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A neutron spectroscopy study of magnetic excitations in uranium oxysulphide

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Abstract. A neutron spectroscopy experiment has been performed to observe the position of the crystal-field levels in both the paramagnetic and antiferromagnetic phases of UOS. The $J = 4$ ground manifold undergoes an overall splitting of about 100 meV in agreement with the predictions of a crystal-field model used to analyse specific heat data. Above T_N, two main peaks are observed at 74 and 82 meV with a shoulder at about 87 meV. Below T_N there is a redistribution of intensity together with a shift of the peak positions to 76, 84 and 92 meV at $T = 30$ K. The experimental observations are interpreted in the framework of a refined crystal-field model.

1. Introduction

Uranium oxysulphide (UOS) is an ionic compound belonging to the tetragonal PbFCl type of crystal structure (space group P4/nmm–D_{4h}^7; $a = 3.845$ Å, $c = 6.685$ Å) [1] whose magnetic and thermodynamic properties have been studied extensively in the past [2–5]. Below $T_N = 55$ K, UOS becomes antiferromagnetic with a magnetic structure characterised by a doubling of the chemical unit cell in the c-axis direction and a P4/mm (C_{4v}^1) space group. The magnetic moments are localised on the uranium sites and aligned along the c axis in a $(+ + - -)$ sequence (figure 1).

The uranium ions are tetravalent with a stable $5f^2$ electronic configuration and $^3H_4$ Russell–Saunders (RS) ground term. The effective moment, which can be deduced from the high-temperature tail of the paramagnetic susceptibility curve [6], is in fact very close to the value of the $J = 4$ multiplet ($\mu_{eff} = 3.58\mu_B$ in RS coupling and $\mu_{eff} = 3.68\mu_B$ in intermediate coupling). However, the curvature shown by this curve in the low-temperature region is indicative of a crystal-field splitting of this multiplet. Moreover, the value of the ordered moment $\mu_{ord} \simeq 2\mu_B$, obtained from neutron diffraction [2], can be interpreted only by assuming a quasi-triplet ground state [3]. More recently, the entropy

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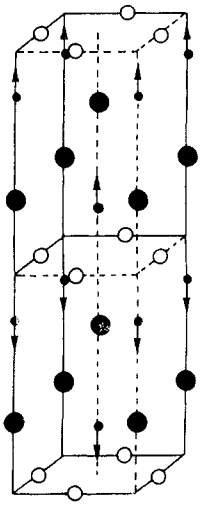

Figure 1. Crystallographic and magnetic structure of UOS, showing the coordination polyhedron of the uranium ions. The arrows on the U sites indicate the direction of the ordered magnetic moments. ●, uranium ions; ○, oxygen ions; ●, sulphur ions.

of ordering and the Schottky contribution to the UOS specific heat have been measured and the results interpreted on the basis of a point-charge crystal-field (CF) model [4]. In reference [3] it is shown that the $J = 4$ multiplet should be split into two groups of states, originating from the cubic Γ_μ ($\mu = 5, 1, 4, 3$) levels, with an overall splitting of the order of 100 meV. Moreover, the ground state and the first excited state, both coming from the splitting of the Γ_5 cubic level, should be a $\Gamma_{t5}^{(1)}$ doublet and a closely lying Γ_{t4} singlet respectively, in agreement with the observed value of the ordered magnetic moment. On the other hand, the huge magnetic anisotropy recently observed [7] for the magnetic susceptibility measured on UOS single crystals could only be explained by assuming an exchange anisotropy within the CF model proposed in reference [3] or by assuming a considerably larger splitting of the CF states.

In this paper we present the results of high-energy transfer neutron spectroscopy measurements performed on polycrystalline UOS both above and below the ordering temperature T_N. The experiment allows direct determination of the position of the CF levels and it gives, as a consequence, a stringent test for the predictions of the CF model developed in reference [4]. We will show here that this model gives a good description of the features of the spectra observed in the paramagnetic phase as well as the change of peak intensities and positions upon ordering below T_N. A brief account of this work has already been published [8].

