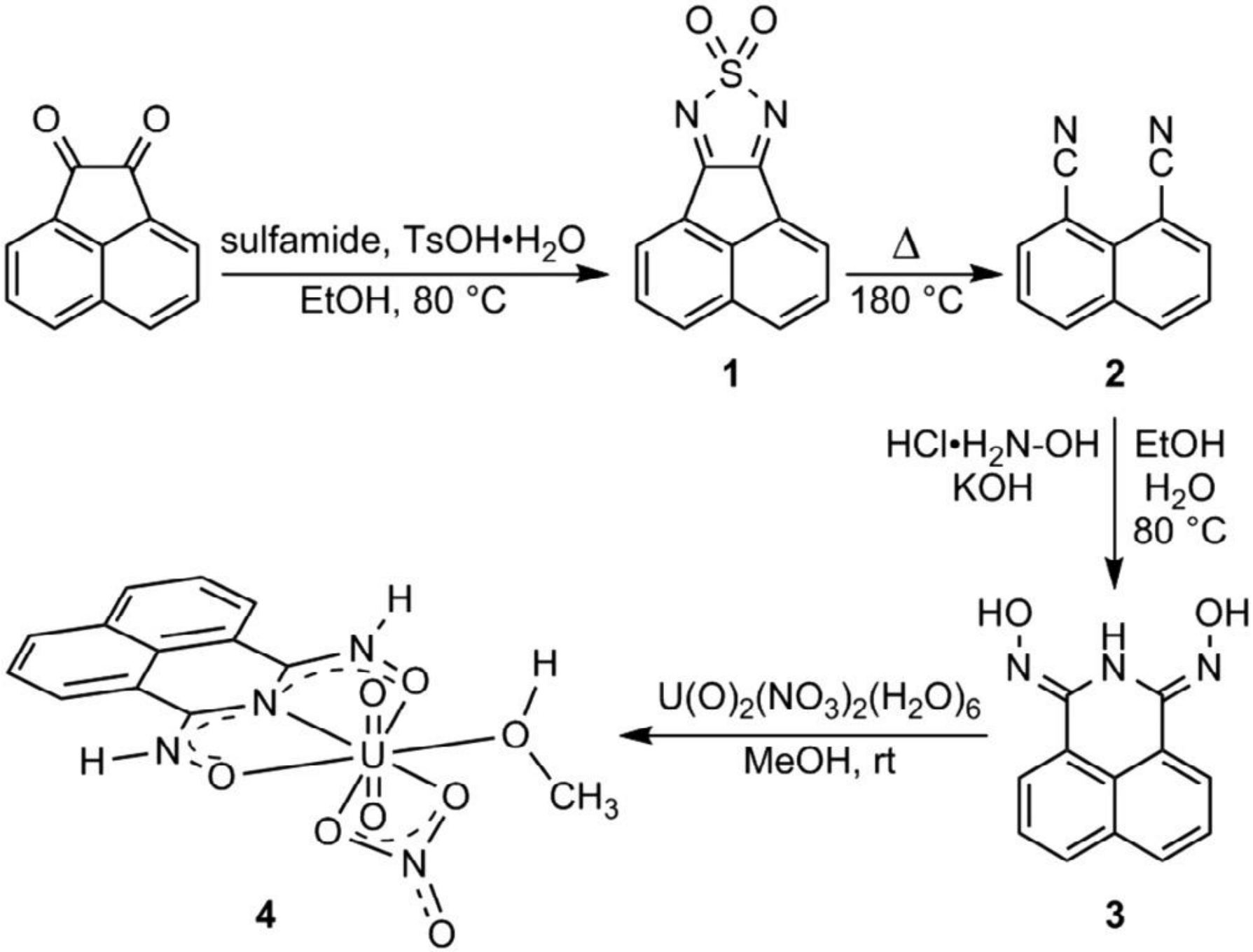

sulfamide, TsOH•H2O
EtOH, 80 °C
Δ
180 °C
1
2
HCl•H2N-OH
KOH
EtOH
H2O
80 °C
3
U(O)2(NO3)2(H2O)6
MeOH, rt
4

Scheme 1. Synthesis of cyclic imide dioxime, **3**, and its uranyl complex, **4**.

