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CeCoAl₄: a collinear antiferromagnet

**P Schobinger-Papamantellos^{1,7}, C Wilkinson^{2,3}, C Ritter³, L D Tung⁴,
K H J Buschow⁵ and O Moze⁶**

¹ Laboratorium für Kristallographie ETH Hönggerberg, CH-8093 Zürich, Switzerland

² Department of Chemistry, University of Durham, South Road, Durham DH1 3LE, UK

³ Institut Laue-Langevin, BP156, 38042 Grenoble Cedex 9, France

⁴ Department of Physics, University of Warwick, Coventry CV4 7AL, UK

⁵ Van der Waals-Zeeman Institute, University of Amsterdam, Valckenierstrasse 65, 1018 XE Amsterdam, The Netherlands

⁶ INFM, Department of Physics, University of Modena and Reggio Emilia, Via G Campi 213/a, I-41100 Modena, Italy

E-mail: penelope.schobinger@mat.ethz.ch

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Abstract

The magnetic ordering of the compound CeCoAl₄ (orthorhombic; *Pmma* space group) has been investigated by means of neutron diffraction from powder and single-crystal samples. Only the Ce moments order antiferromagnetically below $T_N = 12.8(1)$ K, with a propagation vector $\mathbf{q} = (0 \frac{1}{2} \frac{1}{2})$. Their collinear magnetic moments point along the *b* axis of the magnetic unit cell, which is doubled along both the nuclear *b* and *c* axes. At 1.5 K the refined magnetic moment value is 1.29(3) μ_B /Ce atom.

Introduction

The compound CeCoAl₄ crystallizes in the LaCoAl₄ structure type (space group *Pmma*, $Z = 2$, $a = 7.63$ Å, $b = 4.04$ Å, $c = 6.90$ Å at room temperature). This is the same crystal structure as observed for PrCoAl₄ [1, 2]. These two compounds present some rather interesting magnetic properties such as spin flop transitions in high magnetic fields and a large magnetization anisotropy.

We have previously studied the magnetic structure of PrCoAl₄ by means of neutron diffraction for a powder sample as well as for a single crystal [3, 4] in the temperature range 1.5–46 K at the ILL, Grenoble. These data showed that long range magnetic order sets in below about 20 K and is associated with the wavevector $\mathbf{q} = (0 0 q_z)$ where the length of q_z is almost temperature independent, but displays a sample dependence ($q_z = 0.4087(5)$ for single crystal and 0.437(3) for a powder sample). The magnetic structure corresponds to a sine wave

⁷ Author to whom any correspondence should be addressed.

longitudinal moment arrangement with the moments of the two Pr atoms scattering in phase. The refined amplitude of the wave, $\mu_0 = 2.24(4) \mu_B/\text{Pr atom}$, is much less than the free ion Pr^{3+} moment value, $gJ\mu_B = 3.2 \mu_B$, due to crystal field effects. These results are in good agreement with the results of the magnetic measurements and the specific heat [5, 6].

The compound CeCoAl_4 is of particular interest as the Ce atom adopts an unstable 4f shell, which subsequently greatly influences its magnetic and electric transport properties. The stability of the 4f shell may indeed depend on the degree of hybridization with the electronic states of the surrounding polyhedra.

The specific heat and the magnetic properties of CeCoAl_4 studied for a single crystal [7–9] showed that the Co atoms are non-magnetic and that only Ce orders antiferromagnetically below $T_N \approx 13.5$ K. The field dependence of the reciprocal susceptibility in the paramagnetic state indicates that Ce is in a trivalent state. The field dependence of the magnetization is strongly anisotropic, showing a spin flop transition at about 7.5 T along the *c* axis. For fields of 20 T along *b* and *c* the antiferromagnetic coupling is broken, resulting in a parallel (ferromagnetic) moment arrangement. The corresponding moment values at 20 T and 4.2 K are $1.11 \mu_B/\text{Ce atom}$ and $1.53 \mu_B/\text{Ce atom}$ respectively. These results are in good agreement with the bulk properties reported in earlier communications [10, 11].

