Title

OPTICAL AND MAGNETIC PROPERTIES OF URANIUM BOROHYDRIDE AND TETRAKISMETHYLBOROHYDRIDE

Permalink

https://escholarship.org/uc/item/5xq7z1g0

Author

Rajnak, K.

Publication Date

1983-09-01

LBL-16460
Preprint c.2

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

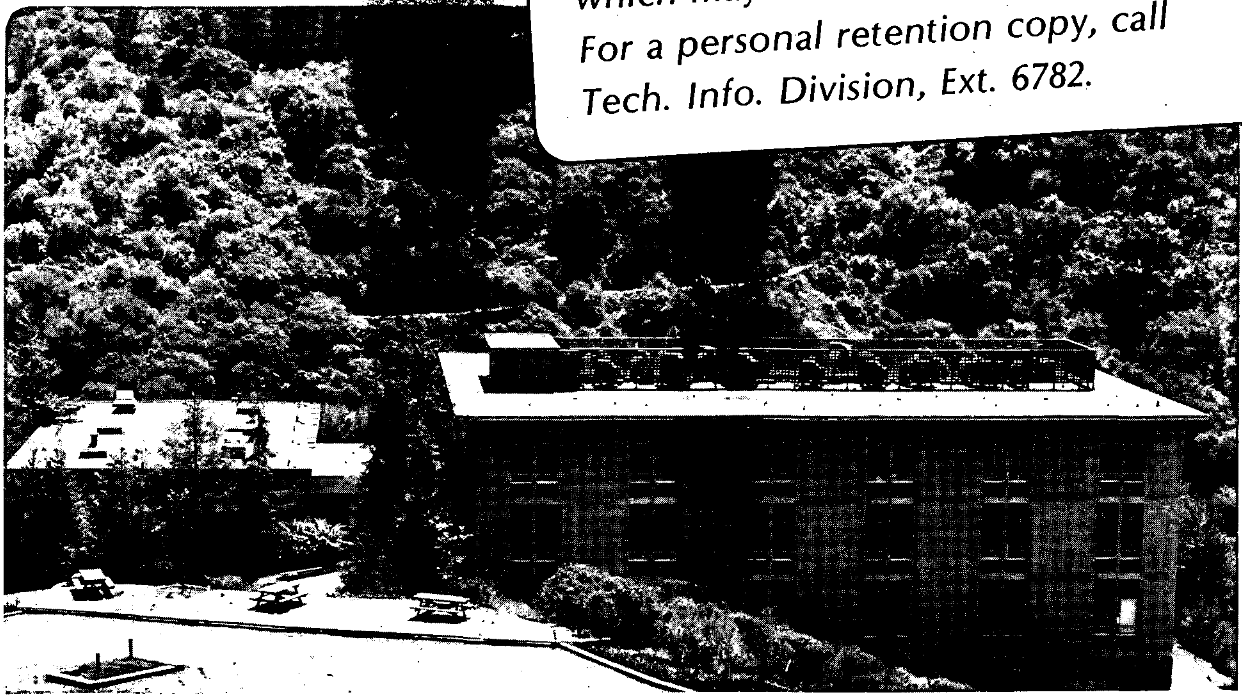
Materials & Molecular Research Division

Submitted to the Journal of Chemical Physics

OPTICAL AND MAGNETIC PROPERTIES OF URANIUM BOROHYDRIDE AND TETRAKISMETHYLBOROHYDRIDE

K. Rajnak, E. Gamp, R. Shinomoto, and N. Edelstein

September 1983



Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

LBL-16460

OPTICAL AND MAGNETIC PROPERTIES OF URANIUM BOROHYDRIDE AND TETRAKISMETHYLBOROHYDRIDE[a]

K. Rajnak,* E. Gamp, R. Shinomoto, and N. Edelstein

Materials and Molecular Research Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720 U.S.A.

Abstract

The $U(BD_4)_4/Hf(BD_4)_4$ optical spectrum reported by Bernstein and Keiderling (J. Chem. Phys. 59, 2105 (1973)) has been reanalyzed. All 19 allowed transitions have been identified. The crystal field is ~ 2.5 times as strong as that of $U^{4+}/ThBr_4$, (Phys. Rev. B, in press) but the values of the F^k and ζ parameters are nearly the same. The magnetic susceptibility of the structurally related molecule $U(BH_3CH_3)_4$ has been measured from 2 - 330 K. Using the eigenvectors from the optical analysis, the magnetic data can be fit with an orbital reduction factor k= 0.85. For $U(BD_4)_4/Hf(BD_4)_4$ $k = \zeta/\zeta_{free\ ion} = 0.91$.

Introduction

The actinide borohydrides possess a number of properties which make them attractive candidates for chemical and spectroscopic studies. The first five members of the series (Th–Pu) have been prepared.[1–3] They are volatile molecules near room temperature which makes it relatively easy to obtain single crystals. High symmetry diamagnetic analogs, Hf or Zr borohydride, are available as diluents.[4,5] In a pioneering study, Bernstein and Keiderling[6] (BK) obtained high resolution optical spectra of $U(BH_4)_4(U(BD_4)_4)$ in single crystals of $Hf(BH_4)_4(Hf(BD_4)_4)$ and fit this data to a parameterized Hamiltonian which included the Slater parameters, the spin-orbit coupling constant, and the two crystal field parameters required for the T_d symmetry of the host crystals. Subsequently, the molecules neptunium and uranium borohydride were synthesized. Preliminary results have been published on the magnetic and optical properties of $Np(BH_4)_4$ $(Np(BD_4)_4)$ diluted in $Zr(BH_4)_4(Zr(BD_4)_4)$.[7,8]

The actinide borohydrides exhibit two structural types. $Th(BH_4)_4$, $Pa(BH_4)_4$ and $U(BH_4)_4$ are isomorphic and increase in volatility with increasing atomic number. $Np(BH_4)_4$ and $Pu(BH_4)_4$ are also isomorphic but closely resemble the highly volatile Zr and Hf borohydrides in structure and properties rather than the earlier actinide molecules.[9] All seven compounds contain triple hydrogen bridge bonds connecting the boron atom to the metal. In addition, the 14-coordinate Th, Pa, and U borohydrides have double-bridged borohydride groups that link metal atoms together in a low-symmetry, polymeric structure.[10] The structures of the other four molecules are monomeric and much more symmetric, the 12-coordinate metal is surrounded by a tetrahedral array of BH_4^- groups.

The polymeric structure of $U(BH_4)_4$ precludes the possibility of obtaining the magnetic susceptibility of this compound with the same symmetry as found in the host $Hf(BH_4)_4$ crystal used in the optical investigations. However, the series of compounds $M(BH_3CH_3)_4$ (M = Th, U, Np, and Zr) have recently been synthesized and structurally characterized.[11] All four molecules are monomeric and for each molecule the metal atom is tetrahedrally coordinated to the four methylborohydride groups through tridentate hydrogen bridge bonds. The Zr and Np tetrakismethylborohydrides belong to the same tetragonal space group with 2 molecules per unit cell. The U and Th compounds are monoclinic and triclinic respectively with 4 molescules per unit cell.

