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The Crystal Structure of *N,N'*-Dimethyl-4,4'-dipyridinium Tetracyanonickelate(II)

BY S. S. BASSON, L. D. C. BOK AND J. G. LEIPOLDT

Department of Chemistry, University of the O.F.S., Bloemfontein, South Africa

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The crystal structure of the compound ($C_{16}H_{14}N_6Ni$) was determined by means of three-dimensional X-ray analysis. The space group is $P2_1/n$ (C_{2h}^5), and the cell dimensions are $a=8{\cdot}08$, $b=7{\cdot}65$, $c=13{\cdot}40$ Å, $\beta=98°30'$, with 2 molecules per unit cell. The corresponding Pd and Pt compounds are isomorphous with the Ni compound. The Ni atoms lie on a symmetry centre. The centre of the dipyridinium ion also lies on a symmetry centre. The bond lengths in the pyridinium rings are in good agreement with those in pyridine. The mean distances in the nickel cyanide ion are: Ni–C, 1·88 Å; C–N, 1·15 Å. The distance between the two nitrogen atoms in the dipyridinium ion is 7·05 Å, while the C–C distance between the rings is 1·47 Å.

Introduction

The crystal structure of *N,N'*-dimethyl 4,4'-dipyridinium nickel cyanide has been determined as part of a programme for the study of complex cyanides with different types of organic cation. In the divalent 4,4'-dipyridinium ion the positive charges on the nitrogen atoms are approximately 7 Å apart. Quite a different arrangement of atoms may be expected from that in a similar compound with an ordinary divalent cation, *e.g.* Ca^{2+} in $CaNi(CN)_4$, where the two positive charges are not separated.

Experimental

N,N'-Dimethyl 4,4'-dipyridinium iodide was prepared by heating 4,4'-dipyridyl under reflux with an equivalent amount of methyl iodide in a methanol medium. A solution of the salt was slowly added to a dilute solution of $K_2Ni(CN)_4$, and left to crystallize slowly. Clear yellow crystals of *N,N'*-dimethyl-4,4'-dipyridinium nickel cyanide were obtained which were sparsely soluble in cold, but reasonably soluble in hot water. The crystals thus obtained tended to form twins, and to obtain single crystals the substance was very slowly crystallized from a solution containing equal volumes of water and dimethylformamide.

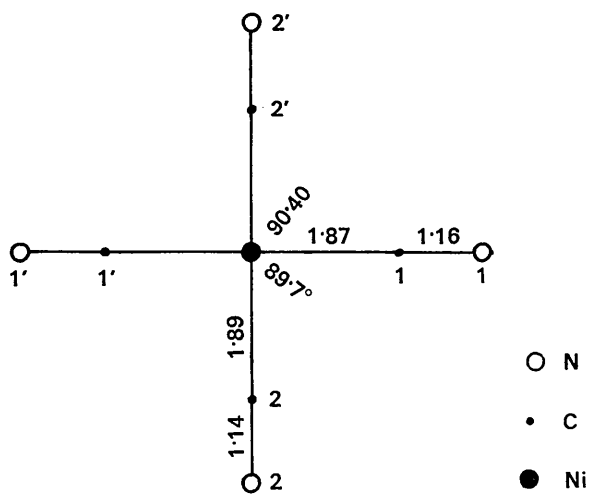

Fig. 1. Bond lengths and angles in $Ni(CN)_4^{2-}$.

The corresponding Pd and Pt compounds (prepared in the same manner) were found to be isomorphous with the Ni salt.

The cell dimensions and space group were determined from oscillation and Weissenberg photographs with Co $K\alpha$ radiation. The camera was standardized with NaCl, $a=5{\cdot}63$ Å.

The crystal data for the three compounds are given in Table 1.

Table 1. *Crystal data*

	$C_{16}H_{14}N_6Ni$	$C_{16}H_{14}N_6Pd$	$C_{16}H_{14}N_6Pt$
M	349·03	396·72	485·41
a	8·08	8·08	8·08 Å
b	7·65	7·71	7·77
c	13·40	13·45	13·45
β	98°30'	98°15'	98°15'
V	820	829	836
d_x	1·43	1·58	1·93
d_c	1·42	1·59	1·94
Z	2	2	2

The space group was found to be $P2_1/n$ from the following systematic absences: $h0l$ for $h+l=2n+1$; $0k0$ for $k=2n+1$.

Intensity data were collected from a needle-like crystal with dimensions $0{\cdot}11\times0{\cdot}13\times0{\cdot}23$ mm, where the **b** direction was the oscillation axis. Intensities were determined for layers 0–4 along the b axis by the multiple film method using a Stoe integrating Weissenberg camera and Cu $K\alpha$ radiation. Ilford industrial G X-ray films were used. Intensities were estimated using a calibrated standard strip. A total of 1068 reflexions was measured of which about 230 were weaker than the background. Intensities varied from 1 to approximately 1500. The usual correction factors (Lorentz, polariza-

