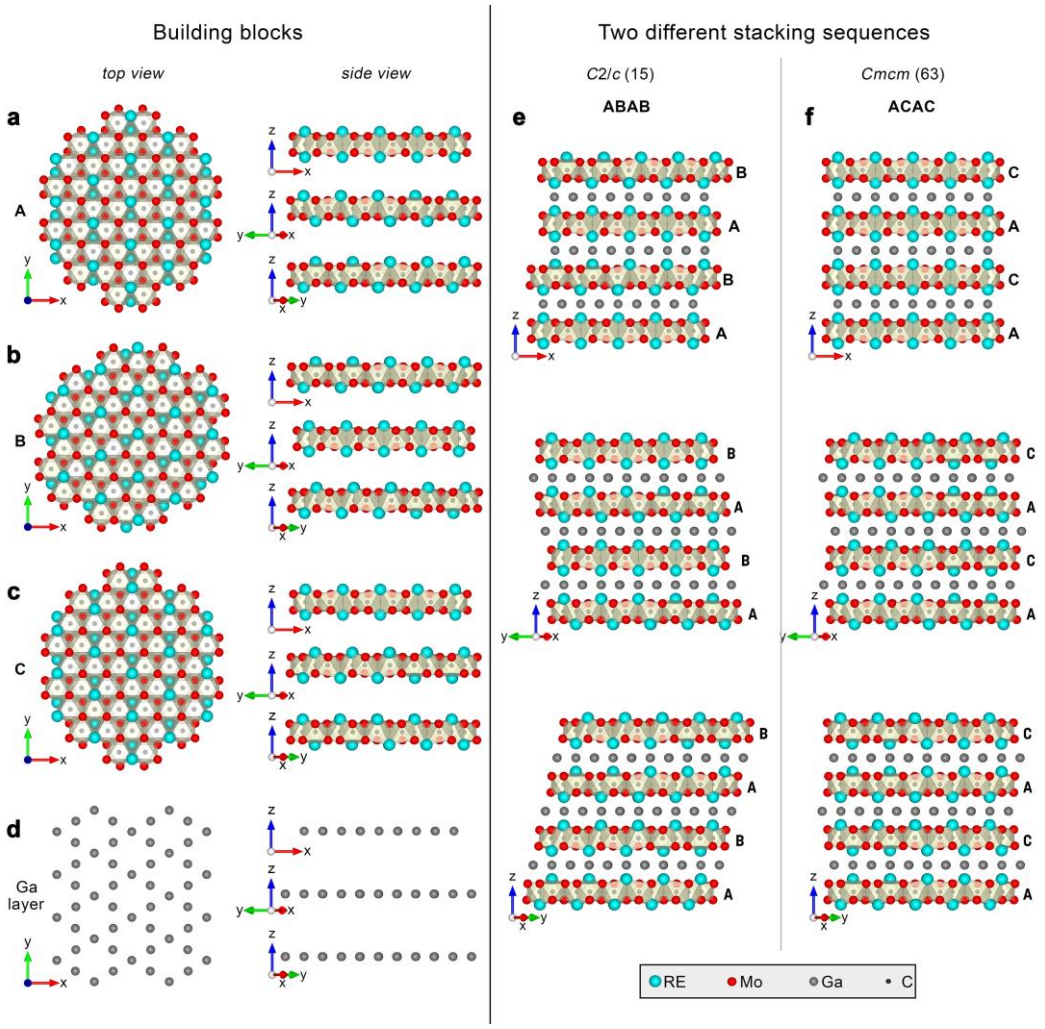
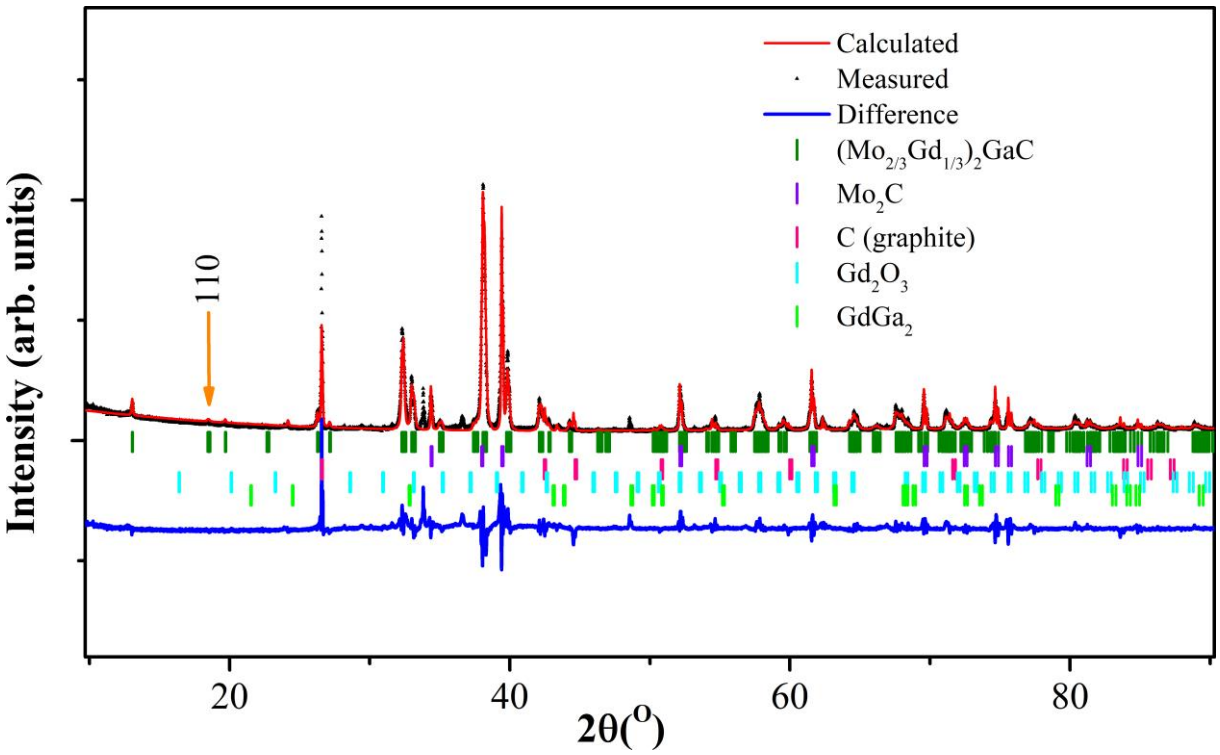