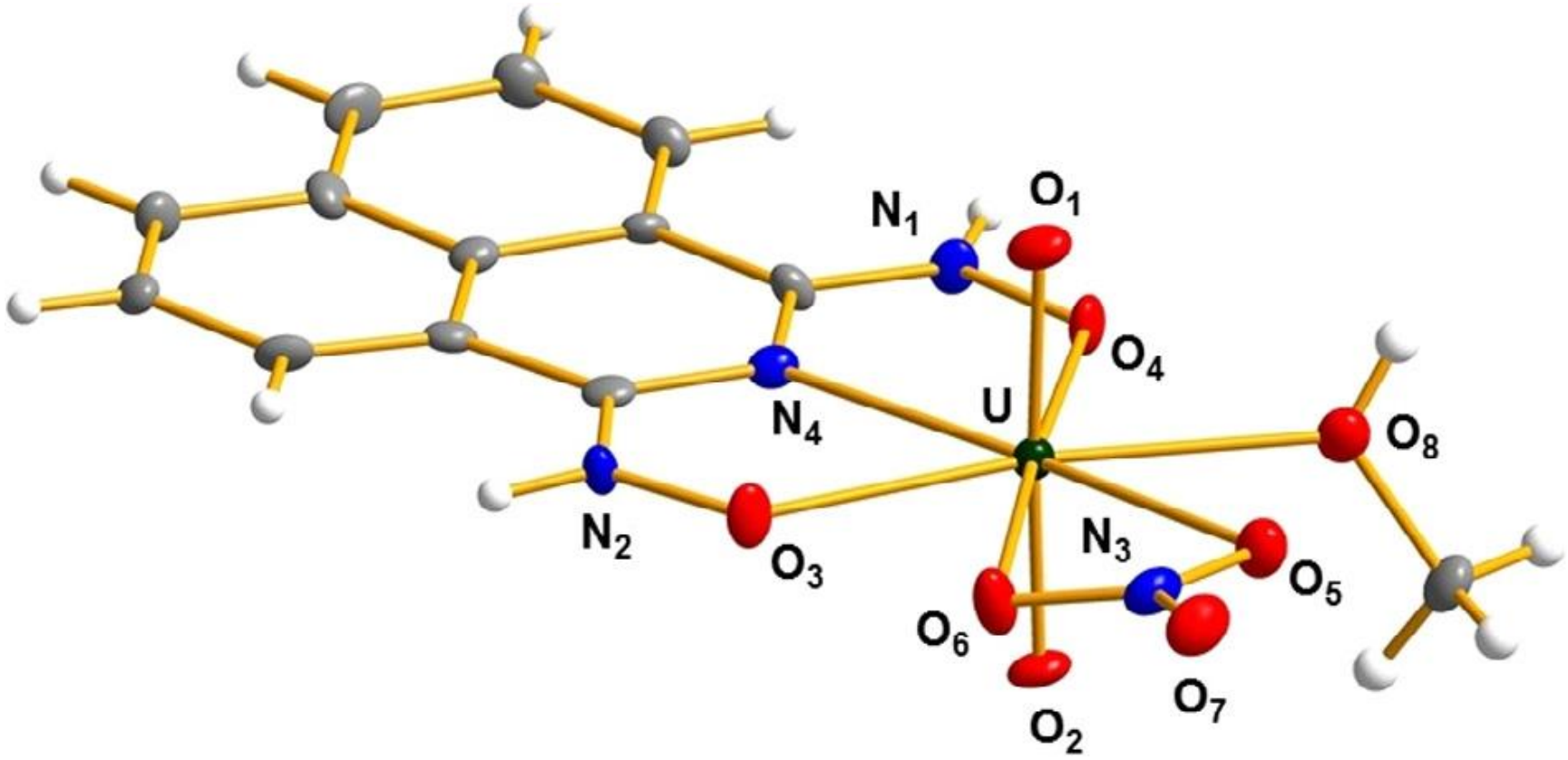
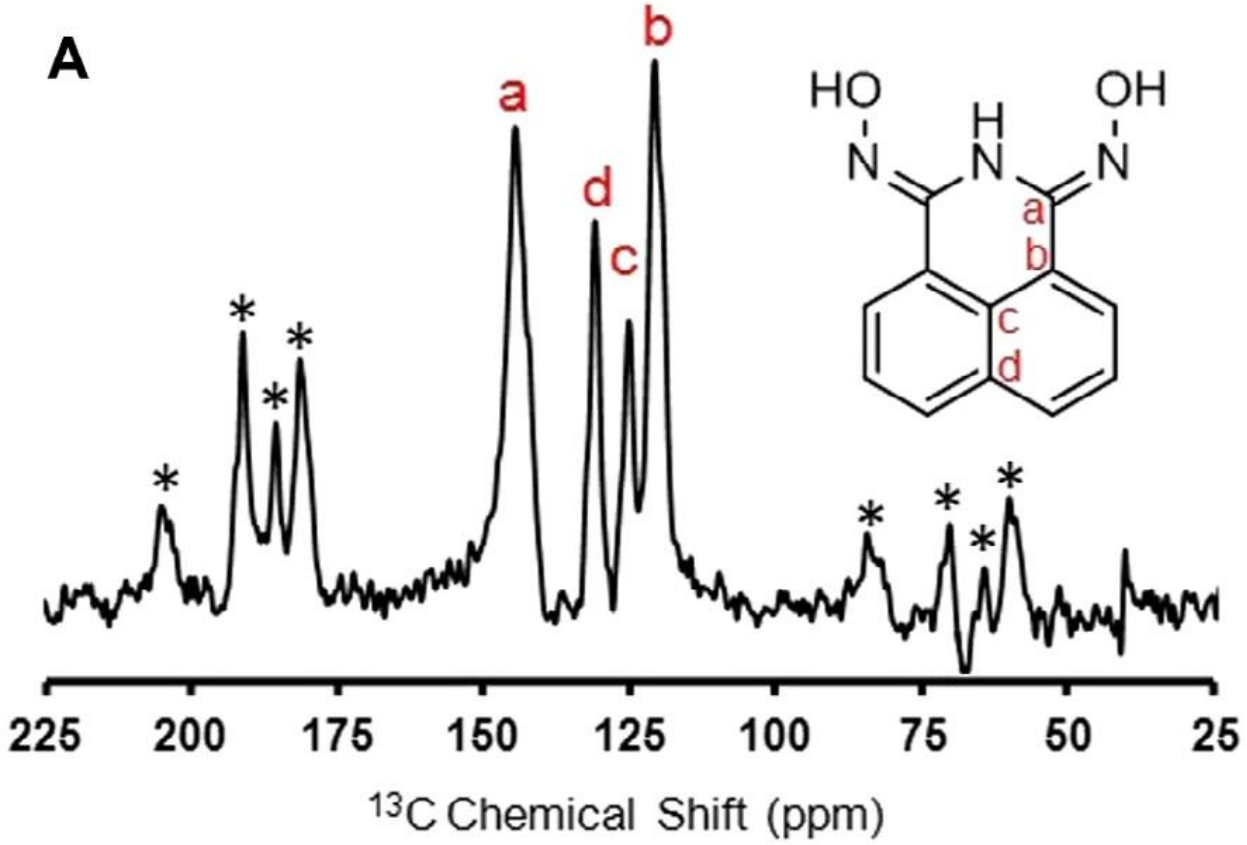


Fig. 1. Crystal structure of (Np-CAO-H_2)U$(O)_2(NO_3)(CH_3OH)$ (**4**) that shows tridentate binding of cyclic amidoxime ligand. Green, blue, red, and gray ellipsoids (50% probability) represent U, N, O, and C, respectively. White spheres represent H. Selected bond distances (Å) and angles (°) are as follows: U-O_1, 1.781(6); U-O_2, 1.767(6); U-O_3, 2.398(6); U-O_4, 2.420(6); U-O_5, 2.589(6); U-O_6, 2.572(6); U-O_8, 2.462(7); U-N_4, 2.514(7); O_1-U-O_2, 177.1(3); O_3-U-O_8, 170.1(2); O_4-U-O_6, 176.8(2); O_5-U-N_4, 169.0(2).