Table 2. *Observed and calculated structure factors*

h k l	F_o	F_c	h k l	F_o	F_c	h k l	F_o	F_c	h k l	F_o	F_c	h k l	F_o	F_c	h k l	F_o	F_c
0 0 2	64.2	60.3	6 0 -2	27.0	24.5	2 1 -12	3.4	-6.7	7 1 4	20.6	19.3	3 2 4	9.1	6.5
0 0 4	7.9	10.0	6 0 -4	20.9	19.4	2 1 -13	17.9	16.9	7 1 5	3.4	-6.2	3 2 5	50.6	47.1
0 0 6	54.8	51.6	6 0 -6	33.8	35.5	2 1 -14	3.4	-5.2	7 1 6	21.0	19.7	3 2 6	17.6	-15.8
0 0 8	56.2	50.3	6 0 -8	5.8	8.4	2 1 -15	13.7	10.4	7 1 7	3.2	4.7	3 2 7	38.3	39.7
0 0 10	36.2	30.6	6 0 -10	11.7	11.8	3 1 1	31.7	24.1	7 1 8	23.5	20.7	3 2 8	20.1	-19.8
0 0 12	11.6	12.2	6 0 -12	20.1	19.1	3 1 2	11.0	13.1	7 1 -1	3.4	2.8	3 2 9	7.2	5.2
0 0 14	17.2	13.8	6 0 -14	18.7	17.5	3 1 3	25.1	-19.6	7 1 -2	34.8	39.2	3 2 10	10.5	9.9
0 0 16	10.2	7.6	7 0 -1	15.0	-13.3	3 1 4	9.7	13.2	7 1 -3	19.3	-17.6	3 2 11	14.9	19.9
2 0 0	10.0	-9.6	7 0 -3	8.0	-6.4	3 1 5	28.4	-26.5	7 1 -4	13.1	11.1	3 2 12	13.5	11.9
4 0 0	46.1	38.2	7 0 -5	10.0	10.6	3 1 6	38.4	36.7	7 1 -5	3.4	1.2	3 2 13	20.4	20.8
6 0 0	39.9	42.2	7 0 -7	10.1	9.8	3 1 7	8.6	8.4	7 1 -6	20.6	20.9	3 2 14	2.2	-1.1
8 0 0	3.4	-3.9	7 0 -9	3.3	2.6	3 1 8	9.2	5.4	7 1 -7	15.8	16.1	3 2 15	7.8	9.9
10 0 0	1.9	6.5	7 0 -11	19.2	16.6	3 1 9	8.0	7.2	7 1 -8	21.4	23.3	4 2 1	11.7	9.1
1 0 1	60.3	58.2	7 0 -13	14.7	12.8	3 1 10	3.4	0.7	7 1 -9	3.4	6.8	4 2 2	39.9	41.8
1 0 3	72.1	78.0	8 0 -2	10.1	8.4	3 1 11	8.4	-7.7	7 1 -10	15.1	14.7	4 2 3	4.0	-1.2
1 0 5	43.6	39.0	8 0 -4	5.8	8.0	3 1 12	20.2	16.0	7 1 -11	3.1	2.1	4 2 4	27.4	29.9
1 0 7	24.4	20.9	8 0 -6	3.3	4.0	3 1 13	3.1	3.0	7 1 -12	12.6	14.5	4 2 5	6.3	6.3
1 0 9	19.7	17.9	8 0 -8	20.8	18.6	3 1 14	6.4	5.6	8 1 1	25.7	26.8	4 2 6	31.0	31.7
1 0 11	5.7	1.1	8 0 -10	13.6	10.7	3 1 -1	7.1	8.8	8 1 2	3.4	-3.6	4 2 7	18.6	-18.1
1 0 13	23.3	19.8	8 0 -12	10.0	12.1	3 1 -2	56.6	64.2	8 1 3	12.9	10.8	4 2 8	24.7	28.6
1 0 15	9.7	15.6	9 0 -1	13.9	11.6	3 1 -3	18.5	12.1	8 1 4	3.2	-0.9	4 2 9	26.9	-24.8
2 0 2	9.7	11.6	9 0 -3	19.8	18.4	3 1 -4	65.9	67.9	8 1 5	13.7	11.9	4 2 10	33.3	30.6
2 0 4	35.9	33.5	9 0 -5	16.2	14.0	3 1 -5	17.9	14.2	8 1 6	2.7	-4.2	4 2 11	8.9	-8.3
2 0 6	64.9	67.6	9 0 -7	14.0	11.8	3 1 -6	24.7	28.4	8 1 7	20.4	19.1	4 2 12	12.1	11.7
2 0 8	58.4	63.6	9 0 -9	12.4	15.1	3 1 -7	6.3	-5.0	8 1 -1	22.2	22.9	4 2 13	2.2	0.7
2 0 10	5.5	5.1	10 0 -2	14.1	14.3	3 1 -8	29.6	29.6	8 1 -2	16.8	-14.7	4 2 14	7.7	8.0
2 0 12	15.5	9.1	10 0 -4	11.8	12.6	3 1 -9	8.9	7.5	8 1 -3	10.3	12.8	5 2 1	37.7	39.5
2 0 14	20.2	17.8	10 0 -6	8.6	10.2	3 1 -10	32.1	30.9	8 1 -4	5.9	5.8	5 2 2	4.4	-1.3
2 0 16	12.6	13.0	0 1 1	22.5	27.0	3 1 -11	18.3	15.2	8 1 -5	10.2	10.2	5 2 3	30.5	28.6
3 0 1	49.6	50.2	0 1 2	39.6	-36.7	3 1 -12	34.6	32.8	8 1 -6	5.9	3.8	5 2 4	8.3	-7.9
3 0 3	60.1	58.6	0 1 3	10.6	6.8	3 1 -13	3.4	2.9	8 1 -7	23.8	29.4	5 2 5	21.0	19.9
3 0 5	24.9	24.8	0 1 4	45.5	39.4	3 1 -14	3.3	5.3	8 1 -8	3.2	-3.3	5 2 6	3.0	0.4
3 0 7	55.3	48.9	0 1 5	28.9	28.6	4 1 1	33.5	33.2	8 1 -9	19.5	22.7	5 2 7	3.0	5.3
3 0 9	57.7	63.8	0 1 6	33.6	27.0	4 1 2	23.0	17.9	8 1 -10	2.8	-4.6	5 2 8	3.0	4.3
3 0 11	49.1	46.8	0 1 7	2.3	4.1	4 1 3	7.5	9.1	8 1 -11	7.3	7.5	5 2 9	23.4	21.9
3 0 13	13.3	10.3	0 1 8	12.6	-9.9	4 1 4	19.4	17.9	9 1 1	2.9	0.1	5 2 10	5.0	-6.2