There has been much recent discussion and speculation concerning the true magnetic ground state of CeCoAl_4 . Dhar *et al* [11] proposed on the basis of electrical resistivity measurements the existence of an incommensurate antiferromagnetic structure, which induces gaps in the conduction electron energy spectrum of CeCoAl_4 , while earlier measurements of the Seebeck coefficient suggest the existence of characteristic positive Kondo contributions [12]. In a more recent publication Koterlin *et al* [10] report on the basis of measurements of electrical resistivity, magnetoresistance and susceptibility that CeCoAl_4 is a magnetic Kondo lattice with a strongly suppressed Kondo interaction of the ground state doublet of the 4f level with the conduction electrons.

The magnetic structure of CeCoAl_4 had not been previously studied by means of neutron diffraction, but from knowledge of the PrCoAl_4 magnetic structure and the results presented in [10–12] it was expected to be rather complex. Preliminary neutron TOF (time-of-flight) data [13] collected by one of us at 5 and 20 K from a powder specimen at the ISIS spallation source at the Rutherford Appleton Laboratory indicated the occurrence of weak magnetic satellites and also showed that the sample contained unidentified impurity phases. This prompted us to extend the temperature window down to 1.5 K and to perform neutron diffraction experiments on a new powder sample and Laue diffraction experiments on a single-crystal sample of CeCoAl_4 . The results obtained in these experiments, which show that CeCoAl_4 is a commensurate antiferromagnet, are reported in the present paper.

1. Neutron diffraction

1.1. Data collection and analysis

Neutron powder diffraction experiments were carried out (a) in the temperature range 1.5–46 K at the Orphée reactor (LLB-Saclay) on the G4.1 (an 800-cell position sensitive detector: PSD) diffractometer using a wavelength of 2.426 Å and (b) on the D2B high resolution diffractometer ($\lambda = 1.594 \text{ \AA}$) at the ILL in Grenoble. The G4.1 data were collected for a full set of temperatures in steps of 2 K in order to study more precisely the magnetic transitions. The step increment in 2θ was 0.1° and the 2θ region 8° – 87° . The D2B data were collected at 1.5, 11.5, 15 and 293 K. The step increment in 2θ was 0.05° and the 2θ interval was from 0° to 161.95° .

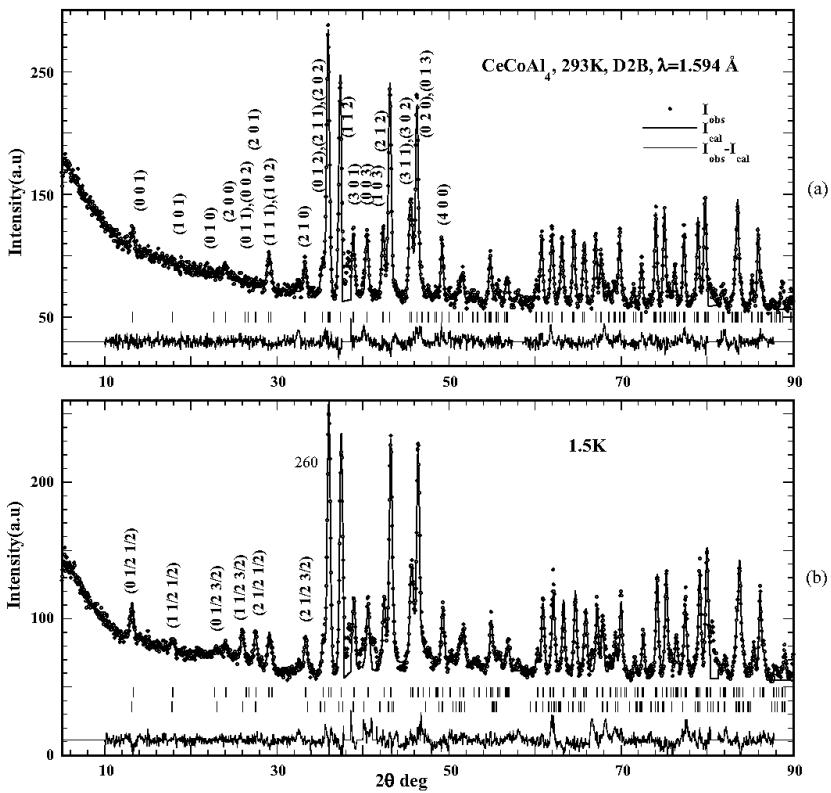


Figure 1. Observed and calculated D2B high resolution neutron powder diffraction patterns for CeCoAl₄, in the paramagnetic state at 293 K (a) and the magnetically ordered state at 1.5 K (b).

