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Magnetic structure of the Kondo lattice compound $CeCu_2$

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The magnetic structure of the Kondo lattice compound $CeCu_2$ has been investigated by unpolarized and polarized neutron diffraction. $CeCu_2$ orders at $T_N = 3.5$ K to an antiferromagnetic structure with the wave vector $\mathbf{k} = (0,0,0)$. Two Ce atoms of the primitive unit cell have oppositely oriented spin directions which are parallel and antiparallel to the c axis. The susceptibility measurements show strong anisotropic magnetic behavior which may be attributed to the crystal field. Only the susceptibility along the c axis shows a broad maximum at the Néel temperature. The magnetization curve with the magnetic field parallel to the a axis shows a metamagnetic behavior at 1.1 T at 2 K.

I. INTRODUCTION

$CeCu_2$ is a Kondo lattice compound which shows magnetic ordering at $T_N = 3.5$ K.[1] The exact nature of the magnetic ordering in this compound is, however, not determined. Neutron-diffraction investigations on polycrystalline samples showed magnetic scattering superimposed on nuclear reflections below T_N.[2] These powder neutron-diffraction investigations failed to determine the magnetic wave vector and spin structure. A long-period-modulated magnetic structure was presumed in analogy to the incommensurate magnetic structure of $CeAl_2$. We planned systematic investigations of the magnetic properties of this interesting Kondo lattice compound by elastic and inelastic neutron scattering and also by susceptibility and magnetization measurements after large good-quality single crystals were successfully grown in our laboratory. In this paper we report the results of unpolarized and polarized neutron-diffraction investigations of the magnetic structure of $CeCu_2$ and also the results of susceptibility and magnetization investigations of the magnetic anisotropy.

II. EXPERIMENTAL RESULTS AND DISCUSSION

Large single crystals of $CeCu_2$ of linear dimension of a few centimeters were grown by the Czochralski method. Small crystals of size of $2\times2\times4$ mm^3 were cut out of large single crystals for neutron-diffraction investigations. For susceptibility and magnetization investigations, crystals of similar size were used. The edges of the rectangular parallelepiped-shaped crystals were parallel to the three orthorhombic crystallographic directions. Unpolarized neutron-diffraction investigations were performed with the D15 diffractometer of the Institut Laue-Langevin, Grenoble. The crystal was attached to the sample stick of a helium cryostat with its a axis parallel to the ω axis of the diffractometer. The detector could be lifted by an angle $-8° < x < 20°$, thus enabling one to measure reflections (hkl) with $h > 0$. A second single crystal with its b axis parallel to the ω axis of the diffractometer was also investigated. Susceptibility and magnetization measurements were performed with a standard SHE 800 SQUID magnetometer.

Figure 1 shows the temperature variation of the intensity of (011) and (110) reflections close to the Néel temperature. Both nuclear and magnetic scattering contribute to the (011) reflection, the magnetic contribution being only about 8% of the nuclear intensity at $T = 2$ K, decreases almost linearly with increasing temperature, and becomes zero at about 3.5 K. The (110) reflection, on the other hand, is purely of magnetic origin because the nuclear (110) reflection is forbidden in the orthorhombic space group *Imma* of the crystal structure ($a = 0.442$, $b = 0.704$, $c = 0.745$ nm at room temperature). The temperature variation of this reflection is consistent with the aforementioned fact. Its intensity decreases continuously with increasing temperature and becomes zero at about 3.5 K. This clearly shows that $CeCu_2$ orders antiferromagnetically at $T_N = 3.5$ K with a magnetic wave vector $\mathbf{k} = (0,0,0)$. The magnetic unit cell is the same as the nuclear unit cell. Since there are only two Ce atoms in the primitive nuclear unit cell, there is only a unique possible magnetic coupling between them, viz., antiferromagnetic. We have measured the temperature variation of several other reflections. The intensities of the magnetic part of these reflections are consistent with the magnetic structure model in which the magnetic moments of the Ce atoms are parallel and antiparallel to the orthorhombic c axis of the crystal (Fig. 2 shows the magnetic structure). However, one should note that the measurement of the relatively weak magnetic contribution to the total intensity is not very accurate and the extinction might change due to the phase transition. The magnetic phase transition in $CeCu_2$ is clearly of second order, and therefore magnetoelastic effects are expected to be small, but cannot be ruled out. It is therefore not quite certain that the orientations of the magnetic moment are parallel and antiparallel to the c axis. A magnetic structure model with the magnetic moments parallel to the b axis which gives the second best fit with the observed magnetic intensities, however, cannot be ruled out by this unpolarized neutron-diffraction investigation.