The magnetic susceptibility of $U(BH_3CH_3)_4$ has been measured in the temperature range 2 - 330 K. We assume in this work that the electronic structures of $M(BH_4)_4$ (with T_d symmetry) and $M(BH_3CH_3)_4$ (M=U or Np) are similar so that we may use the magnetic data for the $M(BH_3CH_3)_4$ compounds to estimate the splitting of the ground multiplet of the tetrahedral $M(BH_4)_4$ systems. With this additional information, plus the vibrational information available from the normal coordinate analysis of $Np(BH_4)_4$ ($Np(BD_4)_4$),[12] we present in this paper a reanalysis of the optical data of BK. In the following paper the optical and magnetic data for $Np(BD_4)_4/(Zr(BD_4)_4)$ and magnetic data for $Np(BH_3CH_3)_4$ are given and analyzed.[13] Finally, in the third paper in this series, a model is proposed to explain the observation in EPR experiments of two magnetically inequivalent molecules in single crystals of $Np(BH_3CH_3)_4$ diluted in $Zr(BH_3CH_3)_4$.[14]

Experimental

The synthesis of $U(BH_3CH_3)_4$ has been described previously.[3,11] Magnetic susceptibility measurements were carried out on a SHE 905 SQUID magnetometer. Powdered samples of $U(BH_3CH_3)_4$ were weighed and sealed into calibrated containers in an inert atmosphere box. Several different samples were measured with weights varying from 75 to 130 mg. All data were obtained with applied fields between 0.5 and 40 kGauss and temperatures from 1.8 to 330 K.

$U(BH_4)_4$ and $U(BH_3CH_3)_4$: Review and Magnetic Susceptibility Data

The U^{4+} ions in $U(BH_4)_4/Hf(BH_4)_4$ and in $U(BH_3CH_3)_4$ occupy a site of T_d symmetry. The ground term of the U^{4+} ion has $J = 4$ (nominally $^3H_4$) which splits in a tetrahedral crystal field into four states: a singlet A_1, a doublet E and two triplets, T_1 and T_2. BK found neither EPR spectra for $U(BD_4)_4/Hf(BD_4)_4$ at 2 or 77 K nor any Zeeman splitting in the 400–750 nm region. This fact, plus the assignment of at least 11 forced electric dipole transitions, led them empirically to assign the ground state as the E state. Their analysis of the optical spectra resulted in a calculated ground state of T_2 symmetry with the E state 14 cm^{-1} above it.

The fact that U^{4+} in $U(BH_3CH_3)_4$ has the same symmetry,

T_d, as $U(BD_4)_4/Hf(BD_4)_4$ allows us to use magnetic susceptibility measurements from 2 K to room temperature to supplement the data of BK. We assume that the electronic structures of $U(BH_3CH_3)_4$ and $U(BH_4)_4$ are similar. The optical spectra of the two compounds, obtained in C_6D_6 at room temperature, are shown in Fig. 1. The spectra are seen to be similar, although most bands appear to be shifted to higher energies for $U(BH_3CH_3)_4$. The susceptibility of $U(BH_3CH_3)_4$ is shown in Fig. 2. The ground state shows temperature independent paramagnetism consistent with the assignment of the E state as the ground state. This data was initially analyzed considering only the $^3H_4$ term. Two fits are shown in Fig. 2 with the splittings given in Table I. From these fits it is clear that the separation between the ground E state and the first excited T_1 or T_2 state must be $\geq$ 150 cm^{-1}. Finally, it should be noted that a reasonable fit could not be obtained without the introduction of an orbital reduction factor.[15]

Optical Analysis

The energy levels within an f^n configuration in T_d symmetry can be written in terms of the atomic free ion (H_{FI}) and crystal field (H_{CF}) Hamiltonians as follows:[16,17]

$$H = H_{FI} + H_{CF}$$

where

$$H_{FI} = \sum_{k=0,2,4,6} f_k F^k(nf,nf) + \zeta_f a_{so} + \alpha L(L+1)$$

$$+ \beta G(G_2) + \gamma(R_7) + \sum_{k=0,2,4,} m_k M^k + \sum_{k=2,4,6} p_k P^k$$

and

$$H_{CF} = B_0^4 \, [C_0^{(4)} + \sqrt{5/14}(C_{-4}^{(4)} + C_4^{(4)})] + B_0^6[C_0^{(6)} - \sqrt{7/2}(C_{-4}^{(6)} + C_4^{(6)})].$$

The $F^k(nf,nf)$'s and ζ_f represent, respectively, the radial parts of the electrostatic and spin-orbit interactions between f electrons, while f_k and a_{so} are the angular parts of the interactions. α, β, and γ are the parameters associated with the two-body effective operators of configuration interaction. The M^k parameters represent the spin-spin and spin-other-orbit interactions while the P^k parameters arise from electrostatic-spin-orbit interactions with higher configurations. The crystal field interaction for T_d symmetry is represented by two parameters B_0^4 and B_0^6 and the tensor operators $C_q^{(k)}$.[17]

BK used a parameterized Hamiltonian which included only the F^k's, ζ, B_0^4 and B_0^6. Their best fit for $U(BD_4)_4/Hf(BD_4)_4$ assigned 11 levels with an rms deviation of 62 cm^{-1} between experimental and calculated levels. However, their calculated ground state was a T_2 level with the E level lying 14 cm^{-1} higher in contradiction with both optical and magnetic data. Furthermore, when they assigned 18 energies, their rms deviation increased to 158 cm^{-1}.

Recently, the U^{4+} free-ion spectrum has been completely analyzed.[18] With this additional information it is now possible to set certain limits for the allowable range of both the free-ion parameters and their ratios. In addition, 26 levels of $U^{4+}/ThBr_4$[19] have been fit with an rms deviation of only 36 cm^{-1}, thus providing some guidelines for the changes in the free-ion parameters in going from the free ion to the crystal. From assignments of the infra-red and Raman spectra of $Np(BH_4)_4(Np(BD_4)_4)$, a normal coordinate analysis of these molecules has been carried out.[12] This resulted in a reliable list of frequencies with which to assign vibronic lines in the optical spectrum of $U(BD_4)_4/Hf(BD_4)_4$. For these reasons a reanalysis of the data of BK has been undertaken.

The values of F^2, F^6 and ζ obtained by BK were 42008, 28048 and 1910 cm^{-1} respectively. The corresponding free-ion parameters are 51938, 27748 and 1968 cm^{-1}.[18] F^k and ζ values are always lower in crystals than in the free ion; thus, in light of the free ion analysis, BK's values of F^6 and ζ are unlikely to be correct. The observed reduction of F^2 to 81 percent of the free-ion value is similar to that found for $U^{4+}/ThBr_4$[19] but in that case ζ was only 1783 cm^{-1}.

In the reanalysis we initially assumed that the ratios $r_{42} = F^4/F^2$ and $r_{62} = F^6/F^2$ should lie somewhere between their free ion values (.82 and .53) and their values for $U^{4+}/ThBr_4$ (.96 and .64). We further assumed that the values of F^2 and ζ should be reduced from the free-ion values in the same ratio as found for $U^{4+}/ThBr_4$. Initial values of α, β, γ, M^k's and P^k's were taken from the $U^{4+}/ThBr_4$ analysis. Even though the data does not allow a determination of all of these parameters, it is important to include them at reasonable values. The distortion of the calculated level scheme due to errors of 20-30 percent in the values of these parameters is less than that caused by setting them equal to zero. This is particularly important for the tetravalent actinides because the spin-orbit coupling and crystal field interactions are both large. The states are so mixed that a number of different sets of parameters will produce moderately good fits (rms deviation ~ 100 cm^{-1}). Only a very good fit that allows further assignments of missing levels can guarantee a unique set of parameters. Finally, in our reanalysis we took account of the magnetic susceptibility data by forcing the first excited state to be greater than 150 cm^{-1} above the ground E state.

