Neutron-diffraction study of UO_2: Paramagnetic state*

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(Received 13 August 1975)

The magnetic form factor of the induced moment in UO_2 has been measured in the paramagnetic state at $T = 64$ K with polarized neutrons in an applied field of 50 kOe. The theoretical form factor is derived with the tensor-operator method and the $\langle j_i \rangle$ integrals for U^{4+} derived from relativistic Dirac-Fock wave functions. Three models for the electronic state are considered: (i) the free-ion U^{4+} ground state, (ii) the $^3H_4$ Γ_5 crystal-field ground state, and (iii) the Rahman and Runciman mixed-J Γ_5 ground state. The calculation of the magnetic scattering in the presence of intermediate coupling and J mixing is discussed. Whereas the experimental data are unable to distinguish between the small angular differences predicted for the three models, the radial dependence of the magnetic scattering is found to be in good agreement with theory and confirms the validity of the Dirac-Fock $\langle j_i \rangle$ integrals.

I. INTRODUCTION

In studies of the electronic structure of actinide metals and compounds one of the major problems is determining the number of $5f$ electrons associated with an actinide ion. Controversy still surrounds questions such as the spatial extent of the $5f$ electrons, the magnitude of the crystalline field in these materials, and the degree of overlap and possible hybridization of wave functions.[1] Measurements of the elastic cross section yield, in principle, information about both the angular character and radial extent of the unpaired $5f$ electrons.[2] To examine the question of the spatial extent of the $5f$ electrons, we have measured the neutron scattering from the compound UO_2, in which the electron configuration is well known.[3] Uranium dioxide is an ionically bonded semiconductor with two unpaired $5f$ electrons surrounding the U^{4+} ion. The properties of the ground-state wave function in UO_2 have been considered by Rahman and Runciman (RR).[4] In a complete diagonalization of the Hamiltonian including Coulomb, spin-orbit, and crystal-field interactions RR showed that the Γ_5 (or T_2) triplet was the lowest-lying level, and was separated by ~2000 K from the Γ_3 (or E) doublet. Measurements of the magnon-dispersion curves with inelastic neutron scattering,[5] of magnetic susceptibility,[6] and infrared spectroscopy,[7] are all consistent with a triplet being the lowest state. The RR calculation, together with more recent work on actinide compounds,[2] emphasizes that the Hund's-rule Russell-Saunders state (in this case $^3H_4$ as in praseodymium compounds with two $4f$ electrons) may be a poor approximation to the ground state of actinide ions. Instead, one must use intermediate-coupling wave functions and take into account the effects of J mixing caused by the strong crystal field.

In this paper we describe polarized-neutron measurements of the induced magnetic moment of the U^{4+} ion in the paramagnetic state of UO_2 ($T = 64$ K). These experiments were undertaken primarily to investigate the large anisotropy in the magnetic form factor that had been observed in the ordered state of UO_2.[8] The source of this anisotropy was not understood; but the absence of similar effects in the paramagnetic state provided an important key to understanding the earlier measurements. An additional aim was to test whether the recently derived relativistic wave functions represent the radial extent of the $5f$ electrons in the solid.[9]

II. EXPERIMENT

The single crystals of UO_2 were cut from a large boule acquired some years ago from Savannah River. The majority of the measurements were performed on a crystal of dimensions $1.6 \times 2.4 \times 5.1$ mm^3, with a $\langle 110 \rangle$ axis parallel to the longest dimension. The magnetic field was applied parallel to this long axis and $\{hhl\}$ reflections in a zone perpendicular to the field were measured. To ensure stoichiometry, the crystal was heated for 6 hr at 1950 °C in a 10^{-6}-Torr vacuum.[10] The lattice parameter of 5.4702(2) Å at 20 °C agrees with values found in the literature.[11] The crystal was extensively characterized[8] at 4.2 and 80 K with unpolarized neutrons of wavelength 1.0 Å. The important point of these crystallographic studies is that they determine the stoichiometry

of the sample, the temperature factors of the uranium and oxygen atoms, the extinction parameter, and the extent to which this parameter accounts for extinction.[8] A second crystal (approximately three times larger in volume) was used only to measure the weak Bragg reflections at high scattering angles, where both nuclear and magnetic scattering amplitudes are small.