A
a
b
c
d
HO
OH
N
H
N
N
a
b
c
d
*
225
200
175
150
125
100
75
50
25
13C Chemical Shift (ppm)

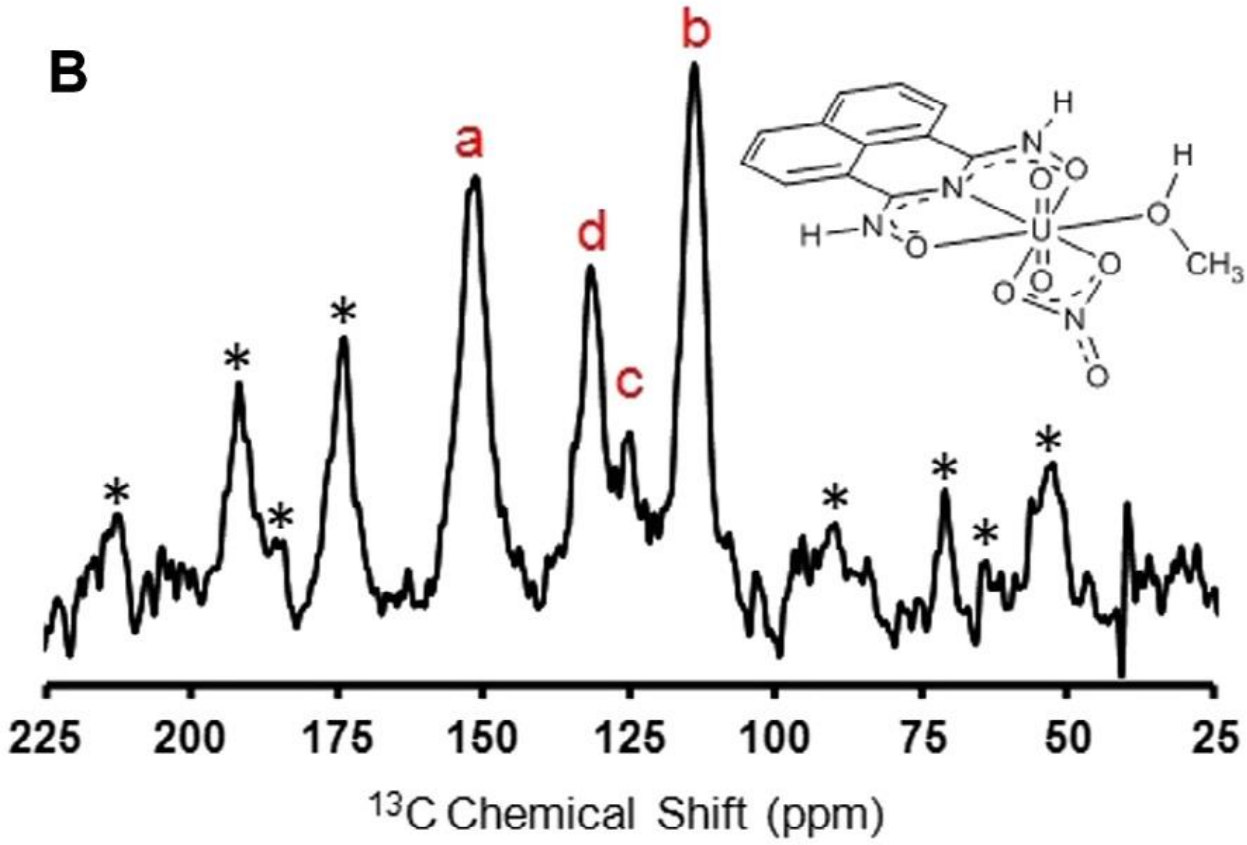

B
a
b
c
d
H
N
O
U
CH3
*
225
200
175
150
125
100
75
50
25
13C Chemical Shift (ppm)

Fig. 2. (A) The ^{13}C CP MAS NMR of **3** with interrupted decoupling exhibiting the resonances of the four quaternary carbons. (B) The ^{13}C CP MAS NMR of **4** with interrupted decoupling revealing the shifted resonances of the four quaternary carbons in the ligand. *Denote spinning sidebands.

