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1992 J. Phys.: Condens. Matter 4 8609

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Anomalous magnetic properties of Pr ions in $\text{Pr}_x\text{R}_{1-x}\text{Co}_5$ ($\text{R} \equiv \text{Y}, \text{Sm}$)

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Received 14 April 1992, in final form 1 July 1992

Abstract. The magnetic hyperfine fields of Pr ions in PrCo_5 are measured by spin-echo NMR at 4.2 K. The magnetic properties for the $\text{Pr}_x\text{Y}_{1-x}\text{Co}_5$, $\text{Pr}_x\text{Sm}_{1-x}\text{Co}_5$ and $\text{Dy}_x\text{Sm}_{1-x}\text{Co}_{5+0.2x}$ compounds are analysed on the basis of the single-ion model under the assumption that the rare-earth ions are triply ionized. The magnetic moment and the hyperfine fields for Pr ions in PrCo_5 are calculated to be much larger than those observed in experiments, while good agreement is observed for Sm and Dy ions in SmCo_5 and $\text{DyCo}_{5.2}$, respectively.

1. Introduction

It is commonly accepted that, in rare-earth–3d metal compounds, most of the rare-earth ions, except Ce, are triply ionized. A number of workers, however, noticed that the properties of Pr ions in PrCo_5 are unusual, and it was assumed that the Pr ion is valence fluctuated [1–5]. Nesbitt and Wernick [1] and Yermolenko [2] found that the magnetic moment of Pr ions in PrCo_5 , obtained as the difference between the magnetic moments of PrCo_5 and YCo_5 , is much smaller than the theoretical value of $g_J J = 3.2\mu_B$, which is in contrast with the other rare-earth ions. Finkel'shteyn and Samsonova [3] observed an additional peak near the normal peak corresponding to the triply ionized state in the L-III x-ray absorption spectrum for Pr ions in PrCo_5 , as was also observed for Ce ions in CeCo_5 . Zhao and co-workers [4] evaluated the values of the R–Co exchange field H_{ex} and the crystalline-electric-field (CEF) parameters $A_{n,m}$ for rare-earth ions in a series of RCo_5 ($\text{R} \equiv$ rare earth) compounds by using the fitting procedure on the assumption that the rare-earth ions are triply ionized. It was found that A_{20} for the Pr ion is opposite in sign and one order smaller in absolute value than those for all the other rare-earth ions [4]. The lattice constant a of PrCo_5 is smaller than that extrapolated from those of heavy rare-earth compounds and LaCo_5 [5].

In this work, the hyperfine fields at the nuclei of Pr ions in PrCo_5 were measured. The magnetic properties of $\text{Pr}_x\text{R}_{1-x}\text{Co}_5$ ($\text{R} \equiv \text{Y}$ and Sm) and $\text{Dy}_x\text{Sm}_{1-x}\text{Co}_{5+0.2x}$, which have been reported by a number of workers, were calculated using the fitted parameters of the rare-earth–Co exchange field H_{ex} and CEF parameters $A_{n,m}$ for Pr, Sm and Dy ions which have been evaluated for the PrCo_5 [4], SmCo_5 [4]

and $\text{Dy}_x\text{Y}_{1-x}\text{Co}_{5+0.2x}$ compounds with the same concentration x [6], respectively. The calculations reproduce the following experimental data well: the temperature dependence of the spontaneous magnetization $M(T)$, the temperature dependence of the cone angle $\theta(T)$ made by the easy direction of magnetization (EDM) and the c axis and the magnetization curves along the crystallographic axes $M(H)$ at 4.2 K, all measured on single crystals of the compounds, and also the magnetic moments and the hyperfine fields of Sm and Dy ions in SmCo_5 and $\text{DyCo}_{5.2}$, respectively. However, the calculated magnetic moment and the hyperfine field for Pr ions in PrCo_5 are much larger than those observed in the experiments.

