TEMPERATURE DEPENDENCE OF ELASTIC CONSTANTS AND THERMAL EXPANSION FOR UO_2 †

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The thermal expansion has been measured as a function of temperature for UO_2 and shows a first-order transition at the Néel temperature. Elastic constants versus temperature also display a first-order transition except for C_{44}, which shows a lattice-spin effect 100° above the transition.

Blume[1] has proposed a theory of the paramagnetic-antiferromagnetic transition which occurs at 30.8°K in UO_2. From neutron diffraction studies[2,3] it appears that the magnetization changes abruptly at the Néel temperature, indicating a first-order transition rather than the usual second-order transition. Blume proposed his theory because there seemed to be no discernible volume change at the transition, thus the order of the phase change would be inherently a consequence of the magnetic properties of the system. The adjustable parameter in Blume's model, the single-triplet splitting of the U^{4+} ion in the crystal field, has been found in Daniel's antiferromagnetic resonance studies[4] to be of the order Blume suggested. However, the far infrared spectra seen by Allen[5] and Aring and Sievers[6] are more complicated than Daniel's.

We should like to report measurements of length change versus temperature in UO_2, which demonstrate directly that the phase change is first order in the lattice properties, and measurements of elastic constant versus temperature, which show that the state of the material above the transition may well be more complicated than anticipated.

The normalized length change, $\Delta L/L$, for the [100] direction in UO_2 is shown as a function of temperature in Fig. 1, curve A. Data have been taken up to room temperature, but only the range 10 to 60°K is shown. No data points are shown since the length was monitored continuously on an x-y recorder. As can be seen, a rather abrupt change in length of the order of 20 ppm occurs at the transition, with the length continuing to change rapidly below the transition. The change is seen more dramatically in Fig. 1, curve B, which shows the temperature dependence of the linear-expansion coefficient, obtained by graphical differentiation of Fig. 1, curve A. A contraction of comparable magnitude has also been observed for the [110] direction. Although it is small, there is thus clearly a volume change at the transition.

The elastic constants as a function of temperature are shown in Fig. 2. Again no points are shown, since what was determined experimentally was a continuous plot of the temperature derivative of the elastic constants. Thus, Fig. 2 was obtained by a numerical integration of the data. The actual experimental method will be published separately.[7] C_{11} was determined from longitudinal wave propagation along [100], C_{44} from shear propagation along [100] and along [110] polarized along [001], and $C_{11}-C_{12}$ from shear wave propagation along [110] polarized along $[1\bar{1}0]$. The curve for C_{12}, shown in Fig. 2, was obtained by subtracting the $C_{11}-C_{12}$ curve from the C_{11} curve.

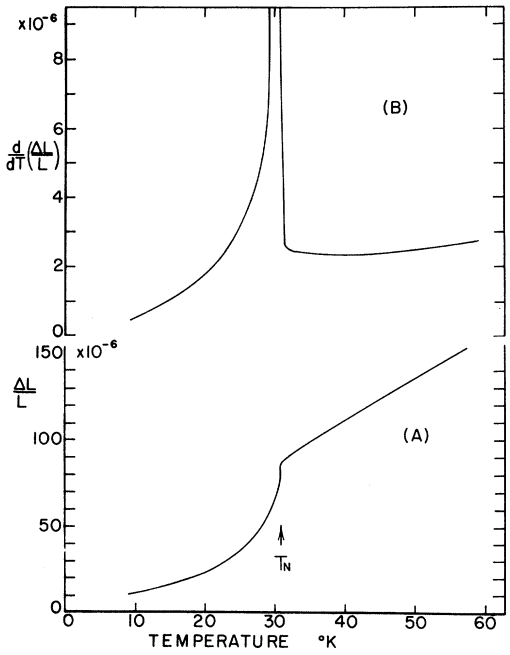

FIG. 1. Normalized length and thermal expansion versus temperature for UO_2. Curve A, change in length per unit length; curve B, thermal expansion obtained by graphical differentiation of A. Néel temperature = 30.8°K.

