Supporting Information

Phase Stability of Nanolaminated Epitaxial $(Cr_{1-x}Fe_x)_2AlC$ MAX Phase Thin Films on MgO(111) and Al_2O_3(0001) for Use as Conductive Coatings

Hanna Pazniak[,§], Marc Stevens[§], Martin Dahlqvist[#], Benjamin Zingsem[§,⁄⁄], Lidia Kibkalo[⁄⁄], Merve Felek[§], Sergey Varnakov[⊥], Michael Farle[§,⊥], Johanna Rosen[#], Ulf Wiedwald[*,§]*

[§]Faculty of Physics and Center for Nanointegration (CENIDE), University of Duisburg-Essen, 47057 Duisburg, Germany

[#]Materials Design, Department of Physics, Chemistry, and Biology (IFM), Linköping University, SE-581 83 Linköping, Sweden

[⁄⁄]Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons (ER-C), Forschungszentrum Jülich, 52425 Jülich, Germany

[⊥]Kirensky Institute of Physics, Federal Research Center KSC SB RAS, Krasnoyarsk, 660036, Russian Federation

Corresponding authors email: hanna.pazniak@uni-due.de; ulf.wiedwald@uni-due.de

Table S1. Identified equilibrium simplex, i.e. set of most competing phases for $(Cr_{1-x}Fe_x)_2AlC$.

x	Equilibrium simplex
0.0 (Cr_2AlC)	Cr_2AlC (in-AFM1)
0.125	Cr_2AlC, Fe_3AlC, C, Fe_5Al_8
0.25	Cr_2AlC, Fe_3AlC, C, Fe_5Al_8
0.375	Cr_2AlC, Fe_3AlC, C, Fe_5Al_8
0.5	Cr_2AlC, Fe_3AlC, C, Fe_5Al_8
0.625	Cr_2AlC, Fe_3AlC, C, Fe_5Al_8
0.75	Fe_3AlC, C, Cr_2AlC, Fe_5Al_8
0.875	Fe_3AlC, C, Cr_2AlC, Fe_5Al_8
1.0 (Fe_2AlC)	Fe_3AlC, C, Fe_5Al_8