2. Experimental details and results

The experiment was performed on the direct-geometry chopper spectrometer HET [9] at the UK spallation neutron source ISIS of the Rutherford Appleton Laboratory. A monochromatic neutron beam is produced at the sample position by phasing a Fermi chopper spinning at 400–600 Hz to the source proton pulse. Scattered neutrons are detected by two arrays of ^{3}He detectors, lying at 2 and 4 m from the sample position and covering a scattering angle range $\Phi = 3°$–$30°$. The scattering function $S(Q, \omega)$ can then be obtained by sorting the signals from the detectors according to the scattering angle and the neutron time of flight.

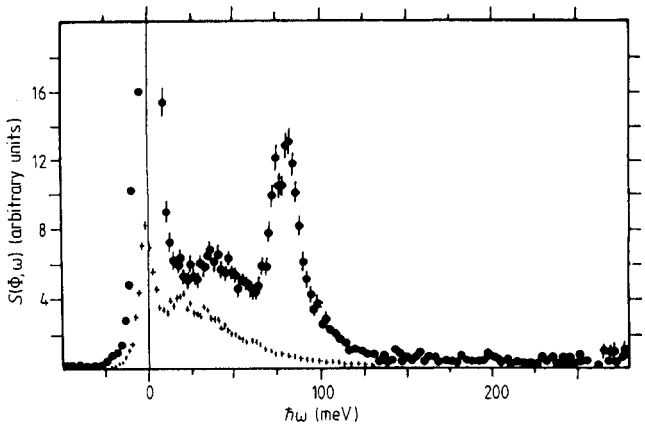

Figure 2. Inelastic neutron scattering cross section (for energy transfer $\hbar\omega$) measured in the paramagnetic phase of UOS at $T = 75$ K with neutrons of incident energy $E_0 = 290$ meV. The spectrum recorded at an average angle $\Phi = 136°$ gives the shape of the phonon density of states. The intensities are not normalised. ●, $\Phi = 5°$; +, $\Phi = 136°$.

For unpolarised neutrons and small values of the momentum transfer $\hbar Q$, the magnetic scattering function for a system of N non-interacting ions is given by

$$S(Q, \omega) = \alpha f^2(Q)\, e^{-2W} \sum_{i,j} p_i |\langle i|J_\perp|j\rangle|^2 \delta[(E_i - E_j)/\hbar - \omega]. \tag{1}$$

Here $\alpha = 0.0724$ b sr^{-1}, $\hbar\omega$ is the energy transfer, $|i\rangle$ are the different CF eigen-functions with energies E_i and thermal occupation probability p_i, $J_\perp$ is the total angular momentum component perpendicular to $\boldsymbol{Q}$, $f(Q)$ is the magnetic form factor of a single ion and e^{-2W} is the Debye–Waller factor. Equation (1) is valid if line broadening due to lifetime effects are negligible; it shows that the inelastic magnetic neutron scattering cross section consists of a sequence of δ-functions, the positions of which are determined by the eigenvalues of the CF Hamiltonian. Information on the CF eigenfunctions is obtained from the scattering intensities via the matrix elements of $J_\perp$. The magnetic scattering can be separated from the contributions of nuclear origin thanks to their different behaviours when the momentum transfer is varied, the former decreasing with Q as $f^2(Q)$ and the latter increasing as Q^{2n} for n-phonon processes.

The sample consisted of about 35 g of a high-purity polycrystalline compound obtained at LEFCA/CEN, Cadarache, France, in the reaction of a stoichiometric mixture of UO_2 and CS_2 at a temperature of about 1300 K. The final product was found to be single-phased without extra lines in the x-ray diffraction pattern [1].

