The Crystal Structure at 120 K of a Salt Formed from Lead(II) Nitrate and 12-Crown-4

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Nicholson, D. G., Sylte, I., Vasudevan, A. K. and Sæthre, L. J., 1992. The Crystal Structure at 120 K of a Salt Formed from Lead(II) Nitrate and 12-Crown-4. – Acta Chem. Scand. 46: 358–362.

The crystal structure of the title compound has been determined from X-ray diffractometer data by Patterson and Fourier methods. Crystals are monoclinic, space group $P2_1/c$, with $Z = 4$ in a unit cell of dimensions $a = 8.165(3)$, $b = 28.208(5)$, $c = 16.123(3)$ Å and $\beta = 93.79(1)°$. The structure was refined to $R = 0.037$ for 7308 observed reflections. There are two lead(II) sites (ratio 1:1); one sandwiched between two 12-crown-4 molecules in a complex cation in which the lead lone pair is not stereochemically active; the other site is occupied by the trinitro(12-crown-4)lead(II) anion.

There is a rich structural chemistry of complex species in solutions that contain macrocyclic polyethers and various main-group acceptor systems.[1–12] To a large extent this can be attributed to the coupling of the fluxional properties of the macrocycles to their multidonor properties, i.e. the oxygen atoms. Such a coupling leads to a number of options for complexation because different conformations can lead to changes in the numbers of the donor sites that can be effectively presented to a potential acceptor. This means that solutions may contain a number of complexed species with varying stabilities. Although some of these species are expected to be only transient it is possible for several of them to coexist on longer timescales, and the most stable of these will aggregate and eventually form crystals under appropriate conditions. The process of aggregation depends on a number of factors, of which effective packing forces are crucial. Another important factor is the so-called macrocyclic or entropy effect, which is an expression for the advantageous displacement of solvent molecules coordinated to the acceptor. Thus, an appropriate conformation of the crown ether towards a potential acceptor will be able to exploit the effect. The effect depends on the size of the crown ether (or, more specifically, on the number of oxygen atoms made available), since the process of liberating a greater number of solvent molecules from the coordination sphere of the metal atoms gives more favourable entropy values.

As a basis for examining some of the species that are present in solutions containing crown ethers (12-crown-4, 15-crown-5 and 18-crown-6) and main-group metal compounds (at least at the point of crystallisation, since caution is needed when extrapolating structural information from the crystalline state to solutions), we have previously characterised[1–10] by means of X-ray crystallography and synchrotron X-ray spectroscopy a number of complexes formed between acceptor compounds containing the heavier main-group elements thallium(I), tin(II), lead(II), antimony(III) and bismuth(III). In addition, Alcock *et al.* have recently reported the structures of 12-crown-4 and 18-crown-6 complexes with bismuth(III) chloride and 18-crown-6 with antimony(III) chloride.[11,12] We extend this work by reporting here the crystal and molecular structure of the lead(II) nitrate complex salt, bisdinitratotris-(1,4,7,10-tetraoxacyclododecane)lead(II), $Pb(NO_3)(CR)_2$-$Pb(NO_3)_3(CR)$, where CR denotes 12-crown-4.

Experimental

Preparation. The complex was prepared by adding 12-crown-4 (1.5 g) to an aqueous solution (5 cm^3) of lead(II) nitrate (1.0 g); diffraction-quality crystals were obtained after several weeks of slow evaporation of the solution.

Crystal data. $C_{24}H_{48}O_{24}N_4Pb_2$, $M = 1190.97$, monoclinic, $a = 8.165(3)$, $b = 28.208(5)$, $c = 16.123(3)$ Å, $\beta = 93.79(1)°$, $U = 3704.25$ Å^3, $Z = 4$, $F(000) = 1960$. MoK_α radiation, $\lambda = 0.710\,69$ Å, $\mu = 88.2$ cm^{-1}. Absent reflections were consistent with space group $P2_1/c$.

Intensity data collection and structure refinement. A crystal was mounted in a general orientation and intensity data collected at 120 K in the ω–2θ scan mode on a Nonius CAD4 automatic four-circle diffractometer out to $\theta = 30°$ using monochromatic MoK_α radiation. The crystal was stable during data collection. The cell dimensions were determined from the refined setting angles of 25 reflections located by a search routine, and the space group deduced from the systematic absences. The data were transferred to

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a VAX 11/750 computer, Lorentz and polarisation corrections applied, systematic absences rejected and equivalent reflections merged. Of the 8900 reflections measured 7308

Table 1. Atomic coordinates ($\times 10^5$ for Pb and $\times 10^4$ for the other atoms) with e.s.d.s in parentheses and equivalent isotropic thermal parameters (in 10^4 Å^2).

