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Antiferromagnetism of the Kondo lattice compound $CeCu_2$ studied by neutron polarimetry

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Abstract. Single-crystal neutron diffraction measurements on the Kondo lattice compound $CeCu_2$, using a zero-field neutron polarimeter have confirmed that $CeCu_2$ orders at T_N = 3.5 K to an antiferromagnetic structure. The sublattice magnetization is not saturated at the lowest temperature, 2.5 K, attained during the experiment. The magnetic moments of Ce atoms are $0.33(2)\mu_B$ at 2.5 K and are oriented parallel and antiparallel to the orthorhombic *c* axis.

1. Introduction

The intermetallic compound $CeCu_2$ has been extensively investigated owing to its interesting Kondo lattice behaviour [1–7]. $CeCu_2$ orders at T_N = 3.5 K and shows highly anisotropic magnetic behaviour due to crystal field effects. The magnetic moments of the Ce atoms are strongly reduced by the Kondo effect from the single-ion value. Neutron diffraction investigations on polycrystalline samples [2] showed magnetic scattering superimposed on nuclear reflections below T_N but failed to determine the magnetic propagation vector and the spin structure. A long-period modulated magnetic structure was presumed in analogy to the incommensurate magnetic structure of $CeAl_2$ [1, 2]. Our recent unpolarized neutron diffraction investigations [8] have clearly established that $CeCu_2$ has in fact a simple antiferromagnetic structure with the propagation vector $\boldsymbol{k} = (0, 0, 0)$, and not a complicated modulated structure as previously conjectured. However, the small Ce magnetic moment and the superimposition of the consequently small magnetic intensities on relatively strong nuclear intensities prevented an unambiguous determination of the spin directions of the Ce atoms. Unpolarized neutron diffraction intensities could not distinguish between the two models in which magnetic moments are oriented parallel to the $\boldsymbol{b}$ and $\boldsymbol{c}$ axes, respectively. However, these data exclude the possibility that the spins are oriented parallel to the $\boldsymbol{a}$ axis (space group *Imma*, $a = 4.43$, $b = 7.05$, $c = 6.45$ Å at room temperature). To determine the spin orientations of Ce atoms uniquely we have studied the magnetic structure of $CeCu_2$ by

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polarized neutron diffraction using the newly developed technique: zero-field neutron polarimetry.

2. Experimental technique

Large single crystals of $CeCu_2$ with linear dimensions of a few centimetres were grown by the Czochralski method. A small single crystal of size of $2 \times 2 \times 4$ mm^3 cut out of a larger crystal was used for the present investigations. The edges of the rectangular parallelepiped-shaped crystal were parallel to the three orthorhombic crystallographic directions. The crystal was carefully aligned and mounted with its [100] axis vertical in the zero-field polarimeter CRYOPAD on the IN20 polarized beam triple-axis spectrometer of the Institut Laue–Langevin, Grenoble. The principle and operation of the CRYOPAD have been fully described elsewhere [9–11]. In short, it allows the input neutron beam polarization to be set to any desired angle and both the magnitude and direction of the polarization in a diffracted beam to be determined. The diffracting crystal is kept in a field-free region and its temperature can be maintained in the range 2–315 K. The Heusler alloy monochromator and the analyser crystal of the IN20 spectrometer were set to a wavelength of 1.532 Å. Although the CRYOPAD allows the input polarization $\boldsymbol{P}_i$ to be set to any desired angle, it is convenient from the point of view of subsequent analysis to make scans in which $\boldsymbol{P}_i$ is varied in the three principal planes defined by the axes X (parallel to the scattering vector $\boldsymbol{\kappa}$, Z (vertical) and Y (completing the right-handed set).

