

## TEM/SAED Study of $\text{Mn}_2\text{CrGaAl}$ and $\text{CoFeCrGe}$ Heusler Alloys

X.-Z. Li<sup>1</sup>, W.-Y. Zhang<sup>2</sup>, Y.-L. Jin<sup>2</sup> and D.J. Sellmyer<sup>1,2</sup>

<sup>1</sup> Nebraska Center for Materials and Nanoscience, University of Nebraska, Lincoln, NE.

<sup>2</sup> Department of Physics and Astronomy, University of Nebraska, Lincoln, NE.

Heusler compounds have attracted much attention recently because of their potential application for spintronic devices. Furthermore, some of them are candidate materials as rare-earth-free hard magnets [1]. Full-Heusler alloys usually crystallize in the  $\text{L}_{21}$  structure and have a stoichiometric composition of  $\text{X}_2\text{YZ}$ , where X and Y are transition-metal elements, and Z is a group III, IV or V element. The structure may degrade into the B2 structure or further to the A2 (bcc) structure due to chemical disorder. Some of the Heusler compounds may crystallize in a tetragonal structure. Quaternary Heusler compounds with elemental composition  $\text{XX}'\text{YZ}$  also exist basically as the derivatives of  $\text{X}_2\text{YZ}$  compounds with one of the X atoms being replaced by another transition-metal element.

Heusler alloys with nominal compositions of  $\text{Mn}_2\text{CrGa}_{1-x}\text{Al}_x$  ( $x = 0.0, 0.2, 0.5, 1.0$ ) and an equiatomic  $\text{CoFeCrGe}$  were arc melted from high-purity (99.95%) elements in an argon atmosphere and then rapidly quenched ribbons were made by ejecting the molten alloy in a quartz tube onto the surface of a copper wheel. TEM/SAED experiments were carried out on JEOL JEM2010 and Thermo Fisher Scientific (formerly FEI) Tecnai Osiris microscopes and analyzed using Landyne software suite [2].

$\text{Mn}_2\text{CrGa}_{1-x}\text{Al}_x$  ( $x = 0.0, 0.2, 0.5, 1.0$ ) ribbons were further annealed at 500 °C for 2 hours to achieve the equilibrium state. The XRD analysis reveals that a disordered cubic phase is present in the alloys with  $x = 0.0$  and 1.0 while a spinodal decomposition with phase separation has been observed in the alloys with  $x = 0.2$  and 0.5. Detailed structural investigation of the alloys with  $x = 0.2$  and 0.5 has been further carried out using TEM/SAED techniques [3]. TEM study confirms a segregation of two crystalline phases, one of them is a cubic phase with the  $\beta$ -Mn prototype structure and the other is a new crystalline phase with a tetragonal structure with lattice parameters,  $a = 0.293$  nm and  $c = 0.874$  nm. The orientation relationship of the two crystalline structures has been determined. The structural model of the tetragonal phase is proposed and confirmed in SAED and HRTEM studies.

Cubic  $\text{L}_{21}$  structure with partial disorder was found in the as-spun  $\text{CoFeCrGe}$  ribbons and those annealed at 300 °C. Phase decomposition was observed when the samples were annealed above 402 °C. TEM/SAED study revealed three compounds in the samples annealed at 500 °C for 2 hours. The primary compound is the  $\text{L}_{21}$ -type structure with partial disorder. The secondary compound has a cubic  $\text{Cr}_3\text{Ge}$  structure with  $a = 0.461$  nm. The tertiary compound is a new tetragonal structure with lattice parameters  $a = 0.76$  nm and  $c = 0.284$  nm. The difference in the magnetic hysteresis loops of the samples annealed at 300 °C and 500 °C has been interpreted as the appearance of the new tetragonal crystalline phase [4].

### References:

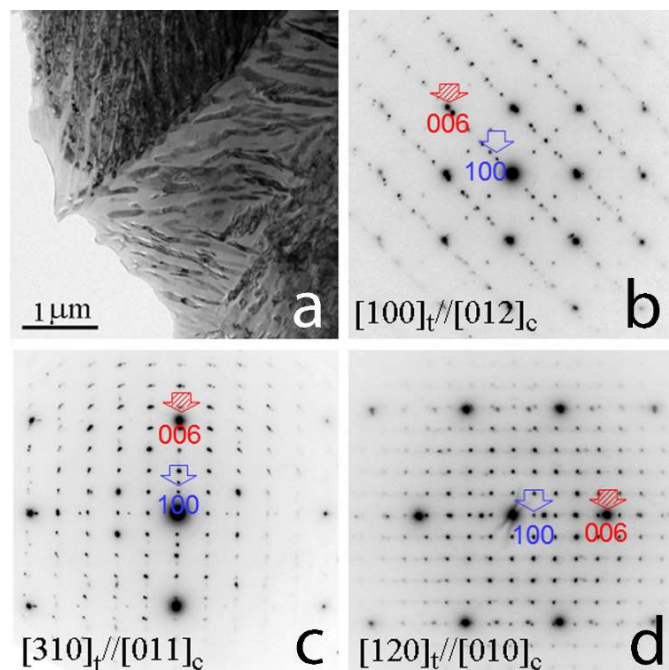
[1] C Felser *et al*, AIP Mater. **3** (2015), p. 041518.

[2] <https://www.unl.edu/ncmn-cfem/xzli/computer-programs>.

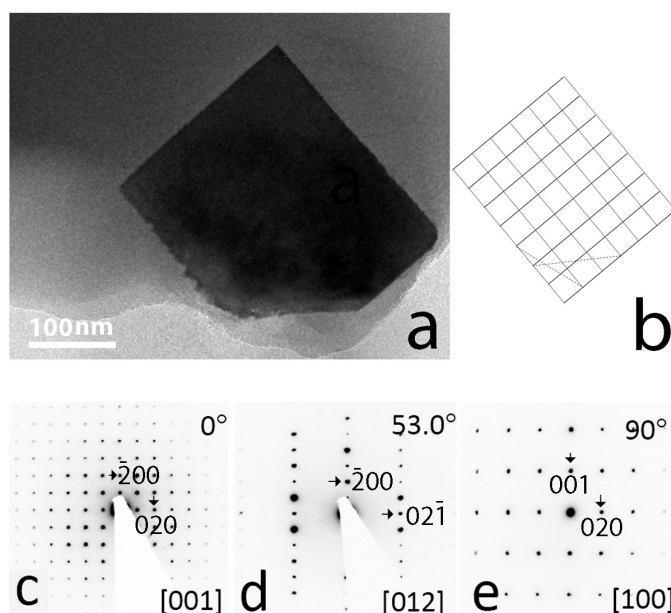
[3] X-Z Li, W-Y Zhang, DJ Sellmyer, Acta Mater. **140** (2017), p.188.

[4] The research was performed in part in the Nebraska Nanoscale Facility: National Nanotechnology

Coordinated Infrastructure and the Nebraska Center for Materials and Nanoscience, which are supported by the National Science Foundation under Award ECCS: 1542182, and the Nebraska Research Initiative. This work is partially supported by NSF-DMREF: SusChEM (Award Number: 1436385).



**Figure 1.** (a) TEM image of the phase segregation in  $\text{Mn}_2\text{CrGa}_{0.5}\text{Al}_{0.5}$  alloy, (b-d) the composite SAED patterns of the Cr-rich cubic phase and a new tetragonal phase.



**Figure 2.** (a) TEM image of the new tetragonal phase in the  $\text{FeCoCrGe}$  alloy, (b) a schematic drawing of the grain shape, (c-e) SAED patterns of the new tetragonal phase.