The experiments were performed on a polarized-neutron diffractometer located at the CP-5 Research Reactor. The neutron wavelength was 1.05 Å. The crystal was contained in a superconducting magnet assembly in a field of 50 kOe. The sample temperature, which was constantly recorded in the data output, reached an equilibrium between a small inner liquid-nitrogen and the main liquid-helium reservoir of between 64 and 69 K. On the basis of the slow drift between these two temperatures and the straight-line behavior of the $1/\chi$ vs T plot[12] we have corrected the experimental data to 64 K. These corrections are less than 4%.

The polarized-beam technique yields the so-called flipping ratio

$$R = (1+\gamma)^2/(1-\gamma)^2 , \tag{1}$$

where $\gamma = M/N$, and M and N are the magnetic and nuclear structure factors, respectively. For uranium dioxide (CaF_2 crystal structure), the structure factors and intensities are as follows: (a) If $h+k+l=4n$, where n is an integer, $N = b_U e^{-W_U} + 2b_O e^{-W_O}$, the reflections have the strongest intensity. Here b_U and b_O are the coherent scattering amplitudes of uranium and oxygen with values 0.853 and 0.580 (10^{-12} cm), respectively. The Debye-Waller factors are represented as e^{-W}, where $W = B(\sin^2\theta)/\lambda^2$. (b) If $h+k+l=4n\pm1$, $N = b_U e^{-W_U}$ the reflections have medium intensity. (c) If $h+k+l=4n+2$, $N = b_U e^{-W_U} - 2b_O e^{-W_O}$, the reflections are weak. The magnetic structure factor M has the same form for all reflections (assuming the magnetic scattering is associated with the uranium atom only—an assumption verified by the present experiment) and $M = 0.2696 \times \mu f(\vec{\kappa}) e^{-W_U}$, ($10^{-12}$ cm), where μ is the magnetic moment per uranium atom and $f(\vec{\kappa})$ is the magnetic form factor associated with the scattering vector $\vec{\kappa}$. From susceptibility measurements[12] on UO_2 the induced magnetic moment at 50 kOe and 64 K is $37.4 \times 10^{-3}\ \mu_B$. Hence, the values of γ_0 (i.e., with $\vec{\kappa}=0$, $f=1$) are 0.0050, 0.0118, and −0.0335 for the three types of reflections discussed above. In practice Eq. (1) has to be corrected for incomplete incident polarization and imperfect spin reversal; but since both the neutron polarization and the flipping efficiency are 0.994(2), these corrections are very small. Other small effects are the correction for diamagnetic scattering[13] and the neutron spin-neutron orbit interaction,[14] but these are at least an order of magnitude below the statistical uncertainties. More important sources of error arise from multiple scattering and extinction. Multiple-scattering effects are difficult to eliminate totally, but the good agreement between equivalent reflections and the use of more than one crystal gives us confidence that the effects are small. In the case of the strong (400) reflection, consistent values could not be obtained for the flipping ratio, and these difficulties were ascribed to multiple scattering. The (400) reflection has not been used in analyzing the data. In view of the excellent agreement (residual of 0.006 and $\chi^2 = 1.2$) between the calculated and observed intensities measured with unpolarized neutrons,[8] we feel confident that the extinction is well represented by the Zachariasen formula[15]

$$I_{obs}/I_{calc} = y = (1 + 2gQ\bar{t})^{-1/2} , \tag{2}$$

where I_{obs} and I_{calc} are the observed and calculated intensities, $\bar{t}$ is the effective path length, g is the extinction parameter, and Q is the crystallographic reflectivity. The value of $g = 936(50)$ and is independent of temperature. The large value of g suggests that the extinction is probably of type II. Studies of[16] TmSb and[17] $Tb(OH)_3$ have shown that the Zachariasen expression can be used to correct polarized-neutron data provided the flipping ratio is independent of the diffracted intensity over a Bragg reflection.

The experimental results in terms of the quantity (μf) are presented in Table I. The $(\mu f)_{obs}$ values were obtained from measurements on at least two equivalent reflections, and [for $(\sin\theta)/\lambda > 0.5$ Å^{-1}] with two different crystals. The good internal consistency of these measurements has been used to derive the standard deviations in Table I. The corrected values of μf are plotted as a function of $(\sin\theta)/\lambda$ in Fig. 1.

III. THEORY

We have used the tensor-operator formalism of Lovesey and Rimmer[18] and Marshall and Lovesey[19] to calculate the elastic neutron cross section from the two unpaired $5f$ electrons in UO_2. The application and advantages of this method for systems with f electrons have been discussed previously[20–22] and only a brief outline will be given here.

