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LOW TEMPERATURE SPECIFIC HEAT AND MAGNETIC PROPERTIES OF Zr(Fe_{1-x}Co_{x})_2

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Abstract.—Specific heat measurements between 1.6 and 10 K of five Zr(Fe,Co)_2 alloys are presented. The results are discussed in connection with the magnetic properties and support the interpretation of the onset of ferromagnetism in terms of weak itinerant electron magnetism.

INTRODUCTION.—Alloys of the solid solution Zr(Fe,Co)_2 crystallize in the cubic MgCu_2 structure. Starting from ZrFe_2 (T_c = 619 K) the magnetic moment and the Curie temperature drops with increasing Co concentration up to x = 0.75, which is the critical concentration for ferromagnetism. The breakdown of magnetic order was discussed by Kanematsu [1] in terms of local environment effects and later [2] in the framework of the Stoner Edwards Wohlfarth (SEW) model [3]. Recently it was shown [4] that from magnetic and resistivity measurements within the restrictions of the SEW model for itinerant electron magnetism an estimation of the density of states at the Fermi level (N(E_F)) can be made from the slope of C/T versus T/ln(T/Theta_C) plots (Arrott plots) [3] and the high temperature deviation of the resistivity from the linear p-T dependence p = p_0 + a_1 T - a_2 T^2 [5] can be correlated with N(E_F) and its derivatives. N(E_F) can be calculated from the experimental data:

\[ N(E_F) = \frac{a_2}{a_1} \frac{6(\pi^2k_B^2N_B^2)}{c} \]  

(1)

with \( N_B \) number of atoms per gram. The aim of this investigation was to compare the estimated N(E_F) values with low temperature specific heat measurements.

RESULTS AND DISCUSSION.—The heat capacity of the unannealed alloys was measured in an automated adiabatic calorimeter using calibrated Ge or Carbon-Glass resistors (heat capacity of sample holder less than 5% of total C_p).

Results of the specific heat measurement are presented in a conventional C/T against T^2 plot (Fig. 1). At low temperatures these curves exhibit a deviation from linearity. The "upturns" of these curves are seen to become more pronounced with higher Co concentration, or become even a maximum for x = 0.9. In view of the paramagnon-cluster controversy [7], [8] for x = 0.9 the upturn of C_p is seen to be the high temperature side of a peak, which excludes the possibility of significant paramagnon contribution. Although for x = 0.8 and x = 0.9 it is found that the upturn of C_p at low temperatures is proportional to 1/T^2, no decision can be made whether this upturn is due to nuclear spin- or Schottky impurity contribution. Especially for binary FeCo alloys it is well known, that the significant upturn of C_p is due to the nuclear specific heat term /9/.
As already mentioned the specially for specific heat measurements prepared 25 g sample were not annealed. Therefore a cluster formation cannot be excluded, but a more detailed analysis of the low temperature upturn requires further experiments. In order to obtain an estimate of the magnetic contribution to \( C_p \), the sample with \( x = 0.72 \) (\( T_c = 30 \) K) was measured up to 70 K: no anomaly was found around \( T_c \). Within the SEW model the magnetic contribution to \( \gamma \) is given by \( \gamma_m = \frac{\chi(2\chi T)^2}{C} \) (for \( x = 0.72 \), \( \gamma_m = 0.15 \) mJ/mole K\(^2\) which is less than 0.3% of measured \( \gamma \)). Additionally it should be noted, that no significant change in the slope of the resistivity versus temperature curve is observed around \( T_c \).

From the concentration dependence of the \( \theta_D \) and the lattice constant (Fig. 2) may be deduced, that the lattice becomes softer with lower Co concentration, while the pressure dependence of \( T_c \) is almost constant in the critical concentration range /11/. From these almost constant \( \frac{3T_c}{\rho_p} \) values it was concluded that the magnetic properties are mainly determined by the lattice parameter, while within the SEW model \( \frac{\Delta T_c}{\rho_p} \) varies as \(-1/T_c\). Tentatively a reason for the constant \( \frac{\Delta T_c}{\rho_p} \) may be that the prediction \( \frac{3T_c}{\rho_p} = -\text{const.} \) is balanced by the more rigid lattice as \( x \rightarrow x_p \). In figure 3 the \( \gamma \) values and the corresponding \( N(E_f) \) values, calculated without any correction due to electron-phonon enhancement or magnetic contribution (\( \gamma_m < 0.3\% \gamma \)), are compared with \( N(E_f) \) values deduced from magnetic and resistivity measurements. The concentration dependence of both \( N(E_f) \) values is similar, however, a significant deviation is found for \( x < 0.66 \). A reason for the significant deviation may be that the system tends to strong ferromagnetism with lower \( x \) values, hence the SEW model is less applicable for \( x < 0.66 \). The susceptibility (determined at 4.2 K and 0.5 T) as a function of \( x \) points also to decreasing \( N(E_f) \) values. Consequently the Stoner enhancement \( S = (1-N(E_f)/I) \) decreases monotonically from \( S = 155 \) to \( S = 37 \) (\( S = 6 \)) for \( x = 0.76 \), \( x = 0.9 \) (\( x = 1.0 \)) respectively.

Although the details of \( C_p \) at low temperatures are unsolved, we conclude from the results in figure 3 that the transition from ferromagnetism to paramagnetism at \( x = 0.75 \) can be interpreted satisfactorily in terms of the SEW model: the paramagnetism for \( x > 0.75 \) is due to the decreasing \( N(E_f) \) values which makes the product \( N(E_f)/I_{\text{eff}} \) smaller than 1.
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