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Spin reorientation phase transitions in RE–Co₅ hexagonal ferromagnets

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Abstract. AC initial susceptibility and static low-field and pulsed high-field magnetisation measurements have been performed on a series of Pr_xNd_{1-x}Co₅ compounds. The results clearly show that the spin reorientation transition temperature from easy plane to easy cone and the low-temperature value of the angle θ between the easy magnetisation direction and the c axis both decrease with increasing Pr concentration. A scaling model of the spin reorientation phase transitions observed in the initial AC susceptibility has been used to obtain the critical exponents of the initial susceptibility. The scaling behaviour of the temperature dependence of θ has also been investigated and the measured critical exponent β is in approximate agreement with a model prediction for systems exhibiting competing axial–planar anisotropies.

1. Introduction

There exist a wealth of data on spin reorientations in rare-earth (RE) intermetallic compounds. The temperature and compositional dependence has been exhaustively documented in the past but the nature of the critical behaviour, if any, of the SR (spin reorientation) phase transition is by no means completely understood. In a SR phase transition, the average magnetisation vector rotates away from an initial easy magnetisation direction (unique axis or plane) to a final fixed easy direction. In a great number of situations the total spin reorientation angle is 90°, but it can also be an incomplete reorientation; giving rise to a conical magnetic structure. The critical behaviour of SR phase transitions in tetragonal Er_xRE_{2-x}Fe₁₄B compounds has been recently investigated by means of low-field AC susceptibility and magnetisation measurements (del Moral *et al* 1989). It was shown that low-field AC magnetic susceptibility measurements are indeed a very sensitive probe of the critical behaviour of the SR phase transition and a simple scaling model of the temperature dependence of the low-field AC susceptibility near the SR was found to be particularly appropriate for these types of compounds.

In order to examine further the nature of the critical behaviour in RE intermetallic compounds, low-field AC susceptibility, and both low- (up to 0.6 T) and high-field (up to 20 T) magnetisation measurements have been extended to Pr_xNd_{1-x}Co₅ compounds. These compounds crystallise in the hexagonal CaCu₅ structure (P₆/mmm) (Wernick

and Geller 1959) and the magnetic properties, in particular the magneto-crystalline anisotropy, of the pure compounds NdCo_5 and PrCo_5 are extremely well documented (Tatsumoto *et al* 1971, Velge and Buschow 1969, Klein *et al* 1975, Algarabel *et al* 1988, Bartholin *et al* 1966, Ermolenko 1980, Ohkoshi *et al* 1976). In NdCo_5 the competition between a large axial anisotropy of the Co sublattice and the planar anisotropy of the Nd^{3+} ion leads to a SR phase transition from an axial easy magnetisation direction above 280 K to a planar one below 235 K. In the temperature interval between the two transitions the easy magnetisation direction takes up a series of values which are strongly temperature dependent. In this case the total SR angle is 90° . In PrCo_5 , however, there occurs a SR phase transition from an initial axial easy magnetisation direction above 107 K to a cone of easy axes persisting down to low temperatures with the final cone angle taking up a value of about 20° from the c axis. Obviously in this case the SR phase transition results in an incomplete reorientation. In a preliminary investigation of the mixed series $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$, measurements of the cone magnetic structure have shown that the SR phase transition temperature from easy axis to easy cone (or plane), and the low-temperature value of the angle θ between the easy magnetisation direction and the hexagonal c axis, both decrease with decreasing Nd concentration (Moze *et al* 1990). Under such circumstances the main mechanism which gives rise to such a complex behaviour is the competition between very large and different anisotropy energies of the Nd, Pr and Co ions.

In this paper are reported low-field AC susceptibility measurements and both low-static-field and high-pulsed-field measurements of the mixed series $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$. A simple scaling model for the low-field susceptibility as applied in the series $(\text{Er}_x\text{RE}_{1-x})_2\text{Fe}_{14}\text{B}$ ($\text{RE} = \text{Dy}, \text{Nd}$) (del Moral *et al* 1989) has been used in this instance to obtain the critical exponents of the SR phase transition for $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$ compounds. The scaling behaviour of the temperature dependence of the easy magnetisation direction has also been investigated and the critical exponent β is in approximate agreement with the prediction of a model of the critical behaviour developed for systems which exhibit axial-planar competing anisotropies (del Moral 1990).

2. Experimental details

Polycrystalline samples of $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$ with $x = 0.8, 0.6, 0.5, 0.4$ and 0.2 were prepared by melting pure elements (Co 4N, RE 4N) in an argon arc furnace under an Ar atmosphere. The resulting buttons were annealed in Ar at 1000°C for six days. Room temperature x-ray powder diffraction patterns of each sample showed that all the observed positions and intensities of the diffraction lines were consistent with the presence of only the CaCu_5 hexagonal structure. The hexagonal unit cell lattice constants were least squares refined by using 12 observed diffraction lines.

The values of the SR temperatures T_{SR} as a function of Pr composition were deduced from the temperature dependence of the low-field AC susceptibility $\chi(T)$. These measurements were performed between 4.2 and 293 K using a modified commercial mutual inductance Hartshorn bridge operating at 15 Hz with AC magnetic fields in the primary coils of approximately 30 mOe peak values. Near T_{SR} , measurements were performed in fine temperature intervals in order to obtain the critical behaviour during the SR phase transition.