3 0 15	11.5	10.5	0 1 9	27.8	26.6	4 1 5	24.8	23.2	9 1 2	2.7	6.4	5 2 11	18.3	22.3
4 0 2	45.2	45.8	0 1 10	11.9	-7.9	4 1 6	7.3	-5.6	9 1 3	2.6	-3.8	5 2 12	2.2	-1.5
4 0 4	47.6	43.7	0 1 11	19.6	16.5	4 1 7	17.2	14.4	9 1 4	2.3	3.1	5 2 13	8.6	12.7
4 0 6	25.6	23.7	0 1 12	3.4	-3.2	4 1 8	3.3	1.3	9 1 -1	5.2	4.4	6 2 1	8.5	-8.6
4 0 8	5.6	-1.4	0 1 13	23.0	19.6	4 1 9	8.3	7.7	9 1 -2	9.1	8.5	6 2 2	25.4	26.2
4 0 10	35.7	31.7	0 1 14	3.3	-5.7	4 1 10	11.9	11.0	9 1 -3	5.3	6.2	6 2 3	2.9	3.9
4 0 12	29.3	29.2	0 1 15	24.8	20.8	4 1 11	19.4	16.5	9 1 -4	12.7	14.5	6 2 4	24.3	25.3
4 0 14	8.2	9.5	0 1 16	2.3	-1.1	4 1 12	3.1	3.2	9 1 -5	2.9	2.0	6 2 5	3.0	-6.8
5 0 1	32.0	31.2	1 1 0	33.1	38.4	4 1 13	17.3	15.3	9 1 -6	13.8	13.3	6 2 6	3.0	-2.2
5 0 3	35.0	33.4	2 1 0	23.3	-18.7	4 1 -1	28.3	28.7	9 1 -7	2.7	-5.1	6 2 7	3.0	-3.9
5 0 5	30.5	30.4	3 1 0	1.8	3.4	4 1 -2	12.2	8.8	9 1 -8	13.5	17.0	6 2 8	11.4	9.6
5 0 7	3.3	4.1	4 1 0	2.2	6.5	4 1 -3	62.2	66.8	10 1 1	9.0	11.1	6 2 9	2.7	0.8
5 0 9	15.5	12.9	5 1 0	29.4	30.2	4 1 -4	13.7	-6.8	10 1 -1	14.6	16.2	6 2 10	15.1	12.4
5 0 11	15.0	11.3	6 1 0	15.0	12.1	4 1 -5	54.4	52.1	10 1 -2	2.0	2.3	6 2 11	1.9	1.8
5 0 13	6.2	6.3	7 1 0	42.9	39.4	4 1 -6	12.9	-8.9	10 1 -3	11.2	12.1	7 2 1	11.7	10.3
6 0 2	25.1	24.9	8 1 0	21.3	-18.2	4 1 -7	25.7	26.5	10 1 -4	2.0	-4.3	7 2 2	5.3	-5.2
6 0 4	18.7	13.6	9 1 0	14.5	13.6	4 1 -8	20.0	19.0	10 1 -5	6.5	7.6	7 2 3	16.7	13.7
6 0 6	5.8	8.8	10 1 0	1.9	3.3	4 1 -9	43.6	45.7	0 2 1	39.0	34.8	7 2 4	7.4	6.9
6 0 8	23.1	19.9	1 1 1	41.5	33.4	4 1 -10	11.4	10.5	0 2 2	41.0	37.5	7 2 5	12.7	12.8
6 0 10	18.0	16.8	1 1 2	19.9	13.8	4 1 -11	37.8	36.8	0 2 3	46.7	45.8	7 2 6	5.0	4.5
6 0 12	12.5	14.6	1 1 3	9.6	8.6	4 1 -12	16.8	-14.1	0 2 4	60.0	64.5	7 2 7	16.2	-16.9
7 0 1	23.0	18.4	1 1 4	75.6	83.2	4 1 -13	20.3	20.6	0 2 5	8.6	-5.8	7 2 8	2.4	-2.2
7 0 3	27.4	27.3	1 1 5	18.1	13.2	5 1 1	4.7	-4.2	0 2 6	27.1	37.7	7 2 9	10.7	11.8
7 0 5	5.8	-7.2	1 1 6	36.2	37.2	5 1 2	27.3	27.0	0 2 7	14.1	-12.0	8 2 1	3.0	-4.6
7 0 7	7.8	4.7	1 1 7	26.0	23.1	5 1 3	5.0	3.4	0 2 8	29.5	27.5	8 2 2	16.9	17.3
7 0 9	15.4	13.8	1 1 8	30.2	28.7	5 1 4	31.3	32.6	0 2 9	6.4	-5.5	8 2 3	2.8	-1.7
8 0 2	14.1	11.0	1 1 9	8.8	6.2	5 1 5	5.4	5.6	0 2 10	21.2	18.7	8 2 4	2.7	2.3
8 0 4	17.5	16.6	1 1 10	16.4	14.7	5 1 6	16.0	13.0	0 2 11	3.0	0.2	8 2 5	8.7	9.1
8 0 6	19.2	16.9	1 1 11	3.3	-0.8	5 1 7	3.3	2.6	0 2 12	3.0	-3.0	8 2 6	10.4	6.8
8 0 8	11.0	9.4	1 1 12	16.8	13.6	5 1 8	15.7	11.4	0 2 13	7.4	8.3	8 2 7	1.9	1.8
9 0 1	11.2	9.7	1 1 13	3.4	-2.0	5 1 9	3.4	-3.0	0 2 14	18.5	13.7	8 2 8	8.6	10.5
9 0 3	6.4	7.0	1 1 14	19.3	15.6	5 1 10	19.2	15.4	0 2 15	2.5	0.2	9 2 1	4.2	5.1
9 0 5	12.4	11.2	1 1 15	2.8	-2.9	5 1 11	10.5	-9.4	0 2 16	13.9	14.1	9 2 2	6.8	-7.3
10 0 2	1.4	-6.5	1 1 16	6.3	8.5	5 1 12	9.0	8.5	1 2 0	97.8	-103.8	9 2 3	2.1	2.4
1 0 -1	43.8	55.2	1 1 -1	17.7	17.2	5 1 13	6.9	-5.4	2 2 0	45.1	46.8	9 2 4	6.5	-6.0
1 0 -3	15.7	-14.6	1 1 -2	46.2	55.6	5 1 -1	6.4	-5.8	3 2 0	17.1	11.7	9 2 5	7.8	10.5
1 0 -5	49.2	57.0	1 1 -3	14.8	-10.2	5 1 -2	24.2	24.3	4 2 0	28.3	27.3	1 2 -1	112.6	119.8
1 0 -7	52.2	49.6	1 1 -4	7.6	-2.8	5 1 -3	16.7	-13.0	5 2 0	7.4	4.2	1 2 -2	59.8	-51.1
