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The Crystal Structure of an *N*-Methylphenothiazine–7,7,8,8-Tetracyanoquinodimethane Complex, *N*-MePZT–TCNQ

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Crystals of the 1 : 1 charge-transfer complex of *N*-MePZT-TCNQ are monoclinic with those lattice constants: a=10.904, b=13.321, c=7.086 Å, and β=91.06°, and with the space group C2/m. X-ray structure analysis showed that the mixed-stacks of *N*-MePZT and TCNQ exist along the c axis. *N*-MePZT is disordered, and its apparent molecular symmetry is C_{2h}, taking four possible conformations. The molecular geometry of TCNQ indicates that the amount of the charge transferred from *N*-MePZT is very small ($<$0.2e).

7,7,8,8-tetracyano-*p*-quinodimethane (TCNQ) is a powerful electron acceptor and forms a number of charge-transfer complexes. *N*-methylphenothiazine (*N*-MePZT) is an electron donor. A neutral *N*-MePZT molecule has a folded structure,[1] while the PZT skeleton is planar in crystals of trinitrobenzene-PZT.[2] As was pointed out by Fritchie,[2] the van der Waals packing requirement of an alternating donor-acceptor stack might suggest that PZT is essentially planar in a 1 : 1 complex. Crystals of *N*-MePZT–TCNQ were subjected to X-ray structure analysis in order to examine the molecular structure of the component molecules.

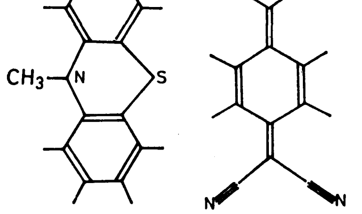

Fig. 1. The molecular structures of *N*-MePZT and TCNQ.

Experimental

Black crystals of *N*-MePZT-TCNQ were obtained by the slow evaporation of an equimolar acetone solution of the components. Oscillation and Weissenberg photographs showed the crystal to be monoclinic. The systematic absence for hkl with $h+k=2n+1$ indicated that the space group is C2/m, Cm, or C2. The unit-cell dimensions, as determined by means of a diffractometer with Mo$K\alpha$ radiation, are a=10.904±0.002, b=13.321±0.002, c=7.086±0.001 Å, and β=91.06±0.04°. The density measured by the flotation method was 1.32 g/cm^3, whereas the calculated density on the assumption of $Z=2$ is 1.347 g/cm^3. The intensity data were collected on a Rigaku automated four-circle diffractometer by employing a ω–2θ scan technique. A crystal with dimensions of 0.25×0.13×0.18 mm was used. Of the 1274 accessible reflections with $2\theta \leqq 55°$, significant counts were recorded for 576 reflections. Measurements of two reference reflections, (2,0,0) and (0,0,2), were repeated every fifty reflections.

Structure and Determination

The trial structure was easily deduced from packing considerations. The space group C2/m was assumed; this was verified at a latter stage. Refinements were performed by means of Fourier synthesis, followed by the block-diagonal least-squares method. Three hydrogen atoms (H1, H2, and H3 in Table 1) were found on the three-dimensional-difference Fourier maps and were refined isotropically. No hydrogen atoms of the methyl group came out. Because of the requirements of the space group, the orientation of *N*-MePZT must be disordered and the apparent molecular symmetry is C_{2h}, which seems to be consistent with the fact that diffuse scattering appears around strong Bragg reflections. Possible models of the disordered structure of *N*-MePZT were easily deduced. The model shown in Fig. 2 is the most plausible one because the difference Fourier synthesis shows nitrogen and sulfur atoms

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[1] N. Wakayama, This Bulletin, **44**, 2847 (1971).

[2] C. J. Fritchie, *J. Chem. Soc.*, *A*, **1969**, 1328.