Spectra were recorded both above and below $T_N = 55$ K. Different incident energies were used to optimise the intensity and the resolution. The results obtained in the paramagnetic phase at $T = 75$ K, with an incident energy $E_0 = 290$ meV and an average scattering angle $\Phi = 5°$, are shown in figure 2. Under these conditions magnetic excitations can be observed only up to 200 meV at most. Comparison with the spectrum measured at $\Phi = 136°$, giving the shape of the phonon density of states, shows that the intense peak centred at about 80 meV is magnetic in origin. Moreover, figure 2 shows that no further transitions are visible above 100 meV. The total energy splitting of the $J = 4$ multiplet is therefore in good agreement with the predictions of reference [4].

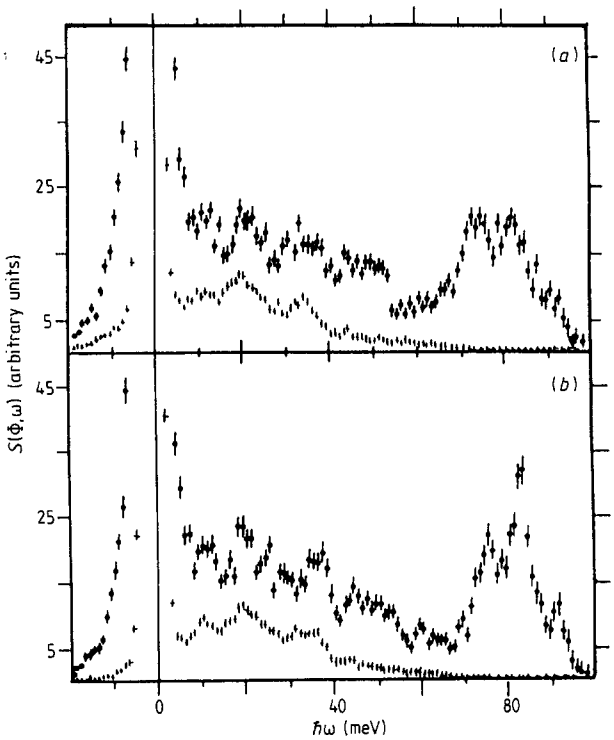

Figure 3. Neutron spectra measured with $E_0 = 110$ meV in (*a*) the paramagnetic phase of UOS at $T = 65$ K and in (*b*) the antiferromagnetic phase at $T = 30$ K. Data are shown for two values of the average scattering angle Φ. The spectra are not normalised. ●, $\Phi = 5°$; +, $\Phi = 136°$.

Higher-resolution scans, obtained by reducing the incident energy to 110 meV, reveal that the magnetic peak is in fact structured and composed of at least two groups of lines. As shown in figures 3(*a*) and (*b*), they are centred at about 74 and 82 meV at $T = 65$ K and at 76 and 84 meV at $T = 30$ K, below T_N. The asymmetric shape and the width of the higher-energy peak in the paramagnetic region suggest the presence of a level at about 87 meV, which appears to move to 92 meV at 30 K. There are signs of low-intensity excitations in the range 40 to 55 meV. However, we are not able, at present, to define unambiguously their origin.

3. Interpretation of the experimental results

The coordination polyhedron of the uranium ions in UOS consists of four oxygen ions at 2.33 Å, four sulphur ions at 2.91 Å and one sulphur ion at 2.97 Å (figure 1). All four sites are equivalent with C_{4v} point-group tetragonal symmetry. The CF Hamiltonian to be considered is then

$$H_{CF} = B_2^0\hat{O}_2^0 + B_4^0\hat{O}_4^0 + B_6^0\hat{O}_6^0 + B_4^4\hat{O}_4^4 + B_6^4\hat{O}_6^4 \qquad (2)$$

where B_n^m are the CF parameters and $\hat{O}_n^m$ the Stevens operator equivalents. The B_n^m may be calculated in the point-charge approximation by assuming the valencies U^{4+}, O^{2-} and