Atom	x/a	y/b	z/c	U_{eq} [a]
Pb(1)	46282(3)	7218(9)	78377(2)	144
Pb(2)	83253(3)	−20077(1)	52393(2)	161
N(1)	3510(8)	−231(2)	6045(4)	191
N(2)	3568(8)	−962(2)	8249(4)	194
N(3)	1948(8)	729(3)	8428(5)	203
N(4)	18738(9)	2561(3)	−1283(5)	245
O(2)	2532(8)	−41(3)	6521(4)	281
O(3)	4973(7)	−283(2)	6325(4)	249
O(4)	3035(8)	−353(2)	5336(4)	255
O(6)	2578(8)	−634(3)	8142(6)	381
O(7)	5028(8)	−864(3)	8203(7)	379
O(8)	3123(9)	−1364(2)	8398(5)	332
O(10)	2451(8)	380(2)	8852(4)	300
O(11)	2430(8)	773(2)	7709(4)	280
O(12)	952(2)	1010(3)	8710(5)	332
O(22)	18457(8)	2822(2)	−672(4)	270
O(23)	20198(8)	2519(2)	−1487(4)	280
O(24)	17608(9)	2347(3)	−1677(5)	373
O(103)	3479(9)	−789(3)	2812(5)	341
O(106)	4460(7)	−907(2)	1209(5)	297
O(109)	3215(8)	−14(2)	782(4)	277
O(112)	2152(8)	117(3)	2403(6)	311
O(201)	10223(7)	3027(2)	1183(4)	246
O(204)	10539(8)	2160(2)	371(4)	284
O(207)	13581(7)	2243(2)	−268(4)	253
O(210)	13470(7)	2742(2)	1248(3)	234
O(221)	7126(7)	−1757(2)	6724(4)	237
O(224)	5175(7)	−1644(2)	5221(3)	207
O(227)	7707(7)	−1144(2)	4537(4)	238
O(230)	9658(7)	−1248(2)	6032(4)	244
C(101)	1769(13)	−295(5)	3170(7)	310
C(102)	1923(19)	−769(5)	2903(10)	670
C(104)	3914(15)	−1275(3)	2283(9)	398
C(105)	3490(18)	−1250(5)	1441(9)	646
C(107)	3902(18)	−701(6)	320(7)	554
C(108)	2675(20)	−387(5)	319(9)	639
C(110)	1737(12)	318(4)	1131(10)	500
C(111)	1036(17)	91(5)	1787(10)	596
C(202)	9067(11)	2654(3)	1273(6)	275
C(203)	9743(12)	2166(3)	1128(5)	264
C(205)	11475(12)	1732(3)	270(6)	302
C(206)	12674(12)	1812(3)	−385(6)	280
C(208)	14963(10)	2250(3)	343(5)	244
C(209)	14510(10)	2333(3)	1227(5)	238
C(211)	12729(10)	2789(3)	2021(5)	234
C(212)	11302(10)	3135(3)	1897(5)	216
C(222)	5369(10)	−1779(3)	6675(5)	229
C(223)	4609(11)	−1469(3)	5992(5)	231
C(225)	4871(10)	−1325(3)	4540(5)	233
C(226)	6119(10)	−932(3)	4548(5)	234
C(228)	9020(11)	−828(3)	4791(5)	266
C(229)	9254(11)	−786(3)	5715(6)	249
C(231)	9529(11)	−1292(3)	6911(5)	276
C(232)	7777(10)	−1343(3)	7122(5)	248

[a] *U*eq is defined as 1/3 of the trace of the orthogonalised U_{ij} tensor.

Table 2. Bond distances (in Å) and angles (in °) with estimated e.s.d.s in parentheses.