Table 1. Final atomic parameters (all × 10^4) and their estimated standard deviations

Temperature factor $=\exp[-(B_{11}h^2+B_{22}k^2+B_{33}l^3+2B_{12}hk+2B_{13}kl+2B_{23}kl)]$

	x	y	z	B_{11}	B_{22}	B_{33}	B_{12}	B_{13}	B_{23}
			N-MePZT						
S	−290	1482	487	135	112	331	2	−59	7
	8	5	12	13	5	29	5	10	9
N1	0	963	0	276	181	422	0	−185	0
		6		10	4	21		9	
C1	1152	524	502	190	95	167	17	−53	10
	5	3	6	6	4	10	4	6	6
C2	2225	1026	983	199	96	212	−22	3	34
	5	4	6	7	4	12	5	7	7
C3	3289	521	1435	147	142	327	−31	81	22
	4	4	7	7	6	14	5	8	7
C4	−171	1808	449	109	154	183	−10	−74	157
	37	15	32	36	18	90	25	45	33
H1	2115	1658	921	9.1[a]					
	43	40	66	1.5					
H2	3964	865	1264	8.2					
	40	36	62	1.5					
			TCNQ						
N2	3686	1645	6407	138	75	398	−13	−12	−4
	3	3	5	5	3	12	3	6	6
C5	604	941	5220	93	43	281	2	22	−2
	3	3	6	4	2	11	3	6	5
C6	1252	0	5449	68	49	210	0	24	0
	5		8	6	4	15		7	
C7	2483	0	5933	115	32	236	0	21	0
	5		8	8	4	17		9	
C8	3166	900	6207	108	52	254	12	0	4
	4	3	6	5	3	11	3	6	5
H3	1033	1603	5335	5.1					
	33	33	54	1.1					

a) Isotropic temperature factor *B*.

separately. The nitrogen atom lies on the twofold axis, while the sulfur atom is in general position. Anisotropic refinement gave an *R*-value of 0.096. Other models were also considered, but they did not give good *R*-values. The final positional and thermal parameters are given in Table 1. The observed and calculated structure factors are compared in Table 2.

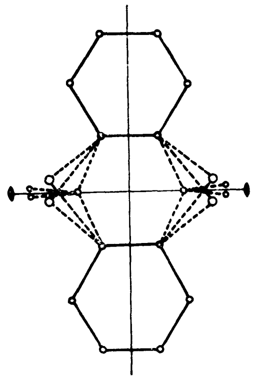

Fig. 2. The model of the disordered structure of *N*-MePZT.

Results and Discussion

The structure is shown in Fig. 3. It consists of mixed stacking columns of alternating donor (*N*-MePZT) and acceptor (TCNQ) molecules. The mode of overlapping is shown in Fig. 4. The average intermolecular distance is 3.44 Å; the least-squares planes of both the molecules are given in Table 3.

The crystal structure analyses of neutral phenothiazine (PZT) and *N*-MePZT have revealed that these molecules have a non-planar structure folded along the N–S direction. A PZT molecule in a crystal of a PZT-3,5-dinitrobenzene complex has a folded structure with a dihedral angle of 156°,[3] and it is planar in a PZT-TNB (trinitrobenzene) complex.[2] Fritchie suggested that a PZT molecule in PZT-TNB is actively folding at room temperature, taking two possible conformations with dihedral angles of about 165—172°.

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[3] C. J. Fritchie and B. L. Trus, *Chem. Commun.*, **1968**, 833.

Table 2. Comparison of observed and calculated structure factors (× 10)