Single-crystal data were collected in the temperature range 4.2–60 K using the Laue diffractometer VIVALDI [14] at the Institut Laue-Langevin (ILL), Grenoble. The crystal is cut in the form of a rectangular pillar with a length of about 9 mm, a width of about 2.5 mm and a thickness of about 2.0 mm. This is the same single crystal as was used previously for magnetic measurements [9]. The Laue results showed that the length is approximately parallel to the *b* crystallographic axis, the width approximately parallel to *a* and the thickness approximately parallel to *c*.

All powder data sets were refined with the program FullProf [15] and the Laue diagrams were analysed using the LAUEGEN suite of programs [16].

1.2. Crystal structure

The structure refinements in the paramagnetic state revealed the presence of a few unidentified reflections, indicating small amounts of impurity phases present in both CeCoAl₄ powder samples. Those which did not overlap with the reflections of the main phase were subsequently excluded from the refinements. The refinement of the 293 and 15 K D2B data led to the fits shown in figure 1 and the parameters given in table 1, confirming that the powder had the CeCoAl₄ structure [1, 2], which is drawn in figure 2. In this structure the Ce atoms form double layers (MgCuAl₂-type slabs) perpendicular to the *c* direction. The Co atoms are located in the centres of the trigonal prisms formed by Al₂ and Al₃ atoms. As can be seen in figure 2, slabs

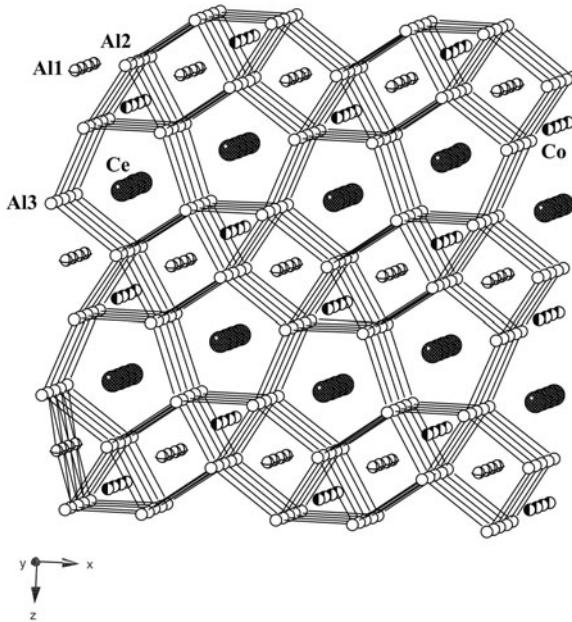


Figure 2. A three-dimensional schematic representation of the CeCoAl₄ structure viewed along [0 1 0], indicating the double-layer character of the Ce arrangement separated by the slabs of Al polyhedra perpendicular to the *c* axis.

Table 1. Refined structural parameters of CeCoAl₄ from powder neutron diffraction D2B data in the paramagnetic state at 15 and 293 K (*Pmma* space group). Ce and Co occupy the 2*e*: ($\frac{1}{4}0z$) positions and Al three sites: Al₁: 2*a* (0 0 0); Al₂: 2*f* ($\frac{1}{4}\frac{1}{2}z$); and Al₃: 4*j* ($x\frac{1}{2}z$). *B*_i is the individual temperature factor. *R*_B, *R*_{wp} and *R*_{exp} are the residuals for the Bragg, the weighted profile and the experimental integrated intensities respectively.