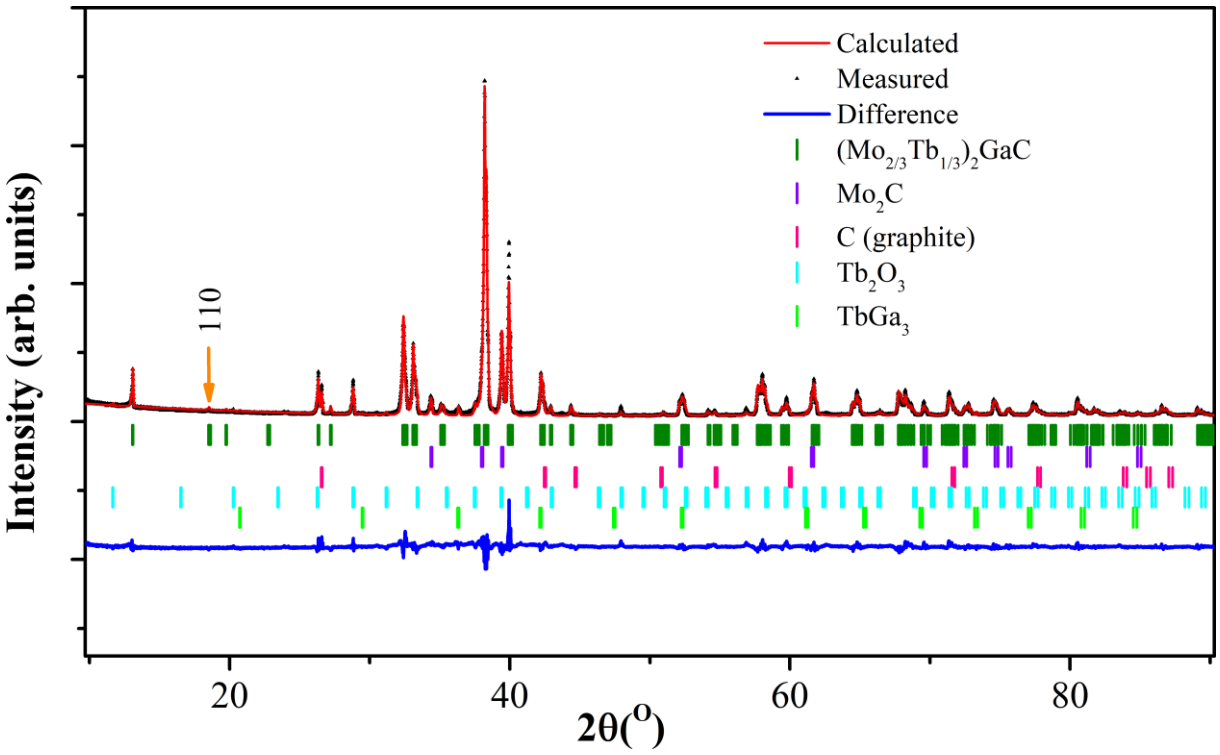


