EVOLUTION WITH COMPOSITION OF THE d-BAND DENSITY OF STATES AT THE FERMI LEVEL IN HIGHLY SPIN POLARIZED Co$_{1-x}$Fe$_x$S$_2$

P.L. Kuhns, M.J.R. Hoch, A.P. Reyes, W.G. Moulton (NHMFL); L. Wang, C. Leighton, (University of Minnesota, Chemical Engineering and Materials Science)

Systems that are close to being half-metallic are currently receiving much attention. We have used low field NMR to probe the electronic properties of ferromagnetic (FM) Co$_{1-x}$Fe$_x$S$_2$, recently established as a highly spin polarized (SP) system, in which $E_F$ and the SP properties can be systematically tuned with $x$. It is shown that, contrary to some misconceptions in the literature, nuclear spin-lattice relaxation in highly SP d-band metals provides a novel means for studying the density of states (DOS) at the Fermi level $\rho(E_F)$. $^{59}$Co NMR $\rho$ measurements in FM Co$_{1-x}$Fe$_x$S$_2$ (0 < $x$ < 0.3) made as a function of temperature $T$ have enabled us to follow changes in $\rho(E_F)$ with $x$. In contrast to transport measurements, such as Andréev reflection and tunneling, NMR spin-lattice relaxation times provide access to the thermodynamic DOS that can be directly compared with electronic specific heat capacity determinations and with band structure calculations. Spin-lattice relaxation rates $1/T_1$ are plotted in Fig. 1 as a function of $T$ for samples with various $x$ values. In all cases Korrninga-type behavior is found with $1/T_1 = \kappa T$. Experimental $\kappa$ values are shown versus $x$ in the inset in Fig. 1. Following an initial decrease, $\kappa$ reaches an asymptotic value for $x > 0.1$. For $T > T_c/2$, the relaxation rate increases more rapidly than predicted by the Korringa relation. Other relaxation mechanisms, involving spin wave excitations for example, become important in this range. The quantity $\kappa$ plotted versus $x$ in Fig. 1 (inset) may be expressed as

$$\kappa = 1/T_1 T = C \left[ \rho_3^2 (x, E_F) + \rho_1^2 (x, E_F) \right]$$

with $C = 3.8 \times 10^{-37}$ J$^2$S$^{-1}$K$^{-1}$ the calculated value of the relaxation constant for this electron-nuclear system. If for $x > 0.07$ we assume $\rho_3^2 (E_F) \to 0$, it follows that $\rho_3 (E_F) \to (\kappa / C)^{1/2}$ and values for $\rho_1 (E_F)$ obtained using this result are plotted in Fig. 2. The results are correlated with theoretical band structure calculations and experimental magnetization and transport measurements that support highly SP behavior in the alloyed materials for $x > 0.07$. The present results are consistent with the density of states at $E_F$ for the majority spin sub-band gradually decreasing with $x$ before reaching an approximately constant value at $x = 0.15$. In the range 0 < $x$ < 0.07, our results suggest that the Fermi surface for the minority spin sub-band shrinks towards zero as $x$ is increased. The DOS at the Fermi level results are in good agreement with values obtained from electronic specific heat measurements and show trends similar to those predicted by LSDA band structure calculations.

References