H	K	FO	FC
L=0			
11	1	40	44
11	3	18	18
11	5	29	28
11	9	24	20
10	0	27	25
9	1	39	48
9	3	41	40
8	4	64	67
8	6	126	126
8	8	23	24
8	10	26	23
7	1	48	50
7	3	66	66
7	5	45	45
7	11	19	19
7	15	27	26
6	0	48	54
6	2	28	30
6	4	35	38
6	8	45	43
6	10	28	34
5	1	116	111
5	3	208	201
5	5	28	35
5	7	87	91
5	11	27	24
4	0	128	127
4	2	59	58
4	4	69	69
4	6	65	59
4	8	66	69
4	10	42	53
3	1	321	315
3	3	255	248
3	5	111	103
3	7	45	34
3	9	59	60
3	11	35	36
3	13	32	22
2	0	143	150
2	2	223	216
2	4	93	91
2	10	55	55
0	2	559	585
0	4	373	375
0	6	52	41
0	8	30	22
L=1			
-14	2	27	3
-11	5	20	10
-10	0	41	44
-10	2	28	12
-10	8	20	5
-9	3	21	3
-9	5	63	69
-9	7	50	54
-8	2	19	19
-8	4	67	73
-8	6	97	96
-8	8	42	31
-7	1	29	33
-7	3	29	41
-7	5	26	24
-7	9	37	30
-6	0	47	38
-6	2	73	74
-6	4	25	29
-6	6	16	17
-6	8	47	43
-6	10	38	40
-5	1	82	79
-5	3	180	172
-5	5	98	91
-5	7	21	24
-5	9	55	65
-5	11	62	71
-5	13	23	16
-4	0	73	74
-4	2	101	95
-4	4	45	43
-4	6	117	113
-4	10	35	27
-3	1	103	101
-3	3	241	233
-3	5	169	166
-3	7	66	66
-3	11	45	43
-3	15	45	42
-2	0	196	192
-2	2	156	144
-2	4	61	69
-2	6	173	178
-2	10	39	29
-2	12	28	41
-2	14	24	20
-1	1	115	106
-1	3	127	120
-1	5	38	45
-1	7	42	31
-1	9	107	107
-1	11	106	107
0	0	83	65
0	4	37	55
0	6	82	70
0	8	83	77
0	10	141	137
0	12	99	109
1	1	110	98
1	3	335	321
1	5	112	108
1	7	102	107
1	9	82	86
1	13	41	34
2	0	84	94
2	2	70	74
2	4	211	198
2	6	125	122
2	8	49	45
2	10	33	25
2	14	55	39
3	3	95	94
3	5	112	110
3	7	118	117
3	9	25	17
3	11	38	38
3	13	36	0
3	15	29	14
4	0	291	284
4	2	176	170
4	4	278	266
4	6	45	51
4	8	37	49
4	10	42	53
4	12	56	53
4	14	22	11
5	1	104	104
5	3	26	18
5	5	61	54
5	9	61	72
5	11	30	34
6	0	111	112
6	4	15	11
6	6	78	83
6	10	33	28
7	1	37	33
7	3	84	93
7	5	76	79
7	7	59	66
7	9	27	13
8	0	61	64
8	2	24	25
8	4	72	72
8	6	101	106
8	8	23	32
9	1	60	57
9	3	29	32
9	9	28	18
10	2	20	15
10	8	17	14
11	1	22	19
11	9	22	11
12	6	26	17
L=2			
-12	0	36	38
-12	4	19	8
-11	1	36	31
-10	2	26	35
-10	6	20	15
-9	3	32	31
-9	5	83	83
-9	7	55	54
-8	2	78	76
-8	4	69	72
-8	6	43	41
-8	8	23	13
-7	1	14	1
-7	3	25	27
-7	7	34	36
-7	9	29	23
-6	0	58	56
-6	2	114	110
-6	4	49	47
-6	6	52	60
-6	10	25	24
-6	14	19	13
-5	1	136	131
-5	3	21	6