The temperature and composition dependence of the angle θ between the easy magnetisation direction and the c axis was determined by two methods. In the first

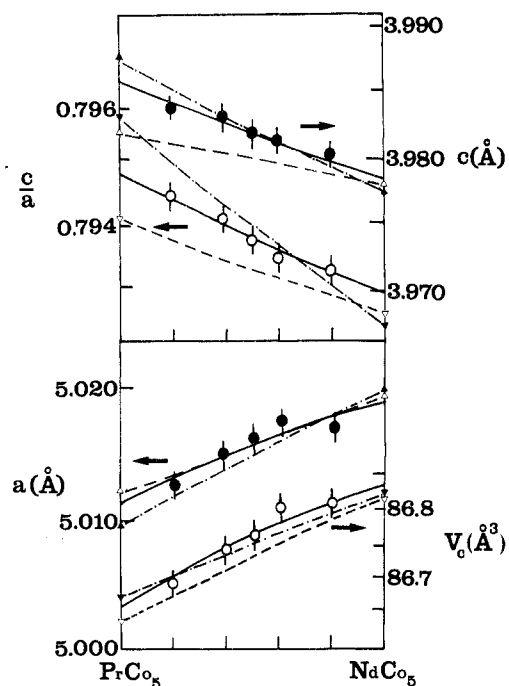


Figure 1. Lattice constants a and c (●), c/a ratio and unit cell volume V_c (○) for the present $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$ compounds. The points for NdCo_5 and PrCo_5 (▽, ▼, △, ▲) are representative points taken from the literature.

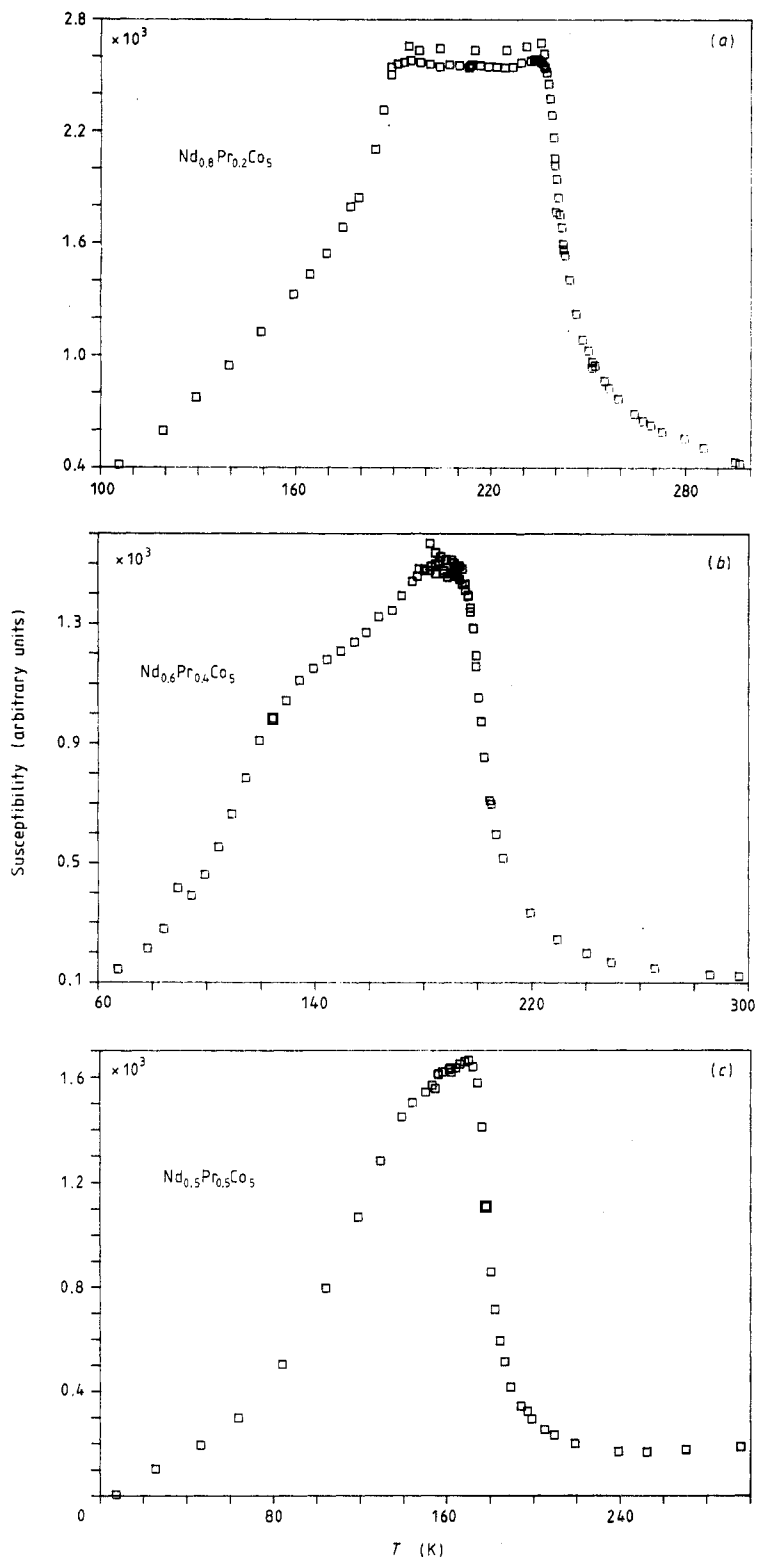
instance, polar plots of the magnetisation as measured using a commercial VSM magnetometer were performed between 293 and 78 K on samples aligned in a magnetic field of 1.9 T. In the second instance θ was determined from 293 to 4.2 K by measuring the parallel and perpendicular components of the magnetisation with the c axis, M_{\parallel} and M_{\perp} , respectively, in an applied magnetic field at 0.6 T. For the second type of measurement the samples were in the form of powders with an average 50 μm grain size aligned in a magnetic field of 2.5 T, fixed in epoxy resin and in the shape of flat disks.

The singular point detection (SPD) technique (Asti and Rinaldi 1972, 1974) was used in order to measure the composition and temperature dependence of the magnetic anisotropy field. Pulsed magnetic fields of up to 20 T were employed for these types of measurements. The magnetic anisotropy fields with the applied magnetic field in the basal plane and along the c axis were measured down to 78 K for various Pr compositions.