2. Experiments

The PrCo_5 sample was prepared by argon arc melting the appropriate amounts of the constituents. The purity of the raw materials is 99.9%. We checked that the powder had the CaCu_5 structure by x-ray diffraction. The spin-echo NMR measurements on ^{141}Pr in PrCo_5 were carried out at 4.2 K. Figure 1 shows the NMR spectrum for ^{141}Pr . Two peaks are observed at 2940 and 3275 MHz. By taking a ^{141}Pr nuclear moment of 4.275 nm [7], these peaks correspond to hyperfine fields of 225.5 T and 251.2 T, respectively. The averaged hyperfine field is 237 T.

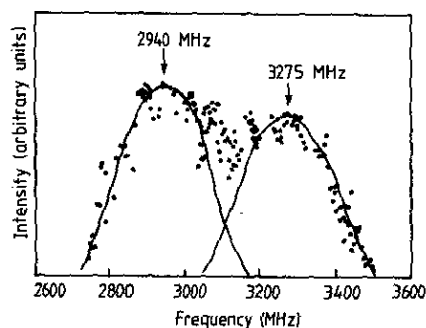


Figure 1. NMR spectrum for ^{141}Pr in PrCo_5 at 4.2 K.

3. Calculation

The Hamiltonian for the rare-earth ion consists of the spin-orbit coupling interaction, the CEF interaction, the rare-earth-Co exchange interaction and the Zeeman energy, i.e.

$$\mathcal{H} = \lambda S \cdot L + \mathcal{H}_{\text{CEF}} + 2\mu_{\text{B}} S \cdot H_{\text{ex}} + \mu_{\text{B}} (L + 2S) \cdot H. \quad (1)$$

Here, S and L are the total spin and orbital angular moments, and H is the applied field. The CEF interaction in the coordinate system with the z and x axes along the c and a axes, respectively, is formulated as

$$\mathcal{H}_{\text{CEF}} = A_{20}C_{20} + A_{40}C_{40} + A_{60}C_{60} + A_{66}(C_{66} + C_{6-6}) \quad (2)$$

where

$$C_{nm} = \sum_j C_{nm}(\theta_j, \varphi_j) = \sum_j \left(\frac{4\pi}{2n+1} \right)^{1/2} Y_{nm}(\theta_j, \varphi_j). \quad (3)$$

$Y_{nm}(\theta_j, \varphi_j)$ are the spherical harmonics, and θ_j and φ_j are the polar and azimuthal angles of the position vector of the j th electron r_j . The rare-earth-rare-earth exchange interaction, which is much smaller than the rare-earth-Co exchange interaction, was neglected. It was assumed that the rare-earth ions are triply ionized. Both H_{ex} and the magnetic moment of the Co sublattice are anisotropic and are represented as

$$H_{\text{ex}}(\theta_{\text{Co}}, T) = H_{\text{ex}}(T)[1 - p'(T) \sin^2 \theta_{\text{Co}}] \quad (4)$$

$$M_{\text{Co}}(\theta_{\text{Co}}, T) = M_{\text{Co}}(T)[1 - p(T) \sin^2 \theta_{\text{Co}}] \quad (5)$$

with $p'(0) = 0.020$ and $p(0) = 0.037$ [8]. $H_{\text{ex}}(T)$ was assumed to be proportional and antiparallel to $M_{\text{Co}}(T)$, and the values of $M_{\text{Co}}(T/T_C)/M_{\text{Co}}(0)$ (T_C is the Curie temperature) and $p(T)$ were taken from those of YCo_5 [9]. The relation $p'(T)/p'(0) = p(T)/p(0)$ was assumed. For a given applied field H and a direction of H_{ex} , the eigenvalues E_i and eigenstates $|n_i\rangle$ ($i = 1, 2, \dots, \sum_J(2J+1)$) are obtained by diagonalizing the $\sum_J(2J+1) \times \sum_J(2J+1)$ matrix of equation (1). The mixing of the first excited J multiplet for the Pr ions with $\lambda = 610$ K [10], the mixing of the lowest two excited multiplets for the Sm ions with $\lambda = 410$ K [10], and no mixing for the Dy ions were taken into account. The free energy for the $\text{R}_x\text{R}'_{1-x}\text{Co}_5$ system is