1 0 -9	39.9	36.5	1 1 -5	27.0	19.0	5 1 -4	42.5	45.7	6 2 0	6.8	6.6	1 2 -3	43.7	43.5
1 0-11	21.3	17.0	1 1 -6	14.9	15.5	5 1 -5	4.9	-5.2	7 2 0	15.7	-16.8	1 2 -4	12.1	10.7
1 0-13	11.7	-9.8	1 1 -7	30.8	24.0	5 1 -6	29.7	29.6	8 2 0	12.8	10.9	1 2 -5	28.7	29.5
1 0-15	16.6	12.8	1 1 -8	22.6	20.3	5 1 -7	3.1	-0.6	9 2 0	6.1	7.6	1 2 -6	5.9	3.9
1 0-17	11.9	13.3	1 1 -9	6.9	3.2	5 1 -8	21.5	22.7	1 2 1	58.5	53.5	1 2 -7	39.5	40.4
2 0 -2	54.7	60.8	1 1 -10	41.6	38.1	5 1 -9	22.3	-20.5	1 2 2	23.1	-20.4	1 2 -8	17.4	-10.4
2 0 -4	27.8	32.0	1 1 -11	3.3	7.1	5 1 -10	27.7	29.9	1 2 3	15.4	16.6	1 2 -9	30.3	31.4
2 0 -6	45.8	43.5	1 1 -12	22.0	23.2	5 1 -11	22.3	-18.6	1 2 4	4.1	3.6	1 2 -10	4.8	-5.8
2 0 -8	31.3	30.7	1 1 -13	3.4	0.5	5 1 -12	13.2	13.0	1 2 5	45.4	41.0	1 2 -11	25.0	22.8
2 0-10	24.4	21.9	1 1 -14	22.6	19.8	5 1 -13	3.2	1.7	1 2 6	2.1	0.5	1 2 -12	3.0	2.5
2 0-12	25.8	21.5	1 1 -15	5.2	-6.0	5 1 -14	14.8	14.6	1 2 7	54.9	57.4	1 2 -13	13.9	14.8
2 0-14	20.7	18.3	1 1 -16	20.0	18.7	6 1 1	25.8	21.7	1 2 8	14.2	12.7	1 2 -14	2.9	5.2
2 0-16	19.4	14.8	2 1 1	8.4	-2.7	6 1 2	3.2	-3.2	1 2 9	41.9	40.2	1 2 -15	17.5	14.9
3 0 -1	33.3	35.1	2 1 2	43.8	-38.5	6 1 3	29.8	31.3	1 2 10	2.9	0.2	1 2 -16	2.0	0.4
3 0 -3	72.0	79.0	2 1 3	49.3	50.7	6 1 4	3.3	3.4	1 2 11	3.0	7.2	2 2 -1	2.5	-0.3
3 0 -5	2.1	4.6	2 1 4	25.6	-20.1	6 1 5	22.8	24.4	1 2 12	3.0	-4.5	2 2 -2	63.4	61.5
3 0 -7	21.4	20.3	2 1 5	55.3	56.7	6 1 6	3.4	7.3	1 2 13	16.4	13.3	2 2 -3	27.1	24.4
3 0 -9	42.5	41.4	2 1 6	2.4	2.5	6 1 7	3.4	15.2	1 2 14	2.7	2.3	2 2 -4	7.8	10.6
3 0-11	32.0	30.7	2 1 7	43.7	42.2	6 1 8	3.4	4.5	1 2 15	12.5	12.1	2 2 -5	18.3	-15.3
3 0-13	20.2	16.5	2 1 8	13.1	-13.0	6 1 9	17.5	13.5	2 2 1	26.7	-23.5	2 2 -6	30.8	30.7
3 0-15	7.2	6.7	2 1 9	5.3	4.6	6 1 10	2.9	4.3	2 2 2	67.4	70.9	2 2 -7	12.2	-7.8
4 0 -2	2.1	3.8	2 1 10	12.7	-10.9	6 1 -1	38.7	39.7	2 2 3	1.7	3.3	2 2 -8	38.7	38.8
4 0 -4	5.6	13.7	2 1 11	14.5	10.9	6 1 -2	22.9	21.2	2 2 4	46.9	48.5	2 2 -9	4.5	3.5
4 0 -6	27.5	24.9	2 1 12	3.4	2.6	6 1 -3	26.0	25.2	2 2 5	2.0	1.4	2 2 -10	31.9	32.4
4 0 -8	31.6	31.1	2 1 13	19.3	15.4	6 1 -4	3.1	-2.1	2 2 6	26.2	23.3	2 2 -11	3.0	2.9
4 0-10	13.7	-10.8	2 1 14	3.0	-2.6	6 1 -5	21.3	22.3	2 2 7	11.1	7.5	2 2 -12	17.5	15.2
4 0-12	5.9	-6.0	2 1 -1	55.3	61.3	6 1 -6	13.8	-8.6	2 2 8	36.4	35.4	2 2 -13	12.9	9.4
4 0-14	10.9	9.2	2 1 -2	15.5	10.0	6 1 -7	18.2	17.3	2 2 9	30.0	27.4	2 2 -14	2.9	-0.2
4 0-16	2.1	-3.1	2 1 -3	44.9	51.8	6 1 -8	3.3	-0.2	2 2 10	28.1	30.2	2 2 -15	2.5	2.2
5 0 -1	17.6	17.7	2 1 -4	13.7	15.3	6 1 -9	10.3	10.4	2 2 11	27.9	24.0	2 2 -16	11.5	11.1
5 0 -3	33.2	29.0	2 1 -5	13.3	17.5	6 1 -10	3.4	4.0	2 2 12	29.3	27.2	3 2 -1	8.0	9.1
5 0 -5	65.5	66.6	2 1 -6	2.2	3.3	6 1 -11	8.3	10.7	2 2 13	2.9	-0.9	3 2 -2	22.0	-15.1
5 0 -7	17.2	15.2	2 1 -7	38.8	39.3	6 1 -12	3.2	6.4	2 2 14	12.3	12.0	3 2 -3	26.6	30.7
5 0 -9	12.5	-9.3	2 1 -8	2.6	1.6	6 1 -13	11.1	11.1	2 2 15	2.0	-2.1	3 2 -4	18.9	-11.7
5 0-11	3.4	3.3	2 1 -9	27.1	24.1	7 1 1	22.0	18.6	3 2 1	28.7	26.7	3 2 -5	25.0	23.3
5 0-13	15.8	14.0	2 1 -10	5.3	-4.8	7 1 2	29.9	30.5	3 2 2	17.3	14.3	3 2 -6	3.8	1.9
5 0-15	2.4	4.9	2 1 -11	32.1	32.4	7 1 3	3.4	-4.0	3 2 3	40.9	34.0	3 2 -7	35.4	35.8