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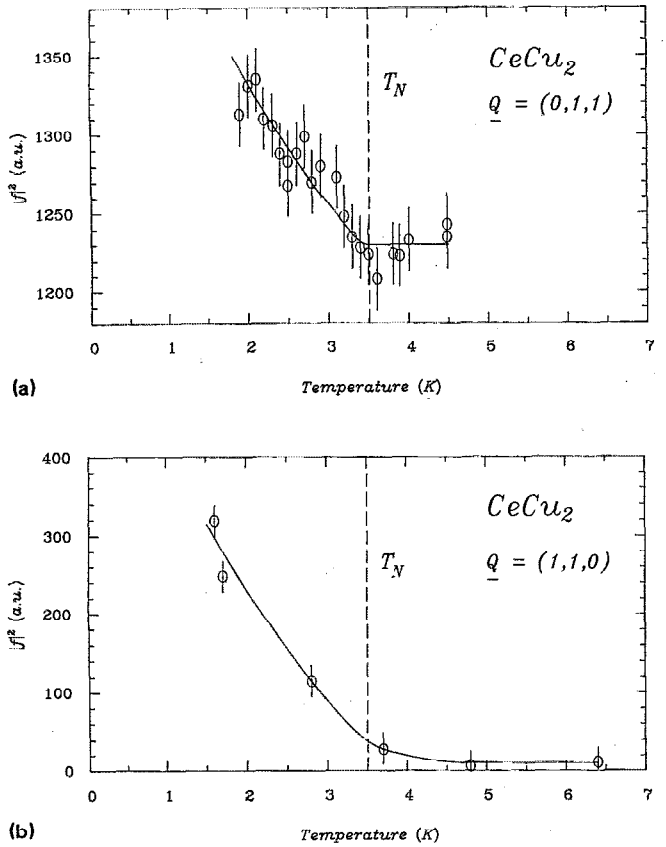

FIG. 1. Temperature variation of the intensity of (a) (011) and (b) (110) reflections close to T_N.

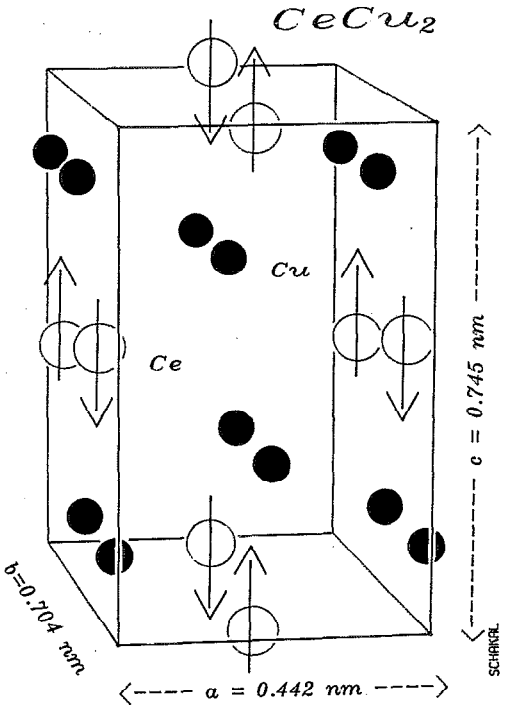

FIG. 2. Proposed magnetic structure of $CeCu_2$.