3. Magnetic scattering from $ClCu_2$

$CeCu_2$ crystallizes [12] in the body centred space group *Imma*. There are four formula units per unit cell. In the primitive unit cell however, there are only two formula units and therefore only two Ce magnetic sublattices. The two Ce atoms of the primitive unit cell related by the centre of symmetry are antiferromagnetically coupled. The magnetic structure factor of $CeCu_2$ is therefore purely imaginary. Assuming that the orthorhombic symmetry is retained in the magnetic structure, the magnetic moments may be parallel and antiparallel to any of the three orthorhombic axes a, b and c. The intensities of the magnetic reflections measured during the unpolarized neutron diffraction investigations preclude the a-axis orientation. However, although these intensities together with the results of susceptibility measurements [8], favour c-axis orientation, they cannot really rule out a structure with magnetic moments parallel to the b axis. In the following we will show that zero-field neutron polarimetry may be used to distinguish between the b- and c-axis models and in fact favours the c-axis model.

With the a axis vertical (parallel to Z) in the polarized neutron experiment, the magnetic structure factors $\boldsymbol{M}(\boldsymbol{\kappa})$ for both the b- and c-axis orientations are in the horizontal X–Y plane. The magnetic interaction vector $\boldsymbol{Q}(\boldsymbol{\kappa})$, which is the projection of the magnetic structure factor $\boldsymbol{M}(\boldsymbol{\kappa})$ onto the plane perpendicular to the scattering vector $\boldsymbol{\kappa}$, is therefore along Y. Table 1 shows the magnetic interaction vectors for a few reflections for both spin orientations. From this table it can be seen that the two models should be distinguishable by analysing the polarization scattered by the 002 reflection for which the magnetic scattering is zero if the moments are along the c axis. In the b-axis model, $\boldsymbol{Q}$ is non-zero, so the polarization should be changed by the scattering

Table 1. Y and Z components of the magnetic interaction vector $\boldsymbol{Q}$ for magnetic moments parallel to the c and b axes, respectively. Re and Im refer to the real and imaginary parts. The X axis is parallel to the scattering vector, Z is vertical and Y completes the right-handed set.

	h	k	l	θ	Re Q_y	Im Q_y	Re Q_z	Im Q_z
Magnetic moments parallel to the c axis	0	1	1	8.27	0.0000	1.1378	0.0000	0.0000
	0	2	0	11.70	0.0000	0.0000	0.0000	0.0000
	0	0	2	11.77	0.0000	0.0000	0.0000	0.0000
	0	2	2	16.72	0.0000	0.4682	0.0000	0.0000
	0	3	1	18.72	0.0000	−1.3186	0.0000	0.0000
	0	1	3	18.81	0.0000	0.3441	0.0000	0.0000
Magnetic moments parallel to the b axis	0	1	1	8.27	0.0000	1.1442	0.0000	0.0000
	0	2	0	11.70	0.0000	0.0000	0.0000	0.0000
	0	0	2	11.77	0.0000	−0.7152	0.0000	0.0000
	0	2	2	16.72	0.0000	0.4708	0.0000	0.0000
	0	3	1	18.72	0.0000	−0.4420	0.0000	0.0000
	0	1	3	18.81	0.0000	1.0381	0.0000	0.0000

process. For scattering from a monodomain sample the polarization is preserved in magnitude but precesses around the magnetic interaction vector by an angle φ given by [11]

$$\cos\varphi = (1-\gamma^2)/(1+\gamma^2) \qquad \text{where } \gamma^2 = \boldsymbol{Q}^*\boldsymbol{Q}/N^*N \tag{1}$$

where N is the nuclear structure factor. If there are 180° domains in the sample, as is possible for a magnetic structure with zero propagation vector, then, in general, depolarization will occur with an accompanying reduction in the precession angle, φ. The direction of the precession for the two domains is opposite giving rise to depolarization of the scattered beam and a reduced precession angle which becomes zero when the two domain populations are equal. The depolarization takes place in the components of $\boldsymbol{P}_i$ perpendicular to $\boldsymbol{Q}$ and is given by

$$\boldsymbol{P}_f = \boldsymbol{P}_i[1-(1-\varepsilon^2)\cos^2\zeta\sin^2\varphi]^{1/2} \tag{2}$$

where φ is given by (1), ζ is the angle between $\boldsymbol{P}_i$ and $\boldsymbol{Q}$ and ε is the difference between the fractional populations of the two domains.