S^{2-} and by taking into account either the nearest neighbours (NN) or the whole lattice (LS) contribution. The relativistic values of the average radii $\langle r^n \rangle$ given in reference [10] and the Sternheimer screening factors quoted in reference [11] have been used in the calculation. The diagonalisation of the Hamiltonian (2) for the calculated values of the CF parameters leads to the following composition of the CF eigen-states (in order of increasing energy):

$$\begin{aligned}
&|\Gamma_{t5}^{(1)}\rangle = \alpha|\pm 3\rangle + \beta|\mp 1\rangle && |\Gamma_{t3}\rangle = (1/\sqrt{2})(|2\rangle + |\bar{2}\rangle) \\
&|\Gamma_{t4}\rangle = (1/\sqrt{2})(|2\rangle - |\bar{2}\rangle) && |\Gamma_{t2}\rangle = (1/\sqrt{2})(|4\rangle - |\bar{4}\rangle) \\
&|\Gamma_{t1}^{(1)}\rangle = \varepsilon(|4\rangle + |\bar{4}\rangle) + \gamma|0\rangle && |\Gamma_{t1}^{(2)}\rangle = (\gamma/\sqrt{2})(|4\rangle + |\bar{4}\rangle) - \sqrt{2}\varepsilon|0\rangle \\
&|\Gamma_{t5}^{(2)}\rangle = \beta|\pm 3\rangle - \alpha|\mp 1\rangle
\end{aligned} \tag{3}$$

where $|M\rangle$ is a short notation for $|J, M\rangle$ and α, β, γ and ε are numerical coefficients the values of which change slightly in the NN and LS cases [4]. The ordered moment corresponding to the $|\Gamma_{t5}^{(1)}\rangle$ ground doublet is (LS case) $\mu_{ord} = 2.32\mu_B$, not too far from the experimental value of $2\mu_B$.

If we assume that the $\Gamma_{t5}^{(1)}$–Γ_{t4} transition is not resolved in the experiment (i.e. that it is embedded in the elastic line), the observed magnetic peaks may be attributed to transitions from the ground $\Gamma_{t5}^{(1)}$ doublet to the CF states of the higher-energy group. In this assumption better agreement with experiment is obtained with the NN rather than the LS CF calculation reported in reference [4], that is, with a level scheme very close to that shown in [4] for UOSe. On the other hand, the fitting of the Schottky peak in the case of UOS would be only slightly affected by assuming NN instead of LS CF parameters. In any case, only a semi-quantitative picture of the CF energy levels and eigen-functions is usually obtained from *ab initio* CF calculations. A considerable improvement may be obtained by using the approach introduced in references [12] and [13] for the study of U^{4+} and Np^{4+} ions in a tetragonal CF. In the present study, it may be noted that the $\Gamma_{t5}^{(1)}$ and the Γ_{t4} states derive from the cubic triplet Γ_5 with a relatively small departure from the cubic case. The strong tetragonal perturbation due to the four sulphur ions at $(0, 0, z)$ positions is partially compensated by the axial potential due to the sulphur ion at the $(\frac{1}{2}, \frac{1}{2}, z)$ position [3]. Moreover, the quadrupolar axial parameter B_2^0 is strongly reduced owing to the Sternheimer shielding [4]. This situation suggests that the tetragonal CF Hamiltonian should be written as the sum of a cubic term H_c and an axial one H_a, acting as a perturbation [12, 13]:

$$H_{CF} = H_c + H_a \tag{4}$$

with

$$H_c = W[x(\hat{O}_4^0 + 5\hat{O}_4^4)/60 + (1 - |x|)(\hat{O}_6^0 - 21\hat{O}_6^4)/1260] \tag{5}$$

$$H_a = Wy[\hat{O}_2^0 + \mathcal{B}_4^0\hat{O}_4^0 + \mathcal{B}_6^0\hat{O}_6^0] \tag{6}$$

where x and W are the Lea, Leask and Wolf parameters and

$$\mathcal{B}_4^0 = (B_4^0 - B_4^4/5)/B_2^0 \qquad \mathcal{B}_6^0 = (B_6^0 + B_6^4/21)/B_2^0. \tag{7}$$

The parameter $y = B_2^0/W$ may be regarded as a measure of the relative strength of the axial term with respect to the cubic one, while the parameter W acts merely as an energy scale factor.