Supplementary figure S1. Schematic structural representation of the different stacking sequences in the *i*-MAX phases with formula $(Mo_{2/3}RE_{1/3})_2GaC$ for space groups *C*2/*c* (15) and *Cmcm* (63). Both polymorphs consist of equivalent $(Mo_{2/3}RE_{1/3})_2C$ and Ga building blocks, with $(Mo_{2/3}RE_{1/3})_2C$ in three different relative orientations; A, B (rotated +60° relative A), and C (rotated +180° relative to A). Different stacking sequences of $(Mo_{2/3}RE_{1/3})_2C$ results in a (*C*2/*c*) structure with ABAB stacking, and a (*Cmcm*) structure with ACAC stacking.

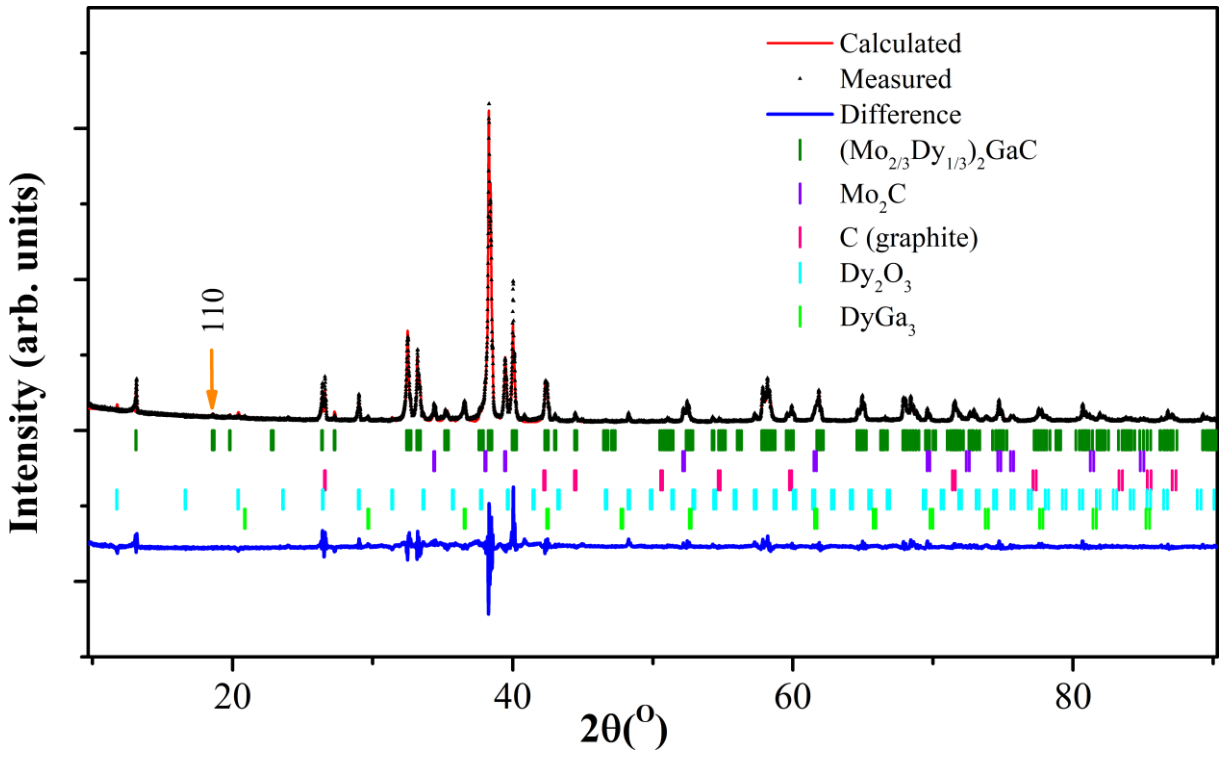
a


Calculated
Measured
Difference
(Mo2/3Gd1/3)2GaC
Mo2C
C (graphite)
Gd2O3
GdGa2
110
Intensity (arb. units)
20
40
60
80
2θ(O)

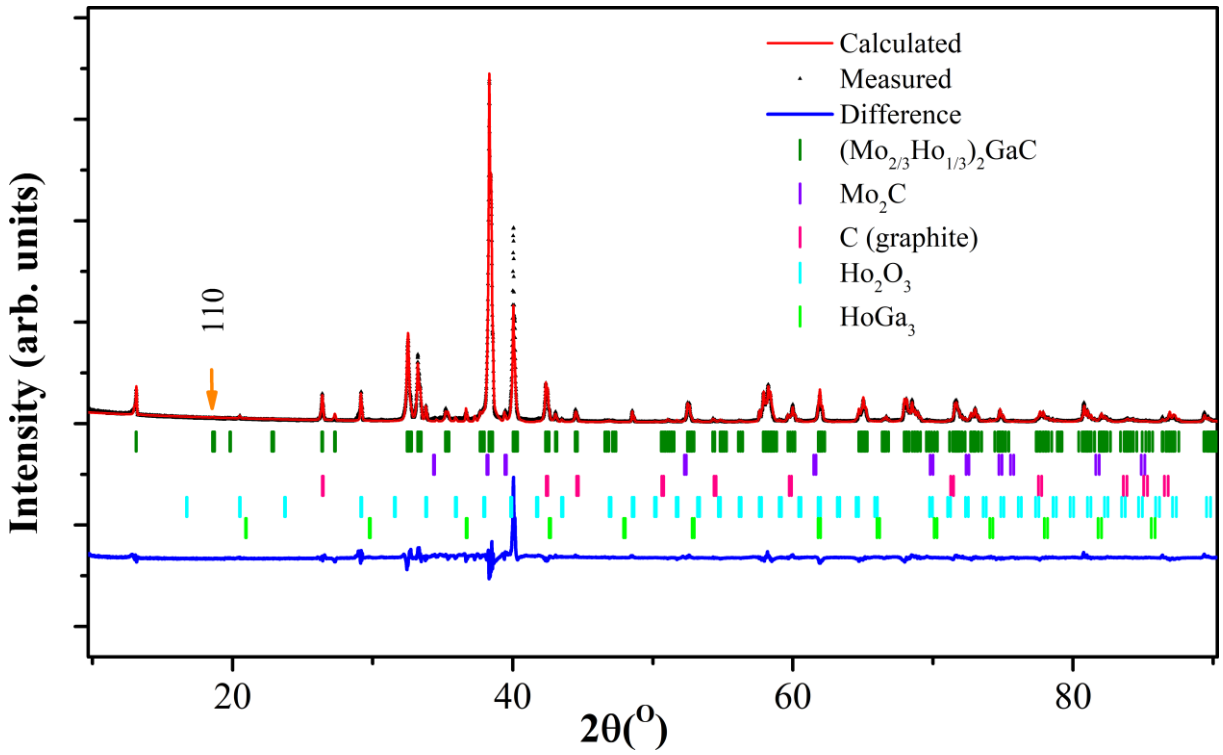
b


Calculated
Measured
Difference
(Mo2/3Tb1/3)2GaC
Mo2C
C (graphite)
Tb2O3
TbGa3
110
Intensity (arb. units)
20
40
60
80
2θ(O)

