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# Magnetic properties of cerium monotelluride

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**Résumé.** — Nous avons effectué, sur des monocristaux de CeTe, des mesures d'aimantation et de susceptibilité magnétique, ainsi que des expériences de diffraction neutronique afin d'étudier les propriétés magnétiques de ce composé à basse température. Nous observons à $T < 2$ K une transition métamagnétique pour un champ magnétique de l'ordre de 18 kOe, associée à un cycle d'hystérésis. Par diffraction de neutrons nous déterminons un moment magnétique $\mu = 0{,}2 \pm 0{,}1\ \mu_B$/Ce étonnamment faible et différent du moment magnétique déduit des mesures d'aimantation en champs forts.

**Abstract.** — Magnetization, magnetic susceptibility and neutron scattering measurements have been performed on CeTe single crystals, in order to study the low temperature magnetic properties. We observe at $T < 2$ K a metamagnetic transition for a magnetic field of about 18 kOe, associated with an hysteresis cycle. By neutron scattering measurements we determine a magnetic moment $\mu = 0.2 \pm 0.1\ \mu_B$/Ce surprisingly low and different from the value deduced from the high field magnetization measurements.

Among the compounds of the trivalent cerium, the monopnictides and monochalcogenides series have great interest. These compounds have a simple crystallographic structure (all are face centred cubic at room temperature) associated with a variety of magnetic behaviour. Previous investigations have principaly been concentrated on the monopnictides. In this last series, the magnetic properties are strongly influenced by the cubic crystal field. In CeX (X = P, As, Sb, Bi), the ground state multiplet of the $Ce^{3+}$ ion, $^2F_{5/2}$, is splitted by the crystal field into a $\Gamma_7$ doublet and a $\Gamma_8$ quartet. This crystal field splitting varies from about 200 K for CeP to approximately 10 K for CeBi and the Néel temperature from 8 K for CeAs to 25 K for CeBi [1]. Thus in CeP and CeAs the crystal field energy is higher than the exchange energy, but in CeSb and CeBi the two energies are similar. This is the origin of the complicated magnetic behaviour of these two compounds.

For the cerium monochalcogenides, very few studies have been done. Loginov *et al.* [2] have shown that polycrystalline samples of CeS, CeSe and CeTe order antiferromagnetically at low temperatures, but for this two last compounds, the reported Néel temperatures are not in agreement with the values reported by Hulliger *et al.* [3]. From specific heat, thermal expansion, magnetic susceptibility and magnetization measurements, these last authors have found that in the monochalcogenides the variation of the crystal field splitting, $\Delta$, is similar to that in the monopnictides series : $\Delta$ varies from 130 K-150 K for CeS to 20 K-40 K for CeTe. However, in these compounds, unlike of the monopnictides, the Néel temperature decreases when the anion size increases. So, in the CeX (X = S, Se, Te) series, CeTe seems to have an abnormal behaviour.

In order to get more information about the magnetic properties of CeTe, magnetization, magnetic suscep-

tibility and neutron diffraction measurements have been performed at very low temperatures and the main results are presented in this paper.

1. **Samples.** — Our samples were obtained by direct reaction of required amounts of cerium and tellurium in sealed molybdenum crucibles under an argon atmosphere. Starting materials are 6N tellurium from Koch-light laboratories and 3N cerium metal from Leico. Single crystals are obtained by sublimation at the top of the crucible during a slow cooling (4 °C/h) from a maximal temperature of 2 000 °C. Synthesis and growth are made in the same crucible to avoid any loss of product. Single crystals of typically $1 \times 1 \times 3\ mm^3$ have been cleaved from the crystallized mass.

From X-ray measurements, our samples are monophase, with a rock-salt structure and a lattice parameter $a = 6.361 \pm 0.005$ Å. Previous published results do not allow to determine the correlation between the lattice parameter and the composition. So, although the lattice constants of our samples are in good agreement with the previous data, no exact information on the composition can be deduced from it. On the other hand, the results of Hulliger *et al.* [3] show no correlation between the lattice parameter and the physical properties as the Néel and paramagnetic Curie temperatures. Density measurements and chemical analysis of samples with various physical properties are needed to understand this problem.

Microprobe analysis of our CeTe samples gives a stoichiometric composition Ce/Te = $1.00 \pm 0.02$ and a good homogeneity. This result is consistent with the susceptibility measurements described below.

2. **Magnetic measurements.** — Susceptibility measurements between 4.2 K and room temperature, in magnetic field lower than 4 kOe, show that CeTe follows a Curie-Weiss law in the entire paramagnetic region. No crystal field anomaly has been detected even at low temperatures. From the high temperature ($T > 100$ K) results we deduce a paramagnetic Curie temperature $\theta_p = -5$ K, in good agreement with previous data [3], and an effective magnetic moment $\mu = 2.57\ \mu_B$/Ce, which is close to the free ion value ($2.54\ \mu_B$/Ce) indicating a good stoichiometry of our sample. This value is somewhat higher than value obtained by Hulliger *et al.* [3]. No magnetic order has been evidenced in the temperature range 4.2 K-300 K in disagreement with the result of Loginov *et al.* [2].