<i>Pmma</i> atom/site	D2B		15 K		D2B		293 K	
	<i>x</i>	<i>z</i>	<i>B</i> _i (Å) ²	<i>x</i>	<i>z</i>	<i>B</i> _i (Å) ²		
Ce: 2 <i>e</i>	0.25		0.3824(3)	0.42(3)	0.25		0.3819(3)	0.55(4)
Co: 2 <i>e</i>	0.25		0.8134(5)	0.02(3)	0.25		0.8134(5)	0.25(6)
Al ₁ : 2 <i>a</i>	0.0	0	0.38(2)	0.0			0	0.57(2)
Al ₂ : 2 <i>f</i>	0.25		0.0427(4)	0.38(2)	0.25		0.0427(4)	0.57(2)
Al ₃ : 4 <i>j</i>	0.0		0.7018(3)	0.38(2)	0.06		0.7018(3)	0.57(2)
<i>a</i> (Å)	7.6456(1)		7.6606(1)					
<i>b</i> (Å)	4.0459(1)		4.04689(1)					
<i>c</i> (Å)	6.8959(1)		6.9150(1)					
<i>R</i> _B , <i>R</i> _{wp} , <i>R</i> _{exp} (%)	5.5, 15, 7		6, 16.6, 13.6					

of the Al polyhedra that link (0 0 1) planes of Ce atoms separate the rare-earth double layers at distances of 6.90 Å along *c*.

1.3. The magnetic propagation vector

Magnetic ordering occurred below \sim 14 K, with the same magnetic peak topology in the neutron diffraction diagrams of both powder samples. The peaks were weak, as expected, but in contrast to the powder diffraction data taken for PrCoAl₄, their shapes were the same as those

of nearby nuclear reflections. Attempts to index the magnetic reflections using a generalized propagation vector did not produce a fit. We therefore used the Laue data to solve this problem experimentally.

In a neutron Laue diffraction diagram the recorded reflection intensities result from a projection of the nuclear and magnetic scattering densities in radial directions in reciprocal space. In the case of incommensurate magnetic structures (e.g. PrCoAl₄), this results in magnetic ‘satellite’ reflections being resolved from nuclear ‘fundamental’ reflections on the diagram. If the magnetic structure is commensurate, however, magnetic reflections will project onto the positions of nuclear reflections.

There was no evidence (by way of additional resolved reflections) in the CeCoAl₄ Laue diagrams taken between 4 and 60 K that the magnetic structure was incommensurate with the crystal lattice. Below about 13 K, however, there was a significant increase in the intensity observed at the positions of some ‘fundamental’ reflections. These increases were limited to $(h k \ell)$ reflections, which had h even and k and ℓ odd. This is consistent with CeCoAl₄ having a commensurate structure with a propagation vector $\mathbf{q} = (0 \frac{1}{2} \frac{1}{2})$, since satellite reflections in the Laue diagram with indices $(h k + \frac{1}{2} \ell + \frac{1}{2})$ overlap precisely with the fundamental reflections $(h' k' \ell') = (2h 2k + 1 2\ell + 1)$, i.e. h' even and k' and ℓ' odd.

Increases at the positions of the nuclear intensities are often difficult to measure accurately when the nuclear (fundamental) reflections are strong, but fortunately there were cases on the Laue diagrams where the nuclear reflections were observed to be weak or absent. This can occur either when the nuclear structure factor is small or the fundamental contribution is absent at a particular crystal setting in the case when there is insufficient flux in the incident neutron beam at the (short) wavelength required for that particular Bragg reflection.

This is illustrated in figure 3, which shows the same small portion of Laue diagrams taken at different temperatures but at the same crystal setting. At 12.9 K, above the Néel temperature, there is no significant intensity at the expected positions (marked by squares) of $(h' 1 1)$ fundamental reflections ($h' = 4$ (extreme left), 5, 6, 7 and 8 (extreme right)). This is because there are practically no neutrons with these wavelengths ($\lambda_{(411)} = 0.66 \text{ \AA}$ to $\lambda_{(811)} = 0.61 \text{ \AA}$) in the VIVALDI beam to excite these reflections at this crystal setting, although they do occur at other crystal settings with wavelengths nearer the peak flux. The superposed satellite reflections $(h \frac{1}{2} \frac{1}{2})$ with $h = 2, 3, 4$ and $\lambda = 1.32, 1.36$ and 1.21 \AA , however, are excited by neutrons near the peak of the incident wavelength distribution. They are absent when the sample is above the Néel temperature but increase in intensity as the temperature is reduced to 4.3 K.