-5	5	30	38
-5	7	16	10
-5	9	53	56
-4	0	187	185
-4	2	32	34
-4	4	92	88
-4	6	73	67
-4	8	43	47
-4	14	20	6
-4	16	21	21
-3	1	69	65
-3	3	135	140
-3	5	27	7
-3	9	60	55
-3	11	19	18
-2	0	30	25
-2	2	50	44
-2	6	60	52
-2	8	30	38
-2	16	21	1
-1	1	983	1000
-1	3	76	77
-1	5	157	150
-1	7	20	19
-1	11	89	101
-1	13	64	47
0	0	577	588
0	2	220	217
0	4	92	96
0	6	49	37
0	8	22	26
0	10	40	50
0	12	51	40
0	14	27	7
1	1	195	201
1	3	63	63
1	7	18	24
1	9	19	18
1	11	43	43
2	0	417	412
2	2	82	79
2	4	232	229
2	8	61	60
2	10	41	30
2	12	33	36
2	16	36	33
3	3	103	101
3	5	112	111
3	9	65	71
3	11	18	31
3	15	21	1
4	0	82	74
4	2	186	182
4	4	110	103
4	6	78	91
4	8	57	59
4	10	22	14
5	1	42	46
5	3	137	146
5	7	15	16
5	9	67	63
6	0	28	31
6	2	15	10
6	4	25	30
6	8	16	6
7	1	24	30
7	5	104	112
7	7	96	88
7	13	25	15
8	0	98	106
8	2	31	35
8	6	40	43
9	1	21	13
9	5	21	14
9	7	22	12
10	0	22	26
10	2	49	46
10	4	20	2
10	6	39	41
10	10	36	30
11	5	21	6
L=3			
-10	0	18	18
-9	1	27	20
-9	5	63	73
-9	7	57	54
-8	6	16	10
-8	8	24	20
-7	1	20	23
-7	3	50	52
-7	5	17	11
-7	9	20	23
-6	0	34	22
-6	4	51	53
-6	6	41	37
-6	8	32	39
-6	10	64	66
-6	12	36	30
-5	1	20	13
-5	3	55	61
-5	5	23	8
-5	13	21	3
-4	0	135	137
-4	2	53	54
-4	4	156	155
-4	6	59	60
-4	10	34	26
-3	1	26	14
-3	5	76	74
-3	7	55	57
-3	13	22	23
-3	15	23	14
-2	0	104	108
-2	2	21	20
-2	4	69	65
-2	8	46	45
-2	10	71	66
-2	12	27	30
-1	1	29	23
-1	3	47	48
-1	5	53	52
-1	7	65	68
-1	9	84	78
-1	11	114	115
-1	13	51	57
-1	15	24	7
0	0	73	76
0	2	109	109
0	4	74	69
0	6	88	88
0	8	26	37
0	10	68	55
1	3	53	52
1	5	101	100
1	7	66	61
1	11	18	6
1	13	27	25
1	15	23	26
2	0	59	53
2	2	28	24
2	4	55	51
2	6	105	103
2	8	50	50
2	10	37	30
3	1	71	71
3	3	270	268
3	5	42	38
3	7	81	85
3	9	22	20
3	11	34	42
3	13	33	14
4	0	131	130
4	2	22	24
4	4	80	79
4	6	36	31
4	8	36	45
4	10	46	57
4	12	33	30
4	14	25	10
5	1	67	60
5	3	19	29
5	5	40	49
5	7	50	52
5	13	23	16
5	15	26	11
6	0	103	107
6	2	42	46
6	4	85	89
6	8	36	25
7	1	74	76
7	3	43	44
7	5	112	111
7	7	68	68
8	0	104	107
8	4	45	48
9	1	35	35
9	5	23	7
10	0	35	32
10	2	17	13
11	3	28	15
L=4			
-12	0	34	27
-10	4	35	37
-10	6	31	38
-9	3	24	31
-9	5	51	49
-9	7	35	30
-8	0	24	15
-8	4	26	22
-7	1	44	40
-7	3	30	33
-6	0	110	108