Table 2 (*cont.*)

h k l	F_o	F_c	h k l	F_o	F_c	h k l	F_o	F_c	h k l	F_o	F_c	h k l	F_o	F_c	h k l	F_o	F_c
3 2 -8	2.5	-4.4	9 3 0	19.9	25.5	1 3 -6	43.6	43.2	0 4 7	14.3	9.5	8 4 3	2.5	-5.1			
3 2 -9	11.6	9.1	1 3 1	31.9	-25.3	1 3 -7	6.4	4.7	0 4 8	32.7	34.4	8 4 4	2.3	1.4			
3 2 -10	19.3	-17.9	1 3 2	2.3	2.8	1 3 -8	15.2	13.9	0 4 9	15.5	14.4	8 4 5	2.0	-1.5			
3 2 -11	5.2	4.6	1 3 3	1.5	2.7	1 3 -9	11.6	8.4	0 4 10	25.3	24.0	8 4 6	5.9	9.7			
3 2 -12	9.1	-8.0	1 3 4	8.7	8.8	1 3 -10	23.2	19.5	0 4 11	3.2	-2.9	9 4 1	10.3	14.8			
3 2 -13	14.8	13.8	1 3 5	30.1	25.0	1 3 -11	4.8	4.6	0 4 12	9.4	9.3	9 4 2	1.5	-1.9			
3 2 -14	2.8	-4.7	1 3 6	17.8	20.2	1 3 -12	12.0	10.1	0 4 13	7.1	-7.1	1 4 -1	53.1	53.1			
3 2 -15	2.4	6.2	1 3 7	19.0	13.6	1 3 -13	2.8	-2.4	0 4 14	16.1	15.9	1 4 -2	11.8	-3.7			
3 2 -16	12.2	-8.6	1 3 8	31.3	28.5	1 3 -14	10.8	9.2	0 4 15	1.9	1.0	1 4 -3	42.7	44.2			
4 2 -1	10.8	-7.1	1 3 9	14.3	-15.7	1 3 -15	2.1	1.2	1 4 0	9.3	8.1	1 4 -4	31.6	-28.8			
4 2 -2	28.0	28.9	1 3 10	4.8	5.3	1 3 -16	12.5	13.8	2 4 0	52.5	53.2	1 4 -5	27.8	27.7			
4 2 -3	5.2	0.2	1 3 11	8.5	-9.8	2 3 -1	39.7	37.4	3 4 0	20.6	-17.8	1 4 -6	2.4	-2.4			
4 2 -4	62.5	64.5	1 3 12	15.5	15.2	2 3 -2	26.0	19.0	4 4 0	23.2	21.7	1 4 -7	24.3	21.3			
4 2 -5	24.7	21.1	1 3 13	2.7	-5.0	2 3 -3	56.0	59.6	5 4 0	23.3	22.2	1 4 -8	9.7	10.1			
4 2 -6	14.6	15.5	1 3 14	19.6	18.3	2 3 -4	26.9	22.7	6 4 0	15.2	14.9	1 4 -9	26.3	23.9			
4 2 -7	4.5	-3.1	1 3 15	8.6	-8.9	2 3 -5	17.2	17.9	7 4 0	3.1	0.5	1 4 -10	5.4	5.5			
4 2 -8	6.7	6.3	2 3 1	10.1	10.6	2 3 -6	18.0	16.0	8 4 0	17.5	19.0	1 4 -11	17.3	16.5			
4 2 -9	19.2	-20.6	2 3 2	7.3	-0.6	2 3 -7	10.1	11.3	9 4 0	1.9	1.7	1 4 -12	5.4	6.1			
4 2 -10	19.3	17.9	2 3 3	32.3	31.3	2 3 -8	18.5	16.2	1 4 1	10.6	8.8	1 4 -13	2.9	5.8			
4 2 -11	10.5	-8.5	2 3 4	34.4	-29.5	2 3 -9	26.2	26.7	1 4 2	18.6	14.3	1 4 -14	2.6	-2.6			
4 2 -12	20.4	19.1	2 3 5	2.0	4.7	2 3 -10	14.0	12.9	1 4 3	37.5	36.8	1 4 -15	10.4	11.0			
4 2 -13	3.0	-0.3	2 3 6	3.2	1.5	2 3 -11	34.0	35.2	1 4 4	2.1	1.5	2 4 -1	66.6	-62.9			
4 2 -14	8.0	9.1	2 3 7	22.7	21.2	2 3 -12	2.8	6.3	1 4 5	40.1	38.5	2 4 -2	67.7	68.9			
5 2 -1	30.1	29.7	2 3 8	7.7	7.5	2 3 -13	10.8	11.0	1 4 6	7.5	-5.2	2 4 -3	36.3	-30.1			
5 2 -2	4.2	4.8	2 3 9	14.1	12.6	2 3 -14	13.9	-9.7	1 4 7	25.6	25.3	2 4 -4	18.8	18.8			
5 2 -3	12.8	12.3	2 3 10	8.9	-5.6	2 3 -15	11.7	10.7	1 4 8	7.1	4.1	2 4 -5	6.9	7.1			
5 2 -4	16.4	13.6	2 3 11	0.8	4.9	2 3 -16	6.5	-6.8	1 4 9	32.5	34.3	2 4 -6	24.7	25.6			
5 2 -5	28.9	27.4	2 3 12	2.8	-5.7	3 3 -1	8.6	7.6	1 4 10	19.6	21.7	2 4 -7	2.7	-2.3			
5 2 -6	6.6	6.0	2 3 13	16.4	14.6	3 3 -2	54.5	54.8	1 4 11	21.2	21.5	2 4 -8	20.9	18.7			
5 2 -7	18.2	17.5	2 3 14	2.1	-0.1	3 3 -3	16.3	14.0	1 4 12	23.3	23.5	2 4 -9	20.7	-22.7			
5 2 -8	13.3	-10.2	2 3 15	9.5	11.3	3 3 -4	34.2	33.2	1 4 13	15.2	14.4	2 4 -10	7.6	8.2			
5 2 -9	16.4	15.4	3 3 1	1.8	0.4	3 3 -5	7.1	7.0	1 4 14	2.3	-0.7	2 4 -11	11.0	-11.9			
5 2 -10	10.5	9.8	3 3 2	51.8	49.4	3 3 -6	18.6	18.5	1 4 15	8.5	9.9	2 4 -12	12.1	11.9			
5 2 -11	3.0	4.5	3 3 3	16.2	12.5	3 3 -7	14.0	12.1	2 4 1	12.6	-9.2	2 4 -13	2.9	-1.7			
5 2 -12	19.4	17.3	3 3 4	36.8	39.0	3 3 -8	46.6	46.9	2 4 2	30.4	26.1	2 4 -14	2.5	3.3			
5 2 -13	12.6	9.2	3 3 5	11.1	-6.0	3 3 -9	21.4	20.1	2 4 3	12.3	9.5	2 4 -15	4.9	-5.6			
6 2 -1	19.2	18.1	3 3 6	31.1	29.5	3 3 -10	31.7	30.7	2 4 4	35.9	32.7	3 4 -1	27.0	26.7			
6 2 -2	2.8	1.9	3 3 7	8.9	-8.6	3 3 -11	2.8	-3.3	2 4 5	2.5	0.1	3 4 -2	2.2	1.4			
6 2 -3	8.4	7.5	3 3 8	16.2	14.8	3 3 -12	17.7	19.9	2 4 6	39.3	36.4	3 4 -3	44.4	45.9			
6 2 -4	18.3	18.6	3 3 9	2.8	-1.4	3 3 -13	10.6	-8.2	2 4 7	4.9	-5.5	3 4 -4	28.5	26.9			
6 2 -5	14.9	-13.6	3 3 10	17.7	15.1	3 3 -14	12.7	13.7	2 4 8	17.3	16.8	3 4 -5	25.7	25.7			
6 2 -6	27.8	29.1	3 3 11	2.8	4.9	4 3 -1	3.6	7.2	2 4 9	9.3	10.4	3 4 -6	11.2	-8.7			
6 2 -7	12.7	-10.3	3 3 12	16.8	16.7	4 3 -2	2.1	-3.0	2 4 10	5.5	6.7	3 4 -7	12.8	12.4			
6 2 -8	3.0	3.3	3 3 13	2.2	3.8	4 3 -3	37.6	36.0	2 4 11	10.8	6.0	3 4 -8	21.7	-19.7			
6 2 -9	7.5	7.8	3 3 14	9.1	10.3	4 3 -4	2.2	-1.5	2 4 12	23.7	22.7	3 4 -9	11.9	11.7			
6 2 -10	3.0	5.4	4 3 1	15.2	15.4	4 3 -5	34.4	32.5	2 4 13	9.8	9.1	3 4 -10	5.4	-7.0			
6 2 -11	14.5	12.1	4 3 2	7.6	4.1	4 3 -6	2.4	2.6	2 4 14	10.8	13.0	3 4 -11	18.1	18.5			
6 2 -12	13.4	10.1	4 3 3	29.5	29.5	4 3 -7	17.5	18.8	3 4 1	35.6	36.1	3 4 -12	3.1	1.5			
7 2 -1	18.8	20.6	4 3 4	2.4	-3.1	4 3 -8	6.5	-5.9	3 4 2	12.7	-11.1	3 4 -13	2.8	5.8			
7 2 -2	3.0	-4.5	4 3 5	39.3	37.4	4 3 -9	15.0	13.6	3 4 3	35.8	34.5	4 4 -1	9.8	7.6			