1.4. Magnetic structure

The wavevector $\mathbf{q} = (0 \frac{1}{2} \frac{1}{2})$ was used to index successfully all magnetic reflections in both the D2B high resolution and G4.1 low resolution neutron powder patterns.

The wavevector $\mathbf{q} = (0 \frac{1}{2} \frac{1}{2})$ is at a high symmetry point in the Brillouin zone and the star of the wavevector (23 in the notation of [17]) has only one arm corresponding to the orthorhombic magnetic lattice A_C (16b) with the non-primitive translation $t = (0 \frac{1}{2} \frac{1}{2})$ and antitranslations $t' = (0 \frac{1}{2} 0)$ and $(0 0 \frac{1}{2})$. In this context, symmetry analysis can be of help for inserting parameter constraints in magnetic refinements. The propagation vector group G_q comprises eight elements. There are eight irreducible representations (*Irreps*) of *Pmma* associated with the wavevector $\mathbf{q} = (0 \frac{1}{2} \frac{1}{2})$ of dimension 1 as given in table 2a. The global magnetic representation calculated for the Wyckoff position 2e can be represented by $\Gamma_m = \Gamma_2 + \Gamma_3 + \Gamma_5 + \Gamma_6 + \Gamma_7 + \Gamma_8$ and contains the six basis functions 2–3 and 5–8. Since the two Ce atoms belong to the same orbit, this means that only uniaxial structures are allowed for Ce

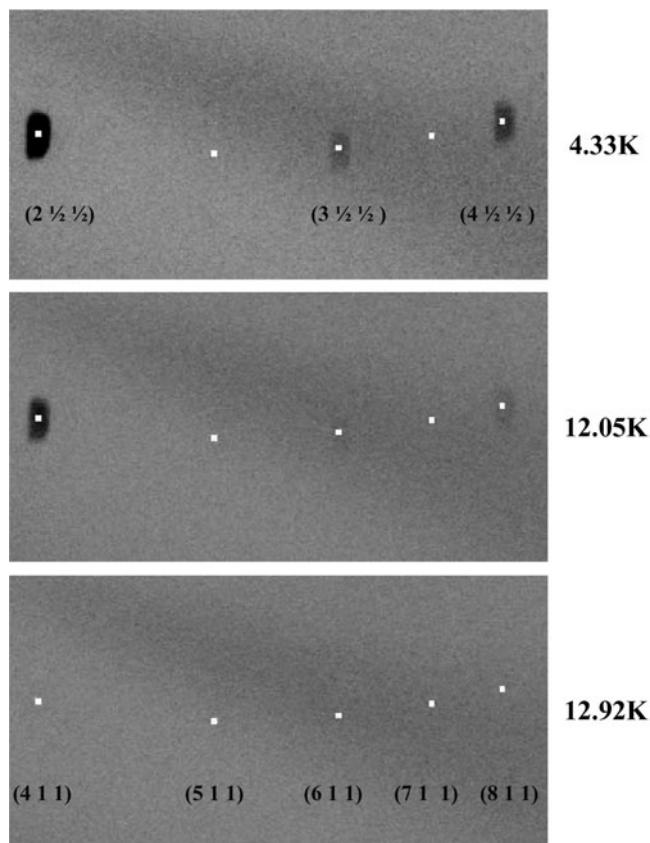


Figure 3. The Laue diffraction pattern of CeCoAl₄ single crystal at temperatures between 4.33 and 12.9 K showing the change in intensity of the magnetic satellite reflections $(2 \frac{1}{2} \frac{1}{2})$ (left), $(3 \frac{1}{2} \frac{1}{2})$ (centre) and $(4 \frac{1}{2} \frac{1}{2})$ (right-hand side). The calculated positions of nuclear reflections $(4 \bar{1} 1)$, $(5 \bar{1} 1)$, $(6 \bar{1} 1)$, $(7 \bar{1} 1)$ and $(8 \bar{1} 1)$, which are ‘tuned out’ at this crystal orientation, are indicated by squares.

Table 2a. Irreducible representations for $\mathbf{q} = (0 \frac{1}{2} \frac{1}{2})$ in *Pmma*.