With the above assumptions it was immediately obvious that some of BK's uncertain origins could not be fit with our parameter values. For example, diagonalization of the Hamiltonian with our estimated free ion parameters and the crystal field parameters of BK resulted in a large gap between 18280 and ~ 20500 cm^{-1}. Consequently, the weak line at 19836 cm^{-1} must be assigned as a vibronic transition rather than an origin. Some of the uncertain origins in the near IR also had to be rejected. We eventually arrived at a list of 12-15 levels which could be fit with $F^2 \sim 42000$ cm^{-1} and $\zeta \sim 1840$ cm^{-1} and $\sigma \sim 100$ cm^{-1}. These values are in better accord with the $U^{4+}/ThBr_4$ results but ζ still seemed large. This led us to reconsider some of the more certain origins in BK's analysis.

The levels at ~ 20700 cm^{-1} are mixtures of $^1I_6$ and $^3P_{0,2}$ levels and are greatly affected by the configuration interaction parameters, which were not included by BK. The two origins at 20674 and 20771 (aa and bb of BK) fit their calculation quite well. Our calculations always placed them 250-300 cm^{-1} apart whereas the observed difference is ~ 100 cm^{-1}. There are four "strong" lines in this region: BK's origin z at 20541 cm^{-1}, aa at 20694, bb at 20771 and cc at 20854 cm^{-1}. The lines z and cc have the calculated separation and aa and bb can be interpreted as vibronic sidebands of z. This leaves unassigned the line at 21221 cm^{-1} (assigned by BK as a vibronic of bb) but which is in good agreement with the calculated position of the missing $^1I_6$ (T_2) level.

Introduction of these levels led to some reinterpretation of the near IR levels and finally resulted in fitting all 19 allowed transitions with σ = 79 cm^{-1}. But ζ remained at 1840 cm^{-1}. The largest deviation was for the T_1 level at 10080 cm^{-1}; this level was always calculated at ~ 10200 cm^{-1}.

Substitution of origin j at 10416 for origin i at 10080 reduced σ only to 71 cm^{-1} but reduced ζ to 1807 cm^{-1}, a value in much better accord with the $U^{4+}/ThBr_4$ results. The largest differences between experimental and observed levels were then for those states in which the $^3F_{3,4}$ and $^1G_4$ are strongly mixed, the same levels with large deviations in $U^{4+}/ThBr_4$. There was now one level (11389 cm^{-1}) with a deviation ~ 2σ. When it was omitted, σ decreased from 71 to 56 cm^{-1} and ζ to 1783 cm^{-1}.

Having arrived at a set of level assignments which could be fit reasonably well with a consistent set of parameter values, we looked to a detailed analysis of the vibronic transitions for confirmation of our choices and resolution of the ambiguities.

Vibrational Frequencies

Banks and Edelstein[12] (BE) have carried out a normal coordinate analysis of the vibrations of $Np(BH_4)_4$ and $Np(BD_4)_4$. Since the mass difference between ^{237}Np and ^{238}U is very small, similar vibrational frequencies are expected for $Np(BD_4)_4$ and $U(BD_4)_4$. Selection rules allow electronic transitions only to T_1 or T_2 states. These can couple with vibrations of any symmetry. Transitions to the forbidden A_1, A_2, and E states become allowed when coupled with T_1 or T_2 vibrations.

BK analyzed the $U(BD_4)_4/Hf(BD_4)_4$ spectrum with more different vibrational frequencies than are allowed by the normal coordinate analysis of BE. If we assume that the vibrational frequency of a given mode may be slightly different when it is associated with different electronic states, it should be possible to assign all of the vibronic transitions to the same normal modes observed in $Np(BD_4)_4$.

Table II shows the energies of the strong vibrations observed in the IR and Raman spectra of $Np(BD_4)_4$[12] along with the corresponding energies derived from the vibronic lines associated with the $Np(BD_4)_4/Zr(BD_4)_4$ level at 5605 cm^{-1} [13]. All of the T_2 vibrations are strong as are two of the A's and the lowest E. Also shown in Table

II are the corresponding vibrational frequencies associated with origin a of $U(BD_4)_4/Hf(BD_4)_4$. The designations are those of BK with ν_β replaced by β, etc. Only the vibration Ξ was not identified by BK.

The same strong vibrations are observed in all three crystals. There are some cases where several closely spaced vibrations were resolved in $Np(BD_4)_4$ and only one is seen in the diluted $U(BD_4)_4$. In such cases the exact correspondence is ambiguous. In both crystals there is a vibration of ~ 300 cm^{-1} which could be interpreted as the IR and Raman forbidden T_1, but for which an interpretation as a combination band is also possible.

Table III shows our assignment of the data of BK with this set of fundamental vibrations. Plausible assignments are possible for all of the lines if we accept small variations in energy for the same vibration coupled to different origins. (A true variation in energy cannot be separated from experimental uncertainty, however.)

Table IV shows the vibrational energies associated with each of the assigned origins. Except for the two lowest origins, the complete set of vibrations was not observed. When the vibrations are closely spaced this leads to some ambiguity in vibrational assignments. Since δ seems to show up more strongly than ϵ and η is the strongest vibration in the 400-500 cm^{-1} region, we have generally assigned these vibrations when some are missing. In assigning combination bands we have used the energies of the fundamentals associated with the origin in question. The discrepancy is generally < 10 cm^{-1} and within the experimental uncertainties.

Only the level at 6046 cm^{-1} (a + 114) cannot be assigned on this basis. There is no place for it in the normal coordinate analysis so it must be attributed to a lattice mode. An interpretation of the data in which this vibration, α, is also coupled to origins j, ℓ, and n, is possible, but, since the δ vibration associated with these origins has lower energy than when it is associated with origin a, we have assumed that the β vibration energy is also lower. In $Np(BD_4)_4/Zr(BD_4)_4$ there are two vibrational frequencies (43 and 138 cm^{-1}) associated with the lower energy levels and which are not found in the normal coordinate analysis.[12,13]

The vibrational analysis is consistent with our earlier choice of origins but does not clearly resolve the ambiguity regarding origin k at 11389 cm^{-1}. That transition is strong, as expected for an origin, but there is a weak transition at 11164 cm^{-1} which fits the calculated level position very well and on which one can also build an assignment of the vibronics. More phonons are involved, however. These assignments, with the origin designated as k', are indicated at the bottom of Table III. There are other cases where the origin is not the strongest transition but the differences in intensities are much less than in this case.

Given calculated energies which are certainly within 100 cm^{-1} of the experimental values, we looked for evidence of vibronics based on the unallowed origins. (These must involve only the T_1 or T_2 vibrations.) Such an interpretation is possible for some transitions but we can find at most three vibrations associated with a single origin. This makes such an interpretation possible but not compelling. We could interpret some of the intensity in origin k at 11389 cm^{-1} as arising from the Γ_3 level calculated at 9933 cm^{-1} plus vibration ξ and/or the Γ_1 at 10850 plus vibration η. This would make the alternate choice, k', at 11164 more likely. A detailed analysis of vibrational intensities is necessary to resolve such ambiguities in interpretation.

Since we lack a clear distinction between the two choices for origin k, both sets of parameters are included in Table V. However, because the relative values of F^k and ζ are more consistent with those of U^{4+}/ThBr$_4$ and the rms deviation is significantly smaller when origin k' is used, we regard k' as more likely to be the correct origin. Thus, the energy levels of calculation B are given in Table VI.