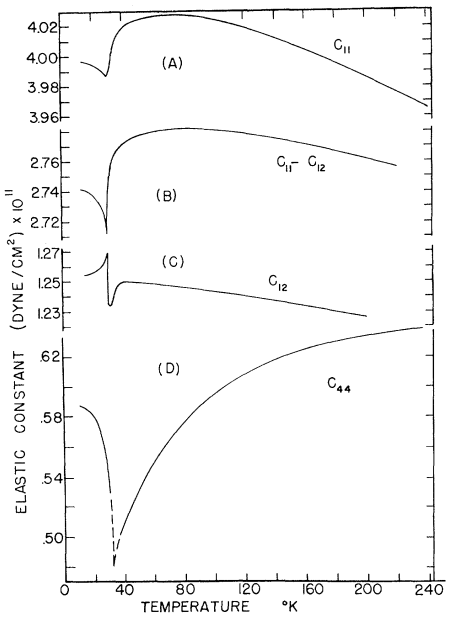

FIG. 2. Elastic constants versus temperature for UO_2. Curve A, C_{11} obtained by integrating dC_{11}/dT data; curve B, $C_{11}-C_{12}$ obtained as in curve A; curve C, C_{12} obtained by subtracting curve B from curve A; curve D, C_{44} obtained as in curve A.

As can be seen, C_{11}, C_{12}, and $C_{11}-C_{12}$ change markedly only near the transition, with $C_{11}-C_{12}$ being particularly abrupt, although even as high as 60°K there is still some memory of the transition. But the temperature dependence of C_{44} is spectacular. The attenuation becomes so great near the transition that the elastic constants cannot be determined in the region indicated by the dashed line. Thus, we cannot tell whether C_{44} actually goes to zero at the transition, nor can we tell how high the slope of the curve actually becomes. But the two most remarkable features are the λ-like shape of the curve, and the fact that 100° above the transition the elastic constant is still changing significantly.

Bean and Rodbell[8] proposed a mechanism by which a paramagnetic-ferromagnetic transition, which ordinarily would be second order, can become first order. In their treatment the interaction between magnetic moments is assumed to depend on the lattice spacing, and an essential feature of their theory is a volume change at the transition. In fact the lattice distortion in the ordered state is what "prevents" the system from having a second-order transition. Our observed volume change at the transition in UO_2 suggests that the question of the mechanism giving the first-order change should be reopened, as the lack of a volume change can no longer serve to eliminate Bean and Rodbell's arguments.

Further, the C_{44} elastic constant data suggest that any interaction between the lattice and the spins is not negligible in UO_2. This latter point has been more directly demonstrated in Dolling and Cowley's deduction[9] from their inelastic neutron data of a lower limit to the magnon-phonon interaction of 9.6°K, in comparison with the Néel temperature of 30.8°K. Thus, the magnon-phonon interaction, which is a measure of the interaction of lattice and spins, is of the same order as the direct magnetic interactions in UO_2.

Garland and co-workers,[10] in a series of five papers, have studied the temperature and pressure dependence of the elastic constants near the order-disorder transitions for NH_4Cl and NH_4Br. The tetrahedral ammonium ions in both crystals have two equivalent positions in the cubic cell of halogen ions. The transition in NH_4Cl is one where the ammonium ions align parallel (quasiferromagnetic) and in NH_4Br the ammonium ions align antiparallel (quasiantiferromagnetic). The analogy to magnetism is not exact, though, since among other features NH_4Cl remains cubic below the transition while NH_4Br becomes slightly tetragonal. Garland, Renard, and Yarnell have calculated the elastic constants for a two-dimensional Ising model, showing a first-order transition, and have applied the results to their data for NH_4Cl.