(a) Lead coordination

Pb(1)–O(10)	2.641(6)	Pb(2)–O(207)	2.628(6)
Pb(1)–O(2)	2.657(6)	Pb(2)–O(23)	2.638(6)
Pb(1)–O(11)	2.668(2)	Pb(2)–O(201)	2.651(6)
Pb(1)–O(6)	2.668(7)	Pb(2)–O(230)	2.688(6)
Pb(1)–O(3)	2.668(6)	Pb(2)–O(22)	2.716(6)
Pb(1)–O(7)	2.722(8)	Pb(2)–O(227)	2.719(6)
Pb(1)–O(112)	2.735(7)	Pb(2)–O(204)	2.732(6)
Pb(1)–O(109)	2.750(6)	Pb(2)–O(221)	2.739(6)
Pb(1)–O(103)	2.792(7)	Pb(2)–O(224)	2.768(6)
Pb(1)–O(106)	2.883(6)	Pb(2)–O(210)	2.817(6)

(b) Nitrate anions

O(2)–N(1)	1.262(9)	O(3)–N(1)–O(4)	122.2(7)
O(3)–N(1)	1.258(8)	O(2)–N(1)–O(4)	120.7(7)
O(4)–N(1)	1.233(9)	O(2)–N(1)–O(3)	117.1(7)
O(6)–N(2)	1.23(1)	O(7)–N(2)–O(8)	121.4(8)
O(7)–N(2)	1.230(9)	O(6)–N(2)–O(8)	121.6(7)
O(8)–N(2)	1.220(9)	O(6)–N(2)–O(7)	117.0(7)
O(10)–N(3)	1.25(1)	O(10)–N(3)–O(12)	120.0(8)
O(11)–N(3)	1.26(1)	O(11)–N(3)–O(12)	122.0(8)
O(12)–N(3)	1.242(9)	O(11)–N(3)–O(10)	118.0(7)
O(22)–N(4)	1.26(1)	O(22)–N(4)–O(24)	121.1(8)
O(23)–N(4)	1.26(1)	O(23)–N(4)–O(24)	120.2(8)
O(24)–N(4)	1.24(1)	O(23)–N(4)–O(22)	118.7(7)

(c) 12-Crown-4 molecules

Molecule 1

C(101)–C(102)	1.41(6)	O(112)–C(101)–C(102)	113.0(11)
C(101)–O(112)	1.71(6)	C(101)–C(102)–O(103)	100.6(11)
C(102)–O(103)	1.29(6)	C(104)–O(103)–C(102)	109.8(8)
C(104)–C(105)	1.38(6)	O(103)–C(104)–C(105)	114.4(10)
C(104)–O(103)	1.67(6)	C(104)–C(105)–C(106)	101.6(11)
C(105)–O(106)	1.32(6)	C(107)–O(106)–C(105)	112.5(8)
C(107)–C(108)	1.34(4)	O(106)–C(107)–C(108)	114.5(11)
C(107)–O(106)	1.59(5)	O(109)–C(108)–C(107)	107.4(12)
C(108)–O(109)	1.35(6)	C(110)–O(109)–C(108)	114.2(8)
C(110)–C(111)	1.39(6)	O(109)–C(110)–C(111)	110.2(8)
C(110)–O(109)	1.65(6)	C(110)–C(111)–O(112)	104.6(11)
C(111)–O(112)	1.30(6)	C(101)–O(112)–C(111)	110.8(9)

Molecule 2

C(202)–O(201)	1.43(1)	C(212)–O(201)–C(202)	116.7(6)
C(202)–C(203)	1.51(1)	C(203)–C(202)–O(201)	114.0(7)
C(203)–O(204)	1.42(1)	C(202)–C(203)–O(204)	109.7(7)
C(205)–O(204)	1.44(1)	C(205)–O(204)–C(203)	112.6(7)
C(205)–C(206)	1.50(1)	C(206)–C(205)–O(204)	109.1(7)
C(206)–O(207)	1.43(1)	C(205)–C(206)–O(207)	113.0(7)
C(208)–O(207)	1.448(9)	C(208)–O(207)–C(206)	118.4(7)
C(208)–C(209)	1.51(1)	C(209)–C(208)–O(207)	114.5(7)
C(209)–O(210)	1.43(1)	C(208)–C(209)–O(210)	109.2(6)
C(211)–O(210)	1.43(1)	C(209)–O(210)–C(211)	112.4(6)
C(211)–C(212)	1.52(1)	C(212)–C(211)–O(210)	108.1(6)
C(212)–O(201)	1.436(9)	C(211)–C(212)–O(201)	113.2(7)

