Crystal Structure of Bis(2-imino-4-amino-4-methyl-pentane)nickel(II) Nitrate

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The aliphatic *Schiff* base complex bis(2-imino-4-amino-4-methyl-pentane)nickel(II) nitrate $(C_6H_{14}N_2)_2Ni(NO_3)_2$ crystallizes in the monoclinic system with the unit cell dimensions $a = 9.05$, $b = 7.36$, $c = 15.40$ Å, $\beta = 105.4°$. The space group symmetry is $P2_1/c$. The crystal structure of $Ni(C_6H_{14}N_2)_2(NO_3)_2$ was determined from three-dimensional X-ray data. The square planar coordination of four nitrogen atoms around the nickel(II) atom corresponds to a low-spin (diamagnetic) d^8 electron configuration of the metal ion. Four nitrogen atoms are at distances of 1.898 (2×) and 1.953 Å (2×). The influence of the electronic structure of the central atom on the molecular structure of the nickel(II) and copper(II) *Schiff* base complexes is discussed.

Complexes of the formula $(C_6H_{14}N_2)_2MX_2$ (X = Cl, Br, I, NO_3, ClO_4, BF_4; M = Cu, Ni) are formed by reaction of polyammine metal salts $M(NH_3)_nX_2$ in acetone or they crystallize from a solution of MX_2 in acetone under the influence of gaseous NH_3 [1—3]. These compounds containe azomethine group —HC=N— characteristic for *Schiff* bases. The X-ray crystal structure analysis of $(C_6H_{14}N_2)_2Ni(NO_3)_2$ was started in connexion with the known structure of the copper(II) complex [3] in order to show the influence of the electronic structure of the central atom on the molecular structure of the metal complex. From the diamagnetism of the nickel complex, a square planar arrangement around central atom could be expected [1, 2].

Experimental

Crystal data

Bis(2-imino-4-amino-4-methyl-pentane)nickel(II) nitrate crystallizes in the monoclinic system with the lattice constants $a = 9.05(6)$, $b = 7.36(4)$, $c = 15.40(8)$ Å, $\beta = 105.4(0.8)°$. From the observed density of 1.36 g cm^{-3}, the number of formula units $Ni(C_6H_{14}N_2)_2(NO_3)_2$ in the above cell was found to be two. The calculated density is 1.38 g cm^{-3}. The space group symmetry $P2_1/c$ was determined from the observed systematic absences in the X-ray pattern.

The three-dimensional X-ray data were obtained from the integrated equi-inclination *Weissenberg* photographs around the b axis using a crystal with the cross section of 0.2 × × 0.3 mm elongated in the direction of the rotation axis. The total of 1315 nonequivalent reflections were measured photometrically. The observed intensity data, after being corrected for absorption (with a cylindrical approximation of the crystal shape) and for *Lorenz* and polarization factors, were brought to an approximately common scale by use of a modified statistical relationship between the intensities and the atomic scattering factors first suggested by *Wilson* [4]. The average values of normalized structure factors

$\langle E_h^2 \rangle$ within the ranges of reciprocal lattice, which are large enough for application of probability, equal

$$\langle E_H^2 \rangle = 1. \tag{1}$$

It means that

$$\langle E_H^2 \rangle = \frac{\langle |F_H|^2 \rangle}{\varepsilon \sigma_2} = \frac{\langle (I_H)_{\mathrm{obs}} \rangle}{\varepsilon \sigma_2} k \exp(BS^2) = 1. \tag{2}$$

H means h, k, l, F_H is a symbol for structure factor, I_{obs} is the observed intensity, ε is an internal factor which corrects the class of reflections (that is, hkl, or $hk0$, or $h00$) for the symmetry elements of the unit cell, k is a scaling constant. The factor $\exp(BS^2)$ describes the effect of isotropic temperature vibrations, where B is the Debye temperature factor and $S = \sin \Theta/\lambda \cdot \sigma_2$ is defined

$$\sigma_2 = \sum_{j=1}^{N} f_j(S)^2 \tag{3}$$

where f_j is the scattering factor for S and N number of atoms in the unit cell.

$\langle (I_H)_{\mathrm{obs}} \rangle / \varepsilon \sigma_2$ may be plotted directly as a function of $\langle S^2 \rangle$ for every set of layer line reflections and the resulting curve will have as intercept $1/k$ when $S = 0$. The shape of the curve will give the statistical temperature corrections which need not be Gaussian. The presence of two formula units in the unit cell implies occupation of the special positions (0, 0, 0; 0, 1/2, 1/2) by the nickel atoms. The heavy atoms in special positions contribute only to those structure factors for which $k + 1 = 2n$. The probability that the sign of these reflections is positive is higher than 90% [5, 6]. The *Fourier* synthesis of the electron density distribution calculated with only positive coeficients F_H for which $k + 1 = 2n$, shows $2(N-2)$ peaks for $(N-2)$ atoms and we have to find $(N-2)$ positions of the basic structure. For the solution of this problem we have used

a) the known shape of the aliphatic *Schiff* base and the NO_3 group [3];

b) a modified *Patterson* synthesis $P(uvw)$, computed from $|F_H|^2$ without heavy atom contribution $(k + 1 = 2n + 1)$. This gave us information on interatomic distances between the atoms in general positions;

c) a modified symbolic addition procedure [6, 7]. For the normalized structure factors E_H without heavy atom contribution, we can use the probability relationship [7]

$$s(E_H) \sim s \sum_K E_K \left(E^2_{\frac{H+K}{2}} - 1\right) \tag{4}$$

where s means „sign of", H and K represent $h_1k_1l_1$ and $h_2k_2l_2$, respectively. E_H, E_K and $E^2_{\frac{H+K}{2}}$ are normalized structure factors without heavy atom contribution. If $E^2_{\frac{H+K}{2}}$ is normalized structure factor with heavy atom contribution, the expression (*4*) has to be replaced by

$$s(E_H) \sim s \sum_K E_K \left(E^2_{\frac{H+K}{2}} - \langle E^2_{\frac{H+K}{2}} \rangle\right) \tag{5}$$

where $\langle E^2_{\frac{H+K}{2}} \rangle$ is the average for all E-values with heavy atom contribution. It differs significantly from unity. Renormalization in (*5*) has a similar effect on the formula (*4*) as subtraction of the heavy atom contribution from E-values in the formula in which $E^2 - 1$

is preserved, would have. Both (E^2-1) and $(E^2-\langle E^2\rangle)$ may be positive or negative. The starting set for the phase determination procedure includes the signs of origin specifying linearly independent reflections.

The structure was then refined by the block-diagonal least-square procedure. A weighing scheme discussed by *Hughes* [8] and individual isotropic temperature parameters have been used. After 10 cycles of this refinment the reliability factor $R = \{\sum||F_o| - |F_c||\}/\sum|F_o|$, had decreased to 0.137 for 1315 non-zero reflections and 0.153 for all 1559 reflections. The refinement was interrupted in the moment when all parameter shifts decreased below submultiple of the standard deviations. The change of the error-square-sum: $\sum w||F_o| - |F_c||^2$ was less than 0.05%. The residual $R' = \sum w||F_o| - |F_c||^2/\sum w|F_o|^2$ dropped to a value 0.038. The weight-analysis showed a satisfactorily uniform distribution of the mean values of $w||F_o| - |F_c||^2$ according to intervals of F_o values and sin Θ. Table 1 shows the final atomic coordinates and isotropic individual *Debye* factors B_j, together with their estimated standard deviations. Table 2 lists the observed and calculated structure factors. Table 3 summarizes the most important bond lengths and bond angles. A general view of the complex is shown in Fig. 1.

Table 1

Final atomic coordinates and the individual isotropic *Debye* factors togehter with their standard deviations (in parentheses)

Atom	x/a	y/b	z/c	B_j
Ni	0	0	0	2.14(3)
O(1)	0.2135(10)	0.6592(14)	0.2067(6)	4.19(17)
O(2)	0.0173(9)	0.7339(14)	0.2556(5)	3.79(16)
O(3)	0.2049(10)	0.5950(15)	0.3440(6)	4.33(18)
N(1)	0.1325(8)	0.0365(12)	0.1214(5)	1.55(12)
N(2)	0.1451(9)	0.6601(13)	0.2702(5)	2.23(14)
N(3)	0.0822(8)	0.2105(13)	—0.0411(5)	1.93(13)
C(1)	0.1905(11)	0.3215(17)	—0.0022(6)	2.42(17)
C(2)	0.3802(14)	—0.0263(19)	0.0861(8)	3.46(22)
C(3)	0.2929(10)	0.1040(16)	0.1338(6)	1.93(15)
C(4)	0.3705(12)	0.1161(19)	0.2350(7)	3.08(19)
C(5)	0.2292(20)	0.4857(24)	—0.0503(11)	5.48(34)
C(6)	0.2801(13)	0.2975(19)	0.0954(8)	3.31(21)

Discussion

The crystal structure analysis of $Ni(C_6H_{14}N_2)_2(NO_3)_2$ showed an atomic arrangement which is in full agreement with the physical properties of the complex compound. The nickel atom is surrounded by four nitrogen atoms in a strictly planar coordination. The metal atom occupies the symmetry centre. Two nitrogen atoms from the imino groups are at distances 1.898(9) Å, another two nitrogen atoms from the amino groups are 1.953(6) Å apart from the central atom. Two oxygen atoms from the NO_3 groups above and below the planar group NiN_4 are at significantly longer distance Ni—O(3): 3.47(1) Å than the four nitrogen atoms in the plane. The planar coordination around the nickel(II) atom corresponds to a low-spin (diamagnetic) d^8 electron configuration of the metal ion. Eight electrons are accomodated in the relatively stable antibonding molecular orbitals derived from metal d_{xy}, d_{z^2}, d_{yz} and d_{xz} orbitals