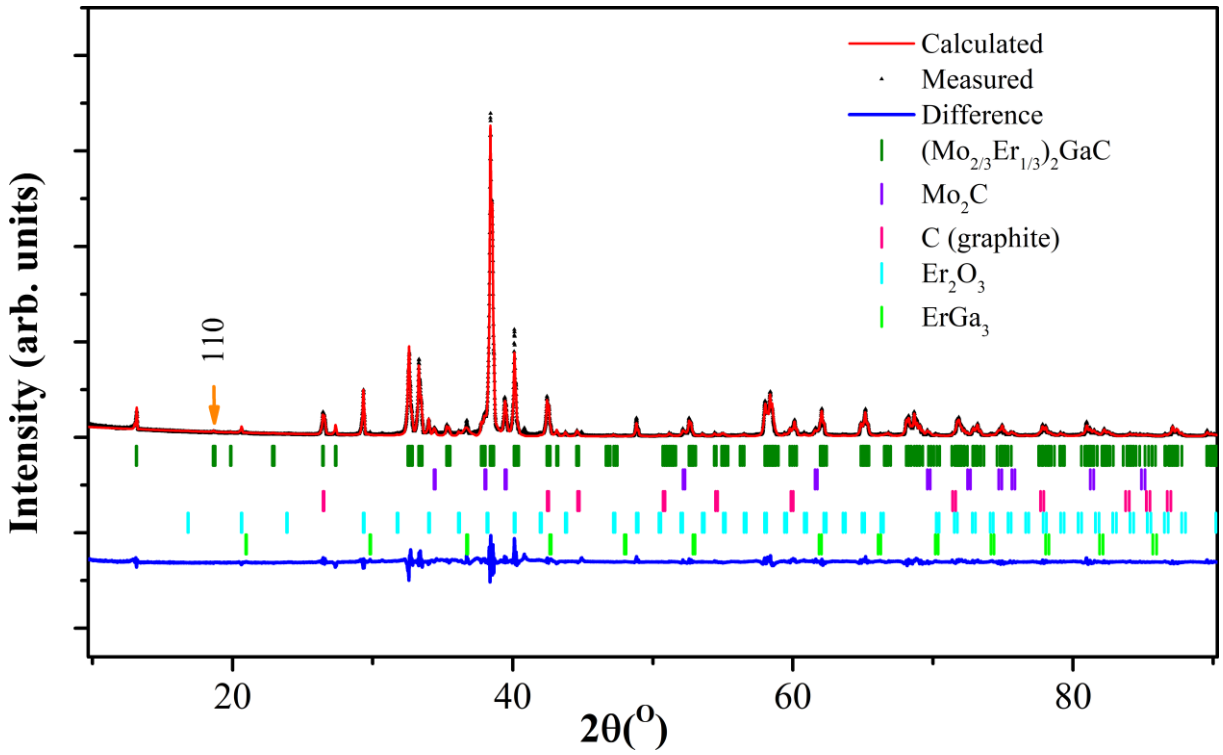
c


Calculated
Measured
Difference
(Mo2/3Dy1/3)2GaC
Mo2C
C (graphite)
Dy2O3
DyGa3
110
Intensity (arb. units)
20
40
60
80
2θ(O)