Molecule 3

C(222)–O(221)	1.43(1)	C(222)–O(221)–C(232)	113.9(6)
C(222)–C(223)	1.51(1)	C(223)–O(224)–C(225)	113.5(6)
C(223)–O(224)	1.441(9)	C(228)–O(227)–C(226)	113.4(6)
C(225)–O(224)	1.428(9)	C(231)–O(230)–C(229)	113.7(6)
C(225)–C(226)	1.51(1)	C(223)–C(222)–O(221)	112.1(6)
C(226)–O(227)	1.430(9)	C(222)–C(223)–O(224)	107.0(6)
C(228)–O(227)	1.43(1)	C(226)–C(225)–O(224)	111.9(6)
C(228)–C(229)	1.49(1)	C(225)–C(226)–O(227)	107.7(6)
C(229)–O(230)	1.43(1)	C(229)–C(228)–O(227)	112.1(7)
C(231)–O(230)	1.43(1)	C(228)–C(229)–O(230)	107.2(7)
C(231)–C(232)	1.50(1)	C(232)–C(231)–O(230)	111.5(7)
C(232)–O(221)	1.42(1)	C(231)–C(232)–O(221)	108.1(7)

were deemed observed, the criterion being $I_{net} > 3.0\sigma(I)$. Scattering factors and dispersion corrections were taken from Ref. 13. The CRYSTALS Issue 9 suite of programs[14] was used for the calculations, and the structure solved by the heavy-atom method, corrected empirically for absorption[15] and refined by least-squares on F in space group $P2_1/c$ with anisotropic temperature factors for all non-hydrogen atoms. The hydrogen atoms were placed in calculated positions, given a common thermal parameter and allowed to ride on their carbon atoms during refinement.

The final stage of the refinement was to assign to each reflection a weight[16] $w = 1/\Sigma^n{}_r A_r T_r(X)$, where n is the number of coefficients, A_r, for a Chebyshev series, T_r is the polynomial function, and X is $F_o/F_o(\max)$. The values of A_r which gave similar values of $w(F_o-F_c)^2$ over ranges of $(\sin\theta)/\lambda$ and F_0 were 2.5, 2.8 and 0.68, giving final values of $R = 3.73$ and $wR = 5.13\%$. At convergence the r.m.s. shift/e.s.d. < 0.3 and the highest peak in the difference Fourier map was 0.6 e $Å^{-3}$. The atomic positions are given in Table 1, and Table 2 contains the interatomic distances and valence angles.

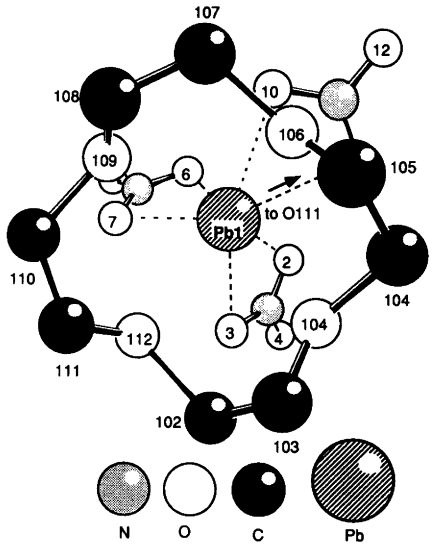

Fig. 2. Structure of the $[Pb(12\text{-crown-4})(NO_3)_3]^-$ anion viewed perspectively through the crown. The dashed lines identify the closest nitrate–oxygen contacts to Pb.

Results and discussion

The crystal structure (Fig. 1) consists of two lead-containing units $PbNO_3CR_2^+$ and $Pb(NO_3)_3CR^-$. We choose to designate these units cation and anion, respectively (although we recognise that the interactions between the nitrate anions and the lead atoms would appear to be similar to those in lead nitrate itself, see below), because the overall view of the structure is consistent with the packing forces between these units (and of course within each unit itself with respect to the nitrates) being the prime driving force behind aggregation and finally crystallisation. The fact that all three crown molecules in the solid-state lead nitrate salt adopt three different conformations and the similar situation observed[11] in the 18-crown-6 complexes with bismuth(III) chloride (see below) lend support to the supposition that the fluxional nature of the 12-crown-4 ligand leads to a range of complexes in solution. Figs. 2 and 3 show the structures of the $Pb(NO_3)_3CR^-$ anion and the $PbNO_3CR_2^+$ cation together with the atom numbering scheme.