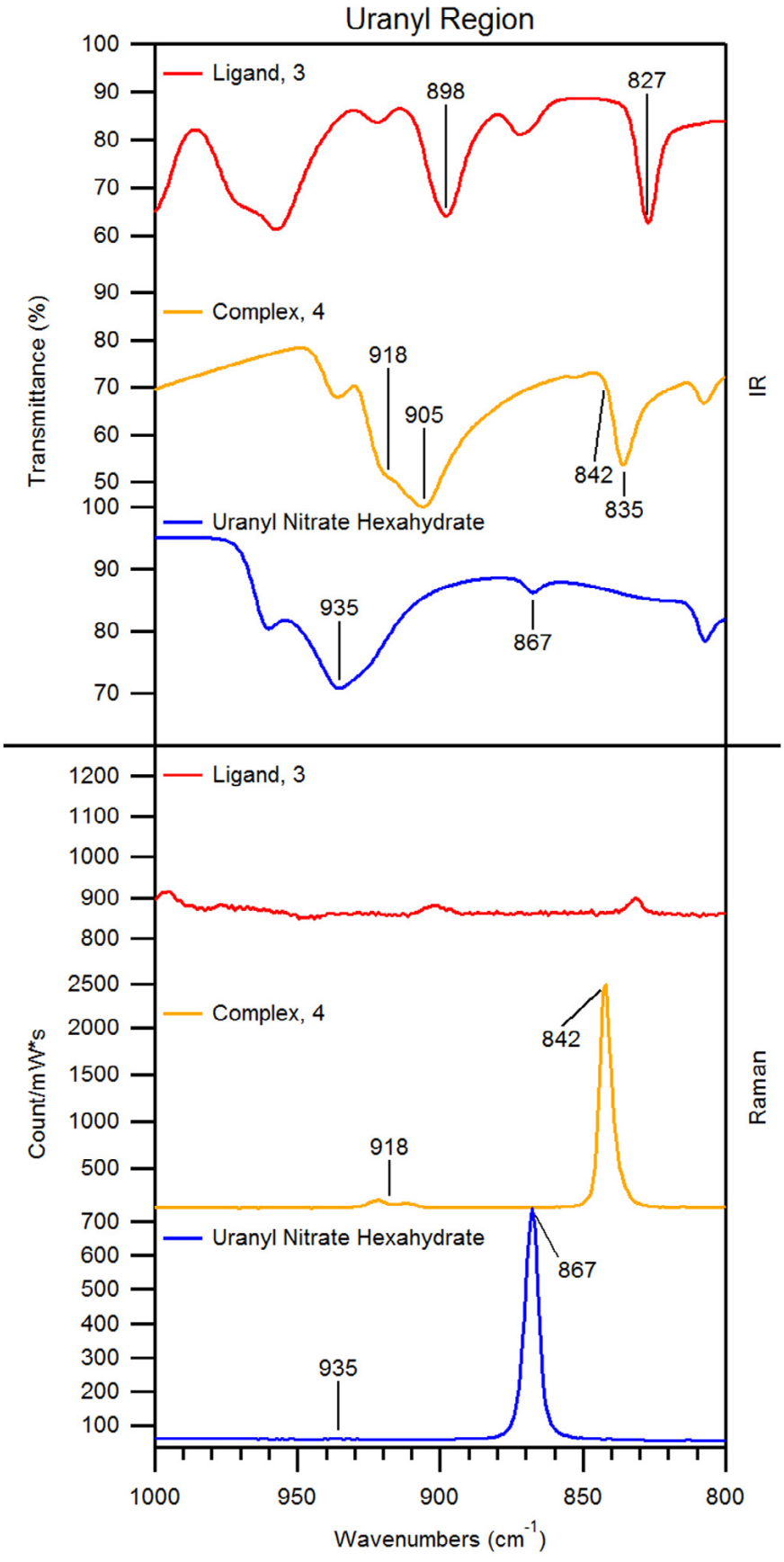

Uranyl Region
Ligand, 3
898
827
Complex, 4
918
905
842
835
Uranyl Nitrate Hexahydrate
935
867
Transmittance (%)
IR
Ligand, 3
Complex, 4
842
918
Uranyl Nitrate Hexahydrate
867
935
Count/mW*s
Raman
Wavenumbers (cm-1)

Fig. 3. The uranyl region ($U(O)_2$) of the vibrational spectra, both IR (top) and Raman (bottom). The spectra of ligand **3**, complex **4**, and uranyl nitrate hexahydrate are shown from top to bottom. The positions, left to right, of the antisymmetric (ν_{as}) and symmetric (ν_s) U=O stretches are labeled for uranyl and the complex while competing vibrations are labeled for the ligand.

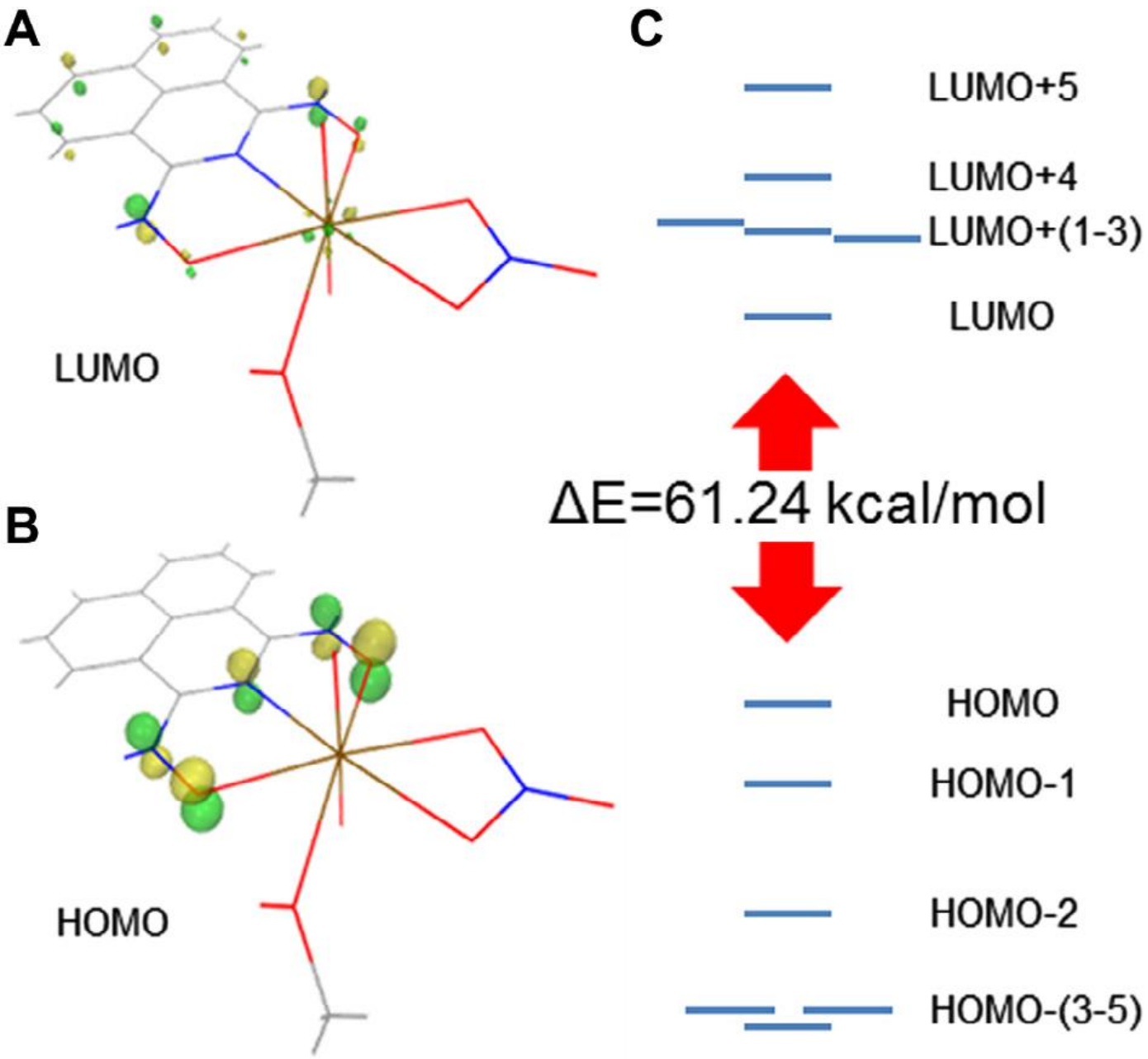

A
LUMO
B
HOMO
C
LUMO+5
LUMO+4
LUMO+(1-3)
LUMO
ΔE=61.24 kcal/mol
HOMO
HOMO-1
HOMO-2
HOMO-(3-5)

Fig. 4. (A) LUMO of (Np-CAO-H_2)U$(O)_2(NO_3)(CH_3OH)$ (**4**). (B) HOMO of complex **4**. (C) Orbital splitting diagram of complex **4** obtained from DFT calculations. All calculations used NWChem/B3LYP/6-311++g**.