4. Results of zero-field neutron polarimetry

Figure 1 shows the temperature variation of the component of polarization scattered without direction change by the 011 reflection for incident polarization set along the X, Y and Z axes defined in section 2. In the X and Z directions the polarization decreases linearly as the temperature is lowered below the Néel temperature $T_N = 3.5$ K. This linear decrease in polarization corresponds to the linear increase of the intensity of the 011 reflection observed in the unpolarized neutron diffraction experiment. The magnitude of polarization does not level off at $T = 2.25$ K, the minimum temperature attained during the experiment. This is consistent with the unpolarized neutron diffraction investigations which also failed to show saturation of the intensity of the 011

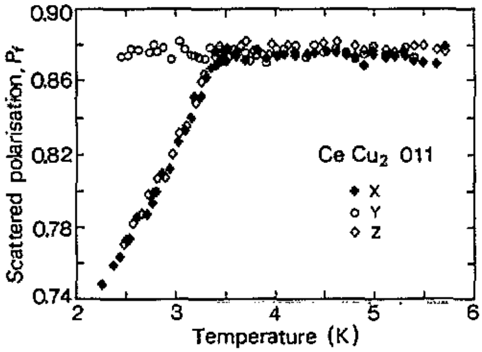

Figure 1. Temperature variation of the component of polarization scattered without change of direction by the 011 reflection for incident polarization set along X, Y and Z axes. The X axis is parallel to the scattering vector, Z is vertical and Y completes the right-handed set.

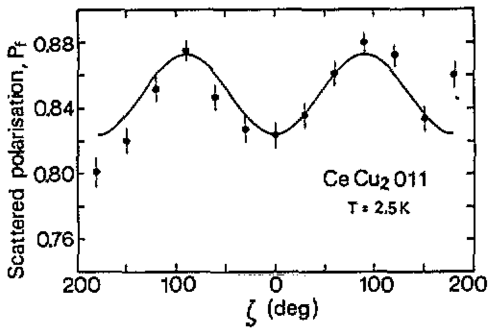

Figure 2. Polarization scattered by the 011 reflection plotted as a function of the angle ζ between P_i and Y for different directions of P_i in the X–Y plane (Z scan). The axes are defined as for figure 1.

reflection even at $T = 1.6$ K. Thus the sublattice magnetization of $CeCu_2$ must saturate at a much lower temperature. The polarization along the Y axis remains constant in the whole temperature range investigated, indicating that the magnetic interaction vector of the 011 reflection is along Y and confirming that the moment direction is in the b–c plane. The temperature variation of the polarization given in figure 1 can be used to find the critical exponent β of the sublattice magnetization m from the equation

$$m = m_0(1 - T/T_N)^\beta \tag{3}$$

noting that the sublattice magnetization m can be related to the magnitude of the initial polarization P_i and the scattered polarization P_f along P_i by the equation

$$m^2 = \gamma^2 N^2/a^2 \qquad \text{with } \gamma^2 = (P_i - P_f)/(P_i + P_f) \tag{4}$$

where $a = \cos\tan^{-1}(lc^*/kb^*)$. A least-squares fit yields a critical exponent $\beta = 0.34(9)$. This value agrees with both the three-dimensional Ising value 0.312 and three-dimensional Heisenberg value 0.38 within experimental error. Figure 2 shows the polarization plotted as a function different directions of P_i in the X–Y plane. The continuous curve is a least-squares fit of the data to (2). The temperature of the sample drifted during this scan from 2.17 K to 2.79 K which explains why the actual minimum values in the polarization increase continuously during the scan. This result also confirms that the magnetic interaction vector Q is parallel to Y.