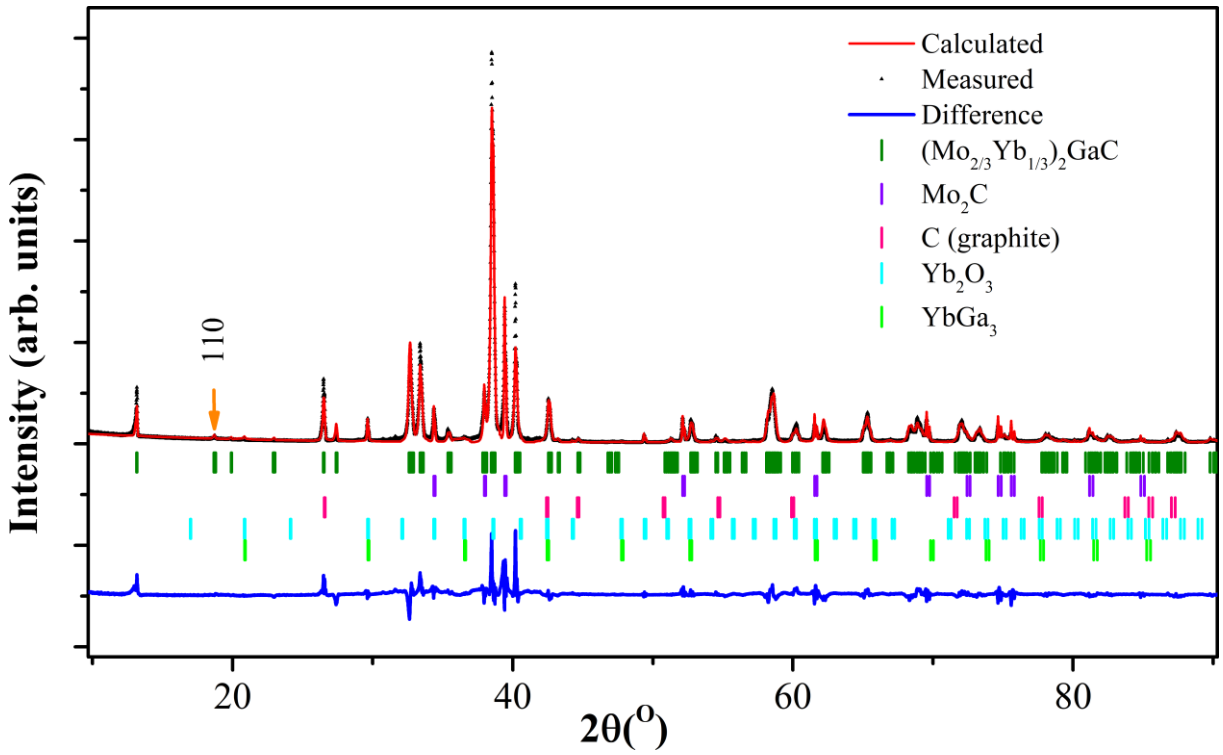
d


Calculated
Measured
Difference
(Mo2/3Ho1/3)2GaC
Mo2C
C (graphite)
Ho2O3
HoGa3
110
Intensity (arb. units)
20
40
60
80
2θ(O)

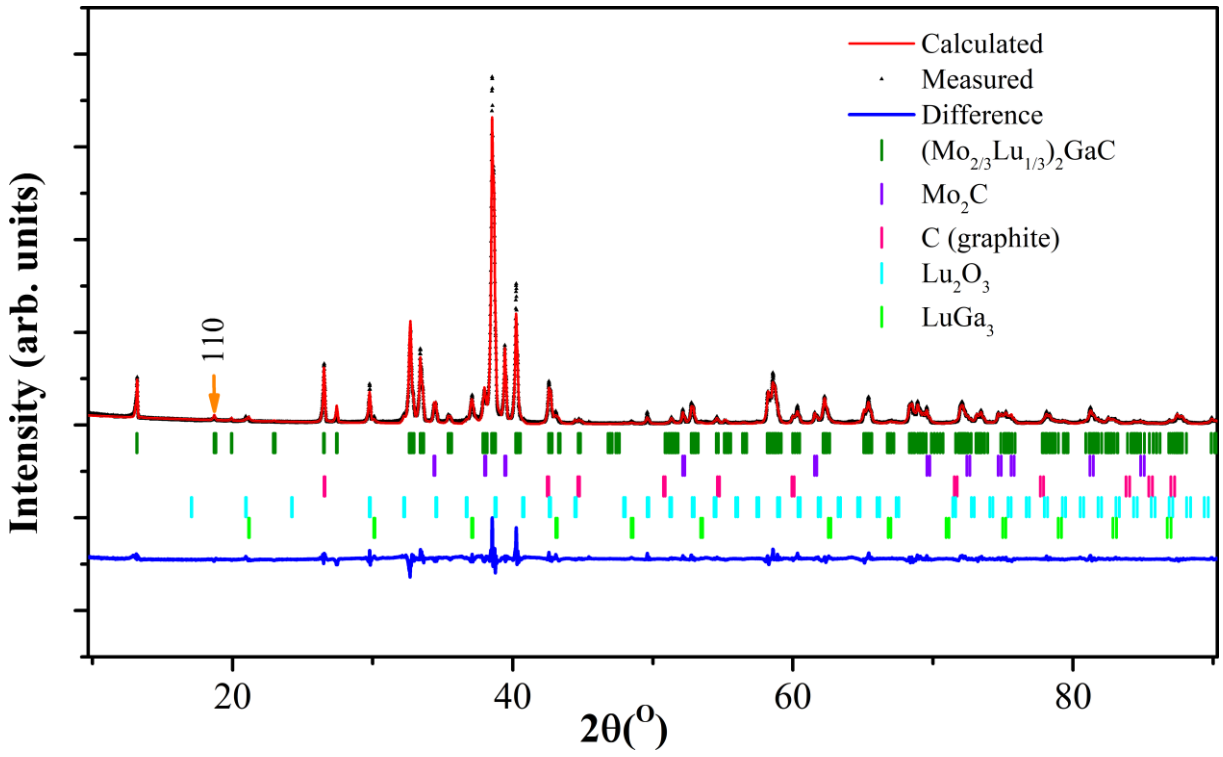
e


Calculated
Measured
Difference
(Mo2/3Er1/3)2GaC
Mo2C
C (graphite)
Er2O3
ErGa3
110
Intensity (arb. units)
20
40
60
80
2θ(O)

f


Calculated
Measured
Difference
(Mo2/3Yb1/3)2GaC
Mo2C
C (graphite)
Yb2O3
YbGa3
110
Intensity (arb. units)
20
40
60
80
2θ(O)