-6	2	44	41
-6	4	20	4
-6	6	20	25
-5	1	21	29
-5	3	22	21
-5	5	33	46
-5	7	47	47
-5	11	26	19
-4	0	16	13
-4	2	42	39
-4	4	37	33
-4	8	32	32
-3	1	69	66
-3	3	22	23
-3	5	24	24
-3	7	16	6
-3	11	21	16
-2	0	410	397
-2	2	187	183
-2	4	58	60
-2	6	59	57
-2	10	45	48
-2	12	43	44
-2	14	27	0
-1	1	291	280
-1	3	33	24
-1	5	52	51
-1	9	24	28
-1	11	51	54
-1	13	24	26
0	0	143	141
0	2	25	26
0	4	25	31
0	8	17	9
0	14	30	1
1	1	53	55
1	3	113	114
1	5	34	29
1	9	45	47
2	0	75	69
2	2	28	22
2	4	108	105
2	6	38	37
2	8	25	31
2	10	35	42
3	1	82	83
3	3	91	88
3	7	61	60
3	9	31	33
3	13	25	3
4	0	52	56
4	2	80	75
4	4	69	73
4	6	43	49
4	8	17	20
4	10	45	39
5	3	15	6
5	9	24	21
6	0	50	47
6	4	22	25
6	6	79	83
6	10	34	31
7	1	84	87
7	5	44	43
7	7	40	41
8	0	51	53
8	4	20	16
8	6	22	7
8	10	19	7
9	3	47	39
11	1	26	7
L=5			
-10	4	36	27
-10	6	42	39
-9	3	19	15
-9	9	19	7
-8	2	27	21
-8	8	24	8
-7	9	40	40
-6	2	16	6
-6	4	27	21
-6	12	27	15
-5	1	40	31
-5	3	63	66
-5	5	57	54
-4	0	48	50
-4	4	53	49
-4	6	38	38
-3	1	20	25
-3	3	20	19
-3	9	24	24
-2	0	46	45
-2	6	43	47
-2	8	41	40
-2	10	58	61
-2	12	54	57
-1	1	37	36
-1	3	22	21
-1	9	33	43
-1	11	23	24
0	4	35	36
0	6	44	45
0	12	20	17
1	1	36	44
1	3	47	44
1	5	49	51
1	7	49	49
1	11	24	21
2	2	72	67
2	4	78	79
2	6	39	42
2	8	40	37
2	12	21	16
3	1	44	44
3	3	47	44
3	5	40	51
3	9	33	36
3	11	31	40
4	0	35	29
4	2	24	16
4	4	17	10
4	6	32	32
4	8	21	12
5	1	32	33
5	3	55	56
5	9	31	28
6	0	89	88
6	4	69	73
6	6	67	65
7	1	61	62
7	3	28	33
7	5	32	31
8	0	48	52
8	2	25	2
9	1	33	25
L=6			
-9	5	18	5
-8	0	26	14
-7	1	32	44
-6	6	20	29
-4	0	58	58
-4	6	19	11
-4	10	18	7
-3	1	98	100
-3	5	33	37
-2	0	148	149
-2	2	84	89
-2	12	28	35
-1	1	30	34
-1	3	37	37
0	2	21	19
1	3	37	37
1	5	43	48
2	0	55	58
2	2	29	33
2	4	23	29
2	8	26	37
3	1	21	25
3	3	44	44
3	11	23	7
4	4	18	11
5	1	19	15
5	5	20	23
6	0	76	71
6	2	28	24
6	6	40	41
10	0	25	10
L=7			
-7	3	23	5
-6	4	32	26
-6	6	18	13
-4	0	19	10
-3	7	28	21
-2	8	20	16
-1	5	19	15
0	0	36	42
0	4	35	32
0	8	20	21
2	4	29	33
3	3	18	19
5	1	29	35
5	5	27	44
5	7	28	22
6	0	38	45
7	1	25	20
L=8			
-4	0	29	28
-3	1	49	58
-2	2	26	13
-1	1	19	22
-1	3	17	4
0	0	20	2
1	1	20	23
2	0	30	32
2	2	25	16
5	1	32	27
6	0	28	27
L=9			
-2	0	21	13