Discussion

The final free-ion parameter values are very similar to those found for U^{4+}/ThBr$_4$. F^2 is ~ 1000 cm^{-1} smaller, ζ is the same and the ratio $r_{42} = F^4/F^2 = .97$ vs. .96 for U^{4+}:ThBr$_4$. The value of r_{62} and, therefore, F^6 is probably too small, but the calculated levels are changed very little by forcing it to be higher.

If we use the measure

$$N_v/\sqrt{4\pi} = \left[\sum_{k,q} \frac{1}{2k+1} (B_q^k)^2\right]^{1/2}$$

of Auzel[20] as an indication of the relative crystal field strengths in crystals of different symmetry, we find $N_v/\sqrt{4\pi} = 3297$ cm^{-1} for $U(BD_4)_4/Hf(BD_4)_4$, 1340 cm^{-1} for $U^{4+}/ThBr_4$ and only 534 cm^{-1} for $U^{3+}/LaCl_3$. This difference probably accounts for the larger σ for $U(BD_4)_4$ than for $U^{4+}/ThBr_4$. With the larger crystal field strength, any deficiencies of the crystal field model are magnified.

The large crystal field strength for $U(BD_4)_4/Hf(BD_4)_4$ is apparent from a comparison of the eigenvectors in Table VI with those given previously[19] for $U^{4+}/ThBr_4$. In the latter case, for 65 of 70 states, it took only two SLJ levels to account for 90 percent of the eigenvector. For $U(BD_4)_4/Hf(BD_4)_4$ only ten states can be similarly represented by two or fewer components.

The larger crystal field strength is also reflected in the fact that for $U(BD_4)_4/Hf(BD_4)_4$ it is the free-ion, not the crystal field, parameters which have large statistical errors and which are sensitive to the choice of levels included in the least squares fit. As in the case of $U^{4+}/ThBr_4$, the largest discrepancies between the calculated and observed energies are for states in which the $^1G_4$ and $^3F_{3,4}$ levels are strongly mixed. Fits of the P^k parameters or calculations with different fixed values of the P^k did not improve the agreement of these levels. Consequently, those parameters were fixed at the values used for $U^{4+}/ThBr_4$. Any significant improvement in the fit can come only from modification of the crystal field Hamiltonian.

The wavefunctions and energies obtained from calculation B have been used to calculate the magnetic susceptibility of $U(BH_3CH_3)_4$ as shown in C of Figure 2. With the introductions of the orbital reduction factor k, obtained from the ratio $k = \zeta_{crys}/\zeta_{free\ ion} = 0.91$, curve D is obtained. Although the spectral shift from $U(BH_4)_4$ to $U(BH_3CH_3)_4$ is generally to higher energies in $U(BH_3CH_3)_4$, it was necessary to lower the energy of the first T_1 state to 215 cm^{-1} as well as the orbital reduction factor to 0.85 to obtain curve E. EPR measurements described in the following paper also show differences in orbital reduction factors between $Np(BH_4)_4$ and $Np(BH_3CH_3)_4$.

Conclusions

While some ambiguities remain, the current analysis of the energy levels of $U(BD_4)_4/Hf(BD_4)_4$ is much more coherent than the previous one. It predicts the right (E) ground state and the calculated position of the first excited state (T_1) is consistent with the magnetic susceptibility of $U(BH_3CH_3)_4$. All but one of the vibrations (assumed to be a lattice mode) have been plausibly assigned to normal modes with nearly the same energies as those observed for $Np(BD_4)_4/Zr(BD_4)_4$. All origins correspond to allowed transitions and all allowed transitions have been identified. Although some vibronics based on unallowed origins cannot be ruled out, there is no strong evidence for their existence. Resolution of the remaining ambiguities in assignments will probably require a detailed analysis of vibronic intensities.

There is some indication that the energies of some normal modes are slightly different in various excited states; experimental uncertainties and ambiguities due to the fact that several normal modes lie close together in energy make it impossible to draw any definite conclusions, however.

While the crystal field model works moderately well for this molecular crystal, there are indications that additional parameters are necessary to account for those covalency effects which are not absorbed by the F^k's: the rms deviation is larger than it is for $U^{4+}/ThBr_4$ and an orbital reduction factor $k = 0.85$ is required to fit the magnetic susceptibility data for $U(BH_3CH_3)_4$. For both $U(BD_4)_4/Hf(BD_4)_4$ and $U^{4+}/ThBr_4$, the largest discrepancies between observed and calculated levels are for those in which the $^1G_4$ and $^3F_{3,4}$ states are badly mixed. Perhaps this fact will provide a clue as to the form of a new parameterization scheme.

References

[a]This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Chemical Sciences Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

*Permanent address: Department of Physics, Kalamazoo College, Kalamazoo, Michigan 49007.

1. H. I. Schlesinger and H. C. Brown, J. Am. Chem. Soc., 75, 219 (1953).
2. H. R. Hoekstra and J. J. Katz, J. Am. Chem. Soc., 71, 2488 (1949).
3. R. H. Banks, N. M. Edelstein, R. R. Rietz, D. H. Templeton, and A. Zalkin, J. Am. Chem. Soc., 100, 1975, (1978).
4. W. E. Reid, Jr., J. M. Bish, and A. Brenner, J. Electrochem. Soc. 104, 21 (1957).
5. B. D. James, R. K. Nanda, and M. G. H. Wallbridge, J. Chem. Soc.(A), 182 (1966).
6. E. R. Bernstein and T. A. Keiderling, J. Chem. Phys. 59, 2105 (1973). This paper will be referred to as BK.
7. R. H. Banks and N. M. Edelstein in Lanthanide and Actinide Chemistry and Spectroscopy, N. Edelstein, Ed., ACS Symposium Series 131, Washington, D.C., 1980, p. 331. When the text refers to both the hydrogenated and deuterated species, the deuterated molecule is given in parentheses.
8. R. H. Banks, Ph.D. Thesis, LBL-10292, 1979.

9. R. H. Banks, N. M. Edelstein, B. Spencer, D. H. Templeton, and A. Zalkin, J. Am. Chem. Soc. 102, 620 (1980).

10. E. R. Bernstein, W. C. Hamilton, T. A. Keiderling, S. J. LaPlaca, S. J. Lippard, and J. Mayerle, J. Inorg. Chem. 11, 3009 (1972).

11. R. Shinomoto, E. Gamp, N. M. Edelstein, D. H. Templeton, and A. Zalkin, Inorg. Chem. (1983), in press.

12. R. H. Banks and N. Edelstein, J. Chem. Phys. 73, 3589 (1980).

13. K. Rajnak, R. H. Banks, E. Gamp, and N. Edelstein, J. Chem. Phys., following paper.

14. E. Gamp and N. Edelstein, J. Chem. Phys., following paper.

15. J. Owen and J. H. M. Thornley, Repts. Prog. Phys. 29, 675 (1966).

16. W. T. Carnall, H. Crosswhite, H. M. Crosswhite, J. P. Hessler, N. M. Edelstein, J. G. Conway, G. V. Shalimoff, and R. Sarup, J. Chem. Phys. 72, 5089 (1980).

17. B. G. Wybourne, Spectroscopic Properties of Rare Earths, John Wiley and Sons, Inc., New York, 1965. Our definition for the crystal field parameters follows Wybourne's convention.