g

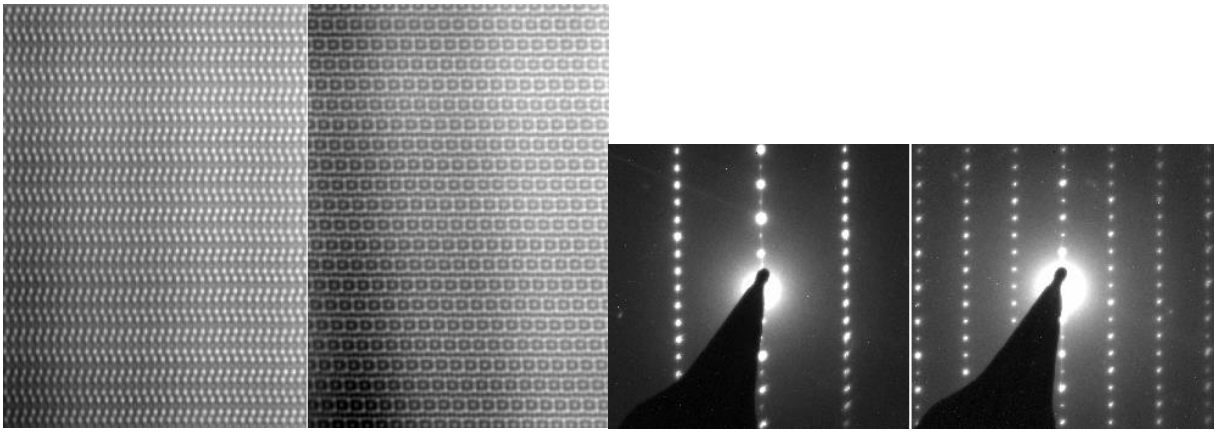


Supplementary figure S2 a-g. X-ray diffraction and Rietveld refinement (calculated) for $(Mo_{2/3}RE_{1/3})_2GaC$, where RE is a) Gd, b) Tb, c) Dy, d) Ho, e) Er, f) Yb, and g) Lu.

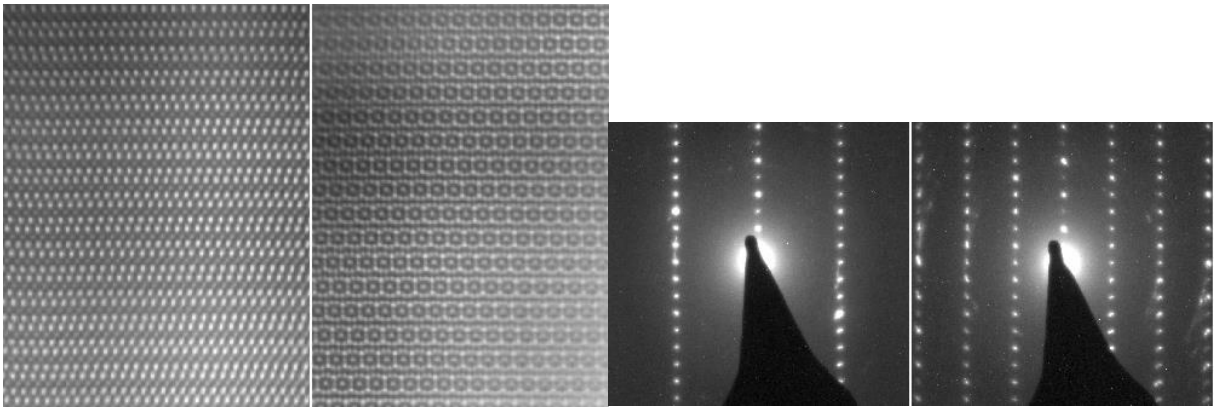
Supplementary table S1. Goodness of fit parameters for the refinement of different samples. χ^2 is the goodness of fit, and RF-factor for the *i*-MAX phase in *Cmcm* symmetry.

The somewhat high χ^2 values and thus RF-factors for some of the samples are most likely related to strong preferred orientation in the sample, arising from large grain size of the powder prepared for the XRD measurement. This preferred orientation is not optimally described in the refinement using Fullprof suite.

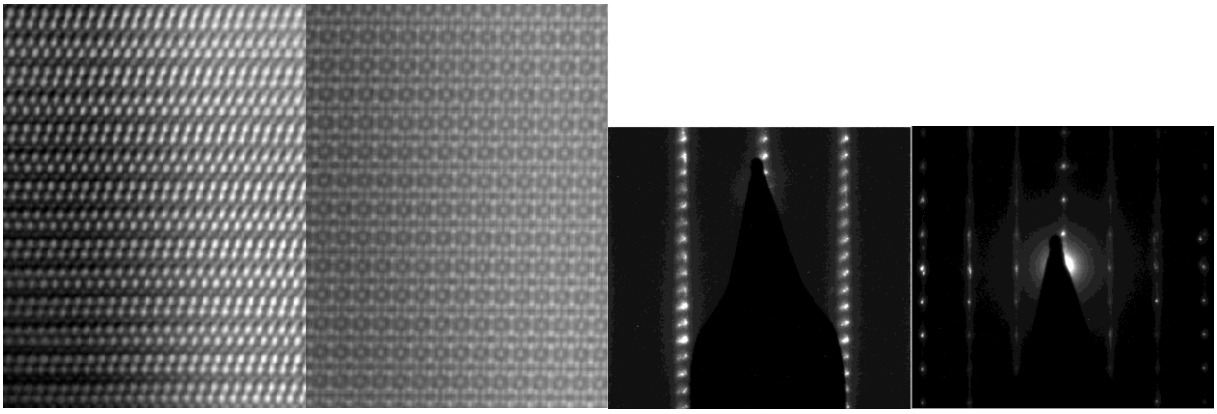
RE element	**χ^2**	**RF-factor (*i*-MAX phase)**
Gd	15.0	-
Tb	11.2	10.3
Dy	10.9	18.1
Ho	20.7	9.2
Er	18.8	8.4
Yb	38.8	37.2
Lu	16.3	6.9



