

## TEM/SAED Study of Mn<sub>2</sub>CrGaAl and CoFeCrGe Heusler Alloys

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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 L<sub>21</sub> structure and have a stoichiometric composition of X<sub>2</sub>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 XX'YZ also exist basically as the derivatives of X<sub>2</sub>YZ compounds with one of the X atoms being replaced by another transition-metal element.

Heusler alloys with nominal compositions of Mn<sub>2</sub>CrGa<sub>1-x</sub>Al<sub>x</sub> (x = 0.0, 0.2, 0.5, 1.0) and an equiatomic 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].

Mn<sub>2</sub>CrGa<sub>1-x</sub>Al<sub>x</sub> (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 β-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 L<sub>21</sub> structure with partial disorder was found in the as-spun 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 L<sub>21</sub>-type structure with partial disorder. The secondary compound has a cubic Cr<sub>3</sub>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:

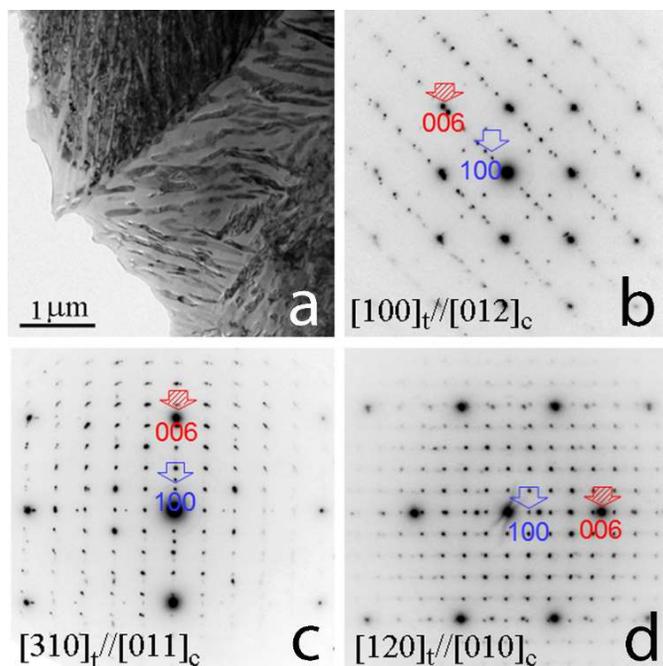
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[2] <https://www.unl.edu/ncmn-cfem/xzli/computer-programs>.

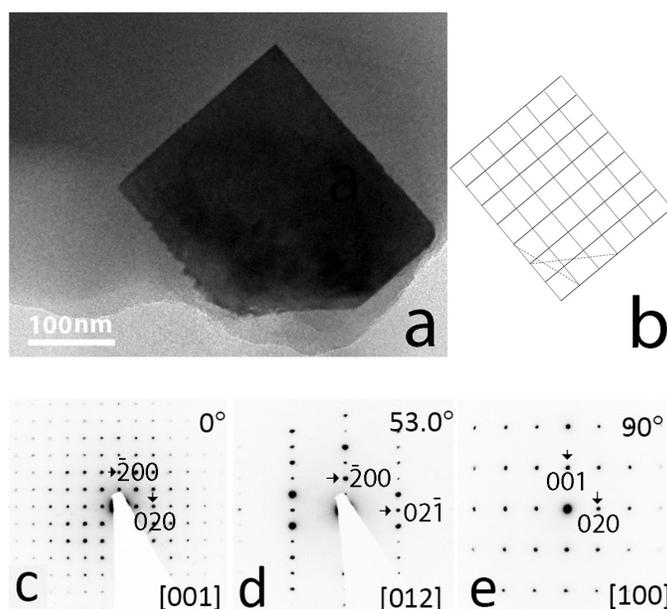
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[4] The research was performed in part in the Nebraska Nanoscale Facility: National Nanotechnology

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.