On the magneto structural transition in \( NdCo_2 \)

Swati Pandya, L S Sharath Chandra, P N Vishwakarma and V Ganesan

UGC-DAE Consortium for Scientific Research, Khandwa Road, Indore (MP) 452017, India
E-mail: swatipandya@csr.ernet.in, vganesan@csr.ernet.in

Abstract. \( NdCo_2 \) undergoes a second order paramagnetic to ferromagnetic transition at \( T_C \approx 100 \, K \) accompanied with a cubic to tetragonal transition. It transforms from tetragonal to orthorhombic structure along with a spin orientation when the temperature is decreased below \( T^* \approx 42 \, K \). The heat capacity and resistivity measurements across the magneto structural transition at \( 42 \, K \) in \( NdCo_2 \) are presented. Effect of magnetic field on the transition is discussed in light of magneto elastic coupling. As field increases, the peak in heat capacity across the transition at \( T^* \) shifts to high temperatures with decreased intensity. The peak height vanishes for fields \( \geq 3 \, T \). Apart from this, we also show the validity of Kadowaki-Woods plot for the present system.

1. Introduction

In recent times, systems with magneto-structural transitions are drawing special attention due to their novel physical properties and possible applications in advance technology \cite{1}. Rare earth Laves Phase Compounds like that of \( RC_2 \), show magneto structural transitions, that are quite interesting. These are potential candidates to study different properties like magneto-caloric, magneto-elastic and magneto-electric phenomena. They also show interesting spin fluctuation behavior \cite{2, 3, 5, 6}. A large magneto volume effect, which accompanies the magnetic order of itinerant electrons, is observed in this family \cite{7}. The crystal symmetry is lowered below the Curie temperature \( T_C \), leading to a large anisotropic magneto-elastic interaction \cite{7}. Among \( RC_2 \) compounds, \( NdCo_2 \) and \( HoCo_2 \) show another structural transition at \( T^* \leq T_C \) along with spin reorientation \cite{10, 11}. In \( NdCo_2 \), the easy magnetization axis below \( T_C \approx 100 \, K \) is \( (100) \). It changes to \( (110) \) below \( T^* \approx 42 \, K \). For temperatures \( \geq T_C \), \( NdCo_2 \) has a cubic structure. It changes to tetragonal below \( T_C \) and changes again to orthorhombic below \( T^* \) \cite{2, 7, 8, 9, 10, 11}. Neutron diffraction results show a strong magneto-elastic coupling in this system\cite{8}. Even though, heat capacity in zero field and resistivity in the presence of fields were reported in literature, detailed investigations are still lacking \cite{4, 5}. Hence, we present here, a detailed heat capacity measurements of the low temperature first order magneto-structural transition(\( T^* \)) and resistivity for both the transitions in presence of external magnetic fields to have further insight in this system.

2. Experimental Details

Polycrystalline sample of \( NdCo_2 \) was prepared using Argon arc furnace, with constituents having purity \( \geq 99.9\% \). The sample was re-melted three times for better homogeneity. It was then vacuum-sealed in a quartz tube and annealed at \( 800^\circ \, C \) for fourteen days. The sample was...
characterized to be in single phase with cubic Lave-phase C15 structure by X-Ray Diffraction using CuKα radiation in a Rigaku diffractometer at room temperature. Resistivity and heat capacity measurements were carried out by conventional four-probe method and relaxation method respectively down to 2 K and up to 14 T using Quantum Design Physical Property Measurement System (PPMS).

Figure 1. (a) C/T versus \( T^2 \) in the temperature range 40-51 K and magnetic fields up to 6.5 T. Inset shows low temperature heat capacity in the temperature range 2-55 K and magnetic fields up to 14 T. (b) Heat capacity peak height of the low temperature first order structural transition versus external magnetic fields. Inset shows the area of peak in different magnetic fields.