To distinguish between the c- and the b-axis models of the magnetic structure of $CeCu_2$, we investigated the polarization scattered by the 002 reflection. If the magnetic moments of the Ce atoms are oriented parallel to the b axis, the beam scattered by the 002 reflection should be depolarized in the same way as was observed for the 011 reflection. However, if the magnetic moments are parallel to the c axis, no depolarization effects are expected. Figure 3 shows the polarization of the 002 reflection in the Z and X scans, respectively. No depolarization of the scattered beam is observed in these scans within the experimental accuracy. From this we conclude that the 002 reflection contains no magnetic scattering and therefore that the magnetic moments of Ce atoms in $CeCu_2$ are oriented parallel to the c axis. If there is a component of moment parallel to the b

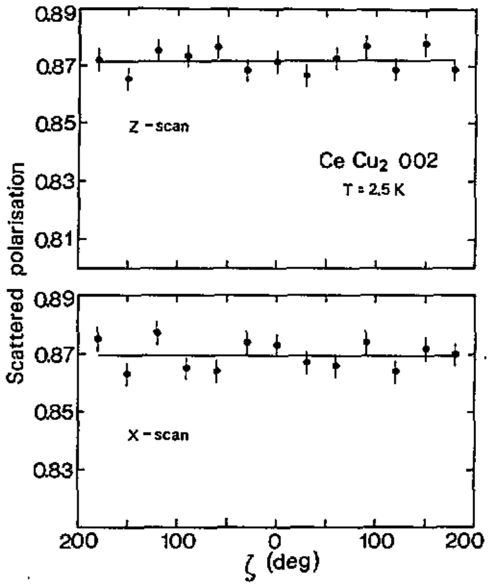

Figure 3. Polarization scattered by the 002 reflection plotted as functions of the angle ζ between $\boldsymbol{P}_i$ and Y in the X–Y plane (Z scan) and also Y–Z plane (X scan).

axis, it must be less than $0.07\mu_B$. Assuming this model, the least-squares fit gives a magnetic moment of $0.33(2)\mu_B$ per Ce atom at $T = 2.5$ K. At this temperature the sublattice magnetization has not attained its saturation value (figure 1). Also, because of the Kondo effect, the saturated magnetic moment of the Ce atoms is expected to be lower than the free-ion value. The magnetic moment determined during the present investigations is in agreement with the results of unpolarized neutron diffraction [13].

5. Conclusions

A study of some magnetic reflections from $CeCu_2$ with zero-field neutron polarimetry shows that the magnetic moments of Ce atoms in the antiferromagnetic structure are parallel and antiparallel to the crystallographic c axis. This experiment demonstrates that zero-field neutron polarimetry can be used to elucidate magnetic structures for which unpolarized neutron diffraction fails to give a unique solution.

References

[1] Gratz E, Bauer E, Barbara B, Zemirli S, Steglich F, Bredl C D and Lieke W 1985 *J. Phys. F: Met. Phys.* **15** 1975
[2] Zemirli S 1985 *PhD thesis* Grenoble
[3] Loewenhaupt M, Prager M, Gratz E and Frick B 1988 *J. Magn. Magn. Mater.* **76, 77** 415
Loewenhaupt M, Gratz E, Pillmayr N and Müller H 1990 *Physica* B **136** 427
[4] Onuki Y, Fukada A, Ukon I, Umehara I, Satoh K and Kurosawa Y 1990 *Physica* B **163** 600
[5] Takayanagi S, Umehara I, Satoh K, Onuki Y and Wada N 1990 *J. Magn. Magn. Mater.* **90, 91** 479
[6] Satoh K, Umehara I, Fukada A, Kurosawa Y, Onuki Y and Maezawa K 1990 *J. Magn. Magn. Mater.* **90, 91** 141
[7] Uwatoko Y, Okita H, Oomi G, Umehara I and Onuki Y 1990 *J. Magn. Magn. Mater.* **90, 91** 485
[8] Trump R, Thierfeldt S, Loewenhaupt M and Chattopadhyay T 1991 *J. Appl. Phys.* **69** 4699

[9] Tasset F 1989 *Physica* B **156, 157** 627
[10] Tasset F, Brown P J and Forsyth J B 1988 *J. Appl. Phys.* **63** 3606
[11] Nunez V 1991 *PhD thesis* University of London
[12] Storm A R and Benson K E 1963 *Acta Crystallogr.* **16** 701
[13] Trump R 1991 *PhD thesis* Köln University