3. Results and analysis

3.1. X-ray diffraction

In figure 1 are displayed the results of the refinements of the lattice constants for $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$ compounds together with some representative points for NdCo_5 and PrCo_5 taken from the literature (Velge and Buschow 1969, Hoffer and Strnat 1966, Ostertag and Strnat 1966). Also displayed in figure 1 are the calculated values of the c/a ratio and the hexagonal unit cell volume V_c . Given the variation in the reported values of a and c which most probably arise from fluctuations in the stoichiometry of the compounds, there is a general increase in a and a decrease in c in going to compositions richer in Nd which is entirely dependent upon size and valency considerations of the Pr^{3+} and Nd^{3+}



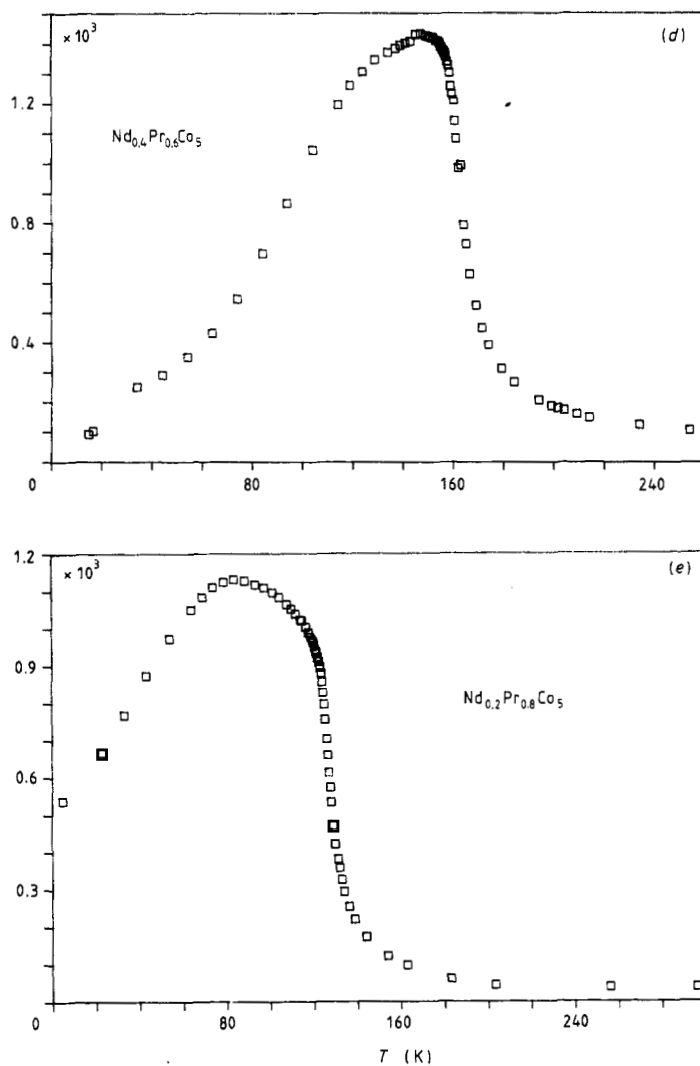


Figure 2. Temperature dependence of the AC initial susceptibility for Pr_xNd_{1-x}Co₅ compounds.

ions. Given the magnitudes of the estimated errors in both a and c , the stoichiometry of the final compounds can also be considered to be very close to the nominal starting compositions.

3.2. Initial AC susceptibility χ and the low-field magnetisation

The temperature dependence of χ for all the measured compounds is displayed in figures 2(a)–(e). There are very clear anomalies in each case and as will be seen they are associated with SR phase transitions. In figures 3 to 5 are displayed the isotherms of the measured parallel and perpendicular magnetisation as a function of θ (with θ the angle between the applied magnetic field and the c axis) for all the measured compounds at some selected temperatures. The relevant minima and maxima in M_{\parallel} and the zero values