7 2 -3	12.8	13.3	4 3 6	2.6	2.7	4 3 -10	9.7	5.8	3 4 4	4.5	1.3	4 4 -2	19.6	18.1			
7 2 -4	3.0	-2.0	4 3 7	2.7	5.7	4 3 -11	4.9	6.5	3 4 5	27.6	25.8	4 4 -3	2.6	0.9			
7 2 -5	9.1	9.6	4 3 8	13.8	10.8	4 3 -12	2.8	-2.7	3 4 6	5.0	5.5	4 4 -4	35.0	34.7			
7 2 -6	3.0	-7.0	4 3 9	4.9	4.7	4 3 -13	19.1	18.5	3 4 7	19.5	17.1	4 4 -5	2.7	3.7			
7 2 -7	21.8	23.3	4 3 10	2.8	-0.8	5 3 -1	22.7	22.8	3 4 8	3.1	0.2	4 4 -6	17.2	16.2			
7 2 -8	3.0	3.6	4 3 11	14.9	12.6	5 3 -2	29.1	30.3	3 4 9	3.2	-0.2	4 4 -7	7.3	-7.4			
7 2 -9	10.3	10.0	5 3 1	15.3	14.3	5 3 -3	2.4	-1.8	3 4 10	3.1	2.4	4 4 -8	15.0	16.2			
7 2 -10	2.8	3.8	5 3 2	9.7	8.5	5 3 -4	31.8	32.4	3 4 11	13.7	11.1	4 4 -9	5.4	0.1			
7 2 -11	10.0	11.3	5 3 3	15.5	14.8	5 3 -5	22.4	-18.3	3 4 12	2.6	-2.7	4 4 -10	18.2	18.7			
8 2 -1	11.7	12.0	5 3 4	22.6	21.2	5 3 -6	26.8	28.0	3 4 13	11.1	11.8	4 4 -11	3.1	3.9			
8 2 -2	5.2	4.1	5 3 5	6.7	8.6	5 3 -7	10.5	-5.9	4 4 1	13.4	12.5	4 4 -12	17.8	17.8			
8 2 -3	14.8	16.4	5 3 6	19.4	22.7	5 3 -8	12.7	11.9	4 4 2	26.0	23.4	5 4 -1	2.9	4.3			
8 2 -4	7.4	7.5	5 3 7	4.9	7.7	5 3 -9	2.8	0.9	4 4 3	2.8	3.2	5 4 -2	14.0	-11.7			
8 2 -5	3.0	5.1	5 3 8	10.9	9.7	5 3 -10	18.4	18.1	4 4 4	25.3	23.8	5 4 -3	16.6	16.9			
8 2 -6	12.5	11.2	5 3 9	12.6	10.1	5 3 -11	4.9	-5.3	4 4 5	14.6	-14.5	5 4 -4	7.2	-10.3			
8 2 -7	2.8	4.5	5 3 10	13.1	10.8	5 3 -12	21.6	25.7	4 4 6	30.1	31.5	5 4 -5	19.4	21.5			
8 2 -8	11.5	12.1	5 3 11	2.2	-0.5	6 3 -1	29.7	30.5	4 4 7	16.3	-13.9	5 4 -6	3.1	-1.2			
9 2 -1	8.8	6.9	6 3 1	41.1	39.7	6 3 -2	8.0	-5.0	4 4 8	15.5	12.0	5 4 -7	3.1	1.3			
9 2 -2	6.3	8.5	6 3 2	15.8	15.8	6 3 -3	12.3	12.5	4 4 9	7.7	7.3	5 4 -8	3.1	-1.4			
9 2 -3	9.9	11.1	6 3 3	26.5	27.6	6 3 -4	15.6	-14.6	4 4 10	13.6	9.0	5 4 -9	17.3	18.2			
9 2 -4	2.5	3.8	6 3 4	2.8	0.6	6 3 -5	30.5	33.7	4 4 11	2.6	4.8	5 4 -10	3.1	-2.1			
9 2 -5	5.0	8.0	6 3 5	9.8	8.6	6 3 -6	9.7	7.7	4 4 12	13.6	13.7	5 4 -11	20.7	21.6			
9 2 -6	2.4	4.9	6 3 6	2.3	5.2	6 3 -7	26.8	30.2	5 4 1	16.8	14.8	6 4 -1	3.1	-3.4			
9 2 -7	6.6	7.3	6 3 7	15.2	14.4	6 3 -8	2.8	5.8	5 4 2	8.9	-8.0	6 4 -2	15.2	15.3			
9 2 -8	2.0	0.2	6 3 8	4.5	8.2	6 3 -9	24.0	22.2	5 4 3	27.7	24.8	6 4 -3	3.1	1.4			
9 2 -9	8.5	11.5	6 3 9	19.0	20.0	6 3 -10	10.8	-10.1	5 4 4	7.6	6.9	6 4 -4	3.1	2.3			
10 2 -1	1.5	3.4	6 3 10	2.0	-1.2	6 3 -11	23.0	27.5	5 4 5	19.6	19.2	6 4 -5	3.1	1.4			
10 2 -2	8.9	11.8	6 3 11	9.1	12.0	7 3 -1	9.8	-5.6	5 4 6	17.3	-17.5	6 4 -6	16.4	14.6			
10 2 -3	1.6	3.0	7 3 1	4.9	-2.7	7 3 -2	11.0	8.9	5 4 7	15.4	15.2	6 4 -7	3.2	4.6			
10 2 -4	10.7	16.9	7 3 2	23.0	21.6	7 3 -3	2.8	1.3	5 4 8	18.5	-20.0	6 4 -8	17.2	17.7			
0 3 1	53.0	57.4	7 3 3	2.8	-2.5	7 3 -4	14.7	15.4	5 4 9	25.1	26.3	6 4 -9	3.1	-1.2			
0 3 2	11.7	9.7	7 3 4	12.8	12.4	7 3 -5	2.8	2.5	5 4 10	6.2	-8.0	6 4 -10	7.1	10.1			
0 3 3	16.5	17.3	7 3 5	2.7	-0.6	7 3 -6	24.0	26.6	5 4 11	12.3	13.0	7 4 -1	13.1	18.6			
0 3 4	23.5	20.3	7 3 6	9.8	7.9	7 3 -7	2.8	6.0	6 4 1	16.2	-17.0	7 4 -2	3.2	4.8			
0 3 5	52.8	49.7	7 3 7	2.3	3.0	7 3 -8	18.0	17.8	6 4 2	13.3	10.9	7 4 -3	15.4	15.1			
0 3 6	12.2	8.3	7 3 8	12.7	11.1	8 3 -1	15.8	17.7	6 4 3	7.7	-7.4	7 4 -4	3.1	1.7			
0 3 7	37.4	38.8	8 3 1	14.5	14.5	8 3 -2	4.8	-7.8	6 4 4	21.2	19.9	7 4 -5	17.2	17.6			
0 3 8	2.4	5.1	8 3 2	2.6	-3.8	8 3 -3	25.2	25.8	6 4 5	5.4	5.2	7 4 -6	3.1	-5.1			
0 3 9	37.1	34.9	8 3 3	10.5	11.7	8 3 -4	2.7	-2.9	6 4 6	10.7	10.9	7 4 -7	18.9	22.9			
0 3 10	8.1	-9.5	8 3 4	12.1	10.9	8 3 -5	12.2	13.0	6 4 7	5.0	7.0	7 4 -8	2.9	3.1			
0 3 11	17.5	19.8	8 3 5	4.2	9.4	8 3 -6	2.6	3.7	6 4 8	14.4	14.8	8 4 -1	2.8	-2.5			
0 3 12	2.8	-7.0	8 3 6	1.9	-0.2	8 3 -7	7.4	8.8	6 4 9	2.3	-6.4	8 4 -2	16.4	19.7			
0 3 13	15.2	12.6	8 3 7	13.4	17.5	9 3 -1	2.1	-7.3	6 4 10	10.9	14.8	8 4 -3	2.8	1.7			
0 3 14	2.5	-0.9	9 3 1	2.0	0.7	9 3 -2	16.9	22.5	7 4 1	16.3	15.4	8 4 -4	6.9	9.5			
0 3 15	11.8	12.7	9 3 2	11.7	14.2	9 3 -3	2.3	5.2	7 4 2	3.1	-3.0	8 4 -5	2.7	3.3			
1 3 0	39.4	42.3	9 3 3	1.7	4.7	9 3 -4	2.1	5.8	7 4 3	15.1	14.3	8 4 -6	10.2	11.6			
2 3 0	21.9	18.4	9 3 4	7.4	11.9	0 4 1	19.5	-16.0	7 4 4	2.9	-3.3	8 4 -7	2.5	-2.7			
3 3 0	26.6	28.5	1 3 -1	36.7	23.1	0 4 2	13.7	13.5	7 4 5	2.3	3.6	8 4 -8	9.6	14.3			
4 3 0	18.1	15.9	1 3 -2	29.8	29.3	0 4 3	9.5	-8.3	7 4 6	2.5	2.6	9 4 -1	6.1	7.9			
5 3 0	36.4	38.1	1 3 -3	24.7	22.3	0 4 4	28.8	27.9	7 4 7	11.7	10.3	9 4 -2	9.4	13.4			
6 3 0	16.8	15.8	1 3 -4	27.6	25.6	0 4 5	2.2	-2.2	8 4 1	2.7	-0.9	9 4 -3	3.5	4.6			
7 3 0	8.5	7.7	1 3 -5	16.2	-11.9	0 4 6	31.4	32.7	8 4 2	2.6	5.0	9 4 -4	9.1	14.1			
8 3 0	2.7	2.1															