	<i>E</i>	2_z	2_y	2_{1x}	$\bar{1}$	a_z	m_y	m_x
Γ_1	1	1	1	1	1	1	1	1
Γ_2	1	1	1	1	-1	-1	-1	-1
Γ_3	1	1	-1	-1	1	1	-1	-1
Γ_4	1	1	-1	-1	-1	-1	1	1
Γ_5	1	-1	1	-1	1	-1	1	-1
Γ_6	1	-1	1	-1	-1	1	-1	1
Γ_7	1	-1	-1	1	1	-1	-1	1
Γ_8	1	-1	-1	1	-1	1	1	-1

at site 2e. The corresponding magnetic modes are given in table 2b. The FullProf refinements of the D2B powder diffraction data led to the representation Γ_6 with the Ce atoms having their moments in parallel along the *b* axis, and moment values of 1.29(3) μ_B /Ce atom and 0.64(4) μ_B /Ce atom at 1.5 and 11 K respectively (table 3). The structure is displayed in figure 4

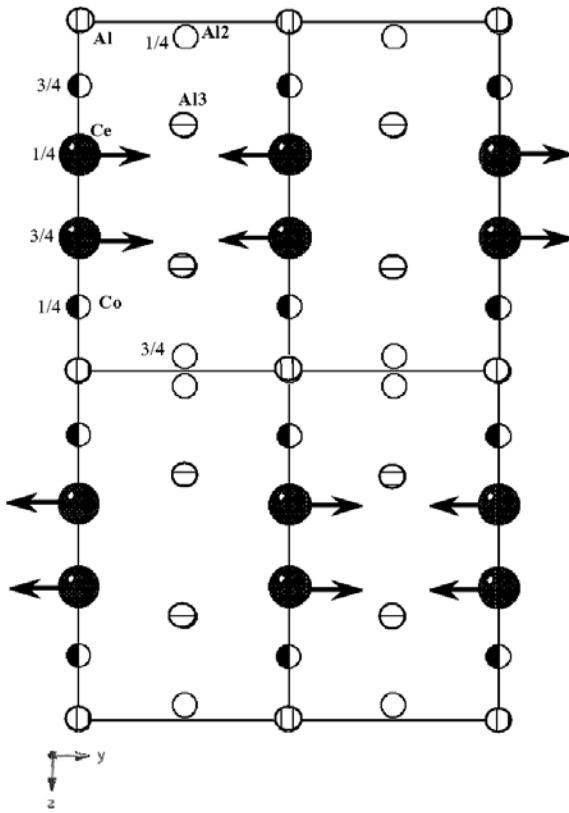


Figure 4. The collinear arrangement of the Ce magnetic moments in CeCoAl₄ associated with the wavevector $\mathbf{q} = (0 \frac{1}{2} \frac{1}{2})$. The magnetic moments point along the b axis and change sign with translations along the b and c directions. The magnetic cell is A_C —centred.

Table 2b. Basis functions for axial vectors bound to the Wyckoff site $2e$ in $Pmma$ for $\mathbf{q} = (0 \frac{1}{2} \frac{1}{2})$.

Atom site	x	y	z	Γ_2	Γ_3	Γ_5	Γ_6	Γ_7	Γ_8	v_0
Ce ₁	0.25	0	0.3815	w	w	v	u	u	v	
Ce ₂	0.75	0	0.6185	w	$-w$	$-v$	u	$-u$	v	

with a unit cell four times larger than the nuclear cell. The collinear magnetic structure can be simply described as an antiferromagnetic stacking of ferromagnetically coupled Ce sheets parallel to (0 1 1) planes. The coupling of the Ce double layers, which are perpendicular to the c axis (separated by a distance of 6.90 Å by Al polyhedra), is also antiferromagnetic.