3. Results and Discussions
Zero field heat capacity \( C \) shows transitions at \( T_C \approx 97 \) K (which is not shown here) and \( T^* \approx 42 \) K. We limit our discussions mainly to the low temperature transition at \( T^* \). Fig.1a shows the \( C/T \) versus \( T^2 \) across the transition at \( T^* \). As the external magnetic field increases the height of the peak decreases and becomes broaden. At 3 T, the peak height vanishes which can be seen in Fig. 1b. We have measured the heat capacity at 6.5 T and 14 T also. Both the curves are almost overlapping. Hence, to throw more light on the effect of magnetic field on this transition, we have calculated the area under the peak in \( C/T \) vs. \( T^2 \) plot (\( A(H) = A(6.5 \) T\)) by considering the heat capacity at 6.5 T as a background. This area decreases as the field increases. The field dependent neutron diffraction (ND) measurement shows that the relative intensity of the reflections for orthorhombic structure at 10 K abruptly drops at \( \approx 0.6 \) T, while it monotonically decreases to zero at 3 T [8]. Our heat capacity data shows a sudden drop in the area for \( H \leq 0.5 \) T which is in agreement with the ND measurement [8]. These observations indicate that relatively a lower field is sufficient to drive the system from orthorhombic to a less distorted tetragonal symmetry. However, a relatively higher field is needed to recover the cubic structure, which implies a weak magnetic exchange striction for orthorhombic \( \text{NdCo}_2 \) and strong magnetic exchange striction for tetragonal \( \text{NdCo}_2 \).

Temperature dependence of resistivity of \( \text{NdCo}_2 \) in zero field as well as in presence of fields up to 14 T are shown in Fig. 2a, which shows two well-separated phase transitions. This is in agreement with our heat capacity data as well as published results [2, 4, 8, 11]. The Curie
transition is clearly visualized in resistivity, but the low temperature structural transition is not that much clear. It shows only a slope change around 42 K. Both the transitions are clearly visible in $d\rho/dT$ (Fig. 2b). The transition temperature, as determined from the maxima of $d\rho/dT$ are $T_C \approx 98$ K and $T^* \approx 42$ K. As shown in Fig. 2, application of fields up to 14 T broadens the high temperature transition ($T_C$) and shifts slightly towards the higher temperature. Kink in resistivity at $T^*$ broadens and slightly shifts towards higher temperature for fields up to 2 T and vanishes at 3 T and above which is in well agreement with heat capacity measurements. We have calculated the $\gamma$ by fitting the heat capacity using $\gamma T + \beta T^3$, in the temperature range 2-30 K. The low temperature resistivity (2-30 K) has been fitted with $\rho_0 + AT^2 + BT^5$, where the phonon contribution is assumed to $(BT^5)$. The value of $B = 1.67 \times 10^{-7} \mu\Omega cmK^{-5}$, which shows that the phonon contribution is negligibly small as compared to other contributions. Conventionally Kadowaki-Woods plot is used to describe the presence of correlations between the coefficient $A$ and $\gamma$ for systems like heavy fermions and spin fluctuating compounds [12, 13]. Such a plot is also valid for our case of $NdCo_2$ which is shown in Fig. 3. It is to be noted that there is no marked difference in the 0 T and 14 T data points. Such a correlation in non-magnetic $RCo_2$ system, has been attributed to spin fluctuations [12]. However, the origin of such correlations in $NdCo_2$ needs further investigation.

4. Conclusion
In essence, the magneto-elastic coupling in $NdCo_2$ is studied through field dependant heat capacity and resistivity measurements. A critical field of 3 T is estimated for the disappearance of the first order magneto-structural transition at $T^* \approx 42$ K. We also establish the presence of correlations between $A$ and $\gamma$ that leads to the Kadowaki-Woods plot in this system.

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Figure 2. (a) Resistivity versus temperature plot for $NdCo_2$ down to 2 K in the presence of magnetic fields upto 14 T. Inset shows the data around 45 K on a large scale. (b) Derivative of Resistivity down to 2 K in the presence of magneti fields upto 14 T.
Figure 3. Kadowaki-Woods plot for NdCo₂ in zero field and 14 T.

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6. References

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