$$F(H, H_{\text{ex}}, T) = -x k_B T \ln[Z(\text{R})] - (1-x) k_B T \ln[Z(\text{R}')] + K_{1\text{Co}}(T) \sin^2 \theta_{\text{Co}} - M_{\text{Co}}(\theta_{\text{Co}}, T) \cdot H \quad (6)$$

where

$$Z(\text{R}) = \sum_i \exp\left(-\frac{E_i(\text{R})}{k_B T}\right) \quad (7)$$

and $K_{1\text{Co}}$ is the magnetocrystalline anisotropy constant of the Co sublattice per formula unit (FU). $K_{1\text{Co}}(T/T_C)/K_{1\text{Co}}(0)$ was taken from that of YCo_5 [9]. The values of $M_{\text{Co}}(0)$, $K_{1\text{Co}}(0)$ and T_C for YCo_5 , PrCo_5 , SmCo_5 and $\text{DyCo}_{5.2}$ are as follows: $7.7\mu_B \text{ FU}^{-1}$, $7.7\mu_B \text{ FU}^{-1}$, $8.33\mu_B \text{ FU}^{-1}$ and $8.92\mu_B \text{ FU}^{-1}$, respectively; 45 K FU^{-1} , 45 K FU^{-1} , 45 K FU^{-1} and 42 K FU^{-1} , respectively [4]; 910 K, 910 K, 980 K and 965 K, respectively [11]. The quantities for the pseudo-binary compounds $\text{R}_x\text{R}'_{1-x}\text{Co}_5$ were treated as linear to x . The equilibrium direction of H_{ex} was determined from minimization of the free energy. The magnetic moments of the R ion and of $\text{R}_x\text{R}'_{1-x}\text{Co}_5$ are given by

$$M_{\text{R}}(T) = -\sum_i \langle n_i(\text{R}) | L + 2S | n_i(\text{R}) \rangle \frac{\exp[-E_i(\text{R})/k_B T]}{Z(\text{R})} \quad (8)$$

$$M(T) = x M_{\text{R}}(T) + (1-x) M_{\text{R}'}(T) + M_{\text{Co}}(\theta_{\text{Co}}, T). \quad (9)$$

The hyperfine field of the rare-earth ion at 0 K consists of two parts: the field produced by the ion itself, and the transferred field induced by the magnetic moments of the Co sublattice, i.e.

$$H_{\text{n}} = H_{\text{n}}(\text{free}) N_{\text{rel}} + H_{\text{n}}(\text{transf}) \quad (10)$$

$$N_{\text{rel}} = |\langle n_1 | N \cdot M_{\text{R}}(0) / M_{\text{R}}(0) | n_1 \rangle | \langle J, -J | N \cdot k | J, -J \rangle | \quad (11)$$

where

$$N = \sum_j \left[l_j - s_j + \frac{3(s_j \cdot r_j)r_j}{r_j^2} \right] \quad (12)$$

H_n (free) is the hyperfine field for the free rare-earth ion the value of which is known from experiment, l_j and s_j are the angular moments of the j th 4f electron, J is the total angular moment for the ground multiplet, $|n_1\rangle$ is the ground eigenstate, and \mathbf{k} is the unit vector along the c axis. The following relations have been used in the calculation:

$$\begin{aligned} \left\langle LSJM \left| \sum_j l_{jq} \right| LSJ'M' \right\rangle &= \langle LSJM | L_q | LSJ'M' \rangle \\ &= (-1)^{L+S-M} [(2L+1)(L+1)L(2J+1)(2J'+1)]^{1/2} \\ &\quad \times \begin{pmatrix} J' & 1 & J \\ M' & q & -M \end{pmatrix} \begin{Bmatrix} J' & 1 & J \\ L & S & L \end{Bmatrix} \end{aligned} \quad (13)$$

$$-s_{jx} + 3(s_j \cdot r_j)x_j/r_j^2 = \sqrt{5}[(C_j^{(2)}s_j^{(1)})_1^{(1)} - (C_j^{(2)}s_j^{(1)})_{-1}^{(1)}] \quad (C_{jm}^{(2)} = C_{2m}(\theta_j, \varphi_j)) \quad (14)$$