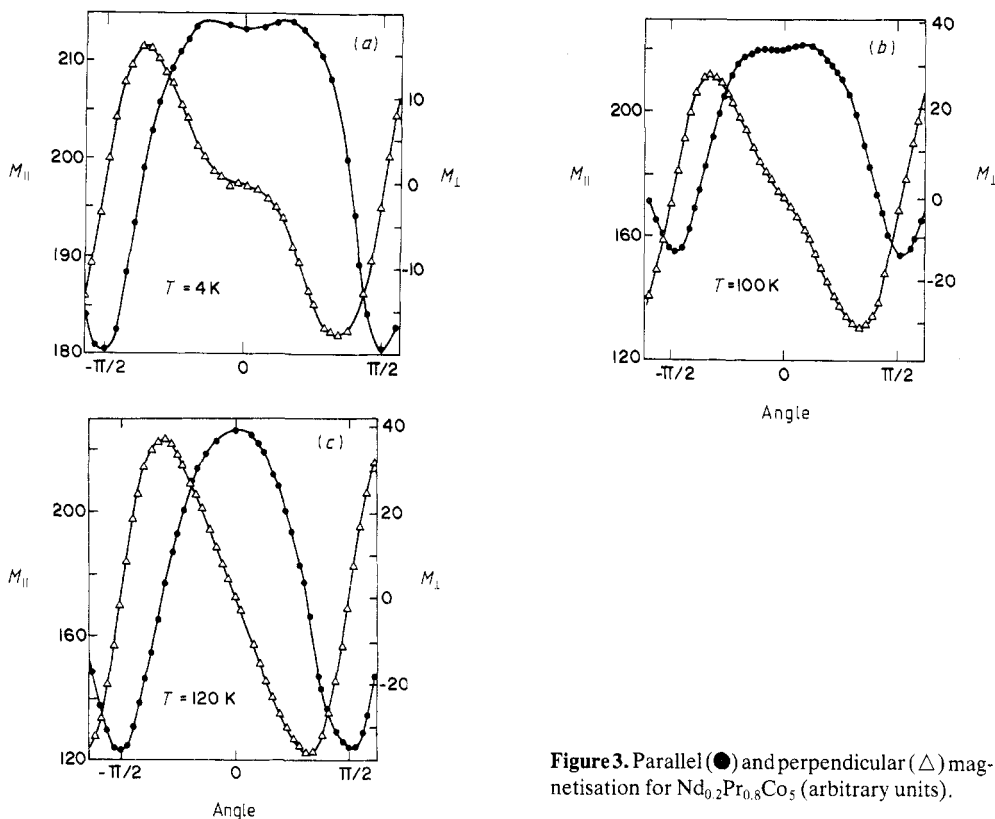


Figure 3. Parallel (●) and perpendicular (△) magnetisation for $\text{Nd}_{0.2}\text{Pr}_{0.8}\text{Co}_5$ (arbitrary units).

of M_{\perp} clearly show the temperature variation of the angle θ . Summarised in figure 6 are the temperature and composition dependence of θ for all the measured compounds obtained from both the polar plot and parallel and perpendicular magnetisation measurements. It is clear that in each case the anomaly in $\chi(T)$ can be associated with a SR phase transition. For the compounds $x = 0.2$ and 0.4 , the anomaly is associated with a complete SR phase transition from axis to plane, whilst for the other three compounds with $x = 0.5, 0.6$ and 0.8 it is associated with an incomplete SR phase transition from axis to cone.

In the case of the compound with $x = 0.2$, the first peak in the $\chi(T)$ curve at $T_{\text{SR1}} = 231$ K is the SR phase transition from axis towards plane which is eventually complete at a temperature $T_{\text{SR2}} = 190$ K. A similar situation exists for the compound with $x = 0.4$. The small shoulder in the curve of $\chi(T)$ for this compound at $T = 120$ K can be considered as the point at which the transition from cone to plane takes place.

3.3. High-field magnetisation

The SPD measurements were performed in two different configurations, with the applied magnetic field parallel and perpendicular to the c axis. The results are summarised in figures 7(a) and 7(b). In figure 7(a), the temperature dependence of the anisotropy field H_a and the critical field H_{cr} for first-order field induced magnetisation processes (FOMP) (Asti and Bolzoni 1980, 1985) are displayed for magnetic fields applied perpendicular

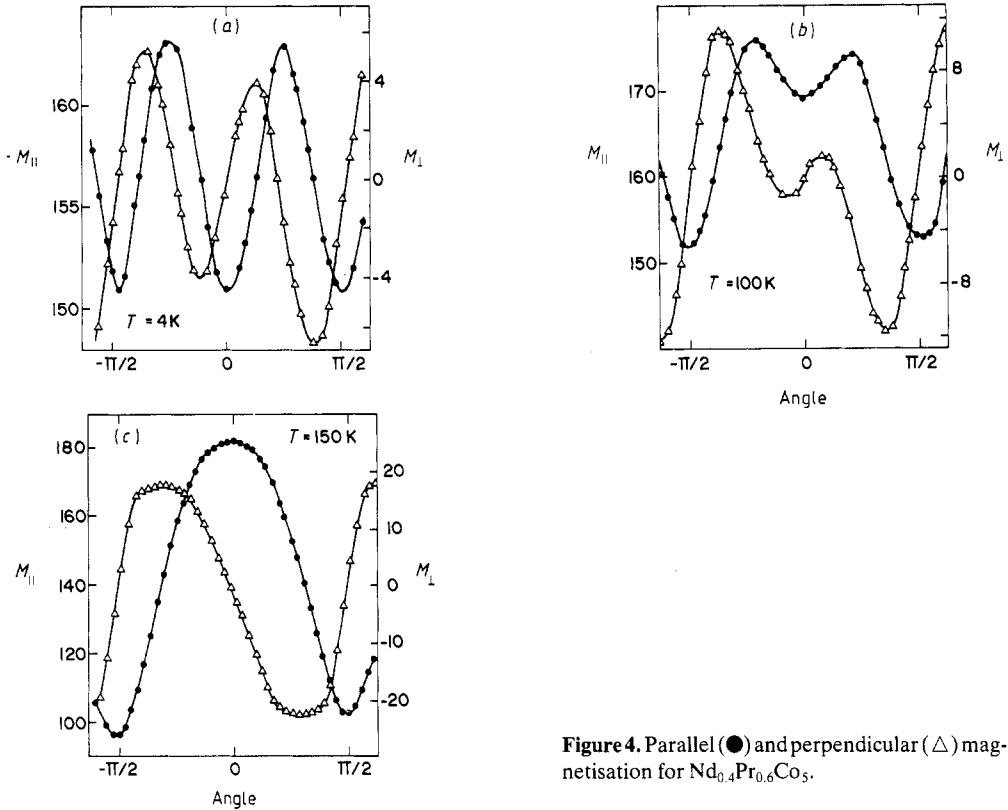


Figure 4. Parallel (●) and perpendicular (Δ) magnetisation for $\text{Nd}_{0.4}\text{Pr}_{0.6}\text{Co}_5$.

to the c axis. In all cases, both the anisotropy and critical fields decrease with decreasing temperature and increase with increasing Pr composition. An analysis in terms of phenomenological anisotropy energy constants (even though not appropriate for a microscopic description of the temperature dependence of the magnetic anisotropy since more suitable and realistic microscopic energy models are available) can give a useful indication for trends in the SR transitions. The macroscopic free energy F_K can be written for a hexagonal crystal up to sixth order as (neglecting anisotropy in the basal plane)

$$F_K = K_1 \sin^2 \theta + K_2 \sin^4 \theta + K_3 \sin^6 \theta - HM_s \cos \theta \quad (1)$$

where K_i are the anisotropy constants, H the applied magnetic field and M_s is the saturation magnetisation.

