A.9: Presence of atomic disorder and its effect on the magnetic and electronic properties of NiCrGa half Heusler alloy

Half-metallic (HM) half Heusler alloys (HHAs),with 100% spin polarized electrons at the Fermi level (E_F), are efficient spin injector materials for applications in spin-polarized electronics. Although in literature, many HHAs are theoretically calculated to be HM in the ordered cubic $C1_b$ structure, the measured spin polarization (SP) in most of these alloys is found to be significantly lower. In most of the synthesized alloys, the structural disorder is reported to be the main reason for the reduced SP. NiCrGa is one such HHA which is predicted to be a HM ferro-magnet with lattice parameter of 5.51 Å, a total magnetic moment of 1 μ_B/f.u. and HM gap of 0.17 eV. Since the material was not synthesized before, our motivation was to synthesize bulk polycrystalline sample and probe its calculated HM property through the experimental study of structural, magnetic and electronic properties.

After melting the sample in arc-melting furnace and subsequent annealing (facility present in SUS), the material was found to possess a significant atomic disorder, as observed in other HHAs also. The measured crystal structure (Figure A.9.1) is different from the ordered cubic $C1_b$ structure, with larger lattice parameter of 5.8 Å. The magnetic ordering, as shown in Figure A.9.2 is observed to be anti-ferromagnetic at very low temperature (μ_{eff}=0.04 μ_B/f.u. and θ=-7.1 K) and paramagnetic at room temperature, which is significantly different from the calculated ferromagnetic property.

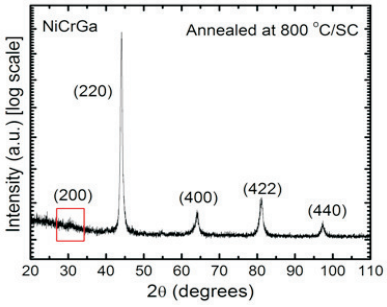

Fig. A.9.1: Room temperature XRD data of NiCrGa sample. The superlattice reflection peak (200) is highlighted in the figure.

To identify the nature of atomic disorder in the material and its effect on the HM property, we have carried out theoretical calculations using Spin Polarized-Relativistic Korringa-Kohn-Rostoker (SPR-KKR) method, by taking into account several types of disorders in the structure. Through systematic comparison of the measured and calculated XRD patterns and the magnetic moments, we could able to identify two most probable types of disorders (types 1 and 3).

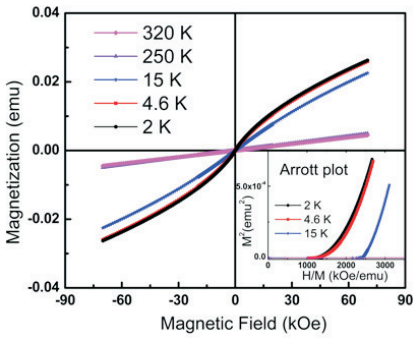

Fig. A.9.2:The isothermal M versus H curves at different temperatures. The inset shows the Arrott plots.

In the type-1 disordered case, there is an intermixing of Cr and Ga atoms and the vacant site is partially occupied by the Ni atom, which results in a lower value of total moment of 0.02 μ_B/f.u. Further, in type-3,the vacant site is occupied by Cr and Ga atoms, resulting a total moment of 0.32 μ_B/f.u. Since the calculated moments for both the disordered structures are comparable with the measured value, it was difficult to identify the type of disorder in the sample at this stage. Thus, the measured valence band (VB) photoelectron spectrum (PES) was compared with the calculated VB for types 1 and 3 disorders (Figure A.9.3) and type-3 matches quite well.

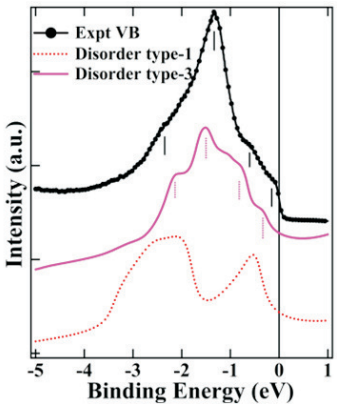

Fig. A.9.3: Comparison of experimental VB spectrum with the calculated total DOS for the type-1 and type-3 disorders.

In conclusion, from detailed experimental and theoretical studies, the possible structural disorder has been identified. For the type-3 disorder, the calculated SP at E_F is only 1%. Hence this disorder significantly disturbed the half metallic property, which eventually limits its potential for device applications. More details can be found in *J. Magn. Magn. Mater. 475, 675-682 (2019).*

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