Table 3. Least-squares planes

	Direction cosines with respect to *a*	*b*	*c**
N-MePZT	−0.2727	−0.0074	0.9621
TCNQ	−0.2386	−0.0046	0.9711

Deviation (Å)								
N-MePZT	C1	−0.004	C2	0.002	C3	0.001	C4[a]	0.341
	S[a]	0.405	N1[a]	−0.010				
TCNQ	C5	−0.011	C6	−0.016	C7	−0.001	C8	0.005
	N2	0.004						

a) The atoms C4, S, and N1 were not used for the the calculation of least-squares plane.

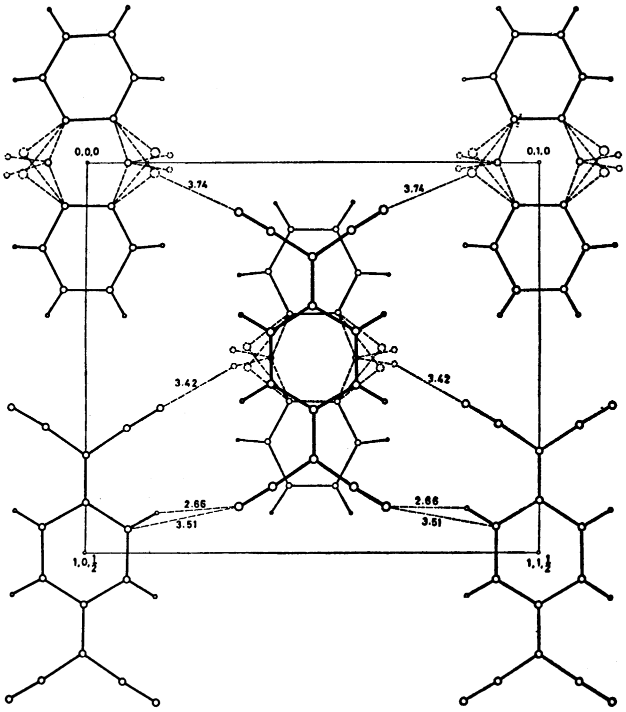

Fig. 3. The molecular arrangement and the short contacts in the plane (102), viewed along the *c* axis.

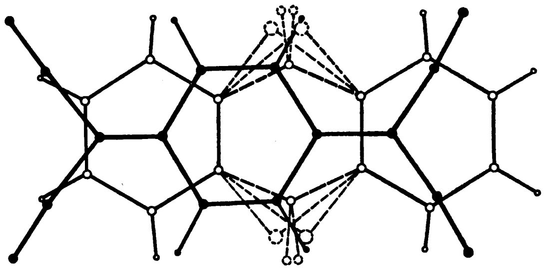

Fig. 4. The molecular overlap as viewed in the direction perpendicular to the mean molecular planes.

In view of the fact that the molecular symmetry of *N*-MePZT is C_{2h} and that the sulfur atom deviates about 0.4 Å from the molecular plane (Table 3), it may be reasonable to conclude that *N*-MePZT is disordered, taking four conformations with dihedral angles of about 160—170 °C with equal probability as is shown in Fig. 5. In addition, the nitrogen atom is on the twofold axis, that is, on the molecular plane; this is consistent with the small deviation of the nitrogen atom from the molecular plane in neutral *N*-MePZT and PZT[1,4] (0.07, 0.03 Å).

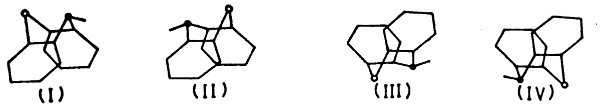

Fig. 5. The accessible structures and orientations of *N*-MePZT.