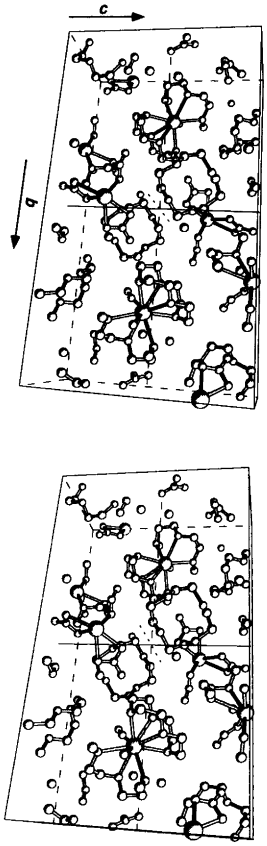

Fig. 1. Stereoscopic drawing of part of $[Pb(12\text{-crown-4})_2(NO_3)][Pb(12\text{-crown-4})(NO_3)_4]$.

Lead coordination. A feature common to both ions in the present structure is that their lead atoms interact with a total of ten oxygen atoms from the crown plus nitrates, cf. Table 2 and Figs. 2 and 3. The lead atom in the $PbNO_3CR_2^+$ cation interacts with all four oxygens $(O)_4$ of each crown. As is the case with all of the crown ether complexes studied in this series, the metal – $(O)_4$ distances are long. The sandwiching of the lead atoms between the two 12-crown-4 molecules is somewhat reminiscent of the tin(II) environment in the $SnCR'_2{}^{2+}$ cation of the 15-crown-5 (= CR') complex with tin(II) chloride (the double cationic charge being compensated by two $SnCl_3^-$ anions) in which the tin atom environment $[Sn(O_5))_2]$ is defined by two pentagonal pyramids with their apices meeting at the tin atom.[6] However, any further similarity stops here; whereas in the tin (II) complex the bases of the pentagonal pyramids are

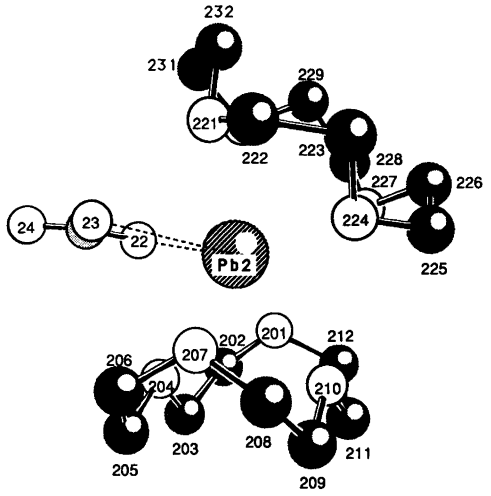

Fig. 3. Perspective drawing of the $[Pb(12\text{-crown-}4)_2(NO_3)]^+$ cation.

staggered and parallel, with the tin atom lying on a *crystallographic* centre of symmetry (special position), the lead atom in the present complex cation is noncentrosymmetric and the $(O)_4$ bases of the square pyramids are not staggered but instead tilted with respect to each other. Another difference is that they do not have identical conformations. Evidently, packing forces play a major role here, with the adjacent nitrate acting in effect as a wedge thereby tilting the crowns away from a parallel (i.e. 'sandwich') arrangement. At the outset we did consider it possible that a $PbCR_2^{2+}$ cation similar to $SnCR'_2{}^{2+}$ might be formed. We still consider this a possibility for the 12-crown-4 and 15-crown-5 complexes with the lead(II) halides.

The anion consists of a single 12-crown-4 ring coordinated to one side of a lead cation, with three nitrate anions being associated on the opposite side.

