Acta Chemica Scandinavica A 28 (1974) 175—184

The Crystal Structure of the Addition Compound *N*-Methylthiocaprolactam-Iodine (1:1)

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The crystal structure of the molecular complex formed by *N*-methylthiocaprolactam (MTCL) and iodine in the mol ratio 1:1 has been determined at 18°C, using X-ray diffraction techniques. The donor molecule is a cyclic thioamide. The crystalline adduct as crystallized from a benzene solution, is monoclinic, the space group being $P2_1/c$ with four molecules per unit cell. The I—S separation [2.688 Å(2)] is significantly shorter than the corresponding I—S distances observed in iodine complexes with thioethers, and the I—I bond length [2.880 Å(1)] somewhat larger. The I—I—S group is nearly linear. The C_1-S and C_1-N bond lengths [1.716(5) Å and 1.302(7) Å, respectively] are close to the corresponding distances observed in pure thioamides. The thioamide group and the methyl carbon atom form an approximately planar grouping with the iodine molecule. Inter-molecular contact distances are of the van der Waals type.

However, crystals grown from solutions containing carbon tetrachloride were found to contain equimolecular amounts of the present adduct and carbon tetrachloride. Crystal data for this compound are given.

The crystal structures of molecular complexes formed by iodine and thioethers have previously been reported.[1a-1d] The structures of related adducts such as thiourea-, merocyanine-, and triphenylphosphine sulfide-iodine are also known.[1a-1d] On the other hand, complexes formed by halogen and thioamides are probably usually too unstable to yield crystals suitable for single crystal work. However, previous studies had shown [2,3] that a relatively stable complex was formed between iodine and *N*-methylthiocaprolactam (MTCL) in the mole ratio 1:1, the acceptor molecule being most likely joined to sulphur. The nitrogen atom in thioamides is expected to be a relatively poor electron donor since the lone electron pair is probably considerably delocalized.

Attempts at obtaining single crystals of MTCL and iodine chloride or bromine, respectively, did not succeed. The latter compound was formed as an unstable, yellow precipitate in pentane.

EXPERIMENTAL

The solid complex was precipitated from a solution of MTCL in pentane by adding an equimolecular portion of iodine (reagent grade) dissolved in pentane.[2] The sample of MTCL was kindly supplied by mag.scient. E. Augdahl. The orange-coloured precipitate was subsequently filtered, washed and dried.

Reddish single crystals (m.p. 97 ± 1°C) in the form of somewhat elongated prisms were found to crystallize from solutions of the adduct in benzene.

When crystallized from solutions containing carbontetrachloride, needle-shaped, orange-coloured crystals of a different molecular complex containing equimolecular amounts of CCl_4 and the former components are formed. Exposed to air, the compound loses rapidly carbon tetrachloride, leaving the MTCL-I_2 adduct as a powder. The latter dissolves easily in polar solvents.

The crystals of MTCL-I_2 were sealed in thin-walled boron-lithium glass-capillaries before exposure to X-ray radiation. A single crystal of about $0.15 \times 0.20 \times 0.25$ mm³ was used in the intensity recording made on an automatic Picker four-circle diffractometer, using graphite monochromated Mo*K*-radiation. 2711 reflections with intensity $> 2.5\sigma(I)$ were used in the structure determination, while 555 reflections with intensity $< 2.5\sigma(I)$ were excluded from the calculations. Further experimental details are: Range: $2° < 2\theta < 60°$; scan: $[2\theta(\alpha_1) - 0.7°] - [2\theta(\alpha_2) + 0.7°]$; scan speed: 1°/min, background counts in 20 sec on either side of scan region; *b*-axis along ϕ-axis; experimental temperature

Table 1. Observed and calculated structure factors.