18. C. H. H. Van Deurzen, K. Rajnak, and J. G. Conway, J. Opt. Soc. Amer., 1983, submitted.

19. P. Delamoye, K. Rajnak, M. Genet, and N. M. Edelstein, Phys. Rev. B, in press.

20. F. Auzel and O. L. Malta, J. Physique 44, 201 (1983).

Table I $U(BH_3CH_3)_4$ parameters and energy levels for fits considering only the $^3H_4$ ground term

	A^a		B^a	
	0	E	0	E
Energies (cm^{-1})	148	T_2	270	T_1
and symmetries	875	T_1	648	A_1
	2099	A_1	978	T_2
B_0^4 (cm^{-1})	-4442		2473	
B_0^6 (cm^{-1})	-2186		-1410	
k	0.79		0.79	

[a] As in Figure 2.

Table II. Fundamental vibrational modes of $U(BD_4)_4$ and $Np(BD_4)_4$

$Np(BD_4)_4$[a]			$Np(BD_4)_4$/ $Zr(BD_4)_4$[b]	$U(BD_4)_4$/ $Hf(BD_4)_4$[c]	Designation[d]
E(cm^{-1})	I(IR,R)[e]	Symmetry	E(cm^{-1})	E(cm^{-1})	
112	s,m	T_2	121	129	β
154	vs,s	E	156	177	δ
(204)	-	A_2	173	195	ε
(413)	-	E	409	419	ζ
(415)	-	T_2			
437	vs	T_2	430	449	η
475		A_1	474	464	θ
795	w	E	793	-	χ
(810)	-	T_1	803	-	κ
860	s,w	T_2	866	853[f]	Ξ[f]
917	s	T_2	914	929	λ
926	s	T_2	929		λ'
1516	-,s	A_1	1516	1520	ξ
1517	vs	T_2	1531	-	ξ'
1548	s	T_2	1542	-	ξ''

[a] Ref. 12. Infrared (IR) and Raman (R) measurements. Energies in ( ) are calculated values for IR and Raman forbidden transitions.

[b] Ref. 13. Energies deduced from vibronics associated with origin at 5605 cm^{-1}.

[c] Energies deduced from vibronics associated with origin a at 5932 cm^{-1}.

[d] Ref 6. ν_β has been replaced by β etc.

[e] I = intensity; vs = very strong; s = strong, m = medium; w = weak.

[f] Not identified as a fundamental vibration in Ref. 6.

Table III. Absorption spectrum of $U(BD_4)_4/Hf(BD_4)_4$ at 2 K, summary of transitions and vibrational assignments[a]

Energy (cm^{-1})	Relative Intensity	Assignment: Origin + $\Delta\nu(cm^{-1})$	Vibrational Identification
5932	95	origin a	
6046	5	$a + 114^{b}$	$a + \alpha^{b}$
6061	15	a + 129	$a + \beta$
6109	75	a + 177	$a + \delta$
6127	5	a + 195	$a + \varepsilon$
6237	5	a + 305	$a + \beta + \delta$
6280	25	a + 348	$a + 2\delta$
6351	2	a + 419	$a + \zeta$
6381	18	a + 449	$a + \eta$
6396	3	a + 464	$(a + \theta)$
6444	6	a + 512	$a + 3\delta$
6557	7	origin b'	
6578	2	a + 646	$a + \eta + \varepsilon$
6735	11	b' + 178	$(b' + \delta)$
6785	2	a + 853	$(a + \Xi)$
6810	2	b' + 253	$(b' + 2\beta)$
6861	12	b' + 314	$(b' + \beta + \delta)$
		a + 929	$a + \lambda$
6910	6	b' + 353	$(b' + 2\delta)$
6932	3	b' + 375	$(b' + 3\beta)$
6962	1	b' + 405	$(b' + \zeta)$
7028	3	b' + 471	$(b' + \theta)$
7070	7	b' + 513	$(b' + 4\beta)$
7119	3	b' + 562	$(b' + 2\varepsilon + \delta)$
7155	2	b' + 598	$(b' + \theta + \beta)$

Energy (cm^{-1})	Relative Intensity	Assignment: Origin + Δν(cm^{-1})	Vibrational Identification
7177	2	b' + 620	b' + 2β + 2δ
		a + 1245	a + λ + β + δ
7252	19	a + 1320	(a + λ + 2ε)
		b' + 695	(b' + 4δ)
7368	4	b' + 811	(b' + 2ε + ζ)
7452	7	a + 1520	a + ξ
7538	4	b' + 981	(b' + β + Ξ)
7585	2	a + 1653	(a + ξ + β)
7631	2	a + 1699	a + ξ + δ
7665	3	b' + 1098	(b' + 2β + Ξ)
		a + 1733	(a + ξ + ε)
7698	22	a + 1776	(a + ξ + 2β)
			(a + λ + Ξ)
			(a + λ + 2δ + η)
		b' + 1141	(b' + β + δ + Ξ)
7809	90	origin e	
7929	8	e + 120	e + β
7952	15	e + 143	(e + δ)
8058	5	e + 250	e + 2β
8098	12	e + 290	(e + 2δ)
8159	10	e + 350	e + 3β
8264	23	e + 455	e + η
8529	7	e + 720	(e + δ + β + η)
8668	5	e + 859	(e + Ξ)
8725	10	origin f'	
		e + 916	e + 2η
			e + λ

Energy (cm^{-1})	Relative Intensity	Assignment: Origin + $\Delta\nu(cm^{-1})$	Vibrational Identification
		origin g'	
8968	5	$f' + 243$	$(f' + 2\beta)$
		$a + 3036$	$(a + 2\xi)$
9148	1	$g' + 180$	$(g' + \delta)$
		$f' + 423$	$(f' + \eta)$
9352	2	$e + 1543$	$e + \xi'$
9589	10	origin g	
		$f' + 864$	$(f' + \Xi)$
9710	3	$g + 121$	$g + \beta$
9787	14	$g + 198$	$g + \varepsilon$
		$f' + 1062$	$(f' + \Xi + \varepsilon)$
10019	4	$g + 430$	$g + \eta$
10080	30	$f' + 1355$	$(f' + \lambda + \eta)$
		$g + 491$	$g + \theta$
		$g' + 1112$	$(g' + \lambda + \delta)$
10210	2	$g + 621$	$(g + \eta + \varepsilon)$
10354	3	$g + 765$	$(g + \beta + \varepsilon + \eta)$
10416	45	origin j	
10490	10	$g' + 1522$	$(g' + \xi)$
10527	20	$g + 938$	$(g + \lambda)$
		$j + 111$	$(j + \beta)$
10837	5	$j + 421$	$(j + \eta)$
10897	8	$j + 480$	$(j + \theta)$
11164	3	$j + 748$	$(j + 3\beta + \eta)$
11389	80	origin k	
11538	5	$k + 149$	$(k + \beta)$

Energy (cm^{-1})	Relative Intensity	Assignment: Origin + $\Delta\nu$(cm^{-1})	Vibrational Identification
11866	9	$k + 477$	$k + \theta$
12322	2	$k + 933$	$(k + \lambda)$
12628	13	origin ℓ	
12740	3	$\ell + 112$	$(\ell + \beta)$
13089	3	$\ell + 461$	$\ell + \eta$
13231	2	$\ell + 603$	$\ell + \delta + \eta$
		$k + 1842$	$(k + \xi + 2\delta)$
13831	100	origin n	
13978	20	$n + 124$	$n + \beta$
14120	2	$n + 268$	$n + 2\beta$
14288	25	origin n'	
		$n + 457$	$n + \eta$
14447	3	$n' + 159$	$(n' + \delta)$
14747	7	$n' + 459$	$(n' + \eta)$
15113	6	$n' + 825$	$(n' + \Xi)$
15373	3	$n' + 1085$	$(n' + \lambda + \delta)$
		$n + 1542$	$n + \xi$
15440	11	$n' + 1152$	$(n' + \lambda + 2\beta)$
15596	2	$n' + 1308$	$(n' + \lambda + 2\beta + \delta)$
		$n + 1765$	$(n + \xi + 2\beta)$
15749	2	$n + 1918$	$(n + \xi + 2\varepsilon)$
15924	1	$n + 2093$	$(n + \xi + 3\varepsilon)$
		$n' + 1636$	$(n' + 2\Xi)$
16011	4	$n' + 1723$	$(n' + \xi + \delta)$
16057	9	origin r	
16182	1	$r + 126$	$r + \beta$
16357	10	$r + 300$	$(r + \beta + \delta)$