Table 2

Observed and calculated structure factors

	F_o	F_c
0 0 L		
2	16.2	18.6
4	64.5	73.4
6	12.9	5.6
8	88.3	68.6
10	14.5	14.6
12	23.5	19.3
14	16.6	17.1
16	17.0	14.3
18	9.8	10.5
1 0 L		
0	124.4	126.3
2	14.3	-2.3
4	77.0	86.8
6	78.9	64.3
8	76.7	84.1
10	3.8	5.1
12	4.2	4.3
14	20.5	16.3
16	11.6	8.1
18	8.1	9.3
$\overline{2}$	76.2	75.2
$\overline{4}$	72.4	79.4
$\overline{6}$	9.9	9.5
$\overline{8}$	4.3	8.6
$\overline{10}$	21.5	21.4
$\overline{12}$	26.3	26.2
$\overline{14}$	6.7	3.3
$\overline{16}$	26.1	25.6
$\overline{18}$	16.8	16.7
2 0 L		
0	42.2	-32.3
2	67.1	69.8
4	85.9	84.8
6	34.1	36.9
8	26.6	30.7
10	20.2	15.1
12	10.6	9.6
14	22.1	17.5
16	8.6	7.0
$\overline{2}$	12.1	-1.9
$\overline{4}$	53.8	65.4
$\overline{6}$	26.8	-20.7
$\overline{8}$	40.3	39.2
$\overline{10}$	8.5	8.8
$\overline{12}$	38.7	44.3
$\overline{14}$	19.1	-15.5
$\overline{16}$	21.9	21.9
$\overline{18}$	10.9	9.8
3 0 L		
0	24.1	22.3
2	26.2	31.8
4	42.8	33.4
6	16.8	19.6
8	24.2	23.4
10	23.4	20.5
12	29.4	24.6
14	3.8	4.7
16	9.5	8.5
$\overline{2}$	36.3	36.7
$\overline{4}$	55.8	47.7
$\overline{6}$	33.4	-26.7
$\overline{8}$	45.9	45.5
$\overline{10}$	3.6	4.9
$\overline{12}$	31.8	30.2
$\overline{14}$	16.4	12.7
$\overline{16}$	12.9	12.7
$\overline{18}$	6.4	5.6
4 0 L		
0	27.2	36.0
2	16.6	14.7
4	39.9	38.8
6	54.6	54.9
8	14.0	13.2
10	16.9	17.4
12	24.3	22.9
14	10.5	7.9
$\overline{2}$	41.9	51.9
$\overline{4}$	37.9	33.9
$\overline{6}$	17.9	20.4
$\overline{8}$	33.5	28.4
$\overline{10}$	18.4	20.0
$\overline{12}$	18.5	18.4
$\overline{14}$	29.9	28.5
$\overline{16}$	17.2	16.3
$\overline{18}$	10.8	10.4
5 0 L		
0	54.9	56.4
2	22.9	19.9
4	59.4	62.3
6	22.5	21.2
8	26.4	25.1
10	21.3	19.7
12	28.6	26.3
14	9.1	9.5
$\overline{2}$	11.7	-4.3
$\overline{4}$	44.7	45.9
$\overline{6}$	30.3	36.8
$\overline{8}$	17.8	15.5
$\overline{10}$	28.4	23.1
$\overline{12}$	32.7	40.7
$\overline{14}$	27.4	29.9
$\overline{16}$	17.0	17.7
$\overline{18}$	9.5	10.9
6 0 L		
0	36.9	31.8
2	25.0	23.8
4	31.9	35.3
6	7.6	-3.1
8	39.8	41.4
10	21.9	17.1
12	19.2	18.1
$\overline{2}$	17.6	-14.3
$\overline{4}$	31.9	29.7
$\overline{6}$	51.2	54.1
$\overline{8}$	22.8	23.2
$\overline{10}$	20.2	19.1
$\overline{12}$	26.8	26.7
$\overline{14}$	27.2	28.5
$\overline{16}$	17.6	17.9
$\overline{18}$	8.5	10.5
7 0 L		
0	23.5	20.7
2	18.8	16.9
4	7.8	4.8
6	5.9	3.3
8	19.4	19.6
10	15.9	11.9
$\overline{2}$	3.9	5.9
$\overline{4}$	13.9	13.5
$\overline{6}$	16.2	13.4
$\overline{8}$	50.8	52.1
$\overline{12}$	17.8	15.4
$\overline{10}$	15.9	19.4
$\overline{14}$	3.9	3.8
$\overline{16}$	17.0	17.7
8 0 L		
0	26.8	23.8
2	9.6	-5.7
4	9.9	3.7
6	6.8	5.9
8	10.2	6.2
$\overline{2}$	21.8	20.8
4	9.9	3.7
$\overline{4}$	32.7	32.6
$\overline{6}$	16.8	-13.8
$\overline{8}$	36.8	36.6
$\overline{10}$	12.9	12.1
$\overline{12}$	20.3	18.0
$\overline{14}$	3.5	0.8
$\overline{16}$	8.7	11.5
9 0 L		
0	12.7	11.8
2	9.4	5.2
4	10.9	8.2
6	14.6	13.7
$\overline{2}$	15.4	16.8
$\overline{4}$	32.1	25.2
$\overline{6}$	16.5	17.8
$\overline{8}$	20.8	17.9
$\overline{10}$	10.3	9.8
$\overline{12}$	11.0	11.5
10 0 L		
0	18.4	17.8
2	14.7	14.9
4	8.3	6.9
$\overline{2}$	15.9	15.6
$\overline{4}$	10.3	10.4
$\overline{6}$	16.2	19.4
$\overline{8}$	15.6	18.5
$\overline{10}$	14.4	14.5
$\overline{12}$	13.5	15.9
11 0 L		
2	16.5	20.9
4	13.9	16.1
6	9.0	10.7
0 1 L		
1	83.6	84.5
2	35.3	-31.7
3	19.9	-11.6
4	20.7	20.3
5	11.3	12.9
6	36.7	34.6
7	52.2	53.3
8	18.2	16.3
9	39.1	39.1
10	9.9	-8.0
11	12.6	9.3
12	16.2	12.9
13	25.1	24.5
14	14.9	-12.9
15	11.0	10.0
16	3.5	4.5
17	16.6	14.3
18	6.9	-7.7
1 1 L		
0	18.6	-16.0
1	37.3	49.7
2	55.3	-57.2
3	17.3	-5.3
4	31.5	27.8
5	23.8	29.7
6	21.7	11.8
7	59.1	59.6
8	6.9	-3.2
9	18.2	20.9
10	6.0	-2.8
11	13.0	12.1
12	3.9	2.8
13	20.6	16.1
14	3.9	-2.0
15	23.4	18.5
16	8.7	-4.0
17	7.7	7.1
$\overline{1}$	53.4	51.6
$\overline{2}$	81.1	-73.4
$\overline{3}$	90.5	82.6
$\overline{4}$	12.2	-8.3
$\overline{5}$	56.7	51.9
$\overline{6}$	14.3	-10.5
$\overline{7}$	26.9	30.9
$\overline{8}$	29.1	-28.5
$\overline{9}$	35.3	40.3
$\overline{10}$	9.7	-3.7
$\overline{11}$	22.3	21.7
$\overline{12}$	8.5	-7.2
$\overline{13}$	8.8	9.1
$\overline{14}$	11.5	8.9
$\overline{15}$	7.1	7.3
$\overline{16}$	4.2	-5.0
$\overline{17}$	24.1	21.4
$\overline{18}$	4.5	5.2
$\overline{19}$	11.9	11.1
2 1 L		
0	27.9	26.1
1	102.3	93.7
2	32.7	-38.5
3	42.9	52.3
4	24.4	23.7
5	59.1	63.2
6	39.5	-34.2
7	13.8	11.7
8	19.7	17.9
9	26.6	27.2
10	6.5	-3.9
11	17.5	15.3
12	6.9	3.6
13	19.1	15.4
14	7.2	3.9
15	21.9	18.9
16	3.0	-3.9
17	10.4	7.7
$\overline{1}$	21.7	21.7
$\overline{2}$	89.5	-100.6
$\overline{3}$	92.3	101.8
$\overline{4}$	17.9	-15.9
$\overline{5}$	19.4	33.5
$\overline{6}$	2.4	0.3
$\overline{7}$	10.8	10.6
$\overline{8}$	7.0	-4.8
$\overline{9}$	31.2	35.5
$\overline{10}$	5.5	-1.8
$\overline{11}$	38.1	42.9
$\overline{12}$	7.9	5.6
$\overline{13}$	12.4	11.8
$\overline{14}$	3.9	1.3
$\overline{15}$	8.5	6.5
$\overline{16}$	3.7	-0.6
$\overline{17}$	14.2	14.4
$\overline{18}$	2.9	2.0
$\overline{19}$	18.2	16.9
3 1 L		
0	13.6	-9.1
1	17.9	23.5
2	3.9	-2.7
3	52.9	53.6
4	11.9	-7.4
5	42.7	50.6
6	14.3	-13.3
7	32.0	29.7
8	17.8	18.1
9	28.7	30.2
10	11.2	4.4
11	22.1	20.0
12	6.5	7.5
13	13.0	13.5
14	5.7	4.3
15	13.3	11.9
$\overline{1}$	75.0	72.4
$\overline{2}$	9.4	8.3
$\overline{3}$	51.5	59.9
$\overline{4}$	16.1	-8.9
$\overline{5}$	30.5	32.6
$\overline{6}$	8.2	-7.9
$\overline{7}$	6.8	6.9
$\overline{8}$	15.5	8.2
$\overline{9}$	21.7	21.4
$\overline{10}$	14.0	12.2
$\overline{11}$	29.3	30.2
$\overline{12}$	20.3	17.3
$\overline{13}$	27.1	28.3
$\overline{14}$	3.9	1.7
$\overline{15}$	23.7	23.5
$\overline{16}$	8.4	6.6
$\overline{17}$	8.9	8.9
$\overline{18}$	2.9	-0.7
$\overline{19}$	10.7	9.6
4 1 L		
0	27.9	-32.6
1	16.8	-8.5
2	2.8	1.7
3	40.2	45.9
4	3.1	-3.8
5	44.8	47.0
6	15.9	10.8
7	33.5	37.3
8	4.9	-0.6
9	5.1	5.9
10	16.5	10.6
11	23.4	21.2
12	3.8	5.5
13	18.5	16.2
14	6.9	2.9
15	12.1	9.3
$\overline{1}$	24.7	30.1
$\overline{2}$	20.7	17.0
$\overline{3}$	24.0	27.9
$\overline{4}$	28.1	34.9
$\overline{5}$	22.2	23.8
$\overline{6}$	12.1	-8.7
$\overline{7}$	43.9	53.7
$\overline{8}$	25.8	25.9
$\overline{9}$	17.2	16.7
$\overline{10}$	3.5	-1.9
$\overline{11}$	26.3	26.0
$\overline{12}$	23.0	19.2
$\overline{13}$	16.7	15.7
$\overline{14}$	3.9	1.9
$\overline{15}$	22.1	23.5
$\overline{16}$	10.9	10.6
$\overline{17}$	8.1	8.1
$\overline{18}$	2.9	-2.1
$\overline{19}$	10.7	11.4
5 1 L		
0	3.9	-1.7
1	27.7	33.3
2	4.2	-2.9
3	28.3	31.3
4	3.5	-0.5
5	18.7	16.1
6	6.9	-3.7
7	7.1	9.2