tion, spot shape, and α_1–α_2 splitting) were taken into account. No correction was made for absorption as the crystal was very much smaller than the optimum size ($\mu = 17{\cdot}5$ cm^{-1}).

Structure determination

As there are only 2 molecules in the unit cell, and as reflexions for which $h+k+l=2n$ are much more intense than those for which $h+k+l=2n+1$ (especially noticeable for the compounds of Pd and Pt, which have a much higher scattering ability than Ni), the Ni must lie on a special position, which for space group $P2_1/n$ must be a centre of symmetry. The Ni could be placed at 000 and at $\frac{1}{2}\frac{1}{2}\frac{1}{2}$. Also, since there are only 2 dipyridinium ions, the ions must have a centre of symmetry which must be situated between the two rings. From the coordinates found, the atoms in the rings lie approximately in the same plane.

The positions of all the atoms could be determined from a 3-dimensional Patterson synthesis. The coordinates thus determined were used for a 3-dimensional Fourier synthesis which gave well defined peaks. The value of $R=\Sigma\,|F_o-F_c|/\Sigma\,F_o=0{\cdot}261$ at this stage. The structure was refined by two cycles of least squares. In the first cycle only the scale factors were varied, bringing R to 0·18. In the second cycle the scale factors, the positional and the thermal parameters were varied, bringing R to 0·125 . The values of F_o and F_c are given in Table 2, and the final coordinates of the atoms in Table 3.

Discussion

The dimensions of the $Ni(CN)_4^{2-}$ and the dipyridinium ions are given in Figs. 1 and 2 and in Tables 4 and 5. The bond lengths in the dipyridinium ion rings [C–C = 1·40 Å (average) and C–N = 1·35 Å (average)] are in good agreement with the bond lengths in pyridine (Palmer, 1967). The distance between the two rings [C(5)–C(5') = 1·47 Å] agrees well with the results for similar compounds [*e.g.* in diphenyl the distance between the two rings is 1.48 Å (Pauling, 1960)]. The two rings are approximately planar as in benzene, but are rather distorted hexagons as in pyridine (Palmer, 1967).

