ABSTRACT

RFe_2 (R = Rare earth) compounds have been known for their multifunctional applications for more than three decades. $CeFe_2$ is an unusual member of the RFe_2 family in the sense that it has a relatively low paramagnetic to ferromagnetic Curie temperature ($T_{C1} \sim 230$ K), an anomalously low lattice constant (7.3 Å), and a relatively small magnetic moment ($\sim 2.3\ \mu_B/f.u.$). Strong hybridization between Ce f and Fe d states has been reported to be the cause of these anomalies. A very small amount (~ 5 %) of certain impurities in $CeFe_2$ is known to cause a loss of ferromagnetism, at a temperature T_{C2} below T_{C1}, to an antiferromagnetic state via a first-order transition. Thus, $CeFe_2$ lies on the verge of a ferromagnetic instability. However, the proposed mechanisms for what drives this instability are still speculative. One such speculation is the impurity induced modification of the f-d hybridization. This thesis aims specifically at investigating the applicability of this very hybridization-based mechanism for the ferromagnetic instability. For the investigations, the following two-fold approach has been adopted: (i) a computational study of a possible systematics of the variation of the f-d hybridization for a number of appropriate impurities spanning the periodic table, and (ii) for a particular impurity, an experimental exploration of the manifestation of a possible connection between the impurity induced changes in the hybridization and the occurrence of the second phase transition. For the first part, elements Ti to Ga from the $3d$ period, Zr to Cd from the $4d$ period, and Hf to Hg from the $5d$ period have been taken as the impurity, and the variation of impurity induced change in the f-d hybridization with the position of the impurity in the periodic table has been monitored. The hybridization strengths were determined semi-quantitatively from the partial electron densities of states of Ce and Fe in all the cases. The hybridization strength is found to decrease on introduction of any impurity in $CeFe_2$. Further, this reduction is more or less successively higher, i.e., the hybridization becomes successively weaker, in going away from the Fe group on either side of the periodic table. Similar variations in local host and impurity moments are also observed. Although no correlation between hybridization and ferromagnetic instability could be derived from this study, a systematics on impurity induced change in hybridization has been inferred. For the second part, the occurrence of the second transition has been explored for Cr, Ag and Au impurities, out of which only Cr is found to induce the second transition. Most importantly, a definite proportionality between the impurity concentration dependence of T_{C2} and the f-d hybridization strength has been revealed by this study. In a supplementary but related experimental study, impurity induced order to disorder phase transition in $Ce(Fe_{1-x}Ni_x)_2$ system has been explored. From this study, the f-d hybridization strength and T_{C1} are found to decrease together with x, and a quantum phase transition is anticipated to take place at $x \sim 0.69$.

Keywords: Laves phase; Ferromagnetic instability; Impurity; Hybridization.