Energy (cm^{-1})	Relative Intensity	Assignment: Origin + $\Delta\nu$(cm^{-1})	Vibrational Identification
16394	2	r + 337	$(r + 2\delta)$
16497	3	r + 440	$(r + \eta)$
16836	2	r + 779	$(r + \eta + 2\delta)$
16973	1	r + 916	$(r + \lambda)$
17266	1	r + 1209	$(r + \lambda + \beta + \delta)$
17622	62	origin t	
17672	1	r + 1615	$(r + 2\beta + \eta + \lambda)$
17741	13	t + 119	$(t + \beta)$
17859	1	t + 237	$(t + 2\beta)$
18016	3	t + 394	$(t + 2\beta + \delta)$ $(t + \zeta)$
18075	26	t + 453	$t + \eta$
18197	7	t + 575	$(t + \eta + \beta)$
18280	35	origin u	
18398	6	u + 118	$(u + \beta)$
18455	6	u + 175	$u + \delta$
18517	3	t + 895 u + 237	$t + 2\eta$ $u + 2\beta$
18575	2	t + 953 u + 295	$t + \lambda$ $(u + \beta + \delta)$
18686	10	u + 406	$u + \zeta$
18753	1	u + 473	$u + \theta$
18785	3	t + 1163	$(t + \lambda + 2\beta)$
18873	2	u + 593	$(u + \theta + \beta)$
18970	3	t + 1348 u + 690	$t + 3\eta$ $(u + \beta + \delta + \zeta)$
19090	4	u + 810	$u + 2\zeta$

Energy (cm^{-1})	Relative Intensity	Assignment: Origin + $\Delta\nu$(cm^{-1})	Vibrational Identification
19185	4	t + 1563	$t + \xi$
19277	1	t + 2015	$(t + \eta + \xi)$
19374	1	t + 2174	$(t + \xi + \delta + \eta)$
19618	1	u + 1338	$(u + 3\zeta + \beta)$
19836	3	u + 1556	$u + \xi$
19899	1	u + 1619	$(u + \eta + 2\beta + \lambda)$
20394	2	u + 2114	$(u + \beta + \eta + \xi)$
20511	2	u + 2231	$(u + 2\beta + \eta + \xi)$
20541	15	origin z	
20694	80	z + 153	$(z + \delta)$
20771	30	z + 230	$(z + 2\beta)$
20854	75	origin cc	
		z + 313	$(z + 2\delta)$
21003	4	z + 462	$(z + \eta)$
		cc + 149	$(cc + \delta)$
21140	1	cc + 286	$(cc + \beta + \delta)$
21221	18	origin z'	
21298	15	cc + 444	$cc + \eta$
21513	3	z' + 292	$(z' + \beta + \delta)$
21578	1	z + 1037	$(z + \lambda + \beta)$
		cc + 724	$(cc + \eta + \beta + \delta)$
21644	3	z' + 423	$(z' + \eta)$
21766	1	cc + 912	$(cc + \lambda)$
21924	2	z' + 703	$(z' + \beta + \delta + \eta)$
		cc + 1070	$(cc + \lambda + \delta)$
22040	1	z + 1499	$(z + \beta + \eta + \lambda)$
		cc + 1186	$(cc + \beta + \delta + \lambda)$

Energy (cm^{-1})	Relative Intensity	Assignment: Origin + $\Delta\nu$(cm^{-1})	Vibrational Identification
22226	2	cc + 1352	(cc + η + λ)
23473	1	cc + 2619	(cc + λ + ξ + δ)
23769	2	cc + 2915	(cc + λ + ξ + η)
23940	2	cc + 3086	(cc + 2ξ)
24312	2	z' + 3098	(z' + 2ξ)
24795	27	origin ee	
24924	10	ee + 129	ee + β
25271	7	ee + 476	ee + θ
25393	3	ee + 598	ee + β + θ
25753	3	ee + 958	(ee + 2θ)
25859	2	ee + 1064	(ee + λ + β)
26219	2	ee + 1424	ee + 3θ
26371	1	ee + 1576	(ee + ξ)
26645	1	ee + 1850	(ee + ξ + 2β)
		Alternate interpretation for origin k	
11164	3	origin k'	
11389	80	k + 234	(k' + 2β)
11538	5	k + 383	(k' + 2β + δ)
11866	9	k + 711	(k' + θ + 2β)
12322	2	k + 1158	(k' + λ + 2β)

---

[a] Energies and intensities are taken from BK. Their notation is retained with ν_β replaced by β, etc. The unprimed origins are those of BK as are the vibrational identifications without parentheses. The primed origins and vibrational identifications in parentheses are those of the current analysis.

[b] This frequency has no place in the normal coordinate analysis and is probably a lattice mode.

Table IV. Vibrational energies (cm^{-1}) associated with the various origins of $U(BD_4)/Hf(BD_4)_4$

Origin[a]	β	δ	ε	ζ	η	θ	Ξ	λ	ξ	ξ'
a	129	177	195	419	449	464	853	929	1520	-
b'	127	178	192	405	-	471	854	936	-	-
e	120	143	-	-	455	-	859	916	-	1543
f'	122	-	198	-	423	-	864	932	-	-
g'	-	180	204	-	441	-	-	932	1522	-
g	121	-	198	-	430	491	-	938	-	-
j	111	-	-	-	421	480	-	-	-	-
k	-	149	-	-	-	477	-	933	-	1544
(k'[b]	117	149	-	-	-	477	-	933	-	-)
	112	142	-	-	461	-	-	-	-	
n	124	-	188	-	457	-	-	-	-	1542
n'	113	159	-	-	459	-	825	926	-	1564
r	126	-	174	-	440	-	-	916	-	-
t	119	157	-	394	453	-	-	953	-	1563
u	118	-	175	406	-	473	-	-	-	1556
z	115	153	-	-	462	-	-	924	-	-
cc	137	149	-	-	444	-	-	912	-	1543
z'	129	-	163	-	423	-	-	-	-	1549
ee	129	-	-	-	-	476	-	935	-	1576

---

[a] Unprimed origins are those of BK; others were assigned in this analysis.

[b] Alternate for origin k.