1.5. Temperature dependence of the magnetic ordering

The temperature variation of the integrated intensity of the $(2 \frac{1}{2} \frac{1}{2})$ and $(4 \frac{1}{2} \frac{1}{2})$ reflections from the Laue diagrams is shown in figure 5, giving an ordering temperature of $T_N = 12.8(1)$ K. There is no evidence in figure 5 of any short range order above the Néel temperature, in contrast to that found in PrCoAl₄ [4]. Furthermore, no change of the peak position or of the relative magnetic intensities has been detected in either the single-crystal and/or G4.1 neutron powder

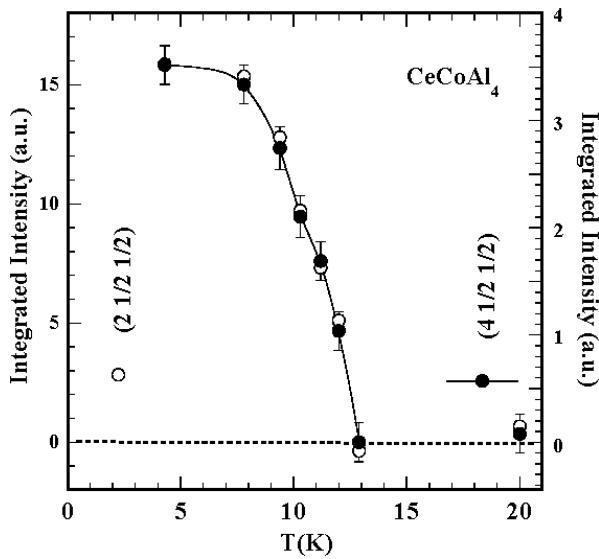


Figure 5. Thermal variation of the intensity of $(2 \frac{1}{2} \frac{1}{2})$ and $(4 \frac{1}{2} \frac{1}{2})$ reflections between 4 and 20 K measured from the Laue diagrams of CeCoAl_4 .

Table 3. Refined parameters of CeCoAl_4 from powder neutron diffraction D2B data in the magnetically ordered state at 1.5 and 11 K ($Pmma$ space group). Ce and Co occupy the $2e$: $(\frac{1}{4}, 0, z)$ positions and Al three sites: Al_1 : $2a$ $(0, 0, 0)$; Al_2 : $2f$ $(\frac{1}{4} \frac{1}{2} z)$; and Al_3 : $4j$ $(x \frac{1}{2} z)$. B_i is the individual temperature factor. R_B , R_m , R_{wp} and R_{exp} are the residuals for the Bragg, the magnetic, the weighted profile and the experimental integrated intensities. μ_y (μ_B) is the refined moment value of the Ce^{3+} atom along the b axis with the wavevector $\mathbf{q} = (0 \frac{1}{2} \frac{1}{2})$.

$Pmma$ atom/site	D2B		1.5 K		D2B		11 K	
	x	z	B_i (\AA^2)	x	z	B_i (\AA^2)		
Ce: $2e$	0.25	0.3836(3)	0.16(3)	0.25	0.3817(3)	0.47(3)		
Co: $2e$	0.25	0.8158(6)	0.03(4)	0.25	0.8133(5)	0.02(5)		
Al_1 : $2a$	0	0	0.23(2)	0	0	0.41(2)		
Al_2 : $2f$	0.25	0.0422(4)	0.23(2)	0.25	0.0429(4)	0.41(2)		
Al_3 : $4j$	0.0	0.7018(3)	0.13(2)	0.0	0.7014(3)	0.41(2)		
μ_y (μ_B)	1.29(3)				0.64(4)			
a (\AA)	7.6518(1)				7.6458(1)			
b (\AA)	4.0488(1)				4.0458(1)			
c (\AA)	6.9010(1)				6.8958(1)			
R_B , R_m (%)	6.8, 9				6.7, 15			
R_{wp} , R_{exp} (%)	16.4, 11				15.2, 7.3			

patterns over the entire magnetically ordered regime. This indicates that the collinear magnetic moment arrangement persists over the entire range.

Using this model, moment values derived from several magnetic satellites in the G4.1 powder data are shown as a function of temperature in figure 6. They demonstrate that long range ordering of the Ce moments sets in below ~ 14 K, in agreement with the magnetization, specific heat and single-crystal Laue measurements.