Table 2 (Continued)

	F_o	F_c
5 1 L		
8	7.3	-5.7
9	12.1	11.5
10	3.9	1.3
11	22.5	22.4
12	9.7	5.3
13	24.6	17.0
$\bar{1}$	10.3	13.5
$\bar{2}$	20.9	-17.4
$\bar{3}$	19.7	19.5
$\bar{4}$	19.9	9.8
$\bar{5}$	26.9	31.8
$\bar{6}$	14.5	12.2
$\bar{7}$	29.8	34.8
$\bar{8}$	25.2	23.2
$\bar{9}$	20.3	17.4
$\overline{10}$	3.7	0.8
$\overline{11}$	23.6	25.7
$\overline{12}$	3.9	-2.3
$\overline{13}$	23.7	26.1
$\overline{14}$	3.9	4.8
$\overline{15}$	6.5	6.3
$\overline{16}$	3.5	-2.2
$\overline{17}$	5.5	5.5
$\overline{18}$	4.6	-1.9
$\overline{19}$	8.9	8.3
6 1 L		
0	20.7	14.5
1	29.9	34.9
2	12.5	-11.7
3	24.7	22.3
4	18.2	-15.3
5	7.9	0.1
6	10.5	-11.1
7	17.5	15.9
8	-3.9	-2.9
9	19.4	22.1
10	7.0	-5.6
11	13.1	13.1
$\bar{1}$	22.7	20.7
$\bar{2}$	4.4	-1.4
$\bar{3}$	16.8	15.5
$\bar{4}$	15.8	-18.1
$\bar{5}$	40.2	53.7
$\bar{6}$	14.4	12.3
$\bar{7}$	20.1	18.5
$\bar{8}$	3.7	-0.2
$\bar{9}$	18.0	14.7
$\overline{10}$	9.9	10.0
$\overline{11}$	3.9	6.0
$\overline{12}$	11.8	-10.6
$\overline{13}$	22.9	25.9
$\overline{14}$	-3.8	-1.1
$\overline{15}$	15.6	15.3
$\overline{16}$	-5.7	-7.8
$\overline{17}$	8.9	8.9
$\overline{18}$	6.5	-6.1
7 1 L		
0	8.6	7.3
1	29.2	32.9
2	19.7	-19.1
3	20.5	17.6
4	-3.9	-0.7
5	16.7	17.5
6	8.7	-11.9
7	17.1	13.9
8	4.6	2.8
9	20.6	17.3
10	11.7	-11.4
$\bar{1}$	20.3	19.6
$\bar{2}$	5.3	5.0
$\bar{3}$	16.9	14.5
$\bar{4}$	8.5	6.6
$\bar{5}$	21.1	20.1
$\bar{6}$	3.8	-0.1
$\bar{7}$	25.5	27.8
$\bar{8}$	9.5	-8.0
$\bar{9}$	27.9	30.0
$\overline{10}$	3.9	2.1
$\overline{11}$	10.3	10.4
$\overline{12}$	3.9	-2.0
$\overline{13}$	6.9	6.9
$\overline{14}$	8.1	-8.4
$\overline{15}$	17.9	20.0
$\overline{16}$	3.0	-4.5
$\overline{17}$	12.3	14.9
8 1 L		
0	3.9	1.8
1	17.8	17.3
2	3.9	-1.4
3	9.5	9.9
4	12.0	11.3
5	17.2	18.3
6	9.9	-8.9
7	8.2	8.6
8	2.8	2.8
9	15.1	14.3
$\bar{1}$	25.7	27.2
$\bar{2}$	3.9	0.9
$\bar{3}$	24.1	22.3
$\bar{4}$	20.5	16.5
$\bar{5}$	8.3	6.1
$\bar{6}$	5.1	2.8
$\bar{7}$	11.4	15.2
$\bar{8}$	-3.9	-1.4
$\bar{9}$	25.0	30.7
$\overline{10}$	3.9	-2.5
$\overline{11}$	19.2	19.0
$\overline{12}$	4.7	-0.5
$\overline{13}$	7.8	10.3
$\overline{14}$	5.3	-3.1
$\overline{15}$	8.3	10.4
9 1 L		
0	9.8	10.3
1	8.2	7.1
2	6.8	7.2
3	8.8	8.2
4	6.1	5.9
5	12.0	10.4
6	2.6	-0.0
7	13.8	12.7
$\bar{1}$	18.4	16.5
$\bar{2}$	3.8	0.2
$\bar{3}$	20.6	20.3
$\bar{4}$	9.4	10.4
$\bar{5}$	16.3	13.3
$\bar{6}$	6.3	-2.9
$\bar{7}$	18.5	19.2
$\bar{8}$	9.9	10.8
$\bar{9}$	10.4	13.1
$\overline{10}$	7.1	-5.9
$\overline{11}$	14.9	16.3
$\overline{12}$	3.2	0.4
$\overline{13}$	7.9	9.4
$\overline{14}$	2.7	-1.4
$\overline{15}$	6.5	5.7
10 1 L		
0	8.9	11.9
1	13.3	12.9
1	8.3	9.9
2	3.3	-0.3
3	11.5	15.4
4	7.8	6.9
5	6.8	6.9
6	7.5	-10.3
7	14.8	15.9
8	6.5	6.1
9	7.2	6.9
10	5.1	-5.6
11	10.7	13.3
12	2.5	1.1
13	7.9	6.9
$\bar{1}$	9.6	10.1
$\bar{2}$	2.3	-1.1
$\bar{3}$	12.5	13.7
$\bar{4}$	2.5	0.2
$\bar{5}$	12.9	12.1
0 2 L		
1	82.3	-90.6
2	67.1	69.3
3	62.7	45.8
4	29.0	-23.3
5	8.9	9.4
6	50.9	47.2
7	18.5	12.1
8	22.1	24.2
9	24.0	-19.5
10	26.9	25.9
11	7.9	1.7
12	13.4	13.8
13	9.4	8.9
14	19.1	20.2
15	3.2	-2.3
16	15.4	15.2
17	6.6	-3.5
18	8.1	8.9
1 2 L		
0	8.7	1.2
1	3.0	-5.1
2	50.9	53.5
3	17.8	18.9
4	9.4	-3.7
5	7.4	4.2
6	65.3	62.8
7	20.5	16.3
8	26.7	28.2
9	2.9	-1.7
10	21.4	22.8
11	11.5	11.7
12	13.6	13.7
13	12.5	11.1
14	18.7	19.3
15	8.4	-8.1
16	12.9	14.7
$\bar{1}$	32.5	28.9
$\bar{2}$	57.2	56.5
$\bar{3}$	12.4	11.1
$\bar{4}$	10.9	3.3
$\bar{5}$	17.9	-5.9
$\bar{6}$	44.0	47.1
$\bar{7}$	4.9	2.1
$\bar{8}$	26.6	27.1
$\bar{9}$	12.2	-10.5
$\overline{10}$	44.7	48.2
$\overline{11}$	15.5	13.6
$\overline{12}$	12.9	12.7
$\overline{13}$	14.9	15.0
$\overline{14}$	17.7	18.5
$\overline{15}$	3.3	-4.7
$\overline{16}$	15.9	13.5
$\overline{17}$	2.7	1.7
$\overline{18}$	15.3	17.8
2 2 L		
0	14.7	18.3
1	29.7	37.4
2	30.9	40.1
3	13.7	-11.5
4	20.8	20.6
5	6.5	5.3
6	34.4	38.9
7	21.9	15.9
8	24.9	19.9
9	20.8	14.5
10	26.6	28.5
11	7.6	-5.0
12	19.3	18.8
13	3.3	6.1
14	20.3	-23.2
15	2.9	-0.2
16	11.5	14.1
17	6.1	5.6
$\bar{1}$	35.7	-38.6
$\bar{2}$	58.4	59.4
$\bar{3}$	8.8	-7.9
$\bar{4}$	27.3	32.2
$\bar{5}$	32.9	34.5
$\bar{6}$	39.2	44.5
$\bar{7}$	5.3	-7.6
$\bar{8}$	28.3	31.3
$\bar{9}$	2.8	-0.1
$\overline{10}$	64.5	71.9
$\overline{11}$	13.2	-11.4
$\overline{12}$	25.4	25.4
$\overline{13}$	28.2	27.9
$\overline{14}$	19.3	18.1
$\overline{15}$	3.3	-6.6
$\overline{16}$	3.1	2.5
$\overline{17}$	2.8	5.4
$\overline{18}$	11.0	14.3
3 2 L		
0	17.7	24.3
1	5.2	6.0
2	9.5	9.9
3	9.1	-8.8
4	25.7	23.4
5	2.6	1.9
6	28.3	26.5
7	2.9	-4.3
8	21.4	18.9
9	3.3	-0.4
10	23.0	22.8
11	3.4	1.4
12	16.1	16.9
13	3.2	0.6
14	17.6	18.9
15	8.3	7.2
16	9.2	10.6
$\bar{1}$	33.1	-37.3
$\bar{2}$	98.1	92.7
$\bar{3}$	-9.5	-12.9
$\bar{4}$	68.9	69.1
$\bar{5}$	14.8	8.5
$\bar{6}$	62.4	60.6
$\bar{7}$	16.9	-16.5
$\bar{8}$	17.4	15.6
$\bar{9}$	21.1	15.0
$\overline{10}$	34.7	43.8
$\overline{11}$	7.8	-5.4
$\overline{12}$	23.8	22.8
$\overline{14}$	26.9	25.9
$\overline{15}$	3.3	2.3
$\overline{16}$	3.1	4.1
$\overline{17}$	2.8	2.1
$\overline{18}$	6.1	7.0
4 2 L		
0	4.1	11.5
1	19.0	-14.5
2	37.7	37.5
3	9.2	-9.8
4	30.4	37.0
5	9.5	5.0
6	27.9	28.3
7	28.3	-27.6
8	7.5	0.4
9	3.4	4.1
10	7.4	7.2
11	9.1	5.6
12	11.9	12.1
13	2.9	-2.2
14	14.2	14.9
$\bar{1}$	21.6	-24.9
$\bar{2}$	50.4	57.1
$\bar{3}$	18.2	11.7
$\bar{4}$	35.6	44.4
$\bar{5}$	12.5	-16.1
$\bar{6}$	67.8	66.8
$\bar{7}$	9.4	-2.9
$\bar{8}$	21.5	24.3
$\bar{9}$	4.5	-0.2
$\overline{10}$	20.4	16.5
$\overline{11}$	8.9	8.7
$\overline{12}$	8.3	7.9
$\overline{13}$	14.7	-12.4
$\overline{14}$	14.6	13.9
$\overline{15}$	3.3	1.9
$\overline{16}$	3.0	6.6
$\overline{17}$	2.7	-1.5
$\overline{18}$	8.7	9.2
5 2 L		
0	26.9	30.5
1	17.5	-16.1
2	46.7	58.2
3	11.9	6.5
4	22.5	18.4
5	19.9	-15.7
6	13.5	13.1
7	3.4	-2.8
8	3.4	-2.3
9	3.4	0.6
10	15.7	14.3
11	6.1	3.2
12	12.1	11.2
13	6.5	-5.4
$\bar{1}$	11.7	14.3
$\bar{2}$	27.9	32.5
$\bar{3}$	11.6	-12.0
$\bar{4}$	20.3	15.3
$\bar{5}$	4.9	-1.9
$\bar{6}$	27.2	30.5
$\bar{7}$	20.1	16.1
$\bar{8}$	16.7	16.5
$\bar{9}$	9.9	-4.6
$\overline{10}$	16.7	14.9
$\overline{11}$	3.3	0.3
$\overline{12}$	19.9	17.1
$\overline{13}$	3.4	2.2
$\overline{14}$	12.9	11.3
6 2 L		
0	23.5	22.4
1	3.1	1.6
2	35.2	37.2
3	3.3	-0.8
4	4.7	5.5
5	15.5	-13.6
6	19.3	18.2
7	20.9	19.5
8	17.3	14.6
9	5.9	-7.6
10	17.2	18.2
$\bar{1}$	26.4	20.4
$\bar{2}$	27.8	28.6
$\bar{3}$	5.3	-2.2