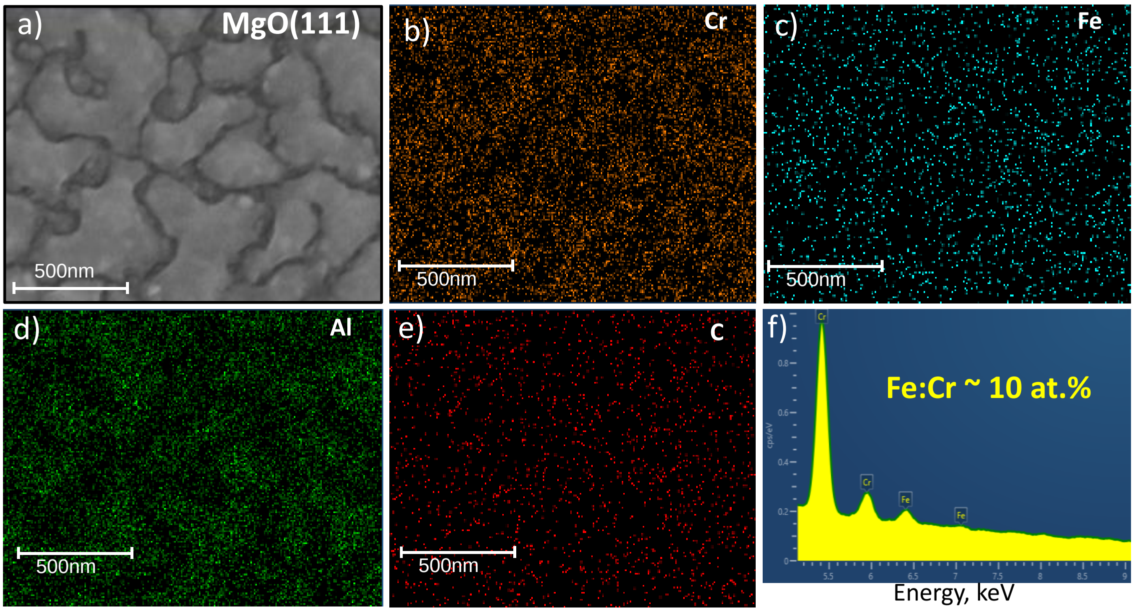

Figure S1. SEM image of the $(Cr_{0.9}Fe_{0.1})_2AlC$ film (a) and corresponding EDX elemental maps of Cr (b), Fe (c), Al (d), and C (e). (f) EDS spectrum of studied region with calculated Fe:Cr ratio.

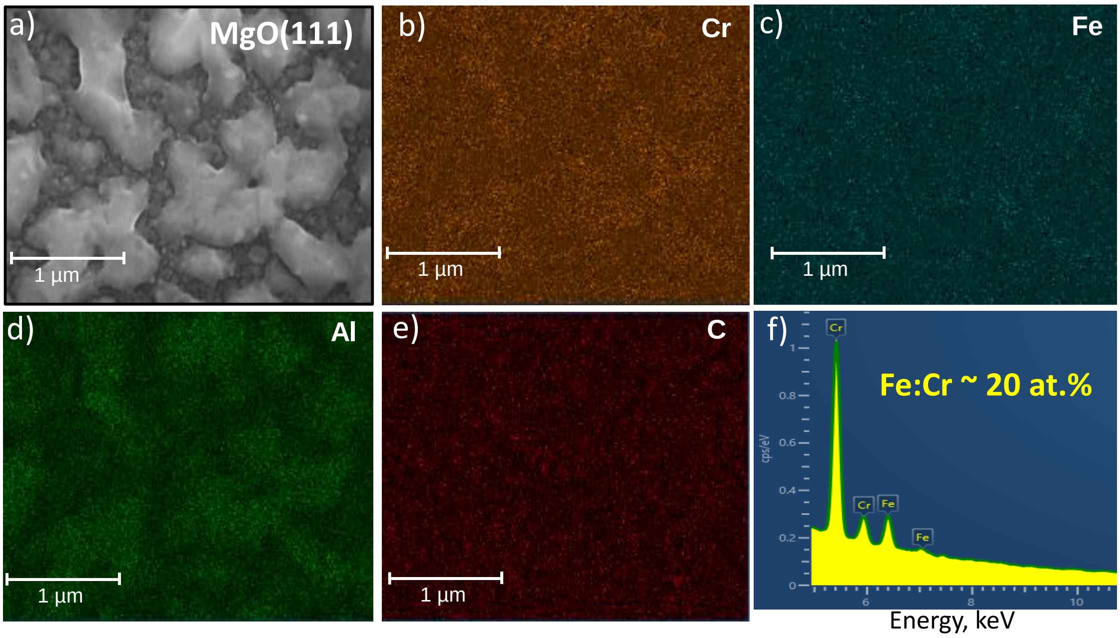

Figure S2. SEM image of the $(Cr_{0.8}Fe_{0.2})_2AlC$ film (a) and corresponding EDX elemental maps of Cr (b), Fe (c), Al (d), and C (e). (f) EDS spectrum of studied region with calculated Fe:Cr ratio.