$$-s_{jz} + 3(s_j \cdot r_j)z_j/r_j^2 = -\sqrt{10}(C_j^{(2)}s_j^{(1)})_0^{(1)} \quad (15)$$

$$\begin{aligned} \left\langle LSJM \left| \sum_j (C_j^{(2)}s_j^{(1)})_q^{(1)} \right| LSJ'M' \right\rangle &= (-1)^{J-M} [3(2J+1)(2J'+1)]^{1/2} \\ &\quad \times \begin{pmatrix} J & 1 & J' \\ -M & q & M' \end{pmatrix} \begin{Bmatrix} L & L & 2 \\ S & S & 1 \\ J & J' & 1 \end{Bmatrix} \langle L || C_j^{(2)} || L \rangle \left\langle S \left\| \sum_j s_j^{(1)} \right\| S \right\rangle \\ &\quad (q = -1, 0 \text{ and } 1) \end{aligned} \quad (16)$$

$$\left\langle S \left\| \sum_j s_j^{(1)} \right\| S \right\rangle = [(2S+1)(S+1)S]^{1/2} \quad (17)$$

$$\langle L || C_j^{(2)} || L \rangle = (-1)^{J-L-S} \frac{\alpha_J J(2J-1)}{2n(2J+1)\sqrt{2L+1}} \begin{pmatrix} J & 2 & J \\ J & 0 & -J \end{pmatrix} \begin{Bmatrix} J & 2 & J \\ L & S & L \end{Bmatrix}. \quad (18)$$

Here, x_j and z_j are the components of r_j on the EDM on the c plane and c axis, respectively, l_{jq} is the q -component of the irreducible tensor operator $l_j^{(1)}$, i.e. $l_{j\pm 1}^{(1)} = \mp(1/\sqrt{2})(l_{jx} \pm il_{jy})$ and $l_{j0}^{(1)} = l_{jz}$, $(C_j^{(2)}s_j^{(1)})_q^{(1)}$ is the q -component of the irreducible tensor operator of rank 1 constructed by multiplying the irreducible operators $C_j^{(2)}$ and $s_j^{(1)}$, and α_J and n in equation (18) are the second-order Stevens coefficient for the ground J multiplet and the number of the 4f electrons for the rare-earth ion, respectively. H_n (transf) induced by the Co sublattice was estimated as follows. It is known that the direction of H_n (transf) is opposite to the magnetic moment direction of the Co sublattice and its value is 10.17 T at 4.2 K for YCo_5 [13]. The values of the hyperfine coupling parameters are roughly 1.7, 3.1 and 4.8 for Y,

La and Lu ions, respectively [14]. Linearly extrapolating the results as a function of the atomic number of the rare earths, the value of $H_n(\text{transf})$ for the rare-earth ion could be estimated. The value of $H_n(\text{transf})$ is one order smaller than the hyperfine field produced by the rare-earth ion itself, and the part of $H_n(\text{transf})$ induced by the other rare-earth ions was neglected.

4. Results and discussion

Table 1 lists the values of the exchange field $2\mu_B H_{ex}$ and CEF parameters used in the calculations for Pr, Sm and Dy ions in $Pr_xR_{1-x}Co_5$ and $Dy_xR_{1-x}Co_{5+0.2x}$ ($R \equiv Y$ and Sm).

Table 1. The values of $2\mu_B H_{ex}(T = 0 \text{ K})$ and A_{nm} for $Pr_xR_{1-x}Co_5$ and $Dy_xR_{1-x}Co_{5+0.2x}$ compounds ($R \equiv Y$ and Sm).

Ions	x	$2\mu_B H_{ex}$ (K)	A_{20} (K)	A_{40} (K)	A_{60} (K)	A_{66} (K)	Reference
Pr	0.0–1.0	1300	25	-75	250	-600	[4]
Sm	0.0–1.0	440	-330	-50	0	0	[4]
Dy	1.0	235	-425	-140	180	0	[6]
Dy	0.5	227	-500	-230	180	0	[6]
Dy	0.35	224	-520	-250	180	0	[6]
Dy	0.2	222	-545	-280	180	0	[6]
Dy	0.1	220	-560	-300	180	0	[6]

A comparison of the calculations of $M(T)$, $\theta(T)$ and $M(H)$ for RCO_5 compounds with experiments has been reported elsewhere [4].