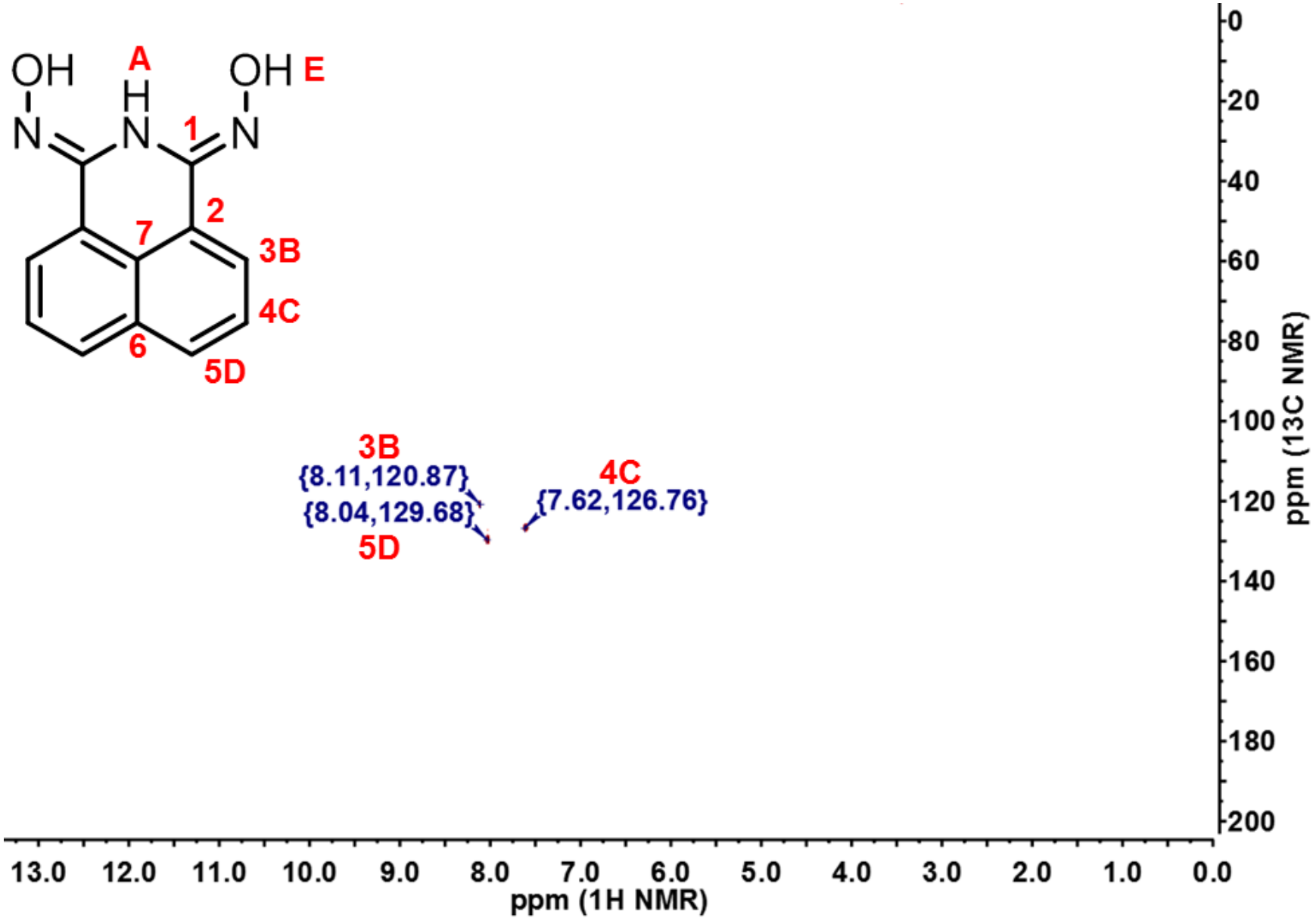


Figure S1. The HSQC NMR spectrum of **3** in DMSO-d_6.