For the applied magnetic field H perpendicular to the c axis the expression for the anisotropy field H_a^{\perp} in the basal plane is

$$H_a^{\perp} = 2(K_1 + 2K_2 + 3K_3)/M_s \quad (2)$$

whilst for the applied magnetic field H parallel to the c axis the anisotropy field along the c axis is

$$H_a^{\parallel} = 2K_1/M_s. \quad (3)$$

When the system undergoes a transition from axis to cone, H_a^{\parallel} becomes zero and K_1 in general changes sign. This is confirmed in figure 7(b) for compounds with $x = 0.2, 0.4, 0.5$ and 0.6 where the extrapolated values of the temperature dependence of H_a^{\parallel} to

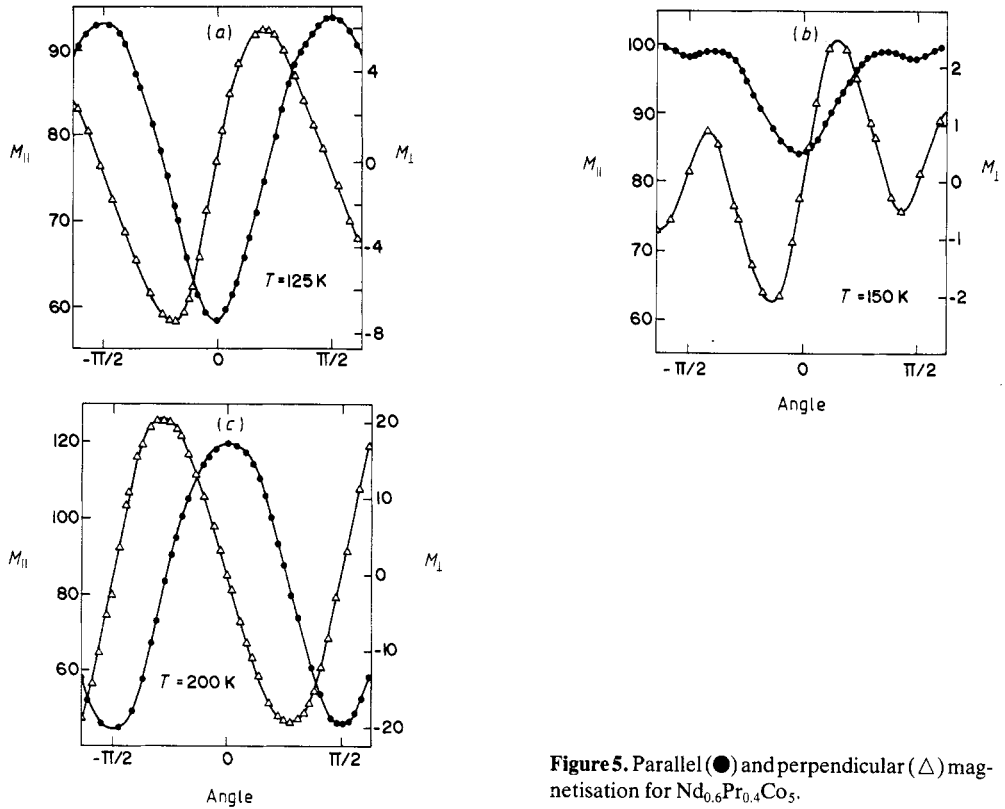


Figure 5. Parallel (●) and perpendicular (△) magnetisation for $\text{Nd}_{0.6}\text{Pr}_{0.4}\text{Co}_5$.

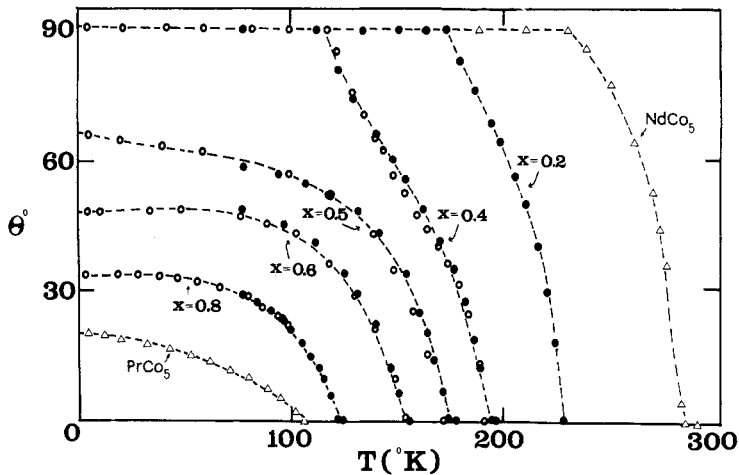


Figure 6. Temperature dependence of the angle θ for $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$ compounds (polar plots ●, perpendicular and parallel magnetisation, ○).

where $H_a^||$ becomes zero give good agreement with the axis-cone transition as measured by the AC susceptibility and low-field magnetisation measurements. When the anisotropy energy perpendicular to the c axis $K_1 + 2K_2 + 3K_3$ changes sign, the system takes on an

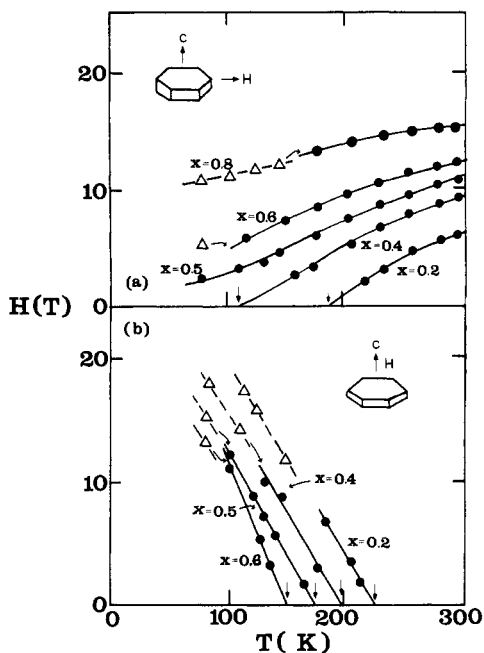


Figure 7. Temperature dependence of the anisotropy field H_a (●) and critical field H_c (△) for $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$ compounds for H applied perpendicular (a) and parallel (b) to the c axis.