It can be shown that $\mathcal{B}_4^0$ and $\mathcal{B}_6^0$ may be determined exactly once the splitting $\Delta = E(\Gamma_{t4}) - E(\Gamma_{t5}^{(1)})$ and the value of the ordered magnetic moment $\mu_{ord} =$

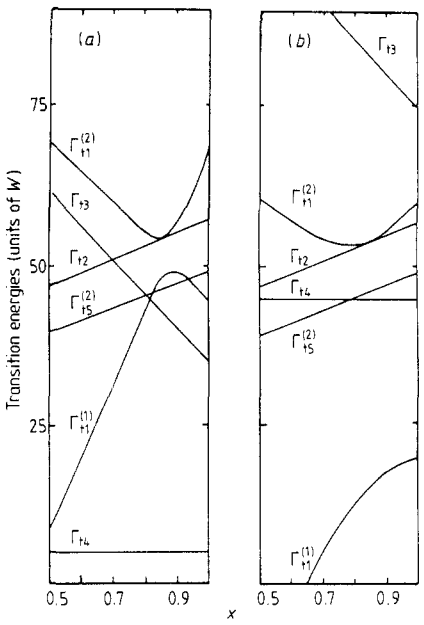

Figure 4. Energies of the crystal-field transitions from the ground to the excited states calculated in the paramagnetic phase of UOS as a function of the cubic Hamiltonian parameter x. The energies are reported in units of the scale parameter W. Fixed values of the ordered magnetic moment ($\mu_{ord} = 2.15\mu_B$) and of the axial parameter ($y = 0.1$) are used. Two positions of the Γ_{t4} level are assumed: (a) $E(\Gamma_{t4}) = 5W$ and (b) $E(\Gamma_{t4}) = 45W$.

$g_J\mu_B(3\alpha^2 - \beta^2)$ are fixed (α and β being the coefficients of the Γ_{t5} wavefunction). The CF levels and the transition probabilities related to the neutron scattering cross section may then be calculated as a function of W, x and y, once Δ and μ_{ord} are fixed to the experimental values. Two typical examples, calculated for $y = 0.1$ and $\alpha = 0.96$ (corresponding to $\mu_{ord} = 2.15\mu_B$, which represents a compromise between the value given in [4] and the quoted experimental value), are shown in figure 4. In this figure the ratios between the transition energies relative to the ground doublet and the parameter W are plotted against x for (a) $\Delta/W = 5$ and (b) $\Delta/W = 45$. It must be noted that the qualitative behaviour of this plot is influenced only slightly by the value and sign of y, providing it remains a perturbative term only. Figure 4 shows that the position of the Γ_{t3} level is strongly dependent on the Γ_{t4} level, moving upwards if the Γ_{t4} energy is increased. Moreover, the range of values of x corresponding to an energy level sequence compatible with the results of the neutron spectroscopy experiment may be identified easily. In this way, we can analyse two possible hypotheses on the position of the most intense transition, namely the $\Gamma_{t5}^{(1)}$–Γ_{t4} transition: (i) its energy is lower than 10 meV and the corresponding peak is not resolved in the present experiment from the elastic line; (ii) it contributes to the neutron intensity in the 80 meV range.