The magnetization has been measured by the extraction method in a magnetic field up to 80 kOe, in an adiabatic cryostat. The magnetic field is along a [001] direction. Figure 1 shows the magnetization curves for two different samples cleaved from the same mass. For clarity the curves for the temperatures lower than 4.2 K are displaced vertically by $0.1\ \mu_B$/Ce (2.17 K), $0.2\ \mu_B$/Ce (1.4 K) and $0.3\ \mu_B$/Ce (0.14 K). At 4.2 K the variation of the magnetization with the applied magnetic field is smooth and is not saturated up to 80 kOe. For this temperature we have performed measurements in field up to 150 kOe and magnetic saturation was not reached, which is different from the result obtained for CeSb in 40 kOe [4]. At 2.17 K, small steps appear on the magnetization curves between zero and 50 kOe. These steps are associated with a very flat hysteresis cycle. When the temperature is lowered down to 1.42 K, the amplitude of the magnetization steps increases. At the same time, an upward curvature of the magnetization curve appears, in the low field region with an inflection point in the vicinity of 18 kOe. The hysteresis cycle extends between zero to 70 kOe. In high field, the magnetization curves at 2.17 K and 1.42 K are identical and parallel to that at 4.2 K. For lower temperatures, this general behaviour is conserved. However, the magnetization steps are much less pronounced. The hysteresis cycle is always centred around 18 kOe and extends to a more limited magnetic field range. At 0.14 K the magnetization in 75 kOe, reaches a value of $0.92\ \mu_B$/Ce.

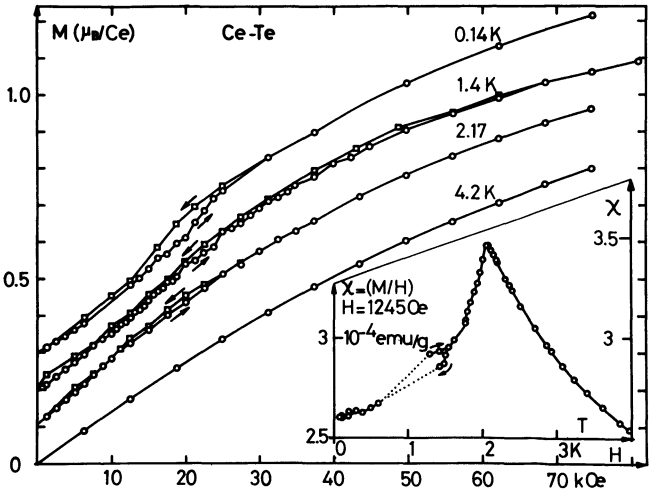


Fig. 1. — Magnetization curves of CeTe single crystals. The magnetic field is along the [100] direction. The insert shows the peaks at $T_N = 2.05 \pm 0.05$ K for the low field (1 245 Oe) magnetization. The curves at $T = 1.42$ K is for an other sample than the curves at $T = 0.14$ K, 2.17 K, and 4.2 K. For clarity the curves at temperature lower than 4.2 K are displaced vertically (see the text).

These low temperature magnetization curves can be analysed as a metamagnetic transition. The amplitude of the magnetization steps associated with the type of transition (like those from Barkausen jumps) increases with the field to 18 kOe and goes through a maximum, around 1.4 K, when the temperature is lowered. During the hysteresis cycle the measured magnetization is either stable as a function of time or rapidly stabilized (< 1 min.).

This magnetization behaviour could eventually be associated with many magnetic phase transitions as in CeSb [4]. Neutron scattering measurements with an applied magnetic field are needed to define the magnetization process.

The low temperature magnetic susceptibility has been deduced from the magnetization measurements performed in weak magnetic fields ($H < 1\,600$ Oe) for a temperature in the range 0.1 K-4.2 K. For a magnetic field of 1 245 Oe, a Néel temperature $T_N = 2.05 \pm 0.05$ K is measured. At lower temperatures small anomalies associated to the steps in the magnetization curves are evidenced. An insert in figure 1 could help to visualize these anomalies. For the same sample these anomalies are also detected in a field of 160 Oe. For these two fields the Néel temperature is the same.

3. **Neutron scattering measurements.** — 3.1 EXPERIMENTAL PROCEDURE. — Neutron scattering experiments have been performed on the $DN_3$ spectrometer of the Siloe reactor at the Centre d'Etudes Nucléaires at Grenoble. This spectrometer is equipped with a counter which can be moved out of the horizontal plane, which allows to scan the non-equatorial reciprocal space, up to an angle of 45°. Neutrons of wavelength $\lambda = 2.4$ Å were used. A graphite filter avoids any $\lambda/2$ contamination.