Figures 2–9 present the comparison for the $R_xR'_{1-x}Co_5$ compounds. The full symbols represent the experimental data and the curves the calculations. Figures 2, 3 and 4 show the temperature dependence of the cone angle, the composition dependences of the spin-reorientation temperature and of the cone angle at 4.2 K, and the magnetization curves at 4.2 K, respectively, for $Pr_xY_{1-x}Co_5$. Figures 5, 6 and 7 show the temperature dependence of the spontaneous magnetization, the temperature dependence of the cone angle, and the magnetization curves at 4.2 K, respectively, for $Pr_xSm_{1-x}Co_5$. Figures 8 and 9 show the composition dependence of the cone angle and the magnetization curves, respectively, both at 4.2 K for $Dy_xSm_{1-x}Co_{5+0.2x}$.

Table 2. The values of the magnetic moment M_R and hyperfine field H_n for Pr, Sm and Dy ions in RCO_5 at 4.2 K.

Ion	$M_R(\mu_B)$		$H_n(\text{free})$		$H_n(\text{transf})$ (T)	$H_n(T)^a$		
	Calculated	Experimental	$g_J J$	(T)		Calculated	Experimental	
Pr	3.07	2.3[2]	3.20	335 ^a	1.009	-19	319	237(this work)
Sm	0.35	0.38[20]	0.71	335 ^a	1.075	-22	338	340[21] 344[22]
Dy	9.97	9.9[19] 8.8[24]	10.0	560 ^a	0.996	25	583	618[23]

^a The nuclear magnetic moments are taken to be 4.2754 nm (^{141}Pr), -0.8148 nm (^{147}Sm), -0.6717 nm (^{149}Sm), -0.4805 nm (^{161}Dy) and 0.6726 nm (^{163}Dy) [7].

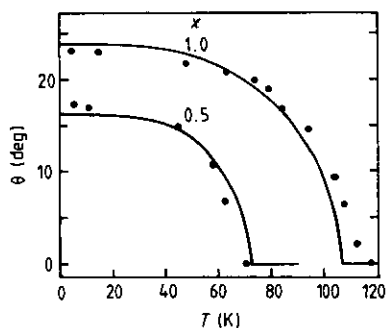


Figure 2. Temperature dependence of the cone angle for $\text{Pr}_x\text{Y}_{1-x}\text{Co}_5$ ($x = 1.0$ and 0.5). The experimental data (●) are from [2].

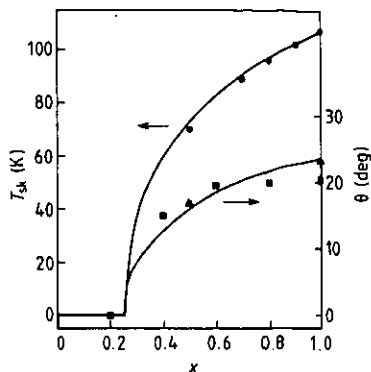


Figure 3. Composition dependences of the spin-reorientation temperature and of the cone angle at 4.2 K for $\text{Pr}_x\text{Y}_{1-x}\text{Co}_5$ ($x = 0.0-1.0$). The experimental data are from [2] (▲), [15] (●) and [16] (■).