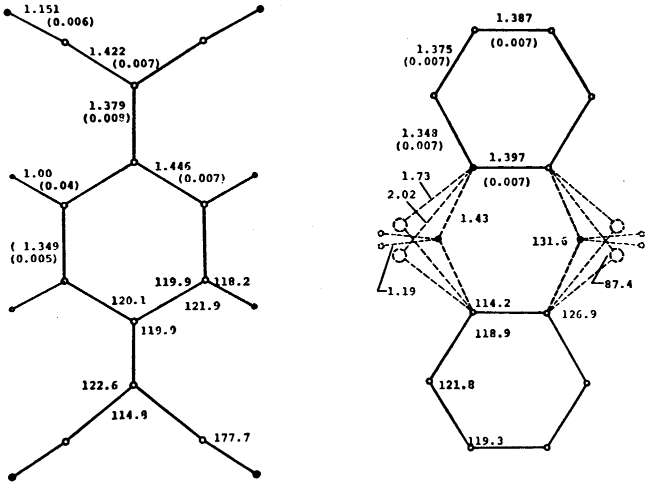

Fig. 6. Bond lengths(Å) and angles(°).
The estimated standard deviations of bond lengths are shown in parentheses. Those of bond angles are about 0.50° for all the angles spanned by non-hydrogen atoms except carbon atom of methyl group and about 4.0° for the angles involving the hydrogen atoms and carbon atom of methyl group.

The bond lengths and angles are shown in Fig. 6. The two C–S bonds are 1.73 and 2.02 Å, giving a mean value of 1.87 Å (Fig. 6), and N–C is 1.43 Å. These values are consistent with those of neutral *N*-MePZT, (1.82, 1.43 Å). The C–C bonds parallel to the twofold axis of the molecule are longer than the others, and the N–C (methyl) bond is very short. These facts are undoubtedly due to the disorder of *N*-MePZT.

The molecular structure of TCNQ is normal. The

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[4] J. D. Bell, J. F. Blount, O. V. Briscoe, and H. C. Freeman, *Chem. Commun.* **1968**, 1656.

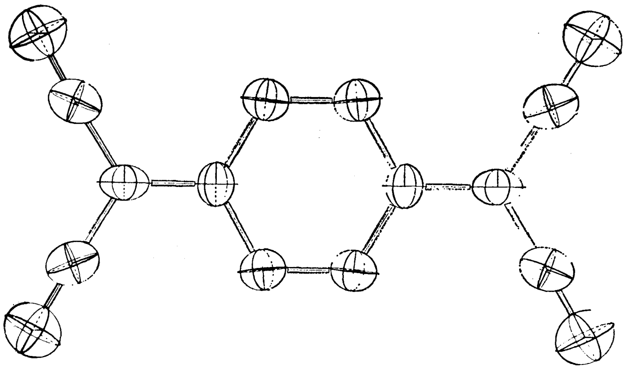
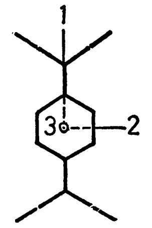


Fig. 7. *ORTEP* plot of the thermal motion ellipsoid of TCNQ.

Table 4. Rigid-body thermal parametres of TCNQ

Principal axes in the form: **La**+**Mb**+**Nc**

axis	L	M	N
1	0.0894	0.0	0.0340
2	0.0	−0.0751	0.0
3	−0.0204	0.0	0.1370



Molecular vibrational tensors

$$\mathbf{T}\times 10^4 \begin{pmatrix} 624 & 0 & 48 \\ & 352 & 0 \\ & & 508 \end{pmatrix} \text{Å}^2 \qquad \boldsymbol{\omega}\times 10 \begin{pmatrix} 323 & 0 & 9 \\ & 9 & 0 \\ & & 41 \end{pmatrix} \text{deg}^2$$

Principal axes of the **T** and $\boldsymbol{\omega}$ tensors relative to the molecular axes.

R.m.s. amplitude	Direction cosines		
0.19 Å	0.0	1.0	0.0
0.22	0.335	0.0	−0.942
0.25	0.942	0.0	0.336
0.9°	0.0	1.0	0.0
2.0	0.030	0.0	0.999
5.7	0.999	0.0	0.030

bond lengths shown in Fig. 6 are uncorrected for thermal libration. Some rigid-body parameters for TCNQ are given in Table 4 and Fig. 7. The translational motion nearly isotropic while the libration is quite anisotropic. The r.m.s. amplitude of 5.7° about the long axis is the largest.