Table 2 (Continued)

	F_o	F_c
$\bar{4}$	24.5	23.9
$\bar{5}$	5.4	-4.1
$\bar{6}$	29.9	31.8
$\bar{7}$	25.2	24.4
$\bar{8}$	10.1	7.9
$\bar{9}$	3.3	-2.2
$\bar{10}$	10.6	10.5
$\bar{11}$	4.8	1.3
$\bar{12}$	9.3	11.2
$\bar{13}$	3.3	0.4
$\bar{14}$	18.8	20.2
$\bar{15}$	8.3	8.2
$\bar{16}$	10.4	9.0
$\bar{17}$	2.3	-2.1
$\bar{18}$	6.5	5.2
	7 2 L	
0	23.5	19.7
1	4.1	4.3
2	27.6	30.5
3	3.4	-2.3
4	18.5	15.7
5	5.8	4.6
6	21.4	20.6
7	7.0	5.1
8	13.8	13.5
9	2.7	-3.8
10	10.9	10.6
$\bar{1}$	9.5	3.6
$\bar{2}$	25.1	24.2
$\bar{3}$	13.9	7.1
$\bar{4}$	14.7	13.9
$\bar{5}$	14.7	8.6
$\bar{6}$	27.7	32.9
$\bar{8}$	24.1	20.3
$\bar{9}$	18.7	12.7
$\bar{10}$	23.9	19.9
$\bar{11}$	3.4	1.0
$\bar{12}$	3.3	-0.6
$\bar{13}$	3.2	-1.6
$\bar{14}$	14.4	13.9
$\bar{15}$	4.7	-5.3
$\bar{16}$	13.9	14.5
$\bar{17}$	6.7	6.2
	8 2 L	
0	26.4	25.4
1	7.2	-3.1
2	22.7	22.3
3	8.9	6.9
4	16.2	15.4
5	3.0	-2.0
6	12.4	13.7
7	3.2	-5.8
8	10.7	12.9
$\bar{1}$	9.7	3.1
$\bar{2}$	27.7	30.9
$\bar{3}$	3.4	-3.3
$\bar{4}$	9.4	6.7
$\bar{5}$	20.0	15.7
$\bar{6}$	16.8	17.1
$\bar{7}$	17.9	-19.9
$\bar{8}$	13.5	13.6
$\bar{9}$	12.1	9.1
$\bar{10}$	21.1	23.0
$\bar{11}$	3.2	-0.2
$\bar{12}$	4.3	3.7
$\bar{13}$	2.9	1.4
$\bar{14}$	11.5	10.5
$\bar{15}$	6.4	-10.3
$\bar{16}$	9.2	11.9
	9 2 L	
0	12.7	14.4

	F_o	F_c
1	3.1	-1.2
2	11.9	11.6
3	2.8	-3.5
4	12.4	12.4
5	2.4	-0.1
6	14.5	14.7
$\bar{1}$	3.2	0.5
$\bar{2}$	21.7	21.7
$\bar{3}$	3.3	1.1
$\bar{4}$	9.7	6.7
$\bar{5}$	4.6	-0.5
$\bar{6}$	13.2	9.4
$\bar{7}$	3.2	-4.2
$\bar{8}$	3.1	5.6
$\bar{9}$	5.3	-6.5
$\bar{10}$	14.8	15.0
$\bar{11}$	2.8	-4.9
$\bar{12}$	7.2	7.6
$\bar{13}$	2.4	-0.9
$\bar{14}$	9.9	9.9
	10.2 L	
0	8.4	6.9
1	2.4	-1.1
2	6.9	6.2
$\bar{1}$	2.6	-0.6
$\bar{3}$	3.4	2.3
$\bar{2}$	8.9	8.7
$\bar{4}$	7.6	6.1
$\bar{5}$	6.2	-5.2
$\bar{6}$	2.7	3.4
$\bar{7}$	2.7	2.1
$\bar{8}$	8.3	4.7
$\bar{9}$	2.5	-1.8
$\bar{10}$	13.6	11.4
$\bar{11}$	2.2	-6.8
$\bar{12}$	9.2	11.5
	0 3 L	
1	57.1	54.2
2	30.7	-30.0
3	27.1	28.9
4	12.7	12.9
5	27.1	23.4
6	13.3	10.0
7	34.5	35.9
8	23.6	-21.3
9	41.9	42.6
10	16.9	-14,1
11	43.5	45.7
12	9.9	7.8
13	19.1	13.6
14	2.7	5.2
15	20.9	23.2
16	2.2	2.1
17	8.1	7.4
	1 3 L	
0	11.5	9.6
1	17.7	-11.3
2	7.4	-5.9
3	10.3	18.3
4	19.3	-13.4
5	22.9	17.7
6	16.5	10.0
7	47.8	43.4
8	21.9	-19.9
9	35.5	36.4
10	9.5	4.2
11	19.3	16.1
12	2.8	3.6
13	18.7	15.2
14	2.6	2.4
15	19.1	18.5

