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Between carbide and nitride MAX phases: Sol-gel assisted synthesis and characterization of the carbonitride phase $Cr_2GaC_{1-x}N_x$

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Structural data of the precursor material

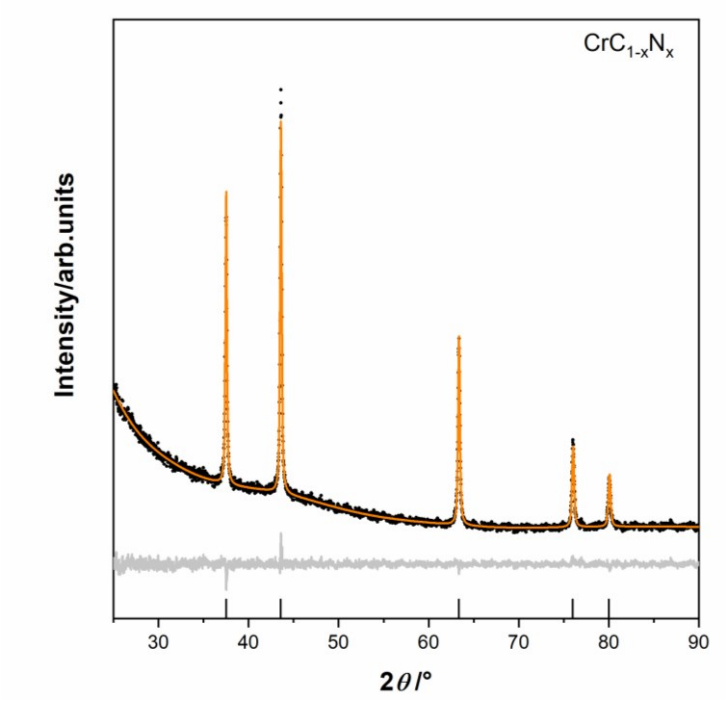

Figure SI-1: Rietveld refinement (orange line), residuum curve (grey) of the X-ray powder diffraction data of the precursor chromium carbonitride (black) based on the structural model of CrN.[1]

Table SI-1: Results of the Rietveld refinement of the $CrC_{1-x}N_x$ precursor.

Phase name	$CrC_{1-x}N_x$
Percentage/weight-%	100
Spacegroup	*Fm-3m*
Lattice parameters *a*/ Å	4.1476(9)
Cell volume/ Å^3	71.35(5)
Crystallite size (Lorentz)/nm	71(5)
Background order	15
R_p	2.93
R_{wp}	3.72
R_{exp}	2.97
GOF	1.25

Experimental details

Table SI-2: Weighing amounts of the prepared MAX phase samples. All amounts are calculated based on the nominal compositions of 0.5 g of the target material.

Phase	*m*(Cr) in g	*m*(Cr_2N) in g	*m*($CrC_{1-x}N_x$) in g	*m*(Ga) in g	*m*(C) in g
Cr_2GaC	0.2800 (1.0 eq.)	-	-	0.1877 g (1.0 eq.)	0.0291 (0.9 eq.)
Cr_2GaN	-	0.3068 (1.0 eq)	-	0.1857 g (1.0 eq.)	-
$Cr_2GaC_{1-x}N_x$	0.1385 (1.0 eq.)	-	0.1716 (1.0 eq.)	0.1857 (1.0 eq.)	-

Table SI-3: Temperature programs of the target materials, heat treated either using the microwave oven (watts and minutes) or the vertical tube furnace (temperature and hours).

Nominal composition	Temperature programs
Cr_2GaC	1300 W, 20 min, 7 g carbon
$Cr_2GaC_{1-x}N_x$	800 W, 30 min, 7 g carbon
Cr_2GaN[157]	740 °C, 5 h, 5 °C/min

DFT calculations

Table SI-4: Results of the ab initio calculation including theoretical lattice constants (*a,c*), the magnetic moment per unit cell and the Fermi energy for $Cr_2GaC_{1-x}N_x$ ($0 \leq x \leq 1$).