Table V. Parameter values (cm^{-1}) for U^{4+} [a]

Parameter	U(BD$_4$)$_4$/Hf(BD$_4$)$_4$			U^{4+}/ThBr$_4$ [b]	U V [c]
	BK[d]	This Work Calc. A[e]	Calc. B[f]		
F^2	42008	41121(236)	41280(175)	42253(127)	51938(39)
F^4	37679	38849(1071)	40013(826)	40458(489)	42708(100)
F^6	28048	21711(827)	22554(625)	25881(383)	27748(68)
α	-	40(3)	38(2)	31(1)	35.5(0.4)
β	-	[-648]	[-648]	-644(75)	-664(25)
γ	-	[1200]	[1200]	[1200]	744(26)
ζ	1910.8	1807(16)	1782(12)	1783(7)	1968(2)
P^2	-	[500]	[500]	[500]	573(66)
P^4	-	[500]	[500]	[500]	524(144)
P^6	-	[500]	[500]	[500]	1173(321)
B^4_0	-3484	-2486(170)	-2445(124)	1316(146)	-
B^6_0	-4240	-5287(113)	-5371(81)	-3170(379)	-
B^4_4	-	-	-	-2230(85)	-
B^6_4	-	-	-	686(246)	-
B^2_0	-	-	-	-1096(80)	-
Number of Levels	11	19	19	26	13
σ	62	71	53	36	9.8
r_{42}	0.90	0.97	0.97	0.96	0.82
r_{62}	0.67	0.55	0.55	0.64	0.53

---

[a] Rms errors are in ( ). Parameters in [] were held fixed. In all cases $M^0 = 0.987$, $M^2 = 0.550$, $M^4 = 0.384$.

[b] Ref. 19.

[c] Ref. 18.

[d] Ref. 6.

[e] With 11389 cm^{-1} as origin k.

[f] With 11164 cm^{-1} as origin (k').

Table VI. Observed and calculated energy levels (cm^{-1}) of U(BD$_4$)$_4$/Zr(BD$_4$)$_4$

State	E_{calc}	E_{obs}[a]	ΔE[b]	Eigenvector[c]
E	0	0	0	$94\ ^{3}H_{4} + 3\ ^{1}G_{4}$
T_1	370	-	-	$76\ ^{3}H_{4} + 11\ ^{3}H_{5} + 7\ ^{1}G_{4}$
T_2	531	-	-	$78\ ^{3}H_{4} + 11\ ^{1}G_{4} + 4\ ^{3}F_{3}$
A_1	2036	-	-	$63\ ^{3}H_{4} + 27\ ^{1}G_{4}$
E	2750	-	-	$57\ ^{3}F_{2} + 31\ ^{3}H_{4} + 9\ ^{1}D_{2}$
T_2	3562	-	-	$58\ ^{3}F_{2} + 20\ ^{3}H_{5} + 10\ ^{1}D_{2} + 3\ ^{3}H_{6}$
T_1	5995	5932	63	$73\ ^{3}H_{5} + 9\ ^{3}F_{3} + 9\ ^{3}F_{4}$
T_2	6545	(6557)	-12	$67\ ^{3}H_{5} + 15\ ^{3}F_{2} + 6\ ^{3}H_{6} + 5\ ^{1}D_{2}$
A_2	6877	-	-	$64\ ^{3}F_{3} + 35\ ^{3}H_{6}$
E	7828	-	-	$54\ ^{3}H_{5} + 18\ ^{3}F_{2} + 9\ ^{1}D_{2} + 7\ ^{3}F_{4} + 7\ ^{3}H_{6}$
T_1	7829	7809	20	$86\ ^{3}H_{5} + 6\ ^{3}H_{4}$
T_1	8697	(8725)	-28	$62\ ^{3}F_{3} + 18\ ^{3}H_{6} + 11\ ^{3}H_{5}$
A_1	8832	-	-	$45\ ^{3}H_{6} + 22\ ^{3}F_{4} + 17\ ^{3}H_{4} + 12\ ^{1}G_{4}$
T_2	9041	(8968)	73	$51\ ^{3}F_{3} + 25\ ^{1}G_{4} + 13\ ^{3}F_{4} + 4\ ^{3}H_{6}$
T_2	9661	9589	72	$28\ ^{3}F_{3} + 24\ ^{3}F_{4} + 18\ ^{1}G_{4} + 12\ ^{3}H_{4}$
E	9933	-	-	$58\ ^{3}F_{4} + 17\ ^{3}H_{6} + 20\ ^{1}G_{4}$
T_1	10356	(10416)	-60	$39\ ^{3}F_{4} + 28\ ^{1}G_{4} + 9\ ^{3}F_{3} + 18\ ^{3}H_{6}$
A_1	10849	-	-	$44\ ^{1}G_{4} + 26\ ^{3}H_{6} + 18\ ^{3}H_{4} + 6\ ^{1}I_{6}$
T_2	11172	11164	8	$82\ ^{3}H_{6} + 5\ ^{3}H_{5} + 5\ ^{1}I_{6}$
A_2	11236	-	-	$61\ ^{3}H_{6} + 36\ ^{3}F_{3}$
T_1	12572	12628[d]	-56	$55\ ^{3}H_{6} + 20\ ^{1}G_{4} + 14\ ^{3}F_{3} + 5\ ^{1}I_{6}$
E	13356	-	-	$42\ ^{3}H_{6} + 20\ ^{1}I_{6} + 14\ ^{1}G_{4} + 10\ ^{3}H_{5} + 8\ ^{3}F_{2}$

Table VI. Continued

State	E_{calc}	E_{obs}[a]	ΔE[b]	Eigenvector[c]
T_2	13809	13839	-30	$44\ {}^1D_2 + 21\ {}^3P_2 + 12\ {}^1I_6 + 11\ {}^3F_2 + 10\ {}^3H_6$
T_2	14289	(14288)	1	$40\ {}^3H_6 + 13\ {}^3F_3 + 18\ {}^1I_6 + 8\ {}^1G_4 + 8\ {}^3F_4 + 5\ {}^3F_2$
E	14796	–	–	$48\ {}^1D_2 + 22\ {}^3P_2 + 11\ {}^3F_2 + 15\ {}^3H_6$
A_1	15632	–	–	$65\ {}^3P_2 + 20\ {}^3F_4 + 7\ {}^1S_0$
T_2	15996	16057	-62	$49\ {}^3F_4 + 24\ {}^1G_4 + 21\ {}^3H_6$
A_1	16655	–	–	$48\ {}^3F_4 + 17\ {}^3P_0 + 24\ {}^3H_6 + 9\ {}^1G_4$
T_1	17649	17622	27	$42\ {}^3F_4 + 42\ {}^1G_4 + 4\ {}^3P_1 + 4\ {}^3H_4$
E	18209	–	–	$27\ {}^1G_4 + 26\ {}^3F_4 + 12\ {}^3P_2 + 15\ {}^1I_6 + 14\ {}^1I_6$
T_1	18283	18280	3	$92\ {}^3P_1 + 2\ {}^3H_6$
T_2	20549	(20541)	8	$57\ {}^1I_6 + 27\ {}^3P_2 + 13\ {}^3H_6$
A_1	20774	–	–	$87\ {}^1I_6 + 4\ {}^3P_0$
T_2	20815	(20854)	-39	$59\ {}^1I_6 + 31\ {}^3P_2$
T_1	21239	(21221)	18	$92\ {}^1I_6 + 6\ {}^3H_6$
A_2	21330	–	–	$96\ {}^1I_6 + 3\ {}^3H_6$
E	21691	–	–	$29\ {}^1G_4 + 39\ {}^1I_6 + 15\ {}^3P_2 + 5\ {}^1D_2 + 4\ {}^3F_4$
E	22791	–	–	$43\ {}^3P_2 + 26\ {}^1D_2 + 23\ {}^1I_6$
T_2	24827	24795	32	$34\ {}^1D_2 + 42\ {}^1I_6 + 19\ {}^3P_2$
A_1	38894	–	–	$88\ {}^1S_0 + 7\ {}^3P_0$

---

[a] Data from Ref. 6. Assignments which differ from theirs are in parenthses.