In the magnetically ordered phase, the contribution $H_{MF} = -hJ_z$ to the Hamiltonian has been estimated self-consistently in the framework of the CF model [14], leading to $h = 1.5$ meV in case (i) and $h = 1.4$ meV in case (ii) at $T \simeq 30$ K. The CF transitions from the ground state and their relative probabilities calculated under assumptions (i) and (ii) are shown in figures 5 and 6 respectively, both in the paramagnetic (figures 5(a) and 6(a)) and in the ordered (figures 5(b) and 6(b)) phases. The set of parameters

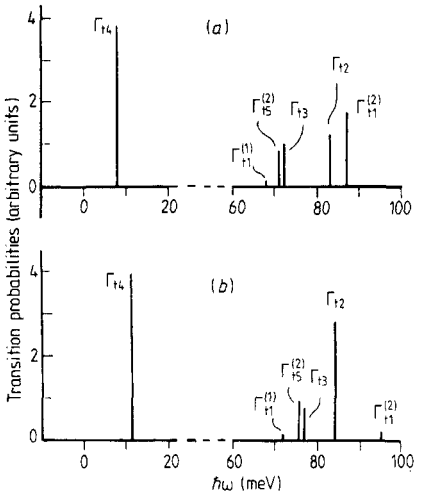

Figure 5. Transition energies from the ground state and probabilities calculated (*a*) above and (*b*) below T_N using the set of parameters $W = 1.6$ meV, $x = 0.8$, $y = 0.1$, $\alpha = 0.96$ ($\mu_{ord} = 2.15\mu_B$) and $[E(\Gamma_{t4}) - E(\Gamma_{t5}^{(1)})] = 8.0$ meV, corresponding to the case (i) discussed in the text.

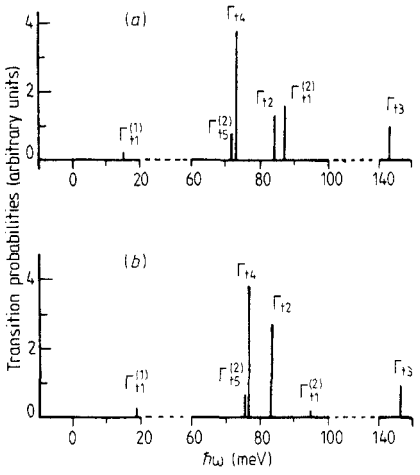

Figure 6. Transition energies from the ground state and probabilities calculated (*a*) above and (*b*) below T_N using the same parameters of figure 5 but for $x = 0.75$ and $[E(\Gamma_{t4}) - E(\Gamma_{t5}^{(1)})] = 72$ meV (case (ii) discussed in the text).

Table 1. Set of crystal-field parameters corresponding to the assumptions (i) and (ii) discussed in the text. The *ab initio* values calculated in the NN and in the LS approximations are also reported for comparison.

	W (meV)	x	y	$60\mathscr{B}_4^0$	$1260\mathscr{B}_6^0$
LS	0.91	0.58	−0.13	−8.91	−1.99
NN	1.31	0.75	0.24	1.98	0.91
(i)	1.57	0.80	0.10	3.74	0.98
(ii)	1.61	0.75	0.10	−3.62	13.11

Table 2. B_n^m crystal-field parameters (in units of W) calculated in the LS and in the NN approximations as well as under assumptions (i) and (ii) discussed in the text.

	B_2^0/W	$10B_4^0/W$	$10^3B_6^0/W$	$10B_4^4/W$	$10^2B_6^4/W$
LS	−0.132	0.293	0.538	0.486	−0.695
NN	0.236	0.221	0.327	0.622	−0.423
(i)	0.100	0.196	0.236	0.667	−0.333
(ii)	0.100	0.065	1.240	0.625	−0.417

corresponding to assumptions (i) and (ii) is given in table 1, where the *ab initio* values are shown for comparison. Table 2 gives the corresponding sets of the CF parameters B_n^m in units of W, from which it is evident that case (i) is very close to the NN calculation. Figures 4 and 6 show that assumption (ii) would lead to a shift of the $\Gamma_{t5}^{(1)}$–Γ_{t3} transition to an energy of about 150 meV. As shown in figure 2, no excitations have been observed

in this energy region in the present experiment. This evidence counts against assumption (ii) but it must be remarked that a peak with the intensity calculated for this transition would be observed with some difficulty at the quoted energy due to the rapid decrease of the form factor. Measurements with higher incident energy are needed to confirm the absence of CF excitations around 150 meV. However, hypothesis (i) seems to us to be more likely and we will restrict the following discussion to this case only.