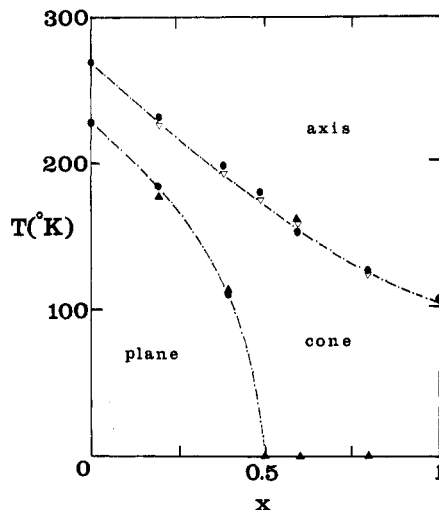


Figure 8. The magnetic phase diagram for $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$ compounds.

easy plane configuration. Again, this is confirmed in figure 7(a) where for compounds with $x = 0.2$ and $x = 0.4$ the system becomes planar at temperatures of approximately 180 and 120 K respectively; again in good agreement with the AC susceptibility and low-field magnetisation measurements. The combination of the three techniques has allowed a rather definite magnetic phase diagram of the $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$ system to be obtained (figure 8).

4. Analysis and discussion

An approach exactly analogous to that based on the scaling behaviour of the AC initial susceptibility already applied to tetragonal $\text{RE}_2\text{Fe}_{14}\text{B}$ compounds (del Moral *et al* 1989) can also be adopted for the hexagonal compounds under consideration here. Following the model of scaling in SR phase transitions the associated initial susceptibility can be written as

$$\chi_H \approx M_s \theta (\partial \theta / \partial H)_T \approx \frac{1}{2} M_s \theta (\partial^2 \theta / \partial H^2) \quad (4)$$

where θ is the SR angle and is taken as the relevant order parameter, and H and M_s are as defined previously, and $\chi_\theta = (\partial \theta / \partial H)_T$ is the associated angular susceptibility. Some important assumptions have been made in order to obtain the above expression. It is implicitly assumed that critical fluctuations in θ are weak so that equilibrium thermodynamics is applicable and that θ is small. Defining t as the reduced temperature $t =$

$(T - T_{\text{SR}})/T_{\text{SR}}$, it is possible now to consider the critical scaling for $t \rightarrow 0$, $H = 0$. The order parameter θ follows the subsequent scaling behaviour

$$\theta = |t|^{\beta_{\text{SR}}} f(x) \quad (5)$$

where β_{SR} and δ_{SR} are the SR critical exponents for $T < T_{\text{SR}}$, and f is an arbitrary scaling function of the reduced magnetic field $x = H/|t|^{\beta_{\text{SR}}\delta_{\text{SR}}}$ which is constant for $H = 0$. The following expression now results for scaling of the susceptibility for $T < T_{\text{SR}}$

$$\chi_H = M_s |t|^{-\omega_{\text{SR}}} f(x) \partial f / \partial x \quad (6)$$

where $\beta_{\text{SR}}(\delta_{\text{SR}} - 1) = \gamma'_{\text{SR}}$ and the auxiliary exponent $\omega'_{\text{SR}} = \delta'_{\text{SR}} - \beta_{\text{SR}}$ is defined.

Therefore for $t < 0$ and $H = 0$, χ_H scales as

$$\chi_H = t^{-\omega'_{\text{SR}}}. \quad (7)$$

Thus χ_H displays scaling behaviour in the regime of $t < 0$ as $t \rightarrow 0$. In the case of $t > 0$ ($T > T_{\text{SR}}$), $\beta_{\text{SR}} = 0$ and therefore $\omega_{\text{SR}} = \gamma_{\text{SR}}$ so that

$$\chi \approx t^{-\gamma_{\text{SR}}}. \quad (8)$$

The angular susceptibility scales as $\chi_{\theta} = |t|^{-\gamma_{\text{SR}}}$, as is expected. When there are two types of transitions (axis to cone at T_{SR1} and cone to plane at T_{SR2}) this analysis applies to both. This approach to critical scaling in SR phase transitions has been applied to the present series of $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$ compounds. It can be seen that from the cone-angle (θ) measurements for $x \leq 0.4$, the system undergoes two successive phase transitions; one from axial to cone structure (at T_{SR1}) and the other from cone to easy plane (at T_{SR2}). For $x \geq 0.5$ there exists, however, only one transition; from axis to cone, and these transitions are clearly reflected in the thermal variation of the initial susceptibility. For the compounds high in Pr content there exists only a rapid increase of χ_H at T_{SR} (figures 2(c)–(e)), but for the low concentrations there are two anomalies associated with the SR transitions at T_{SR1} (axis–cone) and T_{SR2} (cone–plane). For the compound with $x = 0.4$, the second transition manifests itself as an elbow (figure 2(b)). The rounding effects observed for temperatures below T_{SR2} should be associated with the divergence in χ_H but suffers a cut-off which is most likely due to the effects of demagnetising fields or possibly due to the slowing down of the critical relaxation. This cut-off is very apparent for the compound $x = 0.2$ between T_{SR1} and T_{SR2} .