	F_o	F_c
16	7.6	-6.8
17	16.7	16.3
$\bar{1}$	78.8	76.1
$\bar{2}$	25.6	24.7
$\bar{3}$	47.9	42.4
$\bar{4}$	20.9	12.3
$\bar{5}$	58.6	53.7
$\bar{6}$	11.7	6.4
$\bar{7}$	35.9	42.0
$\bar{8}$	3.8	-5.7
$\bar{9}$	31.4	25.6
$\bar{10}$	4.7	1.3
$\bar{11}$	28.2	30.8
$\bar{12}$	18.3	14.2
$\bar{13}$	15.7	11.8
$\bar{14}$	6.8	-4.3
$\bar{15}$	21.6	20.5
$\bar{16}$	8.3	-8.9
$\bar{17}$	8.8	8.4
	2 3 L	
0	19.9	15.3
1	10.3	-7.9
2	8.1	6.5
3	43.1	49.1
4	10.9	-4.9
5	14.4	10.1
6	2.2	-2.4
7	34.7	33.3
8	8.5	4.9
9	3.2	-3.1
10	9.8	-6.3
11	2.8	6.2
12	10.6	-7.9
13	22.6	21.2
14	2.9	1.9
15	16.7	15.2
$\bar{1}$	62.2	62.3
$\bar{2}$	11.7	-18.5
$\bar{3}$	20.6	17.9
$\bar{4}$	35.9	34.7
$\bar{5}$	26.2	32.7
$\bar{6}$	27.8	24.7
$\bar{7}$	28.5	32.3
$\bar{8}$	14.1	-8.8
$\bar{9}$	30.4	30.7
$\bar{10}$	22.0	-16.5
$\bar{11}$	20.7	17.8
$\bar{12}$	9.8	7.5
$\bar{13}$	9.5	9.6
$\bar{14}$	2.7	-0.6
$\bar{15}$	5.8	5.8
$\bar{16}$	5.0	-5.1
$\bar{17}$	8.0	9.2
	3 3 L	
0	10.7	-8.4
1	26.4	29.7
2	28.9	30.5
3	24.5	21.9
4	7.2	1.4
5	22.5	19.1
6	3.2	-2.5
7	16.6	15.1
8	10.1	-7.5
9	8.7	4.5
10	8.3	-4.9
11	12.3	14.4
12	6.7	-4.5
13	18.8	16.2
14	2.2	2.5
15	6.9	4.4
$\bar{1}$	24.9	25.2
$\bar{2}$	11.9	7.1
$\bar{3}$	46.3	45.9

	F_o	F_c
$\bar{4}$	6.9	2.9
$\bar{5}$	24.3	20.7
$\bar{6}$	7.8	7.7
$\bar{7}$	9.8	9.4
$\bar{8}$	8.9	-9.1
$\bar{9}$	24.3	25.8
$\bar{10}$	2.6	-0.8
$\bar{11}$	19.7	20.4
$\bar{12}$	10.7	-10.8
$\bar{13}$	25.7	22.1
$\bar{14}$	3.3	-3.6
$\bar{15}$	3.2	0.9
$\bar{16}$	2.9	1.5
$\bar{17}$	7.4	7.9
	4 3 L	
0	14.4	-13.3
1	39.0	36.6
2	24.0	19.4
3	21.9	16.2
4	15.2	7.2
5	35.2	36.8
6	2.6	-0.5
7	3.4	8.3
8	16.1	-13.5
9	23.9	20.5
10	14.7	10.7
11	12.7	11.2
12	2.4	-3.5
13	10.4	10.6
14	4.7	-4.8
$\bar{1}$	3.7	8.6
$\bar{2}$	18.3	12.9
$\bar{3}$	34.6	40.6
$\bar{4}$	2.5	2.2
$\bar{5}$	49.6	55.3
$\bar{6}$	18.9	-16.5
$\bar{7}$	19.0	15.4
$\bar{8}$	2.4	-2.4
$\bar{9}$	29.5	32.9
$\bar{10}$	2.6	-5.4
$\bar{11}$	16.1	14.7
$\bar{12}$	2.8	-5.0
$\bar{13}$	28.5	29.3
$\bar{14}$	6.7	-6.7
$\bar{15}$	2.5	4.3
$\bar{16}$	4.0	4.7
$\bar{17}$	7.6	8.3
	5 3 L	
0	4.0	6.3
1	35.7	37.1
2	14.1	7.0
3	29.5	27.5
4	2.6	2.5
5	26.9	26.2
6	2.7	0.9
7	9.9	8.8
8	8.7	2.1
9	20.6	17.8
10	9.5	6.5
11	18.6	13.9
12	2.1	-4.0
13	7.4	9.5
$\bar{1}$	30.5	32.4
$\bar{2}$	2.8	-1.9
$\bar{3}$	30.7	33.8
$\bar{4}$	11.8	-14.1
$\bar{5}$	42.8	48.3
$\bar{6}$	4.5	-0.9
$\bar{7}$	29.6	33.7
$\bar{8}$	2.6	1.0
$\bar{9}$	25.5	23.6
$\bar{10}$	3.8	-8.5
$\bar{11}$	21.2	19.3

	F_o	F_c
$\bar{12}$	4.7	-4.6
$\bar{13}$	19.9	18.2
$\bar{14}$	6.7	5.9
$\bar{15}$	11.9	16.8
$\bar{16}$	2.2	-1.3
$\bar{17}$	6.7	5.6
	6 3 L	
0	13.3	8.3
1	31.4	30.2
2	12.6	3.5
3	27.3	26.9
4	13.8	-13.1
5	21.7	18.7
6	22.1	14.7
7	19.9	12.3
8	2.6	5.7
9	12.2	12.9
10	2.2	0.9
11	11.2	11.5
$\bar{1}$	30.5	30.2
$\bar{2}$	11.9	7.4
$\bar{3}$	32.3	35.5
$\bar{4}$	22.4	-23.7
$\bar{5}$	25.9	23.2
$\bar{6}$	12.3	7.9
$\bar{7}$	33.9	40.3
$\bar{8}$	3.3	-1.9
$\bar{9}$	5.9	5.9
$\bar{10}$	2.8	-2.0
$\bar{11}$	16.3	14.2
$\bar{12}$	5.2	-5.8
$\bar{13}$	2.6	4.1
$\bar{14}$	4.3	7.0
$\bar{15}$	18.2	18.1
$\bar{16}$	3.5	4.5
$\bar{17}$	11.5	11.2
	7 3 L	
0	3.4	-2.0
1	26.0	23.5
2	2.7	-0.2
3	26.8	27.9
4	2.7	-0.2
5	9.9	5.8
6	9.6	10.6
7	12.7	15.2
8	2.2	2.1
9	7.8	5.6
$\bar{1}$	23.2	22.2
$\bar{2}$	2.7	0.6
$\bar{3}$	17.5	11.9
$\bar{4}$	2.7	-2.9
$\bar{5}$	2.7	6.2
$\bar{6}$	3.9	-4.3
$\bar{7}$	24.5	21.9
$\bar{8}$	2.8	-1.8
$\bar{9}$	9.5	9.8
$\bar{10}$	4.7	5.3
$\bar{11}$	10.0	9.9
$\bar{12}$	2.6	0.7
$\bar{13}$	2.5	3.9
$\bar{14}$	2.3	-5.9
$\bar{15}$	6.9	7.2
$\bar{16}$	4.2	5.8
	8 3 L	
0	9.4	-9.3
1	13.4	14.8
2	4.6	3.7
3	11.9	11.3
4	2.5	2.7
5	2.3	1.6
6	2.1	-1.5

Table 2 (Continued)

	F_o	F_c
7	16.5	18.1
$\bar{1}$	21.9	19.9
$\bar{2}$	2.7	-1.1
$\bar{3}$	8.7	8.7
$\bar{4}$	2.8	-0.7
$\bar{5}$	2.7	4.3
$\bar{6}$	2.7	-4.9
$\bar{7}$	3.3	5.3
$\bar{8}$	9.3	-9.9
$\bar{9}$	10.3	9.1
$\bar{10}$	9.6	10.8
$\bar{11}$	8.6	9.4
$\bar{12}$	4.7	5.1
$\bar{13}$	9.2	8.7
$\bar{14}$	1.9	-0.7
$\bar{15}$	5.9	10.7

9 3 L

0	6.1	-8.9
1	4.3	5.3
2	2.2	-0.7
3	5.5	6.7
4	1.9	-5.1
5	14.6	14.0
$\bar{1}$	11.6	14.2
$\bar{2}$	3.5	2.3
$\bar{3}$	9.5	9.1
$\bar{4}$	3.1	-2.5
$\bar{5}$	11.9	15.5
$\bar{6}$	3.6	-0.3
$\bar{7}$	3.5	4.9
$\bar{8}$	2.4	-3.8
$\bar{9}$	16.1	16.9
$\bar{10}$	2.2	0.3
$\bar{11}$	3.7	5.7
$\bar{12}$	1.9	3.2
$\bar{13}$	6.1	7.1

10 3 L

0	1.8	-4.8
1	4.1	6.1
2	3.6	-5.7
$\bar{1}$	12.9	15.1
$\bar{2}$	2.0	-1.3
$\bar{3}$	4.9	7.1
$\bar{4}$	2.1	-0.2
$\bar{5}$	10.4	14.4
$\bar{6}$	2.9	1.5
$\bar{7}$	2.8	3.1
$\bar{8}$	2.7	5.1
$\bar{9}$	15.9	18.7
$\bar{10}$	1.7	-0.1
$\bar{11}$	13.0	12.6

0 4 L

2	80.8	60.9
3	52.8	-39.1
4	32.5	28.8
5	17.9	16.9
6	9.1	9.6
7	9.9	-5.0
8	18.7	19.1
9	8.1	-11.1
10	20.8	19.0
11	3.1	-2.8
12	15.3	16.0
13	3.0	2.5
14	2.9	2.5
15	2.6	6.2
16	8.7	11.2