As stated above, the magnetic peak is resolved into two main groups of lines (figure 3). It appears from figure 5 that the first group may be due to the transitions from the ground to the $\Gamma_{t5}^{(2)}$ and Γ_{t3} states, whilst the second group is due to the transitions to the Γ_{t2} and $\Gamma_{t1}^{(2)}$ states. The intensities of these groups are in fact comparable, taking into account that the energy separation between the last two levels should produce a broadening rather than an enhancement of the second sub-peak. By lowering the temperature below T_N, the calculated intensity of the first group of lines remains practically unchanged, while the $\Gamma_{t5}^{(1)}$–Γ_{t2} is strongly enhanced. Because of the simultaneous shift toward higher energy and the intensity decrease of the $\Gamma_{t5}^{(1)}$–$\Gamma_{t1}^{(2)}$ transition, the second sub-peak is expected to become higher and narrower. These effects are observed in practice, together with the shifts of the level positions below T_N (figure 3) which are also fairly well reproduced by our calculation (figure 5).

If the relative energy of the Γ_{t4} singlet is decreased below $\simeq$10 meV, the intensity of the transitions from this state to the excited Γ_{t3} singlet and $\Gamma_{t5}^{(2)}$ doublet becomes non-negligible at $T > T_N$, because the population factor of Γ_{t4} increases. There does appear to be extra intensity just below the main peak at 65–70 meV in the spectrum recorded at $T = 65$ K (figure 3(*a*)). However, the resolution is too poor to allow a definitive assignment of the level positions and a further quantitative refinement of the model calculations. This will only be possible once the position of the first excited level is determined unambiguously. For this reason, the situations considered in cases (i) and (ii) and figures 5 and 6 must only be considered as illustrative of the possible CF transition schemes.

4. Discussion and conclusions

The results of the present neutron spectroscopy experiment show an overall splitting of about 100 meV for the $J = 4$ ground manifold of the U ions in uranium oxysulphide. The sequence of observed magnetic levels and the relative intensities of the corresponding neutron scattering peaks may be accounted for satisfactorily by a CF model introduced to describe the low-temperature thermodynamic properties of the uranium oxychalcogenides UOZ (Z = S, Se, Te) [4]. This model can be refined to improve the fitting of the neutron results. In particular, it must allow for a lower value of the energy of the first excited singlet, which is likely to be embedded in the elastic peak. This condition can be fulfilled by a set of CF parameters that are close to the NN calculation. The agreement with the specific heat data remains unchanged. For instance, the value of the entropy at 140 K (i.e. well above T_N) for the quasi-triplet ground state of case (i) is $S_{calc} = 9.1\ \mathrm{J\ mol^{-1}\ K^{-1}}$. This is in fair agreement with the experimental value at the same temperature $S_{exp} = 8.7\ \mathrm{J\ mol^{-1}\ K^{-1}}$, taking into account the intrinsic difficulty of an accurate determination of the entropy [4], which is often underestimated.

One of the aims of the present work was to determine the overall CF splitting of the $J = 4$ ground multiplet, in order to elucidate the mechanism underlying the behaviour of the magnetic susceptibility of this compound. Measurements on single crystals of

UOS [7] up to 300 K show very different results for $\chi_{\parallel}(T)$ (that is with $\boldsymbol{H} \parallel \boldsymbol{c}$) and $\chi_{\perp}(T)$, the former quantity showing a marked maximum near T_N and a Curie–Weiss behaviour, the latter being weakly temperature-dependent and about five times smaller than $\chi_{\parallel}$ at 150 K. As the CF Hamiltonian entirely defines the energy levels in the paramagnetic phase, the magnetic susceptibility $\chi(T)$ of the system can be calculated within the proposed CF model.