Table 4. *Interatomic distances,*

Ni —C(1)	1·87 ± 0·01 Å
Ni —C(2)	1·89 ± 0·01
C(1)–N(1)	1·16 ± 0·02
C(2)–N(2)	1·14 ± 0·03
N(3)–C(8)	1·51 ± 0·03
N(3)–C(3)	1·34 ± 0·02
C(3)–C(4)	1·42 ± 0·03
C(4)–C(5)	1·39 ± 0·03
C(5)–C(6)	1·40 ± 0·02
C(6)–C(7)	1·39 ± 0·04
C(7)–N(3)	1·35 ± 0·03
N(2)–N(3)	3·65 ± 0·03
C(5)–C(5')	1·47 ± 0·04
N(3)–C(5)	2·79 ± 0·03
N(3)–N(3')	7·05 ± 0·03

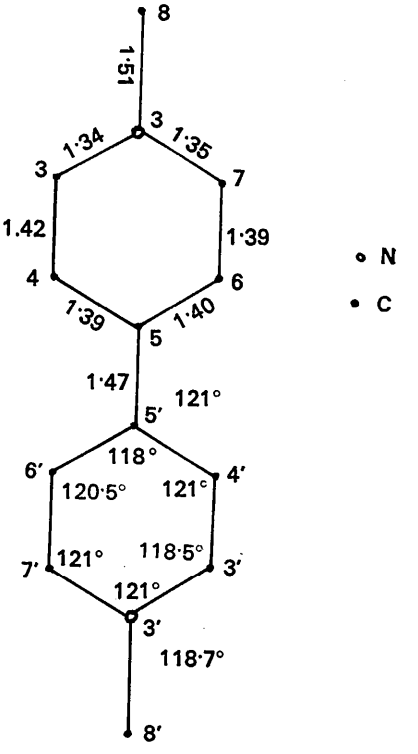

Fig. 2. Bond lengths and angles in the dipyridinium ion.

Table 3. *Atomic parameters with their standard deviations*

	x/a	*y/b*	*z/c*	*σ(x/a)*	*σ(y/b)*	*σ(z/c)*	*B* (Å²)	*σB* (Å²)
Ni	0·0000	0·0000	0·0000				2·15	0·05
C(1)	0·1261	0·0154	0·1285	0·0012	0·0017	0·0007	2·14	0·19
C(2)	−0·1767	−0·1055	0·0580	0·0012	0·0017	0·0007	1·83	0·19
C(3)	0·1540	0·1765	0·5033	0·0012	0·0017	0·0007	1·62	0·19
C(4)	0·2865	0·0974	0·4610	0·0012	0·0017	0·0007	1·71	0·19
C(5)	0·4335	0·0454	0·5214	0·0012	0·0017	0·0007	0·95	0·19
C(6)	0·4465	0·0785	0·6252	0·0012	0·0017	0·0007	1·57	0·19
C(7)	0·3122	0·1513	0·6647	0·0012	0·0017	0·0007	1·96	0·19
C(8)	0·0270	0·2744	0·6490	0·0012	0·0017	0·0007	2·22	0·19
N(1)	0·2025	0·0239	0·2092	0·0010	0·0014	0·0006	3·02	0·17
N(2)	−0·2822	−0·1678	0·0927	0·0010	0·0014	0·0006	2·50	0·17
N(3)	0·1694	0·1944	0·6041	0·0010	0·0013	0·0006	1·54	0·16

Table 5. *Bond angles*

N(3)–C(3)–C(4)	118·5° ± 0·9°
N(3)–C(7)–C(6)	121·0 ± 3·5
C(3)–C(4)–C(5)	121·0 ± 3·2
C(5)–C(6)–C(7)	120·5 ± 1·3
C(4)–C(5)–C(6)	118·0 ± 2·7
C(3)–N(3)–C(7)	121·0 ± 1·4
C(8)–N(3)–C(3)	118·7 ± 0·7
C(4)–C(5)–C(5′)	121·0 ± 2·5
C(1)–Ni –C(2)	89·7 ± 0·4
C(1)–Ni –C(2′)	90·4 ± 0·4

The Ni–C bond length [= 1·88 Å (average)] is in good agreement with the Ni–C distance in $K_2Ni(CN)_4$ and similar complexes (Vannerberg, 1964). The C–N distance in the cyanide group [= 1·15 Å (average)] is the same as in many other complex cyanides, *e.g.* in $K_3Co(CN)_6$, C–N = 1·157 Å (Curry & Runciman, 1959), and also in $K_4Mo(CN)_8.2H_2O$, C–N = 1·15 Å (Hoard & Nordsieck, 1939). The atoms in the $Ni(CN)_4^{2-}$ ion also lie at the corners of a square as can be expected since a dsp^2-hybridization is found for all of Ni, Pd, and Pt in their complexes.

The *N-N* distance in the dipyridinium ion is 7·05 Å, which is thus approximately the distance between the two positive charges. Recent determinations of the structure of the pyridinium ion in solution by means of nuclear magnetic resonance spectroscopy showed that the whole positive charge is not situated on the N atom only, but is distributed over the ring in such a manner that the main portion of the charge does appear at the N atom (Smith & Schneider, 1961). The same can be expected for the dipyridinium ion in the solid state. The present results are not inconsistent with this expectation, as the negative nitrogen of the cyanide group tends to a position in the ring but comes closest to the N atom.

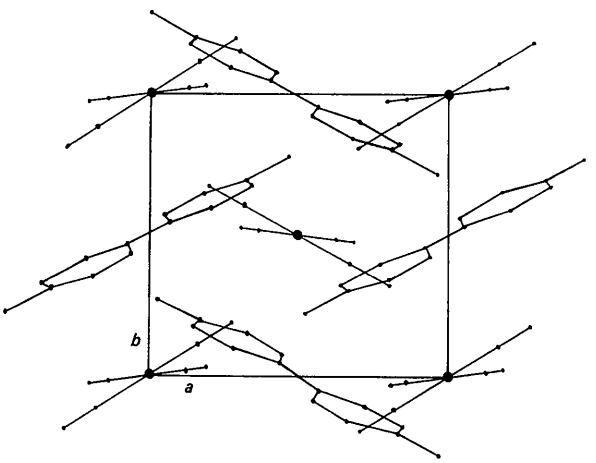

Fig. 3. The structure viewed along the *c* axis.

Fig. 3 is a sketch of the structure viewed along the *c* axis. The dipyridinium ion lies in a plane which is practically the (120) plane. The two methyl groups bound to the N atom are approximately in the same plane. This explains the particularly strong 120 reflexion. The centre of the ion lies on a symmetry centre at $\frac{1}{2}0\frac{1}{2}$. Further, the $Ni(CN)_4^{2-}$ ion lies directly under the dipyridinium ion with two of the cyanide groups in a straight line with the dipyridinium ion, so that the structure as a whole is a layer structure, with the layers being formed alternately of cations and anions.

All calculations were carried out on an IBM 360 computer.

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References

CURRY, N. A. & RUNCIMAN, W. A. (1959). *Acta Cryst.* **12**, 674.

HOARD, J. L. & NORDSIECK, H. H. (1939). *J. Amer. Chem. Soc.* **61**, 2853.

PALMER, M. H. (1967). *The Structure and Reactions of Heterocyclic Compounds*, p. 12. London: Edward Arnold.

PAULING, L. (1960). *The Nature of the Chemical Bond*, 3rd ed. p. 291. New York: Cornell Univ. Press.

SMITH, I. C. & SCHNEIDER, W. G. (1961). *Canadian J. Chem.* **39**, 1158.

VANNERBERG, N.-G. (1964). *Acta Chem. Scand.* **18**, 2385.