1 4 L

0	58.7	49.4

	F_o	F_c
1	3.7	-1.5
2	27.6	21.1
3	25.4	-27.7
4	37.5	39.7
5	14.0	13.2
6	29.7	31.8
7	20.9	-16.6
8	33.2	35.3
9	20.5	-18.5
10	14.5	14.4
11	7.3	-5.2
12	8.7	8.5
13	2.9	-1.9
14	2.7	0.7
15	2.4	0.9
16	10.0	14.3
$\bar{1}$	13.9	-11.5
$\bar{2}$	23.5	28.1
$\bar{3}$	19.8	20.2
$\bar{4}$	39.1	41.3
$\bar{5}$	5.7	3.8
$\bar{6}$	4.2	5.1
$\bar{7}$	2.5	-0.9
$\bar{8}$	28.2	32.7
$\bar{9}$	18.5	-18.5
$\bar{10}$	8.4	3.6
$\bar{11}$	3.1	2.1
$\bar{13}$	5.8	-6.1
$\bar{12}$	15.1	16.2
$\bar{14}$	11.6	13.8
$\bar{15}$	3.1	-4.9
$\bar{16}$	12.3	13.5
$\bar{17}$	5.2	-8.7

2 4 L

0	59.5	60.3
1	7.1	5.4
2	7.6	2.4
3	13.3	10.4
4	43.5	48.2
5	6.9	-4.8
6	32.1	34.6
7	11.3	-10.9
8	29.3	31.3
9	15.1	-10.1
10	3.1	0.9
11	12.4	-8.2
12	20.5	20.2
13	2.8	-1.8
14	12.2	11.6
$\bar{1}$	7.7	-8.7
$\bar{2}$	14.2	14.5
$\bar{3}$	15.5	-11.0
$\bar{4}$	20.4	21.0
$\bar{5}$	15.7	13.4
$\bar{6}$	18.4	18.5
$\bar{7}$	6.9	7.7
$\bar{8}$	31.2	33.4
$\bar{9}$	16.9	-19.8
$\bar{10}$	27.8	25.7
$\bar{11}$	6.2	3.1
$\bar{12}$	8.7	9.6
$\bar{13}$	8.6	-9.3
$\bar{14}$	14.5	16.7
$\bar{15}$	2.7	-2.7
$\bar{16}$	8.4	8.7

3 4 L

0	42.1	42.3
1	10.9	7.5
2	11.5	8.8
3	24.3	24.6
4	33.1	37.0
5	11.4	-13.1
6	20.5	20.9

	F_o	F_c
7	3.5	0.7
8	21.7	21.2
9	3.1	-0.4
10	16.2	14.3
11	2.9	1.7
12	18.3	19.9
13	2.5	-0.2
14	11.3	13.6
$\bar{1}$	15.8	12.8
$\bar{2}$	21.6	24.2
$\bar{3}$	15.1	7.0
$\bar{4}$	46.3	43.0
$\bar{5}$	2.7	0.7
$\bar{6}$	18.5	17.1
$\bar{7}$	7.3	1.6
$\bar{8}$	17.3	14.3
$\bar{9}$	8.2	-6.9
$\bar{10}$	22.6	22.6
$\bar{11}$	12.3	8.7
$\bar{12}$	26.6	26.6
$\bar{13}$	7.2	-2.9
$\bar{14}$	7.6	7.6
$\bar{15}$	2.7	2.4
$\bar{16}$	9.1	10.7

4 4 L

0	25.8	27.8
1	12.8	4.0
2	21.4	20.5
3	17.9	14.5
4	35.8	41.2
5	8.4	6.3
6	6.1	5.0
7	3.1	3.9
8	20.5	25.1
9	11.7	6.4
10	15.2	12.7
11	3.2	3.7
12	9.9	12.4
$\bar{1}$	7.1	2.7
$\bar{2}$	6.3	1.9
$\bar{3}$	25.9	23.9
$\bar{4}$	49.0	52.7
$\bar{5}$	13.3	8.1
$\bar{6}$	20.5	15.5
$\bar{7}$	2.8	1.3
$\bar{9}$	3.5	-3.4
$\bar{8}$	19.1	18.7
$\bar{10}$	3.1	3.7
$\bar{11}$	7.2	-7.1
$\bar{12}$	25.9	29.4
$\bar{13}$	3.0	3.3
$\bar{14}$	7.5	7.8
$\bar{15}$	6.9	6.5
$\bar{16}$	8.9	11.4

5 4 L

0	20.4	18.9
1	18.8	16.5
2	27.9	29.2
3	8.5	5.9
4	22.6	20.4
5	13.8	10.9
6	13.9	12.8
7	3.1	-2.9
8	14.3	15.8
9	6.6	4.8
10	12.7	13.3
11	5.5	-1.3
12	10.3	9.9
$\bar{1}$	5.5	-3.4
$\bar{2}$	12.2	13.1
$\bar{3}$	9.6	-10.4
$\bar{4}$	20.8	18.9
$\bar{5}$	5.7	0.4

	F_o	F_c
$\bar{6}$	14.3	10.1
$\bar{7}$	2.9	4.4
$\bar{8}$	22.5	25.4
$\bar{9}$	8.7	5.3
$\bar{10}$	3.6	7.8
$\bar{11}$	16.8	-17.7
$\bar{12}$	17.7	19.0
$\bar{13}$	2.9	-0.5
$\bar{14}$	13.6	16.2
$\bar{15}$	2.5	1.3
$\bar{16}$	11.6	14.3

6 4 L

0	16.7	14.2
1	17.6	14.2
2	15.1	11.4
3	13.0	-13.5
4	14.0	12.9
5	8.7	1.2
6	18.1	20.5
7	2.9	-0.9
8	9.4	7.6
9	8.6	5.9
10	11.4	15.7
$\bar{1}$	7.0	5.9
$\bar{2}$	15.1	14.0
$\bar{3}$	17.2	-16.6
$\bar{4}$	16.0	20.5
$\bar{5}$	19.3	-19.5
$\bar{6}$	7.9	7.5
$\bar{7}$	8.0	-6.1
$\bar{8}$	29.8	32.0
$\bar{9}$	3.1	-0.6
$\bar{10}$	8.0	8.7
$\bar{11}$	3.0	-4.9
$\bar{12}$	14.6	16.9
$\bar{13}$	2.8	-4.1
$\bar{14}$	9.7	10.8
$\bar{15}$	2.3	-0.2
$\bar{16}$	12.3	19.1

7 4 L

0	15.1	19.1
1	3.1	-5.5
2	10.9	10.1
3	13.6	-12.7
4	13.2	14.7
5	5.7	4.2
6	6.2	6.7
7	6.4	4.9
8	11.5	10.2
9	6.4	3.0
$\bar{1}$	3.1	-4.2
$\bar{2}$	6.3	6.1
$\bar{3}$	3.1	1.1
$\bar{4}$	15.7	17.2
$\bar{5}$	11.2	-13.9
$\bar{6}$	17.5	20.8
$\bar{7}$	5.4	-7.4
$\bar{8}$	20.3	23.2
$\bar{9}$	5.3	-7.7
$\bar{10}$	18.0	17.1
$\bar{11}$	6.7	6.3
$\bar{12}$	12.0	14.5
$\bar{13}$	2.5	-4.3
$\bar{14}$	8.5	9.1

8 4 L

0	17.7	19.2
1	2.9	-6.3
2	12.5	13.0
$\bar{1}$	9.4	-9.9
$\bar{2}$	18.1	17.5
$\bar{3}$	9.6	5.7

	F_o	F_c
$\bar{4}$	13.5	9.1
$\bar{5}$	3.0	-4.1
$\bar{6}$	19.7	22.9
$\bar{7}$	7.6	0.6
$\bar{8}$	11.8	11.4
$\bar{9}$	2.9	-3.3
$\bar{10}$	13.3	15.1
$\bar{11}$	10.5	10.9
$\bar{12}$	11.6	11.4
$\bar{13}$	2.1	0.5
$\bar{14}$	9.2	10.9

9 4 L

0	14.3	13.3
1	2.3	-0.9
2	2.2	3.2
3	2.0	3.1
4	9.9	8.5
$\bar{1}$	2.9	-3.1
$\bar{2}$	13.8	14.4
$\bar{3}$	8.4	5.0
$\bar{4}$	15.3	15.4
$\bar{5}$	2.6	-0.3
$\bar{6}$	11.7	8.4
$\bar{7}$	6.6	4.9
$\bar{8}$	15.7	15.9
$\bar{9}$	2.4	1.4
$\bar{10}$	10.8	9.9
$\bar{11}$	9.1	7.3
$\bar{12}$	9.1	7.4

0 5 L

1	12.2	17.3
2	3.4	-5.2
3	33.3	38.1
4	11.3	9.4
5	21.2	20.4
6	7.1	8.3
7	14.4	11.0
8	2.9	-0.4
9	14.4	14.3
10	3.2	-2.5
11	16.3	12.4
12	10.3	-5.8
13	2.9	3.9
14	2.6	7.3
15	8.7	5.2

1 5 L

0	15.5	-12.8
1	35.8	38.7
2	7.1	3.1
3	21.1	20.8
4	4.7	0.1
5	29.6	31.0
6	13.3	10.7
7	11.6	9.8
8	3.1	5.0
9	22.0	21.2
10	3.2	-1.4
11	10.9	8.0
12	9.9	8.3
13	10.1	6.3
$\bar{1}$	20.1	25.6
$\bar{2}$	19.3	19.5
$\bar{3}$	30.2	31.2
$\bar{4}$	2.0	-1.1
$\bar{5}$	18.0	17.3
$\bar{6}$	5.9	-1.9
$\bar{7}$	22.9	26.7
$\bar{8}$	2.9	-2.7
$\bar{9}$	21.4	18.2
$\bar{10}$	3.2	1.1
$\bar{11}$	6.4	1.1