By way of example the scaling of $\log \theta$ with $\log t$ is displayed in figure 9 for the compound with $x = 0.6$ from which an exponent $\beta_{\text{SR}} = 0.56 \pm 0.01$ is deduced. Also shown in figure 9 is the scaling for $x = 0.4$ for temperatures below T_{SR1} and T_{SR2} where good linearity of $\log \theta$ versus $\log t$ is observed. (Values of $\beta_{\text{SR1}} = 0.48 \pm 0.02$ and $\beta_{\text{SR2}} = 0.79 \pm 0.01$ were obtained in this case.) Very similar plots were obtained for all the remaining compositions. In table 1 are summarised the obtained values of the exponents β_{SR} . These values are, in general, not excessively far from the predictions of a model of critical behaviour based on the spin wave approximation developed for systems with competing axial and planar anisotropies and which predicts $\beta_{\text{SR}} = \frac{1}{2}$ (del Moral 1990). A scaling of the initial susceptibility in the form $\log \chi_H$ versus $\log t$ was also undertaken by use of expressions (7) and (8). The critical exponents are also summarised in table 1.

In these particular types of SR phase transitions, the critical divergence of χ_H is invariably superimposed on a large background susceptibility due to the usual Hopkinson effect (domain wall susceptibility) in the ferromagnetic regime. Thus corrections to χ_H were performed by extrapolations outside the critical regime. In this case the

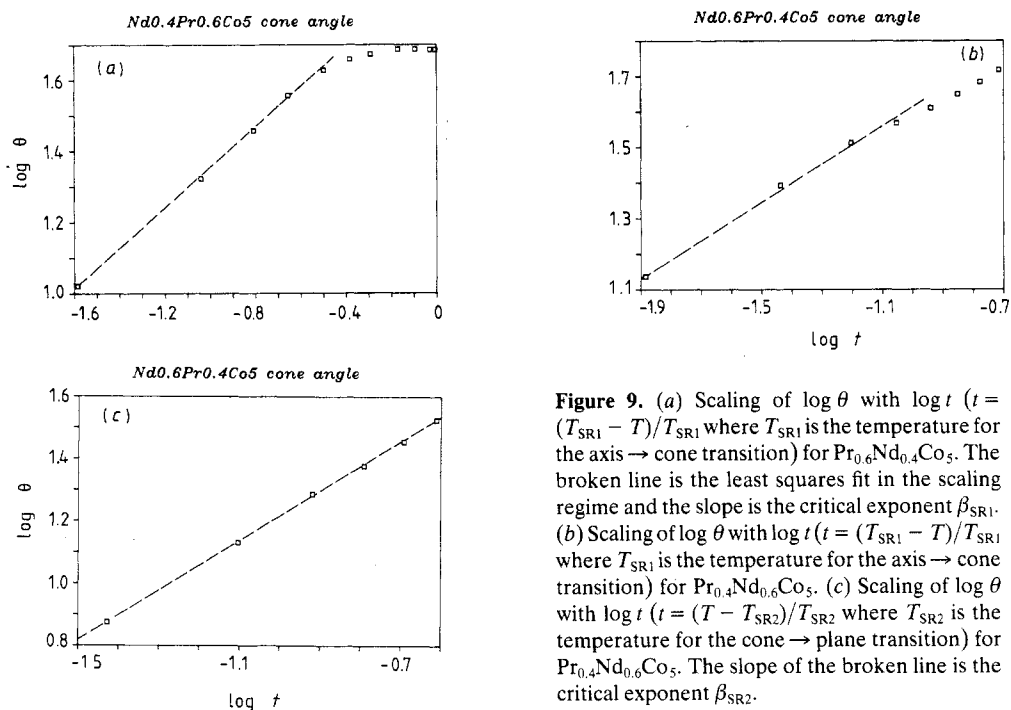


Figure 9. (a) Scaling of $\log \theta$ with $\log t$ ($t = (T_{\text{SR1}} - T)/T_{\text{SR1}}$ where T_{SR1} is the temperature for the axis \rightarrow cone transition) for $\text{Pr}_{0.6}\text{Nd}_{0.4}\text{Co}_5$. The broken line is the least squares fit in the scaling regime and the slope is the critical exponent β_{SR1} . (b) Scaling of $\log \theta$ with $\log t$ ($t = (T_{\text{SR1}} - T)/T_{\text{SR1}}$ where T_{SR1} is the temperature for the axis \rightarrow cone transition) for $\text{Pr}_{0.4}\text{Nd}_{0.6}\text{Co}_5$. (c) Scaling of $\log \theta$ with $\log t$ ($t = (T - T_{\text{SR2}})/T_{\text{SR2}}$ where T_{SR2} is the temperature for the cone \rightarrow plane transition) for $\text{Pr}_{0.4}\text{Nd}_{0.6}\text{Co}_5$. The slope of the broken line is the critical exponent β_{SR2} .

Table 1. Transition temperatures (T_{SR1} : axis \rightarrow cone; T_{SR2} : cone \rightarrow plane) and the critical exponents for SR phase transitions in $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$ compounds.

x (at. % Pr)	T_{SR1} (K)	T_{SR2} (K)	β_{SR1}	β_{SR2}	γ_{SR1}	ω'_{SR2}
0.80	120.0	—	0.66 ± 0.07	—	1.19 ± 0.01	—
0.60	154.5	—	0.56 ± 0.01	—	1.35 ± 0.02	—
0.50	170.2	—	0.69 ± 0.02	—	1.41 ± 0.01	—
0.40	192.0	125.3	0.48 ± 0.02	0.79 ± 0.01	1.21 ± 0.03	1.06 ± 0.08
0.20	230.0	190.8	1.00 ± 0.20	0.87 ± 0.07	1.18 ± 0.01	0.45 ± 0.02

critical exponents, however, are quite sensitive to the particular type of background selected. The scaling produced was however, quite good and examples are shown in figure 10 for different concentrations and types of transition. The values of γ_{SR1} are very sensitive indeed to the values introduced for T_{SR1} . For $T < T_{\text{SR2}}$, $\omega_{\text{SR2}} = \gamma'_{\text{SR2}} - \beta_{\text{SR2}}$. For example, in the case of $x = 0.2$, $\gamma'_{\text{SR2}} = 1.32$ but the comparison gets worse in the case of $x = 0.4$, with $\gamma'_{\text{SR2}} = 1.85$.