One interesting result of their treatment is that the behavior of C_{11} near the transition is dominated essentially by the interaction energy between spins. $C_{11}-C_{12}$ and C_{44}, on the other hand, are dominated by the deformation dependence of the interaction energy. Our plots of C_{11} and $C_{11}-C_{12}$ versus temperature bear a striking resemblance to their data on NH_4Br and one might hope to be able to deduce the deformation dependence of the interaction energy from our data. However, their calculation was made under the assumption of a weak coupling between lattice and spins, a condition which would not appear to pertain for UO_2. The most dramatic discrepancy between our data and Garland, Renand, and Yarnell's treatment lies in C_{44}, as they predict a rather uneventful behavior.

In summary, our data on length versus tem-

perature display a first-order transition in UO_2, but in the lattice properties, in contrast to Blume's hypothesis. This result would suggest a re-examination of the transition mechanism in UO_2. Part of the data on elastic constant versus temperature is in accord with the idea of a first-order phase change, but C_{44} versus temperature would seem to be quite anomalous and indicative of a considerable interaction between lattice and spins well above the transition.

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$\frac{3}{2}^-$ GROUND STATE OF V^{47} †

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Of those nuclei in the $1f_{7/2}$ shell which are readily accessible to experimental investigation, V^{47} is one of the least well studied. Thus, although in studies of the reactions $Cr^{50}(p,\alpha)$,[1] $Ti^{46}(p,\gamma)$,[2] and $Ti^{47}(p,n)$,[3] energy levels have been observed up to about 3 MeV in excitation, in no cases have spin-parity assignments been made. In particular, the assumed[4] $\frac{5}{2}^-$ character for the ground state of V^{47} has been based largely on its observed β^+ decay.

On the other hand, there are now extensive theoretical predictions concerning V^{47} as well as a wide range of other $1f_{7/2}$ nuclei. For example, within the framework of a pure $1f_{7/2}$ shell-model configuration, McCullen, Bayman, and Zamick[5] and Ginnochio[6] have successfully reproduced the excitation energies of the low-lying $\frac{7}{2}^-$ and $\frac{5}{2}^-$ states for many odd-A nuclei. However, their calculations generally predict the $\frac{3}{2}^-$ states arising from this configuration to be as much as 1 MeV higher in excitation than their observed locations. More recently, Federman and Talmi[7] have obtained good agreement for the low-lying states in the Ca isotopes by assuming a deformation of the $1f_{7/2}$ orbital. Malik and Scholz[8] have also shown that the level schemes for a number of odd-A nuclei in this shell can be rather well described in terms of the Nilsson model with Coriolis coupling. According to this model, the relative locations of the $\frac{7}{2}^-$, $\frac{5}{2}^-$, and $\frac{3}{2}^-$ states may be a sensitive measure of the deformations in these nuclei. In this communication we wish to report on a study of the low-lying $\frac{3}{2}^-$, $\frac{5}{2}^-$, and $\frac{7}{2}^-$ states in V^{47} arising from the $(1f_{7/2})^3$ proton configuration. Information concerning the locations of hole states is also presented.

Levels in V^{47} have been studied[9] by means of the reaction $Ti^{46}(He^3,d)$ using a 16.5-MeV He^3 beam from the University of Pennsylvania tandem accelerator. Deuteron spectra were recorded at angles ranging from 7° to 40° with a broad-range magnetic spectrograph and with an over-all resolution <20 keV. Figure 1 shows a partial deuteron spectrum measured at 30° for the transitions leading to the ground and first three excited states at 0.089, 0.147, and 0.258 MeV. The impurity group corresponding to the 1.663-MeV level in V^{49} arose because of the presence of 14.5% Ti^{48} in the target.

In Fig. 2, deuteron angular distributions corresponding to the ground and second excited states are shown. The curves shown were calculated from distorted-wave theory using the code JULIE. Of particular interest here is the unambiguous $l_p = 1$ assignment to the ground-