The magnetic scattering length is defined as a vector $\vec{E}$, with spherical components E_Q given by

$$(2\pi\hbar/m)E_Q = \langle \psi_e | T_Q^K(e, \vec{\kappa}) | \psi_e \rangle \tag{3}$$

[Eq. (3) is identical to Eq. (7.5) of Lovesey and Rimmer[18]], where the electron wave functions are represented by ψ_e, and $T_Q^K(e, \vec{\kappa})$ defines a tensor operator such that

$$\langle\psi_e|T_Q^K(e,\vec{\kappa})|\psi_e\rangle_T=\delta_{K,1}\frac{4\pi\hbar^2}{m}(0.54\times10^{-12})\sum_{K''Q''}(4\pi)^{1/2}Y_{Q''}^{K''}(\hat{\kappa})$$
$$\times\sum_{K'Q'}\Bigg[\sum_{\substack{\theta JM\\ \theta'J'M'}}\Bigg(\sum_{\Gamma}\langle\theta'J'M'|\psi_e(\Gamma_i)\rangle\langle\psi_e(\Gamma_i)|\theta JM\rangle e^{-E_\Gamma/kT}\Bigg)Z^{-1}$$
$$\times[A(K'',K'')+B(K'',K')]\langle K'Q'J'M'|JM\rangle\Bigg]\langle K''Q''K'Q'|KQ\rangle\,. \tag{4}$$

[Eq. (4) is similar to Eq. (4.60) in Ref. 18.] We have now written the wave function as a function of Γ to indicate that it may consist of a number of crystal-field states Γ_i each of energy E_{Γ_i}, and the quantity required is the ensemble average of $\langle T_Q^K\rangle$ at a temperature T. The partition function is denoted by Z. Note that matrix elements between states Γ_i and Γ_j do not appear because we are concerned with the *elastic* cross section only. The evaluation of this expression falls into three parts.

First, we must construct the wave functions of the crystal-field states in the $|SLJM\rangle$ basis, recalling that the use of a single SLJ manifold, which is a good approximation for lanthanide systems, may not be sufficient for actinide ions. To obtain the crystal-field wave functions, the complete set of free-ion eigenfunctions should be used to set up the crystal-field matrix, which is then diagonalized. However, except for the f^1 and f^2 configurations, this process involves an enormous amount of computation. In anticipation of experiments on other actinide systems with more than two $5f$ electrons we have preferred to follow the method developed by Chan and Lam (CL).[1] This method initially considers the free-ion wave function and the strong spin-orbit interaction that leads to the mixing of excited SL components into each J manifold. The next step is to consider the crystal-field interaction that leads to mixed J configurations; J is then no longer a good quantum number. The appropriate quantum number is Γ, the irreducible representation of the point-group symmetry operations. The final states are therefore functions of $|\Gamma M\rangle$, but are only indirectly functions of S and L. This approximation to the full diagonalization procedure is fully discussed by Chan and Lam[1] and is certainly valid for calculations of the magnetic form factor, a quantity that depends on the lowest-lying eigenstates. Table II contains the coefficients of the normalized eigenvectors for the

TABLE I. Observed and calculated values of μf for UO_2 at $H=50$ kOe, $T=64$ K, $(\mu f)_{obs}$ is the observed value, and $(\mu f)_{cor}$ has been corrected for extinction ($g=936$). The calculated values $(\mu f)_{calc}$ are based on models described in the text and all assume $\mu=37.4\times10^{-3}\ \mu_B$.

hkl	$\frac{\sin\theta}{\lambda}$ (Å^{-1})	$(\mu f)_{obs}$ ($10^{-3}\ \mu_B$)	$(\mu f)_{cor}$ ($10^{-3}\ \mu_B$)	$(\mu f)_{calc}$ $^3H_4$ free ion	$^3H_4$ Γ_5	Mixed-J state Γ_5
111	0.159	28.7	34.1(1.3)	32.6	32.7	32.8
200	0.183	29.5	30.8(2.2)	31.1	30.9	31.8
220	0.259	18.2	26.6(1.2)	25.9	26.3	26.3
311	0.304	21.3	23.4(1.1)	22.6	22.5	23.4
222	0.317	19.3	19.8(1.2)	21.6	21.6	21.5
400	0.366	10.2	13.5(1.2)	18.1	18.1	19.9
331	0.399	17.3	17.4(0.9)	15.9	16.1	15.8
422	0.448	7.8	11.4(2.6)	12.9	12.6	13.0
333	0.476	11.1	11.2(0.6)	11.4	10.9	10.2
511	0.476	11.5	11.6(1.0)	11.4	11.6	13.1
440	0.518	8.1	10.6(1.8)	9.3	9.2	9.1
600	0.549	8.5	8.6(0.9)	7.9	8.6	10.4
442	0.549	7.9	8.0(0.6)	7.9	7.5	6.8
533	0.600	5.3	5.7(0.8)	6.0	5.5	5.2
622	0.607	4.0	4.0(0.6)	5.8	5.9	6.8
444	0.634	3.8	4.7(2.3)	5.0	4.1	2.9
551	0.654	4.6	5.1(1.6)	4.5	3.9	3.8
771	0.654	4.6	5.0(0.9)	4.5	5.3	6.7
553	0.703	1.0	1.2(1.6)	3.3	2.5	1.4
800	0.732	3.2	3.6(2.0)	2.8	4.0	5.3