The present data and scaling model analysis indicate very clearly the competing axial and planar anisotropies in this system. Most of the exotic magnetic transitions observed in these types of compounds are primarily due to the different temperature dependences of the RE and Co sublattice anisotropies. In the easy-cone region it can be expected that the RE and Co moments are no longer collinear since the RE-Co exchange coupling cannot dominate the difference in the respective anisotropy energies of the RE sublattices. Classical calculations (Irkhin and Rozenfeld 1974, Rinaldi and Pareti 1979) have

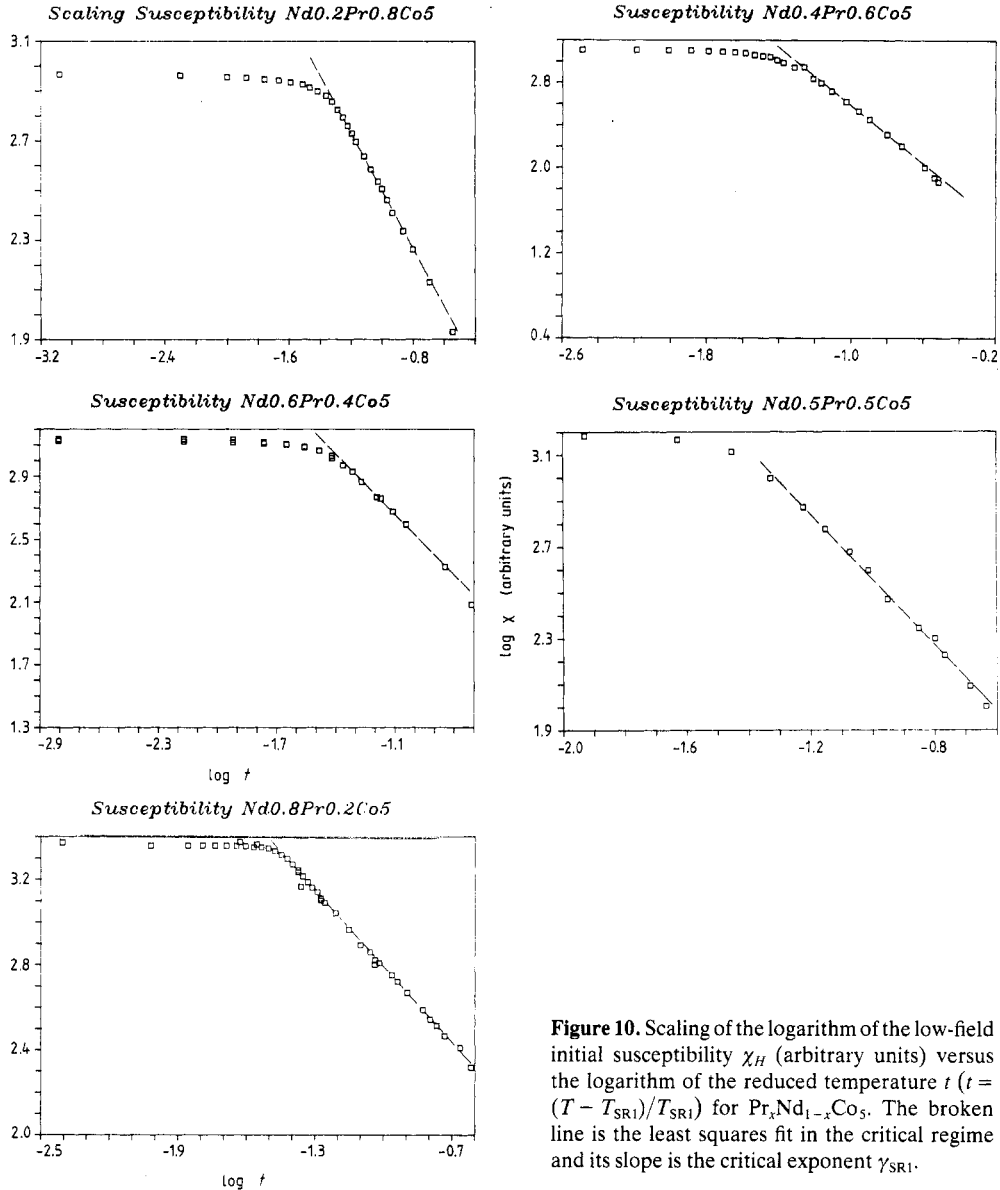


Figure 10. Scaling of the logarithm of the low-field initial susceptibility χ_H (arbitrary units) versus the logarithm of the reduced temperature t ($t = (T - T_{SR1})/T_{SR1}$) for $\text{Pr}_x\text{Nd}_{1-x}\text{Co}_5$. The broken line is the least squares fit in the critical regime and its slope is the critical exponent γ_{SR1} .

suggested that such a non-collinearity can lead to an anomalous field-dependent magnetisation (first-order magnetisation processes, FOMP) and large high-order phenomenological anisotropy constants (Asti *et al* 1980). Such a non-collinearity has for example been observed in HoCo_5 (Decrop *et al* 1982, 1983) and has been very successfully interpreted in terms of a microscopic crystalline-electric-field-mean-molecular-field model with inclusion of non-collinearity between the Ho and Co moments during the SR process. Application of such a model to the present system in order to account for the temperature and composition dependence of θ and the observed transitions in the high-field magnetisation curves with realistic crystal field and exchange parameters is under way and will be presented in a forthcoming publication.

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