$Cr_2GaC_{1-x}N_x$	Lattice constant *a* (Å)	Lattice constant *c* (Å)	Magnetic moment (μB/unit cell)	Fermi energy (eV)
x = 0.0	2.871	12.510	0	7.994
x = 0.1	2.873	12.497	0	7.998
x = 0.2	2.874	12.481	0	8.016
x = 0.3	2.875	12.467	0	8.027
x = 0.4	2.877	12.453	0	8.042
x = 0.5	2.878	12.440	0.001	8.058
x = 0.6	2.881	12.423	0.048	8.060
x = 0.7	2.891	12.394	1.778	8.007
x = 0.8	2.926	12.271	3.653	7.852
x = 0.9	2.928	12.238	3.759	7.851
x = 1.0	2.933	12.196	3.786	7.855

Laboratory-based SXPS details

The laboratory-based SXPS measurements were performed on a Thermo Scientific K-Alpha XPS system. The system employs a 1.4867 keV photon energy (Al Kα) and operates under a base pressure of $2x10^{-8}$ mbar. It is equipped with a 180° double-focusing hemispherical analyser, charge compensating flood gun and 128-channel detector. Survey and core level spectra were collected at pass energies of 200 and 20 eV, respectively. All processing of the data was conducted using the Thermo Scientific Avantage Data System (v5.9925).

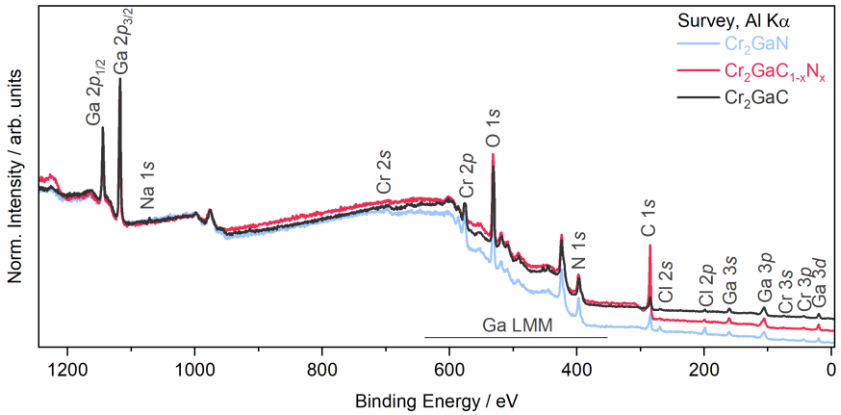

Figure SI-2: Survey spectra collected for the three MAX phase samples with laboratory-based SXPS ($h\nu$ = Al Kα). The spectra are normalized to the maximum height of the Ga $2p_{3/2}$ peak and are offset vertically for clarity.