Polarization analysis of the Bragg scattering of polarized neutrons from superimposed magnetic and nuclear reflections allows the ratio of magnetic to nuclear scattering to be determined uniquely. We have therefore performed neutron-diffraction experiments with polarized neutrons using the generalized polarization analysis device CRYOPAD (Ref. 3) on the triple-axis spectrometer IN20 of the Institut Laue-Langevin in the elastic mode. The ability of CRYOPAD to choose the incident polarization direction independent of the orientation of the crystal on the spectrometer enables the magnetic structure to be determined by measurements of just a few reflections. Details of this investigation will be published elsewhere. This investigation has established that the above-mentioned magnetic structure (Fig. 2) is the correct one.

Figure 3 shows the temperature variation of the magnetic susceptibility of $CeCu_2$ along the a, b, and c axes measured in a SQUID magnetometer by the application of a magnetic field of 0.2 T. Highly anisotropic magnetic behavior of $CeCu_2$ is evident from this figure. The susceptibility along the a axis is by one order of magnitude larger than that along the b and c axes. The susceptibility along the c axis is larger than that along the b axis by a factor of about 3. The susceptibility along the b and a axes still continues to increase as the temperature is lowered below the Néel temperature. Only along the c axis does the susceptibility show a broad maximum at T_N, indicating that the magnetic moments are parallel and antiparallel to the c axis in agreement with the neutron results. However, the susceptibility along the c axis remains finite and does not become zero when extrapolated to $T = 0$. The reason for this is not understood. Figure 4 shows the magnetization of $CeCu_2$ along the a, b, and c axes at $T = 2$ K. At $H = 1.1$ T the magnetization along the a axis shows a change of slope, indicating a metamagnetic phase transition at this magnetic field. The magnetization curves along the b and c axes do not show any change of slope at this field. The magnetization is highly anisotropic in the ordered and also in the paramagnetic state due to crystal-field effects.[4,5] The

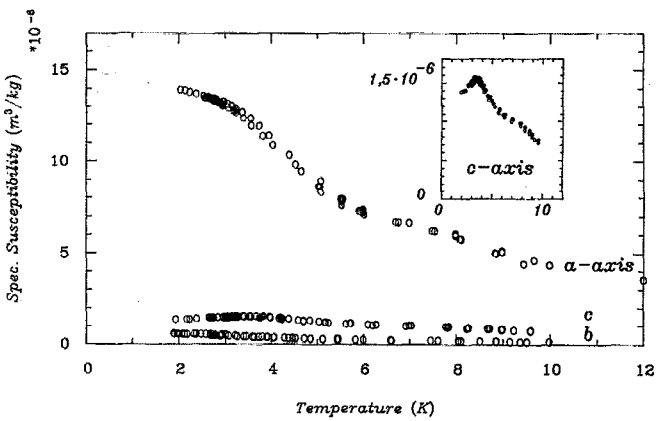

FIG. 3. Temperature variation of the magnetic susceptibility of $CeCu_2$ at 0.2 T.

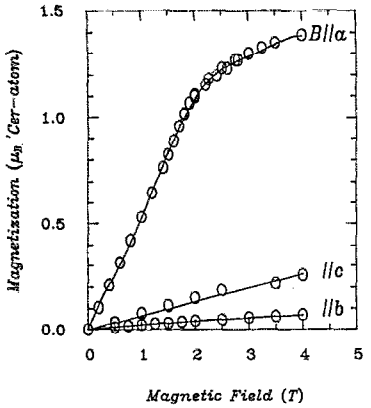

FIG. 4. Magnetization of $CeCu_2$ at 2 K.

present susceptibility and magnetization results agree well with that observed by Ōnuki *et al.*[6]

III. CONCLUSION

The Kondo lattice compound $CeCu_2$ has in fact a simple antiferromagnetic structure and not a modulated complicated structure as previously conjectured. The magnetic properties of $CeCu_2$ are highly anisotropic due to crystal-field effects.

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