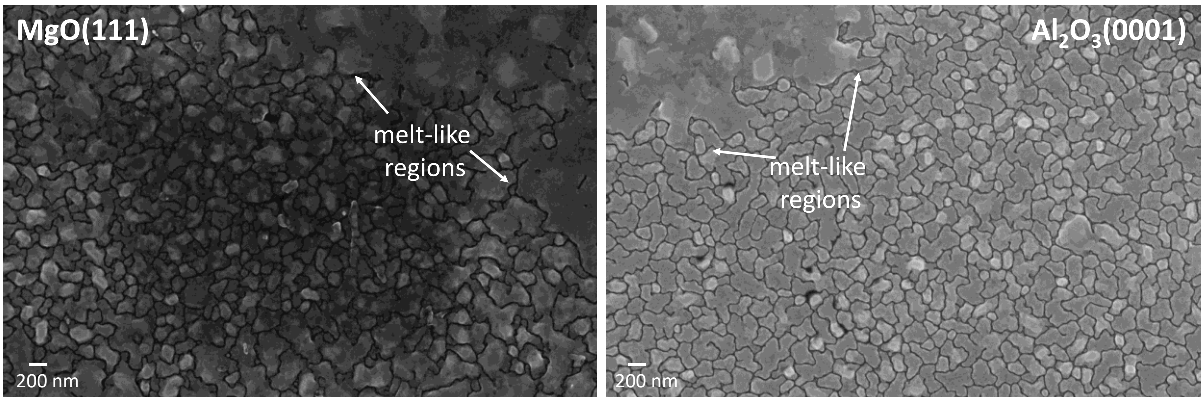

Figure S3. SEM images of the $(Cr_{0.8}Fe_{0.2})_2AlC$ film demonstrating the presence of melt-like regions on MgO(111) and $Al_2O_3(0001)$ substrates.

Table S2. Fe content, at.%, and (Cr+Fe)/Al ratio in $(Cr_{0.9}Fe_{0.1})_2AlC$

	Fe, at%	**(Cr+Fe)/Al ratio**
MAX structure	0.88-3.4 (2.8 average)	1.93-2.13
Intermetallic region	2.61-5.95	1.10-1.34

Magnetometry

Since the signal of the 40 nm $(Cr_{0.9}Fe_{0.1})_2AlC$ on $Al_2O_3(0001)$ film is expected to be small, the substrate signal and eventually a small sample holder signal shall be considered. The best choice for magnetometry is the $Al_2O_3(0001)$ substrate due to the small amount of impurity atoms. Figure S4 (a) presents the raw data for a 2.5x2.5x0.5 mm^3 $Al_2O_3(0001)$ substrate piece after annealing at 600°C for 2h in UHV mimicking the film deposition by PLD. As expected, the magnetic response is diamagnetic and temperature independent. Around zero field, a small jump is obtained which originates from the sample holder or the $Al_2O_3(0001)$ substrate. This is more obvious in Figure S4 (b) after subtraction of the diamagnetic slope. Below fields of about 3 kOe a tiny hysteresis loop appears which has saturation values of $8 \cdot 10^{-6}$ emu and $5 \cdot 10^{-6}$ emu for T = 5 K and T = 300 K, respectively. Importantly, the diamagnetic high-field susceptibility is a constant $\chi^{HF} = 59 \cdot 10^{-6}$ emu mol^{-1} Oe^{-1} as shown in Figure S4 (c). We would like to emphasize that the paramagnetic impurities in MgO(111) single crystals make the analysis of VSM data extremely difficult [S1] and often the data is not conclusive for this substrate. Thus, we restrict ourselves to the $Al_2O_3(0001)$ substrate.

The magnetic response of the 40 nm $(Cr_{0.9}Fe_{0.1})_2AlC$ on $Al_2O_3(0001)$ is displayed in Figure S4 (d). While significant noise is present for larger fields in the hysteresis loop at T = 5 K, the room temperature data appears at high quality and after subtraction of the diamagnetic slope a S-shaped hysteresis is obtained as shown in Figure S4 (e). However, the overall sample magnetic moment is only $1 \cdot 10^{-5}$ emu and thus only a factor of 2 above the bare substrate and holder signal in Figure S4 (b). Ascribing this add-on signal to the film, the saturation magnetization of $(Cr_{0.9}Fe_{0.1})_2AlC$ is about 20 emu cm^{-3}. This value is about 15 times lower as compared with epitaxial $(Cr_{0.8}Mn_{0.2})_2AlC$ MAX phase thin film (330 emu cm^3) [S2]. The magnification around zero field in Figure S4 (f) shows a slightly open hysteresis loops which is temperature independent and likely originates from the holder.