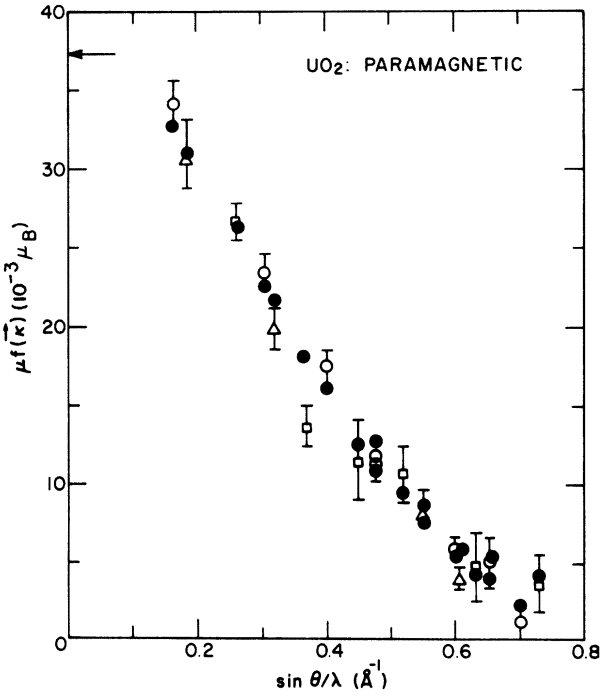

FIG. 1. Values of μf for each reflection plotted vs $(\sin\theta)/\lambda$. Open circles o correspond to reflections with $h+k+l=4n\pm1$; □—$4n$ reflections, Δ—$4n\pm2$ reflections. Solid circles are the theoretical values using the $^3H_4$ Γ_5 model, see Table I. Theoretical value at $(\sin\theta)/\lambda=0$ is 37.4×10^{-3} μ_B as given by susceptibility measurements.

TABLE II. Absolute values of the coefficients a_i for the different SLJ states from the Chan-Lam (CL) and Rahman-Runciman (RR) calculations, truncated after 4 J states. g_{eff} represents the intermediate coupling g values and M_{eff} is defined within the $|\Gamma M\rangle$ representation. The magnetic moment is given by μ in Bohr magnetons.

J	State	CL	RR	R_{eff}	M_{eff}
4	^3H	0.8934	0.874	0.8247	2.5
	^1G	0.3057	0.331		
	^3F	0.0888	0.138		
3	^3F	0.1933	0.195	1.0833	0.5
2	^3F	0.1852	0.214	0.7041	−1.0
	^1D	0.0611	0.044		
	^3P	0.0149	0.055		
5	^3H	0.1103	0.097	1.033	−2.5
$\sum a_1^2$		0.9873	0.9907		
μ (μ_B)		1.7715	1.80		

CL and RR calculations. We have truncated the eigenvector after four J manifolds in the cross-section calculation because the coefficients of additional J manifolds are negligible. If the coefficients are normalized to the same magnetic moment μ, the differences between the CL and RR values are small. Returning to Eq. (4), we may now evaluate

$$\sum_{\theta\theta'} [A'(K'', K', J, J') + B'(K'', K', J, J')]$$

from a consideration of the J manifolds and the intermediate-coupling wave functions within each manifold. The prime notation on A and B indicates that we do not include the $\langle j_i \rangle$ integrals at this stage of the calculation.[21] The SL and $S'L'$ quantum numbers are represented by θ and θ', respectively. For a single J state, ten independent coefficients are needed to specify the A' and B' terms, but this number increases as the square of the number of J manifolds involved.