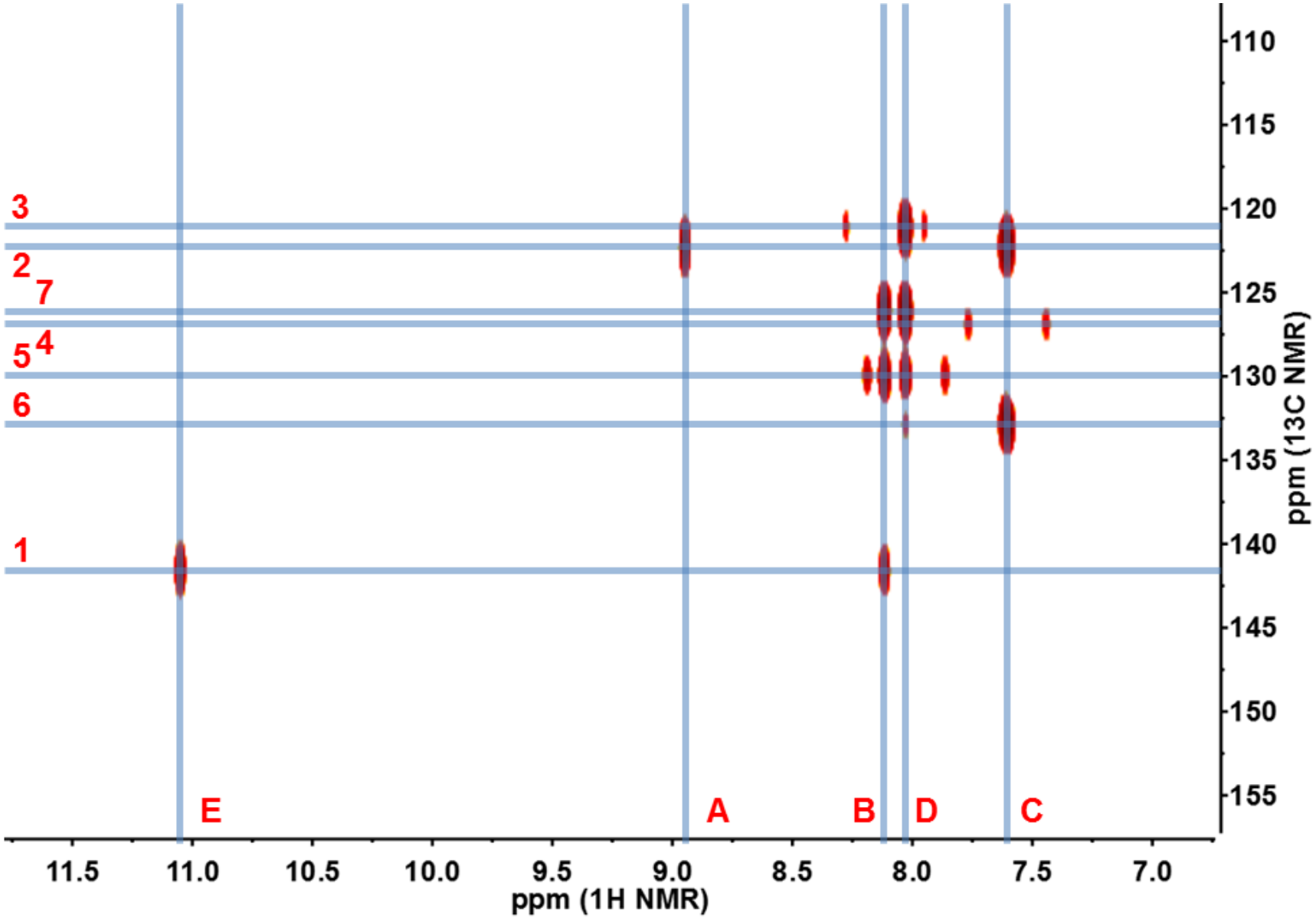


Figure S2. The HMBC NMR spectrum of **3** in DMSO-d_6.

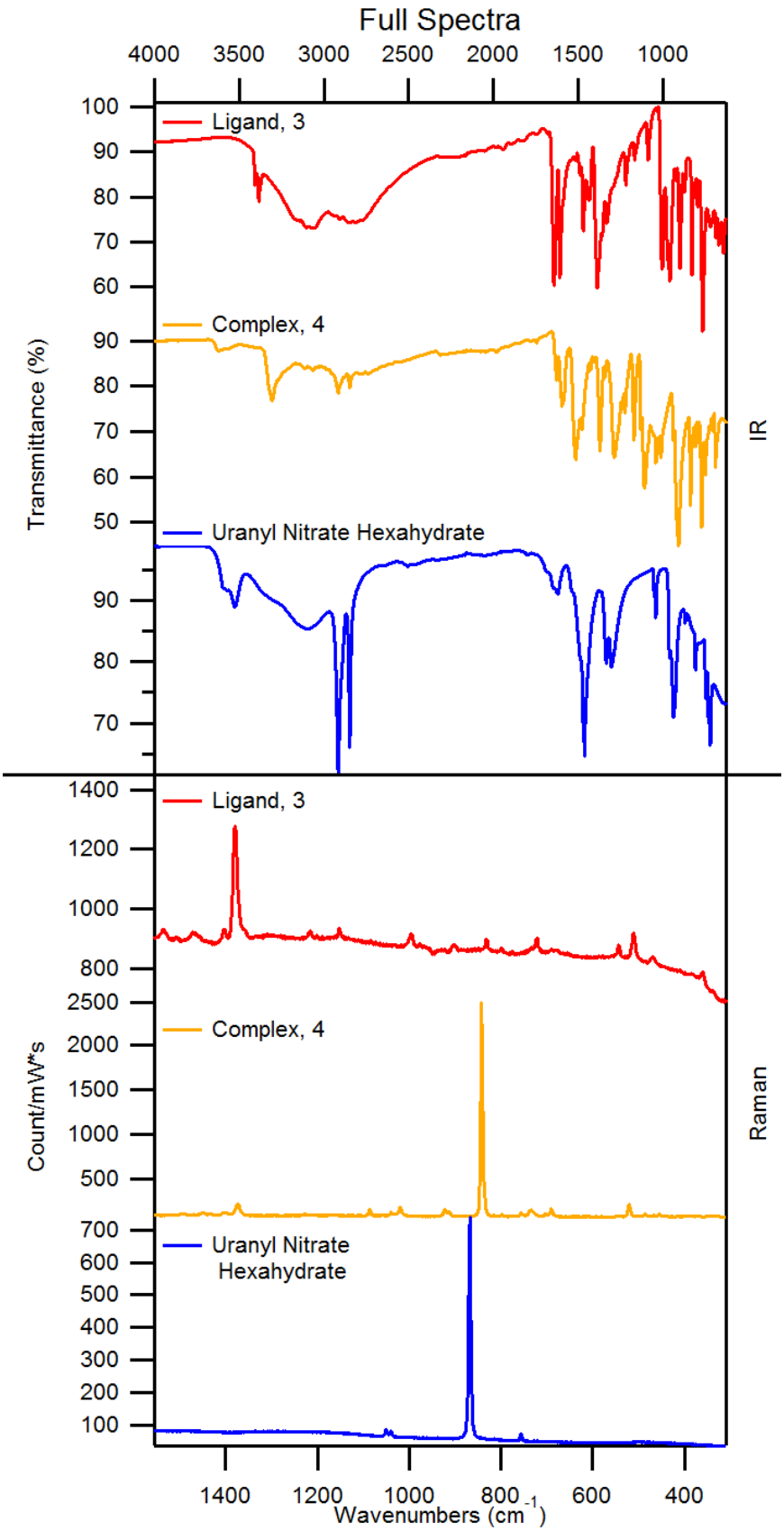

Full Spectra
4000
3500
3000
2500
2000
1500
1000
Ligand, 3
Complex, 4
Uranyl Nitrate Hexahydrate
Transmittance (%)
IR
Ligand, 3
Complex, 4
Uranyl Nitrate
Hexahydrate
Count/mW*s
Raman
1400
1200
1000
800
600
400
Wavenumbers (cm-1)

Figure S4. The full Raman and IR spectra of the samples collected.