These results show that the two units found here constitute at least two of the species present in solution at the given concentration. A similar situation pertains to the 1:1 bismuth(III) chloride:18-crown-6 (CR″) system that produces a solid the crystal structure of which also contains two discrete units (molecules), i.e. $BiCl_3 \cdot H_2O \cdot CR''$ and $BiCl_3 \cdot CR''$,[4] and a 2:1 complex that is ionic and contains two $[BiCl_2 \cdot CR'']^+$ cations and a $[Bi_2Cl_8]^{2-}$ anion.[12]

The 12-crown-4 molecules. The C–C and C–O bond distances in the 12-crown-4 ligands are comparable, within the standard deviations, with those reported for other cyclic polyether complexes.[2-4,6-9] Note in particular that, whereas the two crown molecules are well defined in the cation, definition of the crown is considerably degraded in the anion, as reflected by the large standard deviations and thermal vibrational ellipsoids. We will not speculate as to whether this is due to static or dynamic disorder; certainly the fluxional nature of the crown would give it ample scope for taking a range of conformations that still permit all four oxygen atoms to interact with the metal. Instead of modelling the disorder by attempting to fit a range of constrained and closely related conformations with appropriate partial occupancies, we opt instead to report the parameters as found for an unconstrained and hence less prejudiced model. This decision is also connected to the fact that the very high atomic number of the two crystallographically independent heavy-metal atoms obviously dominates the structure to an extent that this exercise would be unfruitful. This tendency to disorder on the part of the crown ether is not unusual: similar effects have also been observed in other crown ether complexes.[6,9] Note also that the disorder here applies only to *one* of the three crown molecules, the two other crown ethers and all the nitrate anions being well defined.

The conformations of the crown ethers in both the cation and anion are such that all four ring oxygens are directed towards the lead atoms. In this they emulate the 12-crown-4 complex with antimony(III) chloride[1] and the 15-crown-5 complexes with tin(II) chloride,[6] antimony(III) chloride[7,12] and bismuth(III) chloride.[4,11]

The four nitrate anions. All four nitrates are planar [the deviations of the nitrogen atoms from least-squares planes through each nitrate, in logical order, are 0.004(8), 0.001 (8) and 0.017(8) Å] with the bond angles being 120° within the experimental accuracy. The three N–O bond lengths are equal also within experimental error, as is the case in lead nitrate itself.[17] That the nitrate anions retain their structural integrity to such a degree, despite each of them interacting with a lead atom via two of their oxygen atoms, shows that interactions with the lead atoms are relatively weak. Nonetheless, they do have profound effects, the most apparent of which is the structural rôle played by one of the nitrates in the cationic unit (see above).

Some considerations on the properties of the lone pair on the lead atoms. In order to analyse the stereochemical implications of the lead lone pairs in the $PbNO_3CR_2^+$ and $Pb(NO_3)_3CR^-$ aggregates it is necessary to place their behaviour in context with the other complexes in the series; the background for our description for this series of complexes has been expounded upon in the recent literature.[6] With this in mind, it is clear that the influences of the crown ethers on the lone pairs on the lead atoms are central when discussing the stereochemistries of the two aggregates. Whereas it is a trivial matter to establish that the tin and lead lone pairs in the centrosymmetric $SnCR'_2{}^{2+}$ (see above) and $PbCR''^{2+}$ cations[6,9] are necessarily precluded from being sterochemically active, the lower-symmetry point group of the lead cation in the present complex cannot give similar information. However, the crowded environment and strong similarity with the cation lead to the conclusion that the lone pair is not sterically active in the VSEPR sense. The formal Pb^{2+} cation in $Pb(NO_3)_3CR^-$ and in $PbNO_3CR_2^+$ is bonded to the two 12-crown-4 ligands (in essentially the same manner as in the $SbCl_3 \cdot CR$ and $SnCR_2^{2+}$ adducts,[6,7] respectively), but further discussion is

curtailed by referring to the detailed discussion in Ref. 6 (and references therein). Suffice it to state that the lead lone pair in both aggregates can be identified with contributions from the 6s and 6p orbitals and is explicitly represented by the sum of antibonding molecular orbitals with their varying lead or ligand character. The geometry adopted reflects the desire of the system to attain a lower total energy through maximum population of the lead lower 6s valence orbital. This end is achieved through distortions which are appropriate in the context of all the other energies that are germane to the system as a whole.

Acknowledgements. We are grateful to Dr. Reidar Lie, Head of Research, Borregaard Industries Ltd., Sarpsborg, Norway for donating the 18-crown-6. We also thank Dr. David Watkin, The Chemical Crystallography Laboratory, University of Oxford, UK for kindly supplying us with the CRYSTALS package and the Nansen Foundation, Oslo for an equipment grant.

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Received October 3, 1991.