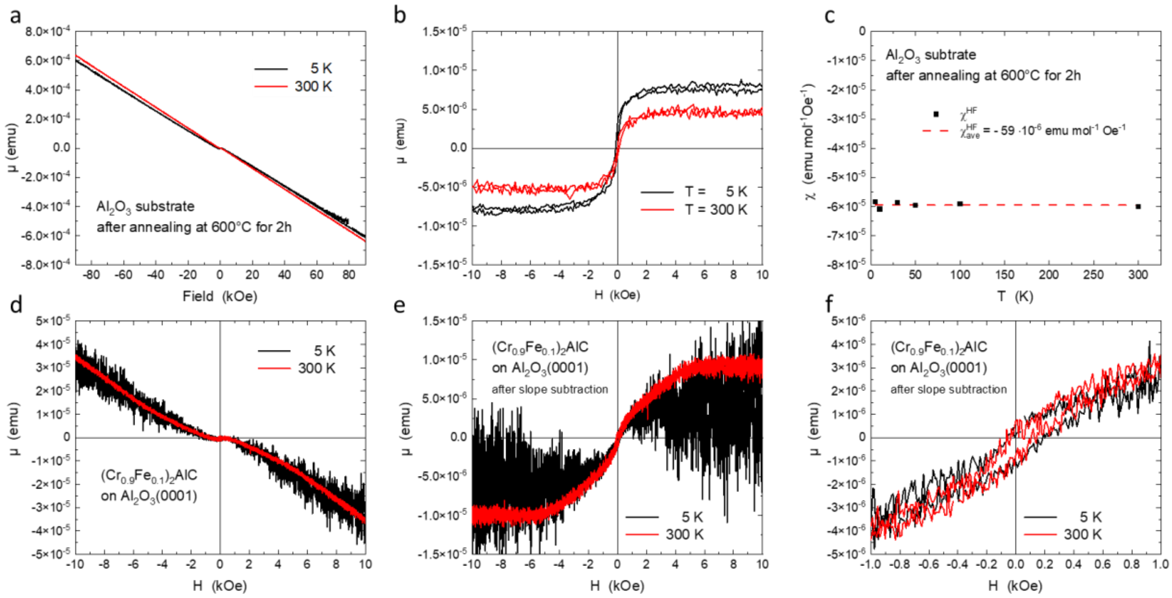

Figure S4. Vibrating Sample Magnetometry of a 2.5x2.5x0.5 mm^3 Al_2O_3(0001) substrate after annealing at 600°C for 2h in the PLD deposition chamber mimicking the deposition process (a-c). The Al_2O_3(0001) substrate shows the expected diamagnetic slope with a small jump around zero field. After slope subtraction, a small ferromagnetic signal remains (b). The high field susceptibility $\chi^{HF} = 59 \cdot 10^{-6}$ emu mol^{-1} Oe^{-1} of the Al_2O_3(0001) substrate is constant (c). Field dependence of the total sample magnetic moment μ of the 40 nm $(Cr_{0.9}Fe_{0.1})_2AlC$ on Al_2O_3(0001) at 5 K and 300 K (d) and after subtraction of the diamagnetic slope (e). Panel (f) shows a magnification around the origin.

References:

S1. Novoselova, I.; Petruhins, A.; Wiedwald, U.; Weller, D.; Rosen, J.; Farle, M.; Salikhov, R. Long-term stability and thickness dependence of magnetism in thin $(Cr_{0.5}Mn_{0.5})_2GaC$ MAX phase films. *Materials Research Letters* **2019**, *7*, 159–163, DOI:10.1080/21663831.2019.1570980.

S2. Mockute, A.; Persson, P. O. Å.; Magnus, F.; Ingason, A. S.; Olafsson, S.; Hultman, L.; Rosen, J. Synthesis and characterization of arc deposited magnetic $(Cr,Mn)_2AlC$ MAX phase films. *physica status solidi (RRL) – Rapid Research Letters* **2014,** *8* (5), 420-423, DOI: 10.1002/pssr.201409087.