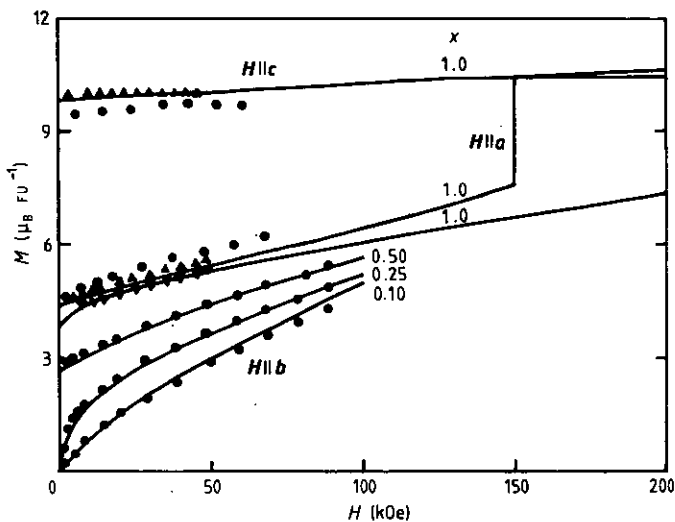


Figure 4. Magnetization curves along the crystal axes at 4.2 K for $\text{Pr}_x\text{Y}_{1-x}\text{Co}_5$ ($x = 1.0, 0.5, 0.25$ and 0.1). The experimental data are from [2] (●) and [16] (▲).

Figure 10 shows $M_{\text{Pr}}(0)$ and N_{rel} as functions of θ_{Co} calculated for Pr ions in PrCo_5 . It can be seen that they are approximately proportional to each other. At 4.2 K, the cone angles for M , M_{Pr} and M_{Co} are 24.6° , 27.2° and 23.5° , respectively. With $H_n(\text{free}) = 335$ T [7] and $H_n(\text{transf}) = -19$ T, therefore, $M_{\text{Pr}}(0)$ and H_n in a domain are calculated to be $3.07\mu_B$ and 319 T, respectively, both of which are larger than the experimental values by 35%, as shown in table 2. In contrast, the calculations of $M_{\text{R}}(0)$ and H_n coincide well with experiments on Sm and Dy ions in SmCo_5 and $\text{DyCo}_{5.2}$, respectively (table 2).

Smaller values of $M_{\text{Pr}}(0)$ and H_n are expected for the free Pr^{4+} ion than for the free Pr^{3+} ion. $M_{\text{Pr}}(0)$ is $2.14\mu_B$ for Pr^{4+} compared with $3.2\mu_B$ for Pr^{3+} . The ratio of the hyperfine fields for the ions is

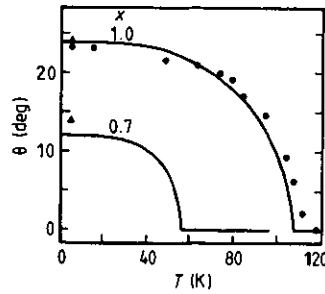
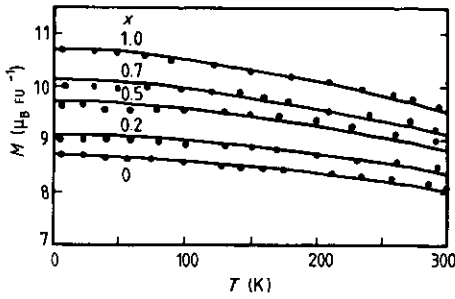


Figure 5. Temperature dependence of the spontaneous magnetization for $Pr_xSm_{1-x}Co_5$ ($x = 1.0, 0.7, 0.5, 0.2$ and 0.0). The experimental data (●) are from [17].

Figure 6. Temperature dependence of the cone angle for $Pr_xSm_{1-x}Co_5$ ($x = 1.0$ and 0.7). The experimental data are from [2] (●) and [17] (▲).

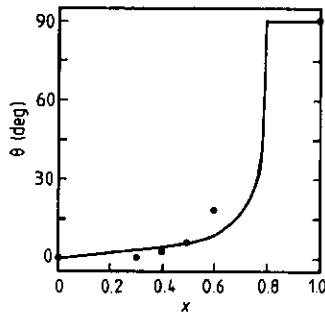
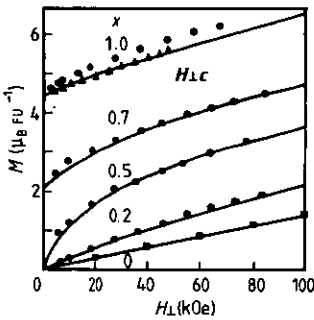


Figure 7. Magnetization curves along the crystal axes at 4.2 K for $Pr_xSm_{1-x}Co_5$ ($x = 1.0, 0.7, 0.5, 0.2$ and 0.0). The experimental data are from [2] (●), [16] (▲) and [18] (■).

