ABSTRACT

The multifunctionality such as half-metallicity, spin-gapless semiconducting property, topological insulating property, thermoelectrics and magnetocaloric effect of Heusler compounds originates from their highly correlated 3d electrons. For exploiting the multifunctionality of these compounds in device applications, a detailed understanding of the underlying physics of each function, particularly in the context of the electronic band structure is essential. Any type of atomic disorder in their crystal structure is one of the main hindrances in any application. This thesis presents a comparative study on the physical properties of the disordered Co_2FeAl (CFA) and Fe_2CoAl (FCA) Heusler nanoalloys via theory and experiment. The structural characterizations reveal that both CFA and FCA nanoalloys are crystallized in A2-disordered phase. Magnetization data divulge that they are soft ferromagnets with high saturation magnetization (M_s) and Curie temperature (T_c). It is demonstrated that the saturation magnetic moment and Curie temperature can be controlled by the shape and size of the nanoparticles. It is found that the origin of higher saturation magnetic moment is related to the disorderness of the Heusler nanoalloys. From the measurement of isothermal magnetizations, the high entropy change ($-\nabla S_M$) is observed near the magneto-structural phase transition (MST) temperature under a moderate magnetic field. The Arrott plots of CFA and FCA nanoalloys confirm the first-order nature of magnetostructural transition (MST). A comprehensive density functional theory (DFT) study has been carried out on both systems, to understand any correlation between structural disorder and their magnetic properties. From the theoretical findings, it is predicted that the half-metallic CFA alloy is energetically stable in its XA (inverse) phase but not in $L2_1$ (conventional) phase, while its total magnetic moment is consistent with the Slater Pauling rule in all phases. On the other hand, FCA alloy is found to be a metal and the half-metallicity is not restored in its any hypothetical ($L2_1$ and XA) structures. The pronounced deviation from the Slater Pauling value and integer magnetic moment is found to be the reason for the loss of half-metallicity in FCA alloy. Such various fascinating characteristics of Cobalt and Iron based Heusler nanoalloys are depicted in this thesis.

Keywords: Heusler nanoalloys, Saturation magnetization, Curie temperature, magnetocaloric effect, electronic band structure, Slater-Pauling behavior, Half-metallicity, exchange and correlation.