Second, the three components of the rank-one tensor operator ($K=1$, $Q=0, \pm 1$) are evaluated to give coefficients that depend on both the spherical harmonics $Y_Q^{K''*}(\hat{\kappa})$ and the $\langle j_i \rangle$ radial integrals, which are functions of κ. Spherical harmonics up to rank 8 are necessary to specify the cross section. The results for this part of the calculation with the single SLJ manifold $^3H_4$ Γ_5 crystal-field ground state, and with the RR model are given in Table III.

The third part of the calculation consists of evaluating the radial integrals $\langle j_i \rangle$, and the unit scattering vector $\hat{\kappa}$ in terms of the spherical coordinates Θ and Φ. In this experiment, the quantization axis of the magnetic system $\vec{H} \| [1\bar{1}0]$, is perpendicular to the scattering vector (i.e., $\Theta = \frac{1}{2}\pi$), which leads to the special case $E_1 = E_{-1} = 0$. The cross section then reduces to

$$\frac{d\sigma}{d\Omega} \propto N^2 - 2NPE_0 + \tfrac{1}{4}E_0^2 , \tag{5}$$

where P is the neutron polarization. Equation (5) has the same form as the usual expression for polarized neutrons

$$\frac{d\sigma}{d\Omega} \propto N^2 + 2PMN + M^2 . \tag{6}$$

If $\Theta = 90°$, we define the magnetic moment μ (parallel to the field direction) and a form factor $f(\vec{\kappa})$ in Eq. (5) so that $E_0 \propto \mu f(\vec{\kappa})$ and

TABLE III. Coefficients for the magnetic cross section ($Q=0$ term only) for UO_2 at $T=64$ K with $\vec{H} \| [1\bar{1}0]$ and $\mu = 37.4 \times 10^{-3}$ μ_B. The coefficients (all multiplied by 10^3) modify terms $Y_Q^{K''*}(\Theta, \Phi)\langle j_i \rangle$, where Θ and Φ are defined by $\vec{\kappa}$ and $\vec{H}$ and $\langle j_i \rangle$ are the radial integrals. For the free-ion calculations, the only nonzero entries are $(12.47\ Y_0^0 - 5.57\ Y_0^2)\langle j_0 \rangle + (20.50\ Y_0^0 - 9.17\ Y_0^2)\langle j_2 \rangle$.

		$^3H_4$ Γ_5				4 J States Γ_5			
K''	Q''	$\langle j_0 \rangle$	$\langle j_2 \rangle$	$\langle j_4 \rangle$	$\langle j_6 \rangle$	$\langle j_0 \rangle$	$\langle j_2 \rangle$	$\langle j_4 \rangle$	$\langle j_6 \rangle$
0	0	12.47	20.49	0	0	12.47	21.48	0	0
2	0	−5.57	−9.26	−0.136	0	−5.57	−9.56	+0.160	0
2	2		−0.204	−0.277	0		+0.058	+0.222	0
4	0		0.074	−0.048	−0.946		−0.033	−0.216	−1.529
4	2		0.176	−0.055	−1.878		−0.044	−0.761	−2.35
4	4			0.085	0.542			+0.825	+0.788
6	0			0.124	0.911			+0.080	+1.290
6	2			0.253	1.563			+0.195	+1.918
6	4			−0.088	−0.620			−0.061	−1.329
6	6				−0.019				−0.784
8	0				−0.108				−0.015
8	2				0.039				+0.035
8	4				0.055				+0.056
8	6				0.022				−0.055

TABLE IV. Coefficients c_i in Eq. (7) for selected values of Φ. All entries are for $\Theta = 90°$ (i.e., $q^2 = 1$) and Φ is the angle between $\vec{\kappa}$ and the [001] axis. For the free-ion $^3H_4$ state $c_2 = 1.6445$, $c_4 = 0$, $c_6 = 0$ for all Φ.

		c_2		c_4		c_6	
		$^3H_4$	$4J$	$^3H_4$	$4J$	$^3H_4$	$4J$
[hkl]	Φ	Γ_5	Γ_5	Γ_5	Γ_5	Γ_5	Γ_5
[001]	0.0	1.603	1.732	0.022	0.272	0.483	0.537
[113]	25.2	1.622	1.727	−0.009	0.042	0.124	0.232
[112]	35.3	1.638	1.723	−0.023	−0.090	−0.112	−0.120
[111]	54.7	1.672	1.713	−0.022	−0.192	−0.407	−0.717
[221]	70.5	1.695	1.707	0.005	−0.115	−0.415	−0.581
[110]	90.0	1.707	1.704	0.026	−0.034	−0.356	−0.240

$$f(\vec{\kappa}) = \langle j_0 \rangle + c_2 \langle j_2 \rangle + c_4 \langle j_4 \rangle + c_6 \langle j_6 \rangle \ . \tag{7}$$