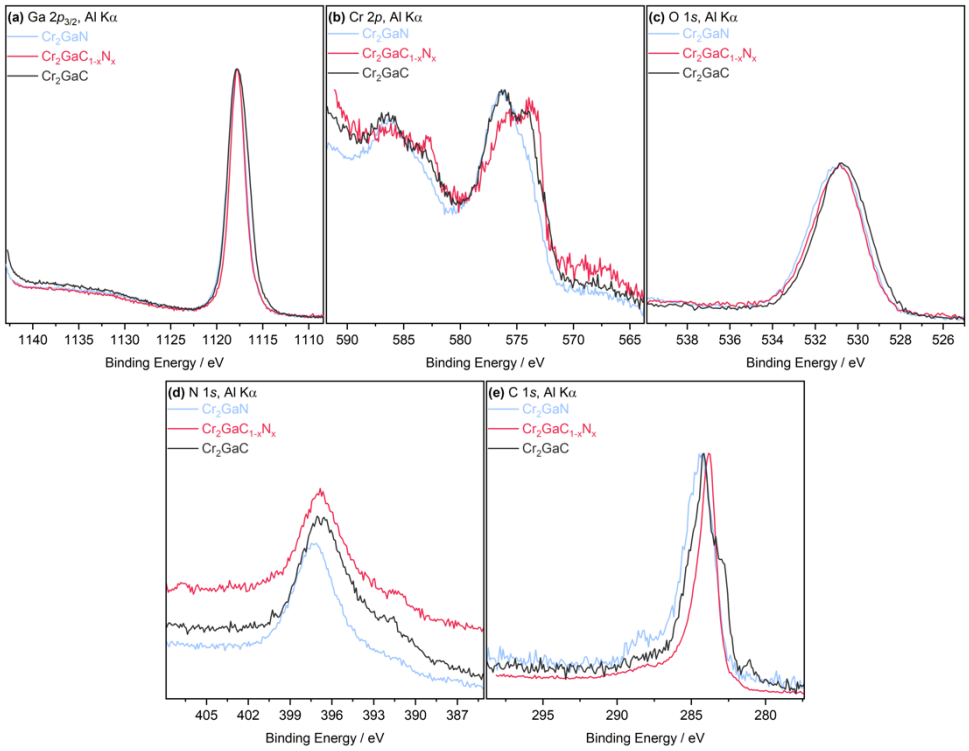

Figure SI-3: SXPS ($h\nu$ = Al Kα) core level spectra collected for the three MAX phase samples including the (a) Ga $2p_{3/2}$, (b) Cr $2p$, (c) O $1s$, (d) N $1s$ and (e) C $1s$ spectra. All spectra except for the N $1s$ are normalized to their maximum height. The binding energy scale was aligned to the lowest Ga $2p_{3/2}$ main peak position as no valence band spectra were collected.

SXPS/HAXPES survey spectra

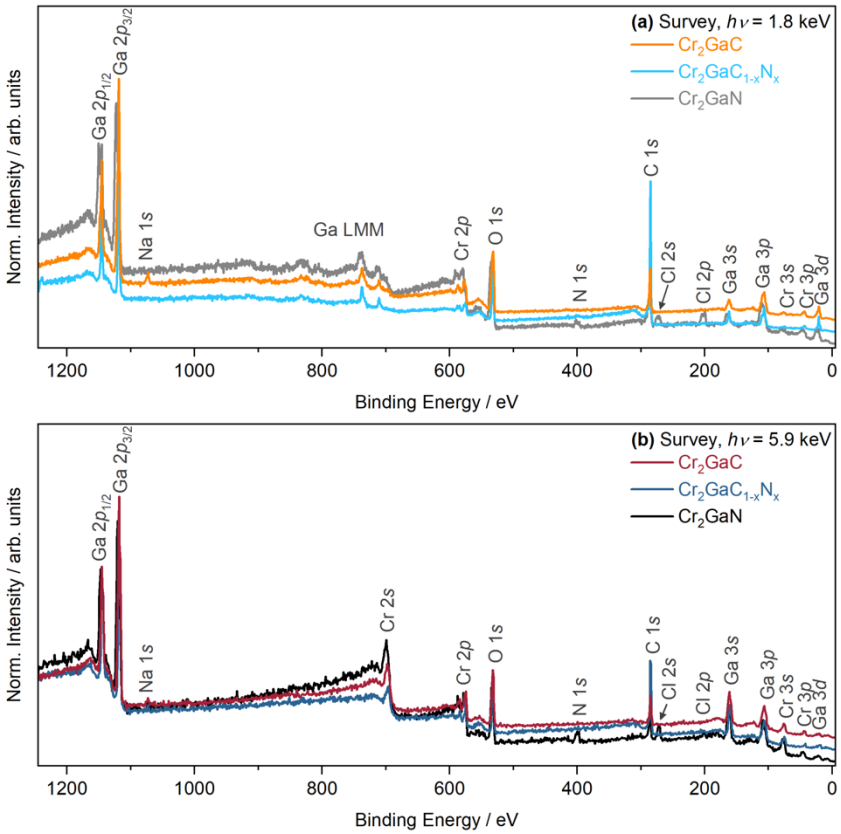

Figure SI-4: Survey spectra collected for the three MAX phase samples with synchrotron-based (a) SXPS ($h\nu$ = 1.8 keV) and (b) HAXPES ($h\nu$ = 5.9 keV). The spectra are normalized to the maximum height of the Ga $2p_{3/2}$ peak and are offset vertically for clarity.