Table 2 (Continued)

	F_o	F_c
$\bar{12}$	12.4	12.2
$\bar{13}$	10.4	10.1
$\bar{14}$	2.7	0.3
$\bar{15}$	7.6	6.9
	2 5 L	
0	16.1	-23.8
1	39.5	41.1
2	12.1	12.2
3	22.6	20.3
4	4.5	-1.3
5	29.0	33.2
6	14.9	12.7
7	27.5	29.7
8	3.2	-4.3
9	13.7	11.9
10	3.2	0.1
11	13.5	10.6
12	12.3	8.5
13	11.7	12.5
$\bar{1}$	52.4	56.2
$\bar{2}$	10.5	5.2
$\bar{3}$	24.6	19.3
$\bar{4}$	9.2	6.2
$\bar{5}$	11.9	10.9
$\bar{6}$	9.6	9.4
$\bar{7}$	18.8	18.3
$\bar{8}$	5.9	-3.5
$\bar{9}$	23.2	23.4
$\bar{10}$	5.5	1.2
$\bar{11}$	12.8	11.4
$\bar{12}$	3.1	3.3
$\bar{13}$	14.6	13.2
$\bar{14}$	2.8	3.6
$\bar{15}$	10.1	10.5
	3 5 L	
1	24.3	27.6
2	2.9	2.8
3	25.2	22.3
4	9.9	4.7
5	32.3	37.4
6	6.2	-10.4
7	28.8	29.5
8	3.2	-4.8
9	14.1	16.6
10	3.0	1.9
11	13.1	14.2
12	8.3	6.7
13	10.5	12.3
$\bar{1}$	26.2	24.9
$\bar{2}$	14.4	-15.8
$\bar{3}$	34.9	35.7
$\bar{4}$	5.8	3.8
$\bar{5}$	19.7	18.4
$\bar{6}$	2.7	1.2
$\bar{7}$	11.5	9.7
$\bar{8}$	9.3	-7.1
$\bar{9}$	5.4	2.4
$\bar{10}$	3.7	1.9
$\bar{11}$	23.9	27.6
$\bar{12}$	3.1	-1.8
$\bar{13}$	15.9	15.7
$\bar{14}$	10.6	6.5
$\bar{15}$	12.9	14.4
	4 5 L	
1	16.5	16.7
2	11.5	-12.6
3	35.4	33.8
4	13.9	5.7
5	20.0	22.9
6	15.2	-16.9
7	17.7	15.8
8	10.8	7.1
9	18.2	19.7
10	2.8	-1.0
11	12.2	11.0
$\bar{0}$	5.6	5.9
$\bar{1}$	7.4	-6.1
$\bar{2}$	21.2	-13.5
$\bar{3}$	7.8	11.9
$\bar{4}$	4.9	-2.9
$\bar{5}$	28.4	29.6
$\bar{6}$	8.4	6.9
$\bar{7}$	16.7	15.3
$\bar{8}$	6.3	-8.7
$\bar{9}$	9.0	8.7
$\bar{10}$	3.2	-0.6
$\bar{11}$	8.3	16.6
$\bar{12}$	5.3	-9.2
$\bar{13}$	15.1	16.4
$\bar{14}$	11.3	9.1
$\bar{15}$	11.5	9.9
	5 5 L	
0	8.3	4.9
1	15.2	14.1
2	6.3	-5.7
3	16.9	19.7
4	6.4	3.5
5	5.5	2.7
6	10.9	-9.0
7	11.1	11.1
8	4.1	5.5
9	13.0	12.9
10	2.7	-2.5
11	13.2	13.5
$\bar{1}$	11.9	11.9
$\bar{2}$	13.5	-8.5
$\bar{3}$	3.0	4.3
$\bar{4}$	9.3	-7.4
$\bar{5}$	9.4	7.3
$\bar{6}$	8.5	4.1
$\bar{7}$	18.3	16.5
$\bar{8}$	12.9	10.1
$\bar{9}$	17.6	20.3
$\bar{10}$	3.2	-0.5
$\bar{11}$	15.0	12.9
$\bar{12}$	2.9	-3.6
$\bar{13}$	10.7	10.9
$\bar{14}$	2.9	-1.2
$\bar{15}$	11.4	12.8
	6 5 L	
0	14.3	12.1
1	9.8	7.9
2	8.3	-3.1
3	3.2	0.1
4	3.1	-3.2
5	14.8	14.5
6	2.9	-3.5
7	9.3	8.3
8	4.2	5.3
9	11.0	11.4
$\bar{1}$	16.5	15.7
$\bar{2}$	6.4	0.6
$\bar{3}$	22.1	22.7
$\bar{4}$	6.4	4.8
$\bar{5}$	9.7	5.9
$\bar{6}$	8.3	-7.9
$\bar{7}$	15.5	14.0
$\bar{8}$	13.5	15.2
$\bar{9}$	17.8	16.1
$\bar{10}$	3.1	-1.4
$\bar{11}$	11.3	11.5
$\bar{12}$	10.1	5.9
$\bar{13}$	9.9	11.7
$\bar{14}$	3.2	-3.9
$\bar{15}$	9.5	13.5
	7 5 L	
0	8.0	4.2
1	10.6	10.5
2	3.0	-1.7
3	12.6	11.3
4	2.8	-4.1
5	12.7	14.6
6	8.2	6.4
7	10.1	6.5
$\bar{1}$	13.6	10.1
$\bar{2}$	6.3	4.5
$\bar{3}$	12.6	11.3
$\bar{4}$	11.5	8.8
$\bar{5}$	20.9	22.6
$\bar{6}$	8.2	6.4
$\bar{7}$	17.0	16.8
$\bar{8}$	11.1	7.4
$\bar{9}$	16.7	16.4
$\bar{10}$	2.8	-0.5
$\bar{11}$	10.2	10.8
$\bar{12}$	6.0	1.7
$\bar{13}$	11.5	16.1
	8 5 L	
0	2.8	0.6
1	18.5	18.8
2	6.2	-0.9
3	9.7	7.9
$\bar{1}$	12.2	11.4
$\bar{2}$	3.3	2.4
$\bar{3}$	9.6	7.6
$\bar{4}$	2.9	2.5
$\bar{5}$	11.7	14.6
$\bar{6}$	2.8	-5.2
$\bar{7}$	18.3	22.3
$\bar{8}$	2.7	-0.4
$\bar{9}$	12.6	13.5
$\bar{10}$	6.2	2.0
$\bar{11}$	12.5	14.8
	9 5 L	
0	2.0	-3.3
1	7.7	10.1
$\bar{1}$	10.7	10.9
$\bar{2}$	3.2	2.6
$\bar{3}$	12.3	16.6
$\bar{4}$	6.1	0.9
$\bar{5}$	6.6	6.7
$\bar{6}$	2.3	-1.3
$\bar{7}$	10.8	14.0
$\bar{8}$	2.1	-8.7
$\bar{9}$	9.9	14.2

[9]. The short Ni—N bonds correspond to the similar bond lengths in diamagnetic nickel complexes having the configurations $(t_{2g})^6(d_{z^2})^2$. For example, four nitrogen atoms around the central metal atom in nickel(II) dimethylglyoxime are at distances of 1.85 Å [10], the Ni—N distances in nickel(II) methyl-ethylglyoxime are 1.83 Å (2×) and 1.88 Å (2×) [11], respectively. The Ni—N distances in paramagnetic octahedral complexes having the configuration $(t_{2g})^6(d_{z^2})^1(d_{x^2-y^2})^1$ are significantly longer. This can be illustrated by interatomic distances Ni—N in the crystal structure of triethylenediamine nickel(II) nitrate $Ni(NH_2CH_2CH_2NH_2)_3(NO_3)_2$. In the structure each nickel atom has around it six nitrogen atoms, two from each ethylenediamine molecule, with Ni—N = 2.12 Å [12]. In the case of d^9 complexes, any magnitude of distortion is possible. Four short bonds Cu—N in the structure of $Cu(C_6H_{14}N_2)_2(NO_3)_2$ are 2.01 Å (2×) and 1.98 Å (2×). The octahedral coordination around Cu is completed by two oxygen atoms from the two nitrate groups at the distances 2.75 Å [3] (Fig. 1*a*). The elongation of the Cu—O bond lengths in the distorted octahedron CuN_4O_2, compared to those of Cu—N makes then 0.74 and 0.77 Å; in $Ni(C_6H_{14}N_2)_2(NO_3)_2$, this elongation is 1.52 and 1.57 Å, respectively.