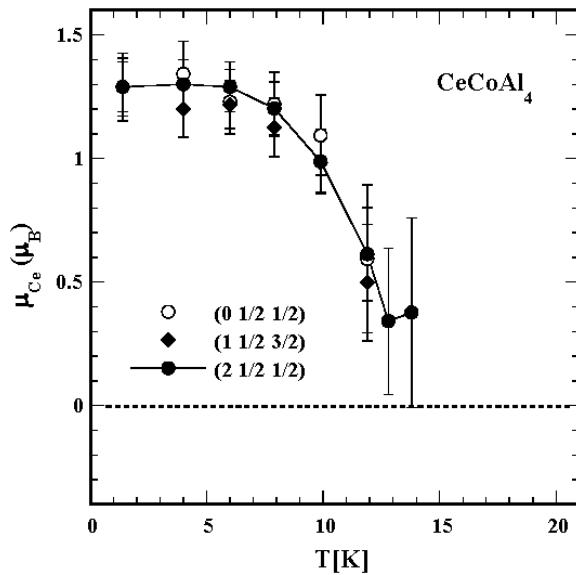


Figure 6. The thermal variation of the amplitude of the magnetic moment of the Ce atoms derived from the $(0 \frac{1}{2} \frac{1}{2})$, $(1 \frac{1}{2} \frac{3}{2})$ and $(2 \frac{1}{2} \frac{1}{2})$ satellite reflections in the G4.1 powder diffraction data, showing the onset of magnetic order at ~ 14 K in CeCoAl₄.

2. Discussion

Within the Ce double layer normal to the *c* axis this arrangement corresponds to antiferromagnetic nearest neighbour interactions between atoms at a distance of 4.05 Å along the *b* axis. Next nearest neighbour Ce atoms, which are related by a centre of symmetry, are separated by 4.15 Å in a direction making an angle of 24° with the *a* axis in the *a*–*c* plane. Their coupling is ferromagnetic, as is the coupling between Ce atoms which are separated by 7.63 Å along the *a* axis. This is the difficult axis of magnetization observed in the magnetic measurements [9].

In a previous investigation Dhar *et al* [11], on the basis of electrical resistivity measurements, proposed the existence of an incommensurate antiferromagnetic structure for CeCoAl₄. The present investigation has clearly shown that this compound has a commensurate collinear magnetic structure, with the moments oriented along the *b* direction in a cell four times larger associated with the wavevector $\mathbf{q} = (0 \frac{1}{2} \frac{1}{2})$. The refined magnetic moment value at 1.5 K is 1.29(3) μ_{B} /Ce atom, which is significantly smaller than the Ce³⁺ free ion $gJ \mu_{\text{B}}$ value of 2.14 μ_{B} and could indeed be taken as an indication of the presence of a Kondo lattice as suggested by Koterlin *et al* [10]. On the other hand, this value could also be interpreted as arising through crystal field effects. Since Ce³⁺ is a Kramers ion, the associated point symmetry indicates a crystal field splitting of the ground state manifold into three doublets. Of these, the $|\pm 3/2\rangle$ doublet has an ordered moment value equal to 1.29 μ_{B} /Ce atom, which is identical to the experimentally observed ordered moment value. A vital clue for a more complete comprehension of the magnetic ground state of this interesting system is via a measurement of the dynamic magnetic response at low temperatures. For this purpose, inelastic neutron scattering measurements of the crystal field excitations and associated energy levels are under way.

Finally we wish to discuss the results of magnetic measurements made on a CeCoAl₄ single crystal [9]. These measurements showed the presence of a spin flop transition at a

critical field of 7.5 T at 4.2 K when the field was applied along the *c* direction. It is well known that spin flop transitions are generally observed for applied field directions that are parallel (and antiparallel) to the moment directions in antiferromagnets. As a consequence, the *c* direction has previously [9] been identified as the preferred moment direction in CeCoAl₄, which is in contrast with the present neutron diffraction data (the *b* direction is the preferred moment direction; see section 1.4). Fortunately, the present neutron diffraction data were obtained from the same single crystal as was used for the aforementioned magnetic measurements. In the magnetic measurements the spin flop transition was observed when the field was applied in the length direction of the crystal, a direction that was formerly erroneously attributed as the *c* direction from x-ray Laue data. However, the present neutron data show that the length direction actually corresponds to the *b* direction. Physically, this is a most satisfactory result. It means that a spin flop transition was observed for an applied field direction parallel to the main antiferromagnetic moment direction, as expected.

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