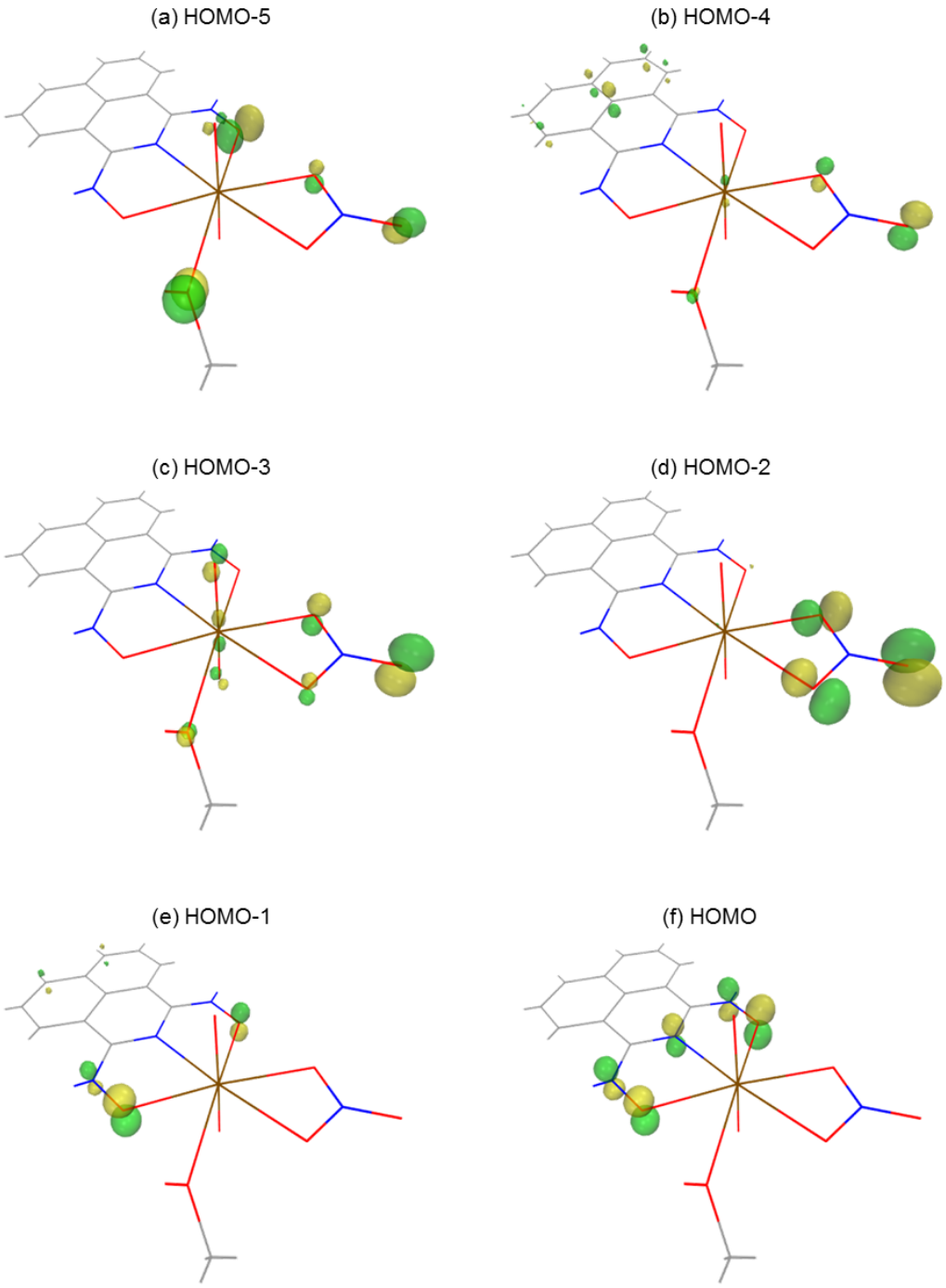


Figure S5. Selected molecular orbital plots from HOMO-5 (a) to HOMO (f).

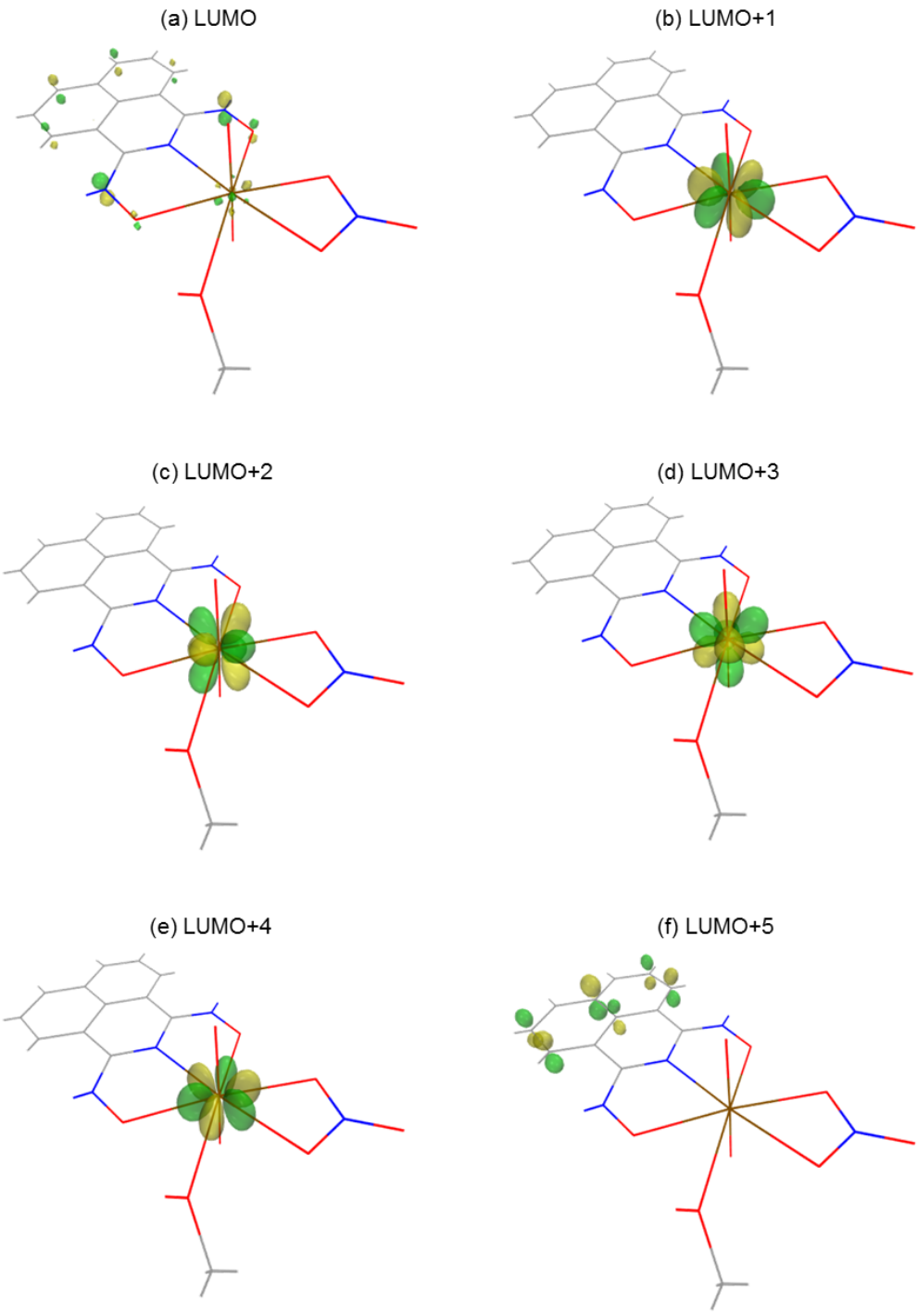


Figure S6. Selected molecular orbital plots from LUMO (a) to LUMO+5 (f).