Figure 8. Composition dependence of the cone angle at 4.2 K for $Dy_xSm_{1-x}Co_{5+0.2x}$. The experimental data (●) are from [19].

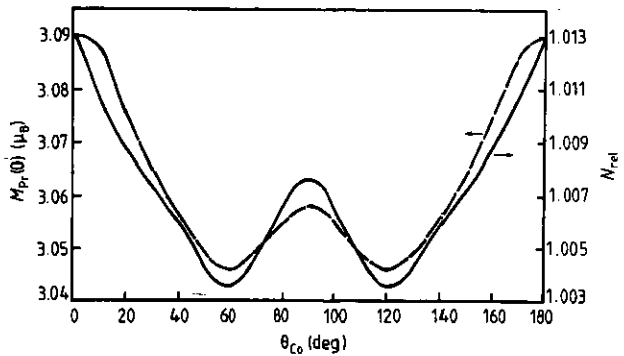
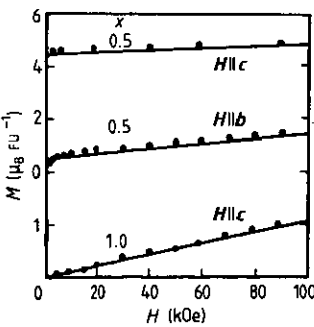


Figure 9. Magnetization curves along the crystal axes at 4.2 K for $Dy_xSm_{1-x}Co_{5+0.2x}$ ($x = 1.0$ and 0.5). The experimental data (●) are from [19].

Figure 10. $M_{Pr}(0)$ (---) and N_{rel} (—) as functions of θ_{Co} .

$$\frac{H_n(Pr^{4+})}{H_n(Pr^{3+})} = [2\mu_B \langle r^{-3} \rangle J \langle J || N || J \rangle + 4]_{Pr^{4+}} / [2\mu_B \langle r^{-3} \rangle J \langle J || N || J \rangle + 8]_{Pr^{3+}} \quad (19)$$

where 4 T and 8 T are the contributions from the core polarization, and

$\langle J||N||J \rangle = \frac{48}{35}$ and $\frac{296}{225}$ for the Pr^{4+} and Pr^{3+} ions, respectively [12]. By taking the hydrogen-like atomic approximation, the 4f radial wavefunction is proportional to $\exp(-Z_{\text{eff}} r/2a_0)r^3$, where Z_{eff} is the number of effective nuclear charges and a_0 is the Bohr radius. It is known that $\langle r^{-3} \rangle = 5.0a_0^{-3}$ for Pr^{3+} from the extrapolation of the data in [25]; so we can easily calculate that $Z_{\text{eff}} = 23.8$ for Pr^{3+} . For $Z_{\text{eff}} = 23.8 + 1$, then $\langle r^{-3} \rangle$ is calculated to be $5.66a_0^{-3}$. Since Z_{eff} for Pr^{4+} should be between 23.8 and $23.8 + 1$, the value of the ratio $H_n(\text{Pr}^{4+})/H_n(\text{Pr}^{3+})$ should be between 0.64 and 0.74. The higher limit should decrease for the ions in intermetallic compounds because of the 4f-conduction electron admixture.

The above results, therefore, support the suggestion that Pr ions in the PrCo_5 and $\text{Pr}_x\text{R}_{1-x}\text{Co}_5$ compounds are valence fluctuated, as are Nd ions in $\text{Nd}_x\text{Y}_{1-x}\text{Co}_5$ [26].

Acknowledgments

Part of this work has been supported by the Magnetism Laboratory, Institute of Physics, Chinese Academy of Sciences, Beijing, People's Republic of China. Thanks are due to Mr M Mizukami for his assistance with the NMR measurements.

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