[b] $\Delta E = E_{calc} - E_{obs}$.

[c] Percent of SLJ state. Enough components are given, at least 2, to include 90 percent of the state.

[d] There was a misprint in Table IV of Ref. 6.

Figure Captions

Figure 1. Optical spectra of $U(BH_3CH_3)_4$ and $U(BH_4)_4$ in C_6D_6 at room temperature.

Figure 2. Paramagnetic susceptibility of $U(BH_3CH_3)_4$;

A,B: calculated considering only $^3H_4$ from parameters in Table 1.

C: calculated from the parameters of the optical analysis (calculation B in Table V)

D: same as C, but with an orbital reduction factor $k = 0.91$.

E: same as C, but T_1 ($^3H_4$) moved to 215 cm^{-1} and orbital reduction factor $k = 0.85$.

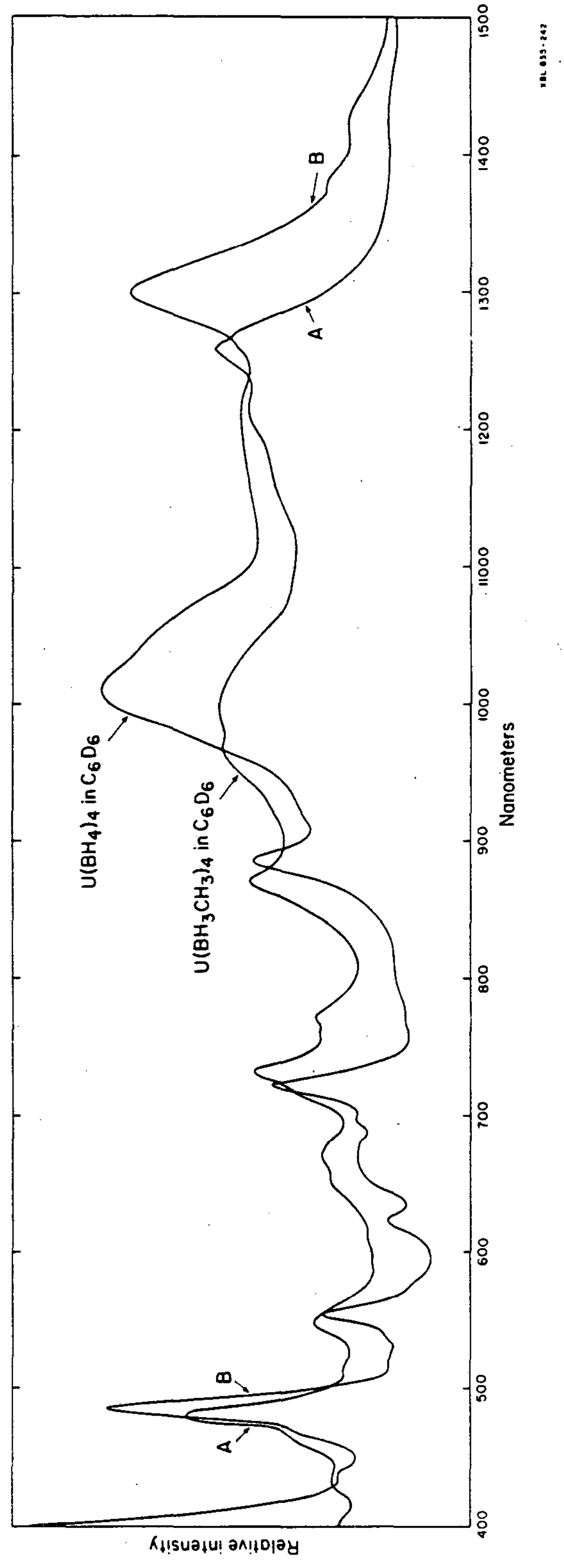

Relative intensity
Nanometers
400
500
600
700
800
900
1000
1100
1200
1300
1400
1500
$U(BH_4)_4$ in C_6D_6
$U(BH_3CH_3)_4$ in C_6D_6
A
B
A
B
XBL 835-242

Figure 1

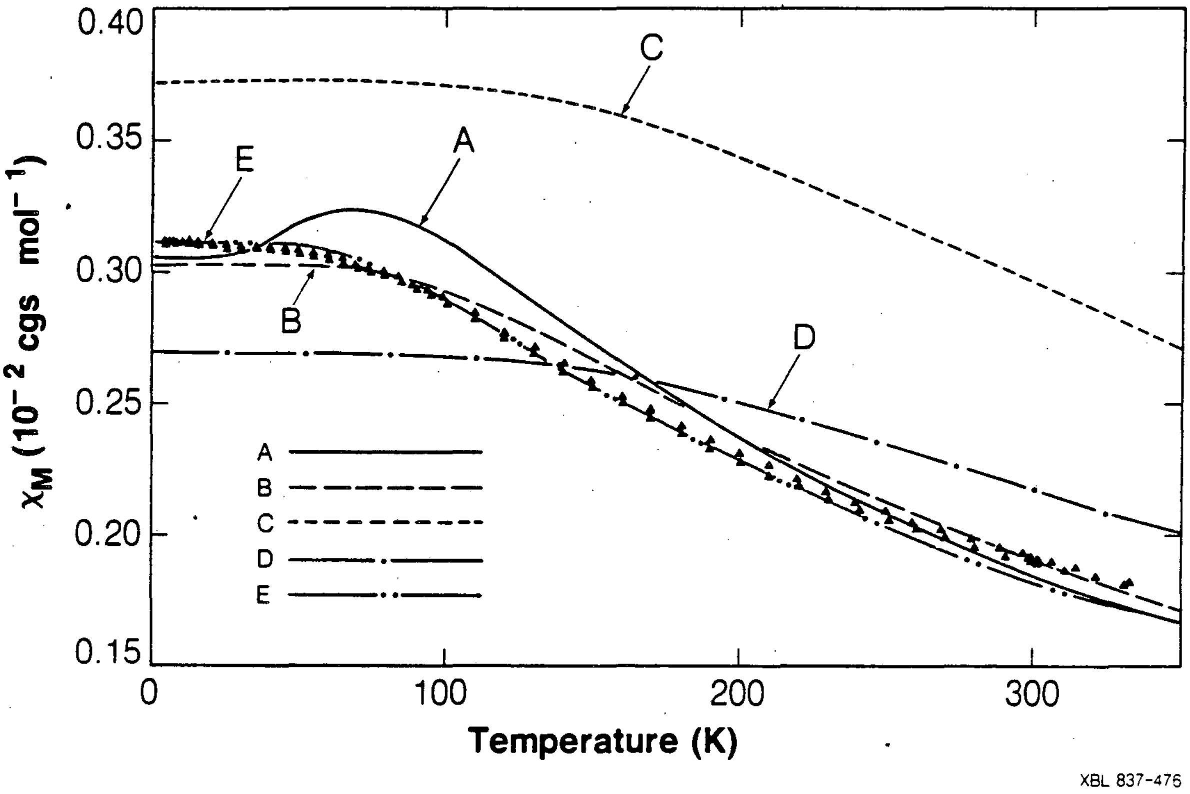

0.40
0.35
0.30
0.25
0.20
0.15
χ_M (10^{-2} cgs mol^{-1})
0
100
200
300
Temperature (K)
A
B
C
D
E
XBL 837-476

Figure 2

TECHNICAL INFORMATION DEPARTMENT
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720