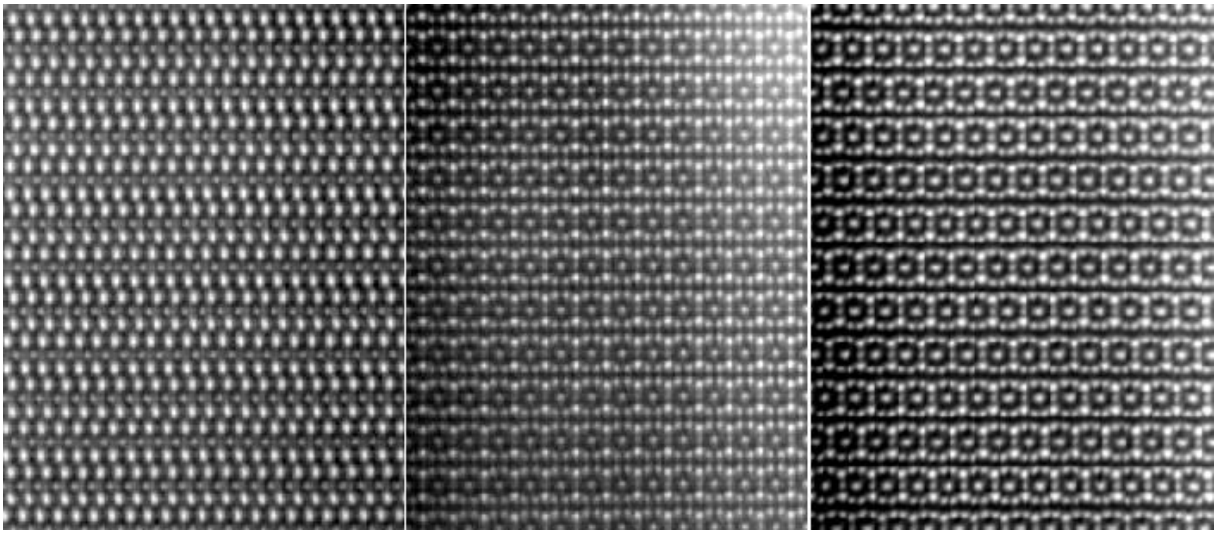
Supplementary figure S3. STEM and SAED of $(Mo_{2/3}Gd_{1/3})_2GaC$. Shown zone axis are [100] and [110] for both STEM and SAED.



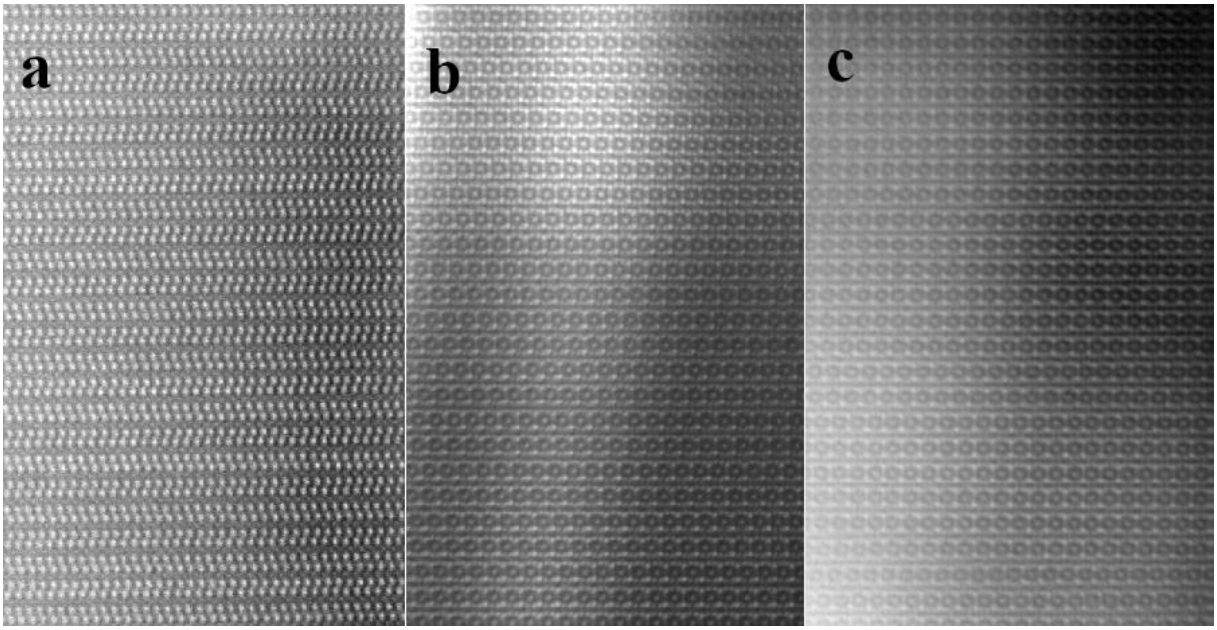
Supplementary figure S4. STEM and SAED of $(Mo_{2/3}Tb_{1/3})_2GaC$. Shown zone axis are [100] and [110] for both STEM and SAED.