Table 3

Bond lengths and bond angles in $Ni(C_6H_{14}N_2)_2(NO_3)_2$. Standard deviations are given in parentheses

Bond length in (Å)	Bond angle in (°)
Ni—N(1): 1.953(6)	N(1)—Ni—N(3): 90.7(3)
Ni—N(3): 1.898(9)	N(1)—Ni—O(3): 67.4(3)
Ni—O(3): 3.47(1)	N(3)—Ni—O(3): 113.2(3)
N(1)—C(3): 1.50(1)	Ni—N(1)—C(3): 119.7(6)
N(3)—C(1): 1.29(1)	Ni—N(3)—C(1): 132.6(7)
C(1)—C(5): 1.51(2)	N(1)—C(3)—C(2): 109.5(9)
C(1)—C(6): 1.52(1)	N(1)—C(3)—C(4): 108.4(8)
C(2)—C(3): 1.55(2)	N(1)—C(3)—C(6): 106.7(8)
C(3)—C(4): 1.53(1)	N(3)—C(1)—C(5): 121.7(9)
C(3)—C(6): 1.53(2)	N(3)—C(1)—C(6): 121.7(9)
N(2)—O(1): 1.29(1)	C(1)—C(6)—C(3): 117.0(1)
N(2)—O(2): 1.24(1)	C(2)—C(3)—C(4): 111.4(8)
N(2)—O(3): 1.22(1)	C(2)—C(3)—C(6): 112.9(9)
O(1)..N(1): 3.08(1)	C(4)—C(3)—C(6): 107.8(9)
O(2)..N(1): 2.98(1)	C(5)—C(1)—C(6): 117.0(1)
O(2)..N(3): 3.21(1)	O(1)—N(2)—O(2): 118.5(8)
O(3)..N(1): 3.26(1)	O(1)—N(2)—O(3): 121.4(8)
O(3)..N(3): 3.23(1)	O(2)—N(2)—O(3): 120.0(9)

Different electron configurations of the central atoms also influence bond angles within the coordination polyhedra. While bond angles inside the distorted octahedron CuN_4O_2 in $Cu(C_6H_{14}N_2)_2(NO_3)_2$ are close to right angles ($<$ N—Cu—N: 88 and 92°, $<$ O—Cu—N: 84, 88, 92 and 96°) in $Ni(C_6H_{14}N_2)_2(NO_3)_2$ only the Ni—N bonds in the planar coordination are perpendicularly oriented, with bond angles 89.4(3) and 90.6(3)°. The O—Ni—N bond angles are: 67.4(3), 112.6(3), 113.2(3) and 66.8(3)°, respectively (Fig. 2*b*). The position of the NO_3 group is mainly influenced by a system of hydrogen bonds. The oxygen atom O(3) of this group is in contact with the amino and imino groups from different complex ions in distances 3.26(1) and 3.23(1) Å. The remaining oxygen atoms from the nitrate group make the following contacts with the nitrogen atoms of the amino and imino groups: O(1)—N (3.08(1) Å), O(2)—N (2.98(1) and 3.21(1) Å). There is no doubt about a weak attraction of the Cu^{2+} ion and the two oxygen atoms of the nitrate groups in the apices of the elongated octahedron CuN_4O_2.

The organic ligands of the nickel(II) complex establish the chelate ring structure in the same way as it was found in the detailed structural analysis available for copper(II) complex [3]. The amino and imino groups are clearly distinguished through different carbon-to-nitrogen distances, the C—NH and C—NH_2 lengths being 1.29(1) and 1.50(1) Å, respectively. The corresponding bond lengths in the copper(II) complex are 1.30 and 1.51 Å, respectively. The mean single bond distance C—C in the organic part of the aliphatic *Schiff* base complex is 1.525 Å for $Ni(C_6H_{14}N_2)_2(NO_3)_2$ and 1.510 Å for $Cu(C_6H_{14}N_2)_2(NO_3)_2$ [3]. The bond angles in the six member chelate ring of the nickel complex are: N(1)—Ni—N(3): 90.6(3)°; Ni—N(3)—C(1): 132.6(7)°; N(3)—C(1)—C(6): 121.7(9)°; C(1)—C(6)—C(3): 117.1(9)°; C(6)—C(3)—N(1): 106.7(8)°; C(3)—N(1)—Ni: 119.7(6)°. All six carbon atoms of the aliphatic chain are situated above (or below) the plane defined by the nickel and four nitrogen atoms. The six tetrahedral bond angles around C(3) are: 106.7(8), 107.8(9), 108.4(8), 109.5(9), 111.4(8) and 112.9(9)°.

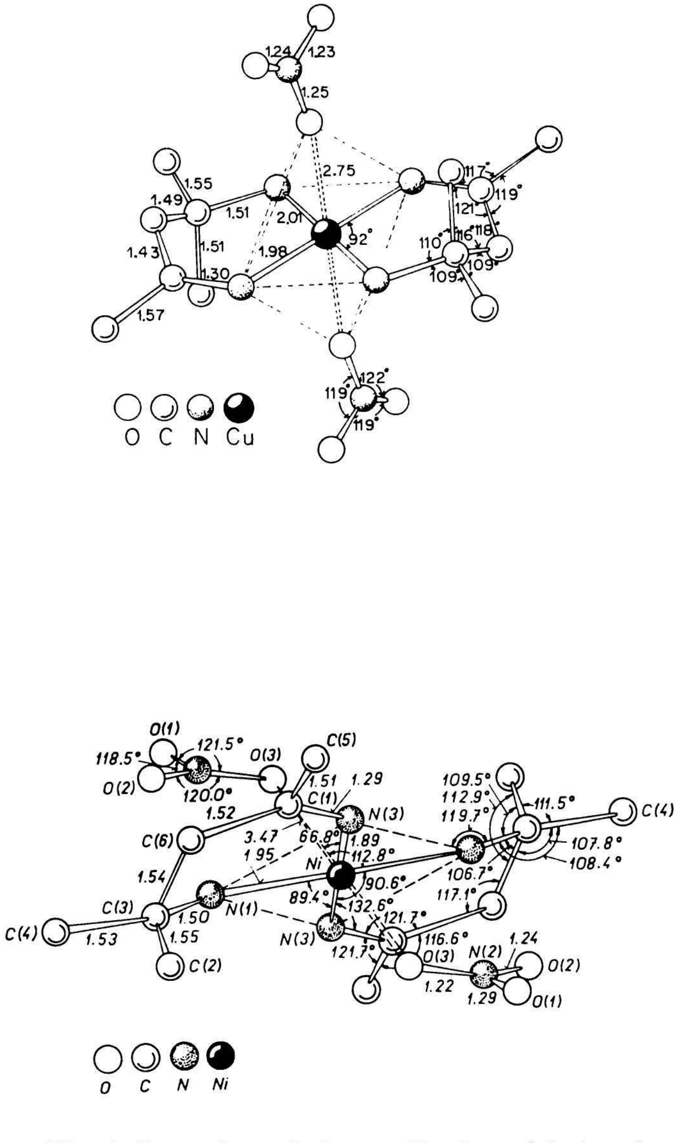

Fig. 1. Comparison of the coordination polyhedra of $Cu(C_6H_{14}N_2)_2(NO_3)_2$ (upper) and $Ni(C_6H_{14}N_2)_2(NO_3)_2$ (lower). Only one structural unit is shown.

The NO_3 group is of usual shape with the mean N—O distance 1.25 Å. The three bond angles O—N—O are: 118.5(8), 120.0(9) and 121.5(8)°.

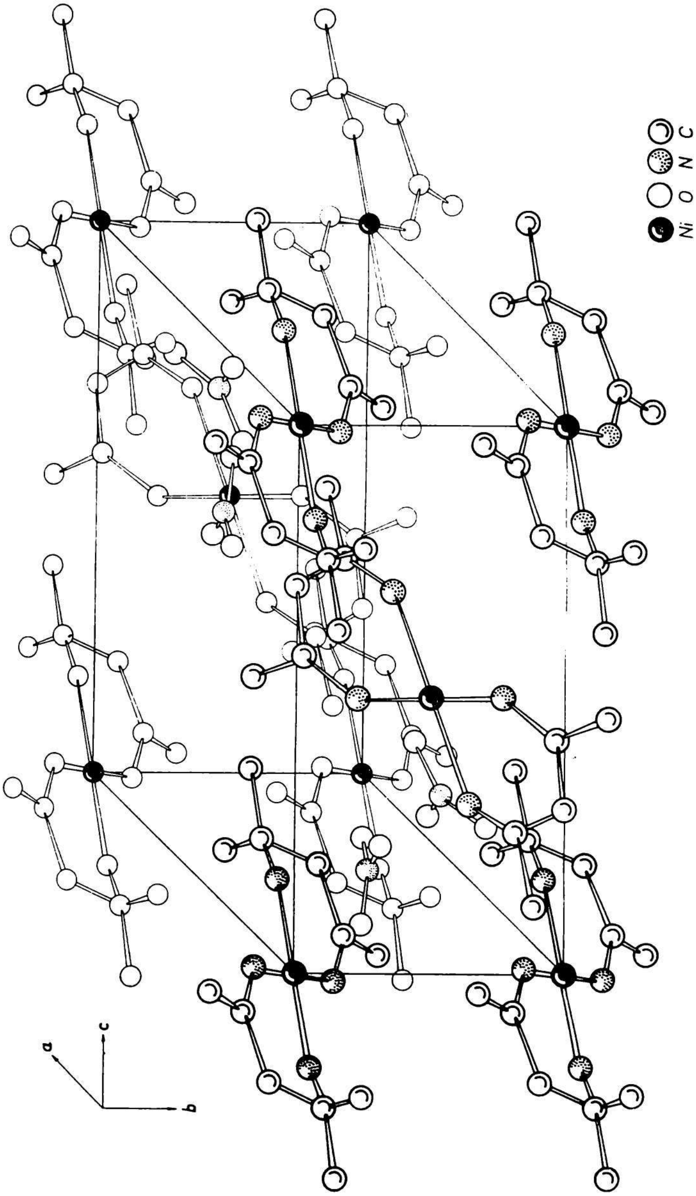

Fig. 2. Unit cell of $Ni(C_6H_{14}N_2)_2(NO_3)_2$.

Crystals of $Ni(C_6H_{14}N_2)_2(NO_3)_2$ *were kindly supplied by Dr. W. Jehn from the Institute of Inorganic Chemistry of the Jena University. The Fourier synthesis and least square refinement were calculated with a set of programs written in the Chemical Institute of the Aarhus University by Dr. R. Hazzel, Dr. J. Nyborg, Dr. J. Danielsen and Dr. S. Lauesen. All calculations were performed on a GIER computer in the Computing Centre of the Slovak Academy of Sciences in Bratislava.*

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