Recent studies of TCNQ anion radical salts have revealed a dependence of the molecular geometry on its formal charge ($TCNQ^0$, $TCNQ^{-1/2}$, $TCNQ^-$).[5,6] In a charge-transfer complex, TCNQ may be charged to some extent according to the magnitude of the charge-transfer interaction. If the bond lengths obtained are sufficiently accurate, they will show values intermediate between those of $TCNQ^0$ and $TCNQ^-$. The bond lengths of TCNQ in the charge-transfer complexes so far reported are summarized in Fig. 8. The standard bond lengths of $TCNQ^0$, $TCNQ^{-1/2}$, and $TCNQ^-$ are taken mainly from Refs. 5 and 6. This figure can be regarded as a diagram which relates bond length to the amount of transferred charge, δ ($TCNQ^{-\delta}$). The "a" bond lengths of $TCNQ^0$ (neutral molecule), $TCNQ^{-1/2}$, and $TCNQ^-$ are 1.346, 1.354, and 1.358 Å respectively. The "a" bond elongates with an increase in the charge transferred from a donor to TCNQ. Thus, the bond lengths seem to be a rough measure of the amount of charge-transfer. For example, the "a" bond seems to show the formal charge, δ, of TCNQ ($TCNQ^{-\delta}$) of about 0.25 in an *N*-MePZT-TCNQ complex. The dots corresponding to the observed bond length are scattered, probably because of the inaccuracy of the experiments. Despite the inaccuracy, the diagram seems to indicate that the average bond lengths of TCNQ in *N*-MePZT, anthracene-TCNQ,[7] and hexamethylbenzene-TCNQ[8] correspond to those of molecules with a small formal charge ($\delta<0.2$). In the case of *N*-MePZT-TCNQ, the amount of transferred electrons seems to be about 0.15e. The ESR, IR, and electronic spectra indicate that an *N*-MePZT-TCNQ complex is not an ionic type but almost neutral,[9] and they support the above conclusion. In tetramethyl-*p*-phenylenediamine (TMPD)-TCNQ,[10] the distribution of the bond lengths differs from the distributions of those of the other nonionic complexes. The electron transferred from TMPD to TCNQ seems to be more than one, which is a rather unexpected situation.

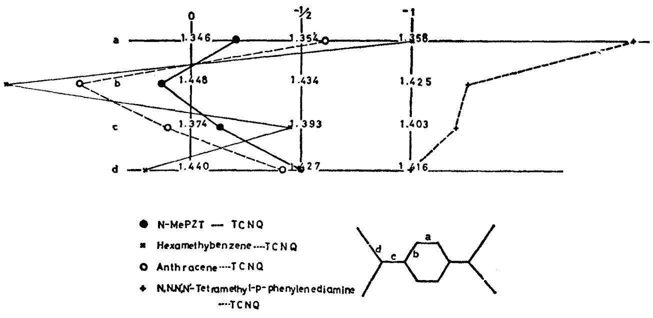


Fig. 8. The bond lengths of TCNQ.
The values are corrected for thermal vibration.

The crystal structure of *N*-MePZT-TCNQ is similar to that of the average structure of PZT-TCNQ.[11] The main difference between the two complexes is that the former has the orientational disorder of *N*-MePZT, while the latter is a sinusoidally modulated structure. In the crystal of PZT-TCNQ, PZT are held by hydrogen bonds of N–H⋯N type. On the other hand, in *N*-MePZT-TCNQ, *N*-MePZT are packed loosely by van der Waals forces. Thus, the orientation of *N*-MePZT is likely to be easily disordered.

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6) P. Goldstein, K. Seff, and K. N. Trueblood, *ibid.*, *B*24, 778 (1968).

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8) R. H. Colton and D. E. Henn, *J. Chem. Soc.*, *B*, **1970**, 1532.

9) N. S. Bhat and M. Kinoshita, private communication.

10) A. W. Hanson, *Acta Crystallogr.*, **19**, 610 (1965).

11) H. Kobayashi and Y. Saito, to be published.