If isotropic exchange is assumed, the whole behaviour of $\chi(T)$ up to 850 K in the powder can be accounted for by assuming an overall CF splitting much higher than 100 meV and a doublet–singlet ground state with $\Delta \simeq 20$ meV [6]. However, this model cannot account for the measured anisotropy in the temperature region up to 300 K, unless a greater value of Δ ($\simeq$80 meV) is assumed. In addition, both the specific heat measurements and the present neutron experiments are not in favour of this interpretation. In particular, if the level scheme were of the type proposed in [6], the peaks observed in the neutron scattering should originate from the transitions between a split $\Gamma_{t5}^{(1)}$ doublet and the Γ_{t4} singlet. Above T_N a doublet splitting would imply the existence of a lattice distortion, which has not been observed [2]. Below T_N a splitting could arise from the molecular field, but we estimate a value much smaller than 8 meV.

On the other hand, the susceptibility measurements may be reproduced satisfactorily with the energy level scheme adopted in our model by assuming that the exchange interaction is anisotropic. In a phenomenological description this means that two different exchange shifts $\lambda_{\parallel,\perp}$ must be introduced in order to obtain the two components of the inverse susceptibility from the corresponding Van Vleck terms $\chi_{\parallel,\perp}^{(VV)}$ relative to the system of non interacting ions:

$$\chi_{\parallel}^{-1}(T) = (\chi_{\parallel}^{(VV)})^{-1} - \lambda_{\parallel} \qquad \chi_{\perp}^{-1}(T) = (\chi_{\perp}^{(VV)})^{-1} - \lambda_{\perp}. \tag{8}$$

The susceptibility for the powdered UOS sample can then be expressed as

$$\chi(T) = \tfrac{1}{3}\chi_{\parallel}(T) + \tfrac{2}{3}\chi_{\perp}(T). \tag{9}$$

Using the CF parameters determined from the neutron experiment, $\chi_{\parallel,\perp}^{(VV)}$ can be calculated and $\lambda_{\parallel,\perp}$ can be obtained by fitting equation (9) over the whole temperature range. The values $\lambda_{\parallel} = 18$ and $\lambda_{\perp} = -240$ (in molar units) give the best fit for both the single-crystal and the powder experimental data, when the CF parameters of case (i) are assumed. The single-crystal measurements and a detailed discussion of the interpretation of the magnetic susceptibility of UOS will be the subject of a separate paper [15]. Another argument in favour of the present interpretation is provided by the recent inelastic neutron experiments on UO_2 [16, 17]. The magnetic peaks at $\simeq$150 and 170 meV first observed in [16] were finally attributed [17] to transitions between the ground Γ_5 triplet and the excited Γ_3 doublet and Γ_4 triplet, because no other transitions were found up to 700 meV. Thus the observed CF splitting of the ground multiplet was close to the point-charge estimate and also to the prediction of the superposition model analysis [18, 19] for U^{4+} ions. Moreover, in this case the assumed CF level scheme accounts for the extra contribution to the specific heat of UO_2 with respect to the non-magnetic isomorphous ThO_2.

In conclusion, the present interpretation of the neutron experiment confirms the CF model proposed for UOS in reference [4] and supports the interpretation of the observed magnetic anisotropy in terms of anisotropic exchange effects. A further improvement would require the experimental determination of the Γ_{t4} level position and a better refinement of the ordered magnetic moment. A lower-energy neutron spectroscopy experiment and a powder neutron diffraction study are planned to settle these points.

Acknowledgments

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