The single crystal, prepared as described above, was cleaned with the edges along the (100) planes of the rock-salt structure. To avoid any atmospheric contamination, the sample was closed in a sealed silica tube, with an helium pressure of about 300 torrs. This container was put in a $^3$He cryostat. The sample was oriented with a $\langle 100 \rangle$ axis vertical and cooled down to 0.45 K.

3.2 RESULTS. — The propagation vector of the CeTe magnetic structure has been determined by performing scans along the symmetry directions of the Brillouin zone. Magnetic peaks have been detected only at the boundaries of the Brillouin zone. Thus the structure is described by a propagation vector $\mathbf{k} = \langle 1/2\ 1/2\ 1/2 \rangle$. As no intensity was detected at the reciprocal point [1/2 1/2 1/2] the magnetic moments are parallel to the propagation vector, i.e. to a $\langle 111 \rangle$ axis. CeTe is, therefore, a type II antiferromagnet, which corresponds to a stacking of ferromagnetic (111) planes with the magnetic moments lying along the [111] direction.

In order to determine the value of the magnetic moment, the integrated intensities associated with the four antiferromagnetic domains have been measured at $T = 0.45$ K. To obtain the scaling factor, the nuclear intensities were measured. However a serious difficulty occurs. Using the Fermi lengths $b_{Ce} = 0.482 \times 10^{-12}$ cm and $b_{Te} = 0.548 \times 10^{-12}$ cm, the calculated ratio of the intensities of the strong reflections with even Miller indices to the weak reflections with odd Miller indices is not in agreement with the measured value. This discrepancy is essentially due to severe extinction effects. Thus to get the calibration factor, it should be better to use the odd reflections. However, the calculated intensities of these reflections are strongly dependent of the stoichiometry and the Fermi length values. These uncertainties give an under-estimation of the magnetic moment value found equal to $\mu = 0.15 \pm 0.05\ \mu_B$/Ce. On the other hand the scaling factor deduced from the even reflections gives a magnetic moment $\mu = 0.3\ \mu_B$/Ce. In this case, due to the extinction phenomena, this value is over-estimated. These results are similar to those of Ott *et al.* [5], who have reported a magnetic moment $\mu = 0.3\ \mu_B$/Ce, deduced from neutron scattering measurements, at 1.5 K.

The temperature dependence of the [3/2 1/2 1/2] reflection, reported in figure 2, gives a Néel temperature of $T_N = 1.95 \pm 0.1$ K in good agreement with the value deduced from the magnetic susceptibility measurements. For a sample with the same lattice parameter ($a = 6.361$ Å), Hulliger *et al.* [3] measured a lower Néel temperature $T_N = 1.5$ K.

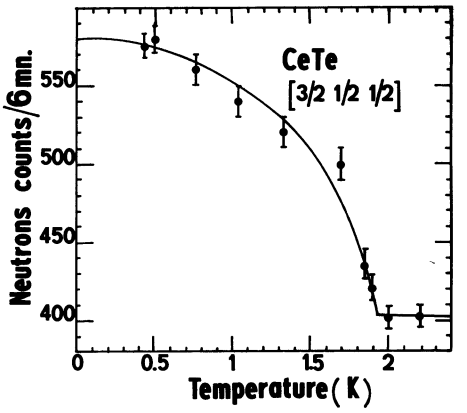


Fig. 2. — Thermal variation of the intensity of the [3/2 1/2 1/2] peak of CeTe.

4. **Conclusion.** — These experiments allow us to conclude that CeTe orders antiferromagnetically below $T_N = 2$ K with a type II antiferromagnetic structure. The magnetic moments are parallel to the propagation vector, i.e. along a [111] direction. However, at $T = 0.45$ K, the ordered magnetic moment deduced from neutron scattering experiments is abnormally low ($0.15 < \mu < 0.3\ \mu_B$/Ce) in contrast to the value $\mu = 0.57\ \mu_B$/Ce published for CeSe by Ott *et al.* [5] and for CeS by Schobinger-Papamantellos *et al.* [6].

This magnetic moment value cannot be compared to that deduced from the high magnetic field measurements, because in high field the mixing with the $\Gamma_8$ level is not negligible. The non saturation of the magnetization curves up to 150 kOe is a good proof of this mixing.

On the other hand, CeTe shows at low temperature and in a field of about 18 kOe, a metamagnetic transition. It seems that the magnetic field induces a transition from the antiferromagnetic state to, probably, a ferromagnetic state. The transition is asso-

ciated with a large hysteresis cycle. This magnetization process, with steps on the magnetization curves, remains rather obscur and need additional neutron experiments to be understood.

However the main puzzling result of this study is the low magnetic moment value in the antiferromagnetic state, while the $Ce^{3+}$ ground state is the $\Gamma_7$ doublet which has a moment value of about 0.7 $\mu_B$. At present, we are not able to understand why this crystal field picture explains quite well the high field magnetic properties and not the zero field moment value. Some unusual mechanisms related to the proximity of 4f electrons to the Fermi energy may explain this result.

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