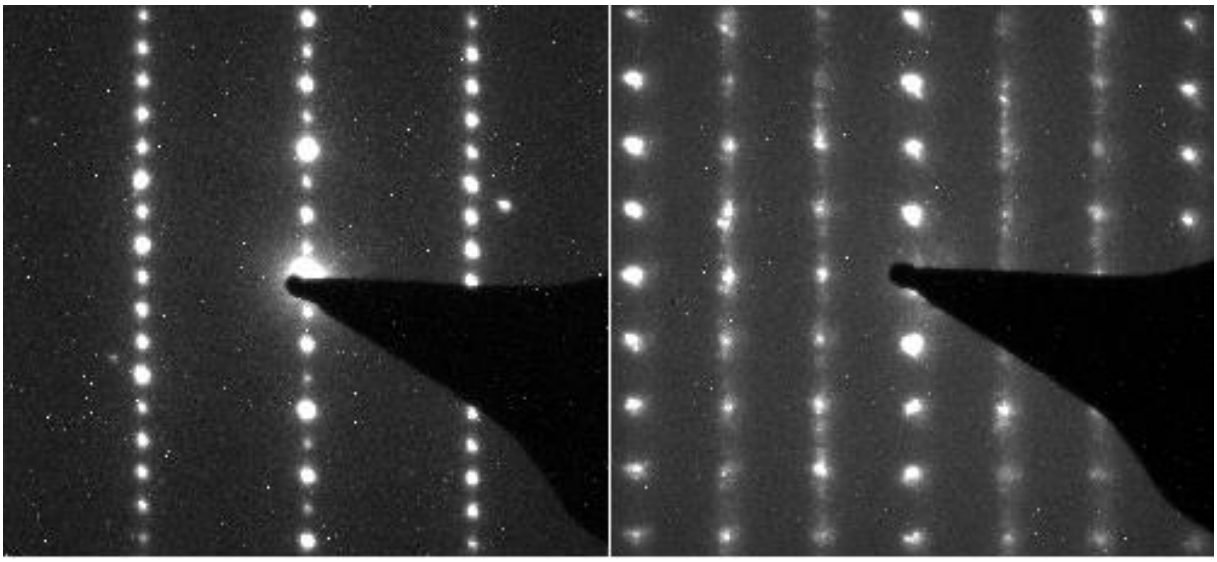
Supplementary figure S5. STEM and SAED of $(Mo_{2/3}Dy_{1/3})_2GaC$. Shown zone axis are [100] and [010] for both STEM and SAED.



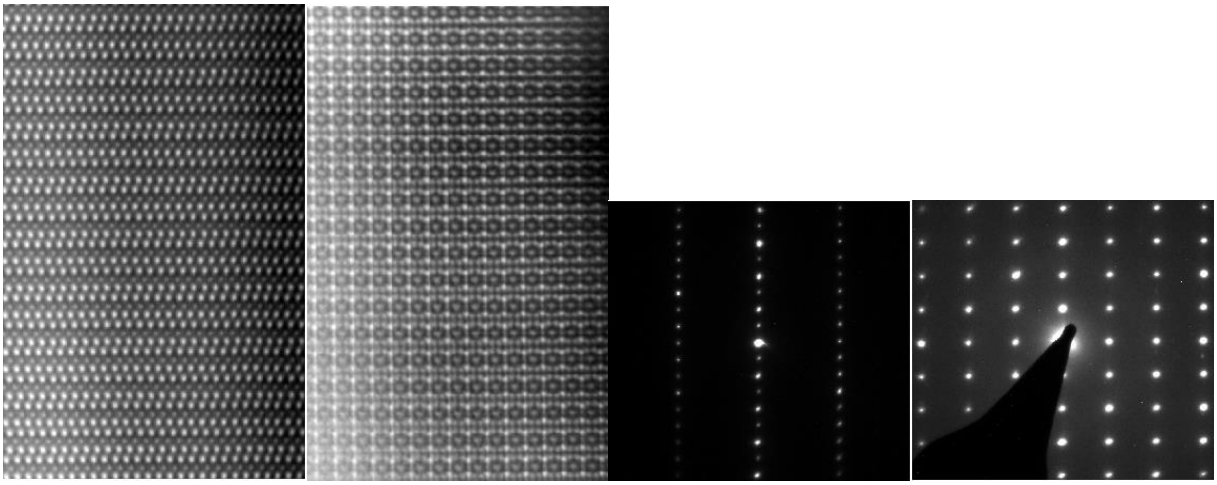
Supplementary figure S6. STEM of $(Mo_{2/3}Ho_{1/3})_2GaC$. Shown zone axis are [100], [010] and [110].



Supplementary figure S7. STEM of $(Mo_{2/3}Er_{1/3})_2GaC$. Shown zone axis are [100] in a) and [010] in both b) and c). It should be noted that both (*Cmcm*) and (*C*2/*c*) crystal structure are seen in panels b) and c).



Supplementary figure S8. SAED of $(Mo_{2/3}Er_{1/3})_2GaC$. Shown zone axis are [100] and [010], showing both crystal structures – (*Cmcm*) and (*C*2/*c*).



Supplementary figure S9. STEM and SAED of $(Mo_{2/3}Lu_{1/3})_2GaC$. Shown zone axis are [100] and [010] for both STEM and SAED.

Supplementary table S2. Summary of the relative metal content of targeted $(Mo_{2/3}RE_{1/3})_2GaC$ grains, obtained from SEM-EDX analysis. These values are in good agreement with the *i*-MAX composition, within experimental error bars of about ±2 at. % for EDX analysis.

RE element	**Relative elemental composition, at. %**		
	Mo	**RE**	**Ga**
Gd	43	23	34
Tb	45	22	33
Dy	45	21	34
Ho	46	22	32
Er	44	23	33
Tm	44	22	34
Yb	45	22	33
Lu	43	23	34