Additional SXPS/HAXPES spectra

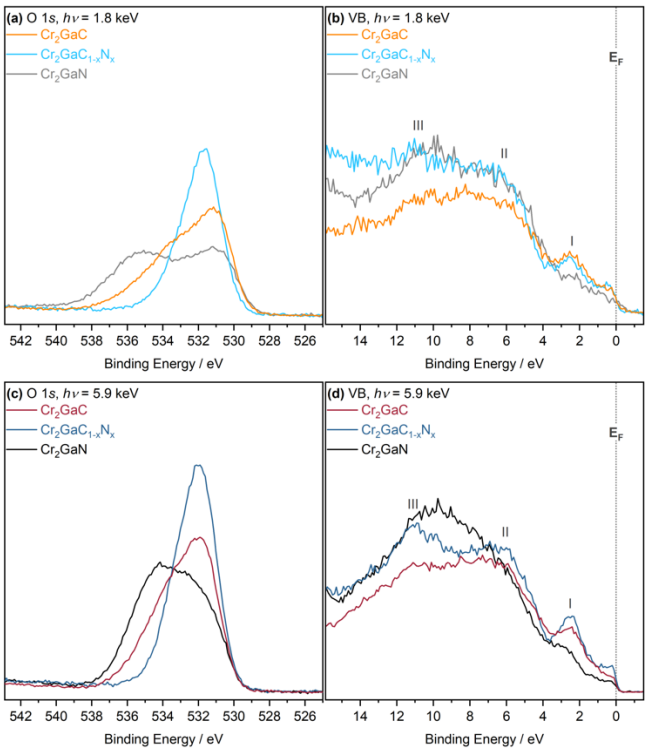

Figure SI-5: Additional SXPS ($h\nu$ = 1.8 keV) and HAXPES ($h\nu$ = 5.9 keV) spectra collected for the three carbonitride MAX phase samples including the (a/c) O 1s and (b/d) valence band (VB) spectra. The spectra are normalized to the total spectral area of the Ga $2p_{3/2}$ core level and the binding energy scale is calibrated to the intrinsic Fermi energy (E_F) of the sample.

Resistivity measurements

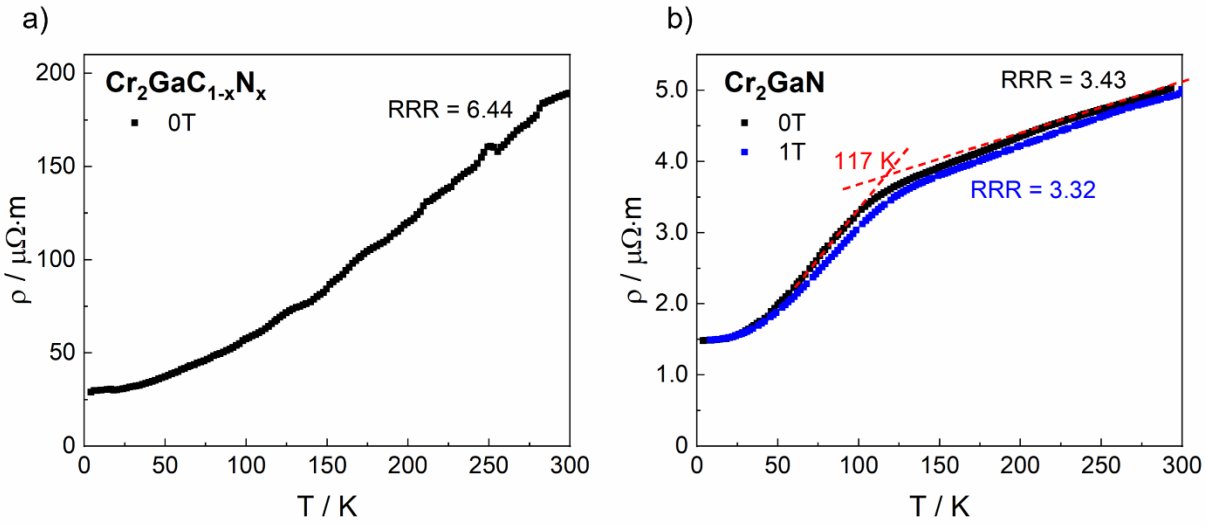

Figure SI-6: Temperature dependence of the electrical resistivity of the carbonitride $Cr_2GaC_{1-x}N_x$ in zero magnetic field (a) and the nitride Cr_2GaN (b) which was measured at both, zero magnetic field and a field of B = 1 T.

References:

(1) Blix, R. Röntgenanalyse Des Chrom—Stickstoffsystems Nebst Einer Orientierenden Konstitutionsuntersuchung Des Stickstoffhaltigen Ferrochroms. *Zeitschrift für Phys. Chemie* **1929**, *3B* (1), 229–239. https://doi.org/https://doi.org/10.1515/zpch-1929-0317.