Vector	Occupancy	E (kcal/mol)	Orbital
113	0	-41.77785877	LUMO+5
112	0	-56.05737593	LUMO+4
111	0	-62.86012399	LUMO+3
110	0	-64.62625088	LUMO+2
109	0	-65.68347973	LUMO+1
108	0	-77.99359441	LUMO
107	2	-139.2314168	HOMO
106	2	-151.9577588	HOMO-1
105	2	-172.4153379	HOMO-2
104	2	-187.5987581	HOMO-3
103	2	-187.7764689	HOMO-4
102	2	-190.3751761	HOMO-5

Table S1. Selected molecular orbital data from HOMO-5 to LUMO+5.

checkCIF/PLATON report

You have not supplied any structure factors. As a result the full set of tests cannot be run.

THIS REPORT IS FOR GUIDANCE ONLY. IF USED AS PART OF A REVIEW PROCEDURE FOR PUBLICATION, IT SHOULD NOT REPLACE THE EXPERTISE OF AN EXPERIENCED CRYSTALLOGRAPHIC REFEREE.

No syntax errors found. CIF dictionary Interpreting this report

Datablock: U

```
Bond precision:  C-C = 0.0127 A          Wavelength=0.71073

Cell:          a=7.1221(15)       b=10.601(2)      c=13.683(3)
               alpha=102.880(2)   beta=99.789(2)   gamma=98.454(2)
Temperature:   100 K
```

	Calculated	Reported
Volume	973.9(3)	973.9(4)
Space group	P -1	P -1
Hall group	-P 1	?
Moiety formula	C13 H12 N4 O8 U, 2(C H4 O)	?
Sum formula	C15 H20 N4 O10 U	C15 H20 N4 O10 U
Mr	654.38	654.38
Dx,g cm-3	2.231	2.232
Z	2	2
Mu (mm-1)	8.397	8.397
F000	620.0	620.0
F000'	601.18	
h,k,lmax	9,14,18	9,14,18
Nref	5034	4526
Tmin,Tmax	0.664,0.777	0.679,0.787
Tmin'	0.651	

```
Correction method= MULTI-SCAN

Data completeness= 0.899          Theta(max)= 28.680

R(reflections)= 0.0465( 4098)     wR2(reflections)= 0.1325( 4526)

S = 1.073                 Npar= 289
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The following ALERTS were generated. Each ALERT has the format
test-name_ALERT_alert-type_alert-level.
Click on the hyperlinks for more details of the test.

Alert level B

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PLAT417_ALERT_2_B Short Inter D-H..H-D       H2N    ..  H9O    ..       1.83 Ang.
```

Alert level C

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PLAT342_ALERT_3_C Low Bond Precision on  C-C Bonds ..............     0.0127 Ang.
PLAT417_ALERT_2_C Short Inter D-H..H-D       H8O    ..  H8O    ..       2.12 Ang.
```

Alert level G

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PLAT002_ALERT_2_G Number of Distance or Angle Restraints on AtSite         10
PLAT005_ALERT_5_G No _iucr_refine_instructions_details  in the CIF          ? Do !
PLAT083_ALERT_2_G SHELXL Second Parameter in WGHT Unusually Large.      12.48
PLAT154_ALERT_1_G The su's on the Cell Angles are Equal ..........    0.00200 Deg.
PLAT710_ALERT_4_G Delete 1-2-3 or 2-3-4 Linear Torsion Angle ... #         54
            N2  -O3  -U1  -O8   -166.40  1.10   1.555   1.555   1.555   1.555
PLAT710_ALERT_4_G Delete 1-2-3 or 2-3-4 Linear Torsion Angle ... #         63
            N1  -O4  -U1  -O6   -175.00  4.00   1.555   1.555   1.555   1.555
PLAT710_ALERT_4_G Delete 1-2-3 or 2-3-4 Linear Torsion Angle ... #         67
            C13 -O8  -U1  -O3     57.10  1.50   1.555   1.555   1.555   1.555
PLAT710_ALERT_4_G Delete 1-2-3 or 2-3-4 Linear Torsion Angle ... #         89
            N3  -O6  -U1  -O4      9.00  4.00   1.555   1.555   1.555   1.555
PLAT860_ALERT_3_G Note: Number of Least-Squares Restraints .......          6
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0 **ALERT level A** = Most likely a serious problem - resolve or explain
1 **ALERT level B** = A potentially serious problem, consider carefully
2 **ALERT level C** = Check. Ensure it is not caused by an omission or oversight
9 **ALERT level G** = General information/check it is not something unexpected

1 ALERT type 1 CIF construction/syntax error, inconsistent or missing data
4 ALERT type 2 Indicator that the structure model may be wrong or deficient
2 ALERT type 3 Indicator that the structure quality may be low
4 ALERT type 4 Improvement, methodology, query or suggestion
1 ALERT type 5 Informative message, check

It is advisable to attempt to resolve as many as possible of the alerts in all categories. Often the minor alerts point to easily fixed oversights, errors and omissions in your CIF or refinement strategy, so attention to these fine details can be worthwhile. In order to resolve some of the more serious problems it may be necessary to carry out additional measurements or structure refinements. However, the purpose of your study may justify the reported deviations and the more serious of these should normally be commented upon in the discussion or experimental section of a paper or in the "special_details" fields of the CIF. checkCIF was carefully designed to identify outliers and unusual parameters, but every test has its limitations and alerts that are not important in a particular case may appear. Conversely, the absence of alerts does not guarantee there are no aspects of the results needing attention. It is up to the individual to critically assess their own results and, if necessary, seek expert advice.

Publication of your CIF in IUCr journals

A basic structural check has been run on your CIF. These basic checks will be run on all CIFs submitted for publication in IUCr journals (*Acta Crystallographica*, *Journal of Applied Crystallography*, *Journal of Synchrotron Radiation*); however, if you intend to submit to *Acta Crystallographica Section C* or *E*, you should make sure that full publication checks are run on the final version of your CIF prior to submission.

Publication of your CIF in other journals

Please refer to the *Notes for Authors* of the relevant journal for any special instructions relating to CIF submission.

PLATON version of 01/06/2013; check.def file version of 24/05/2013

Datablock U - ellipsoid plot