In this expression the angular parts of the form factor are represented by the coefficients c_i, which depend on Θ and Φ only, and the radial parts by the functions $\langle j_i \rangle$, which depend on κ only. The coefficients c_i for selected values of Φ are tabulated in Table IV for the single-J and mixed-J calculation. The entries in Tables III and IV show that the effects of J mixing are readily apparent in the coefficients c_4 and c_6 of Eq. (7). In particular, the coefficient c_4 is at least an order of magnitude greater in the presence of J mixing and is caused by the increased number of nonzero matrix elements in the full calculation. Another point of interest is that the Φ dependence of c_2 in the mixed-J calculation is reversed in sign from the $^3H_4$ Γ_5 calculation, but because of terms in $\langle j_4 \rangle$ and $\langle j_6 \rangle$ the final Φ dependence is the same for the two calculations.

IV. RESULTS AND DISCUSSION

Since the exchange parameters in UO_2 are unknown, the internal magnetic field used in the calculation is such that a magnetic moment of 37.4×10^{-3} μ_B is induced at 64 K (see Table II). Thus the magnetization at a temperature T is given by

$$M(T) = \mathrm{Tr}(\mu e^{-H/kT})$$

$$= Z^{-1} \sum_m \langle \psi_m | \mu | \psi_m \rangle e^{-E_m/kT} \ , \tag{8}$$

where H represents the sum of the exchange and applied magnetic fields, and $\mu = \sum_i l_i + g_s \sum_i s_i$, where s_i and l_i are the spin and orbital angular momenta, respectively, and g_s is the gyromagnetic ratio. The sum is over all f electrons.

The values of μf for the free-ion, $^3H_4$ Γ_5, and the CL mixed-J state model are given in Table I. Differences between the calculated μf values from the three models are small. In most cases, these differences are smaller than experimental errors. The free-ion form factor is a function of θ and contains terms in $\langle j_0 \rangle$ and $\langle j_2 \rangle$ only,[21] since $gM_J \mu H \ll kT$. On the other hand, if the triplet is well separated from the next state, the form factor will be essentially independent of temperature, i.e., the form factors in the ordered and paramagnetic states will be identical.

A comparison of the experimental data with the $^3H_4$ Γ_5 calculation is given in Fig. 1. The good agreement between theory and experiment suggests (a) that the extinction corrections are reliable, because for $h+k+l = 4n$ these corrections are larger than for the $4n+2$ reflections, and (b) that any magnetic moment located at the oxygen site is extremely small. We emphasize that the agreement between theory and experiment contains no adjustable scale factor. Conversely, we may ask what is the value for μ determined by comparing the experimental values of $(\mu f)_{\mathrm{cor}}$ and the theoretical form factor? The answer is $(37.5 \pm 1.0) \times 10^{-3}$ μ_B, compared to the value of 37.4×10^{-3} μ_B from susceptibility measurements.[12] In contrast, no such agreement is found for metallic US. For this compound, neutron experiments give 1.70 μ_B, whereas the bulk magnetization value is 1.55 μ_B.[2] This discrepancy in US has been attributed to the conduction-electron polarization arising predominantly from the occupied $6d$ band.[2,9] Since the conduction band is unoccupied in UO_2, we would not anticipate any discrepancy between the neutron and magnetization magnetic moments to arise from this source. Effects owing to covalency may also give rise to an apparent discrepancy in the magnetic moment, but any such effects must be small in UO_2. We should note that the agreement between theory and experiment in the paramagnetic state is in marked contrast to the initial examination of the magnetic scattering in the ordered state.[8] The anisotropy observed in the ordered state cannot be understood on the basis of simple magnetization density considerations. A report of these investigations is in preparation.[23]

Finally, the experimental data are not of sufficient accuracy to distinguish between the three

models given in Table I. However, the agreement in Fig. 1 shows that the radial integrals $\langle j_i \rangle$ derived from the relativistic Dirac-Fock calculations[9] are a good representation of the spatial distribution of the 5*f* electrons in UO_2.

ACKNOWLEDGMENTS

We would like to thank D. J. Lam and S. K. Chan for advice on the subject of strong crystal-field interactions, and R. L. Hitterman for experimental assistance.

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*Work supported by the U. S. Energy Research and Development Administration and the National Science Foundation.

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