l	Fo	Fc
	h=0,k=0	
2	45.9	40.40
4	54.9	47.63
6	7.0	5.93
8	119.1	119.72
12	61.5	62.64
	h=0,k=1	
0	60.2	58.77
1	122.5	120.25
2	57.8	59.09
3	15.9	17.10
4	134.6	132.60
5	134.0	135.78
6	44.4	47.78
1	127.4	173.47
	h=0,k=2	
0	46.6	51.05
4	133.9	132.43
5	83.7	81.70
6	108.3	100.61
7	192.5	186.66
8	66.0	66.98
9	4.0	3.94
10	46.2	44.26
11	60.5	57.73
12	40.2	38.53
13	21.6	21.57
14	7.7	7.48
15	64.4	64.95
	h=0,k=3	
14	82.6	80.03
13	17.1	17.40
12	20.3	18.90
11	5.5	3.71
9	20.4	20.37
8	80.9	78.06
7	17.0	17.12
6	101.4	98.45
5	31.7	30.82
4	200.3	226.40
3	38.2	40.14
2	35.8	40.71
1	10.9	11.08
	h=0,k=4	
0	103.3	117.82
1	9.3	6.43
2	68.0	75.36
3	117.3	135.81
4	48.5	50.35
5	77.6	78.85
6	3.8	1.28
7	79.4	77.40
8	64.0	62.64
9	41.0	39.96
10	45.3	44.16
11	17.3	17.70
12	14.8	14.64
13	38.7	37.06
14	9.4	10.04
	h=0,k=5	
12	9.3	10.78
11	73.5	69.24
10	36.8	36.90
9	14.2	14.51
8	55.0	52.68
7	74.2	74.35
6	20.1	22.13
5	19.9	20.20
4	49.5	54.28
3	73.6	81.91
2	61.8	67.87
1	71.2	80.34
	h=0,k=6	
0	112.5	136.86
1	43.1	47.19
2	9.1	8.69
4	32.8	34.71
5	14.8	15.00
6	32.7	34.56
7	5.5	7.49
8	48.7	48.69
9	43.2	42.10
10	53.6	50.65
12	37.0	37.01
	h=0,k=7	
10	5.9	6.07
9	8.9	9.85
7	36.7	38.06
6	19.8	19.96
5	28.5	29.61
4	41.6	45.04
3	5.5	3.43
2	27.2	29.06
1	61.6	67.66
	h=0,k=8	
0	25.1	27.72
1	12.9	13.48
2	21.7	22.51
3	30.4	33.62
4	23.4	24.76
5	19.5	20.15
6	36.8	37.94
7	30.3	30.83
	h=0,k=9	
2	18.0	19.22
	h=1,k=9	
-2	11.0	11.38
0	26.5	28.53
1	25.2	25.65
3	29.5	29.83
	h=1,k=8	
6	19.5	19.61
5	18.8	20.18
4	8.3	9.42
3	29.9	31.60
2	45.2	49.08
1	40.0	44.64
0	15.5	15.33
-1	13.6	15.23
-2	11.3	11.05
-3	10.4	11.20
-4	23.8	24.38
-5	8.5	7.60
-6	19.6	20.86
-7	34.2	35.40
	h=1,k=6	
9	40.6	40.09
8	77.8	79.83
7	12.3	11.76
6	30.7	31.68
5	22.6	23.94
4	37.0	40.58
3	19.6	21.02
2	11.2	13.33
1	19.0	21.23
0	60.7	66.13
-1	53.7	59.26
-2	61.4	67.60
-3	28.7	30.84
-4	100.8	107.25
-5	6.5	5.12
-7	23.2	23.63
-8	12.7	12.81
-9	5.4	6.21
-10	59.5	58.59
-11	18.7	17.89
-12	20.6	19.38
	h=1,k=5	
-13	28.5	27.57
-12	45.9	45.02
-9	5.7	4.65
-8	31.7	31.98
-7	24.5	23.22
-6	56.4	55.04
-5	4.8	1.22
-3	116.0	127.96
-2	71.3	80.29
-1	70.6	81.83
0	68.9	79.56
1	49.6	56.79
3	22.5	24.45
4	39.3	43.22
5	64.2	70.27
6	30.9	31.84
7	83.5	83.73
8	16.5	16.97
9	15.7	15.69
10	44.5	42.87
11	24.9	24.53
12	16.5	15.00
	h=1,k=4	
	6.5	6.51
11	82.3	78.66
10	37.3	35.89
9	8.9	8.03
7	5.1	3.99
6	21.7	22.11
5	104.2	109.90
4	97.2	107.04
3	120.1	137.02
2	17.8	18.68
1	88.2	106.64
0	101.9	121.24
-1	54.3	60.66
-2	15.8	16.79
-3	14.2	15.81
-4	63.5	63.82
-5	63.2	62.56
-6	74.8	71.92
-7	154.7	150.34
-8	33.5	32.64
-10	33.5	33.22
-12	23.4	23.07
-13	22.8	22.78
-14	42.6	42.90
	h=1,k=3	
-15	13.9	13.12
-14	31.1	29.86
-13	10.9	11.10
-12	10.8	11.05
-11	21.6	21.22
-10	68.0	65.43
-9	33.4	33.45
-8	146.2	140.49
-7	30.1	28.44
-6	147.7	140.96
-5	36.7	36.75
-4	91.0	87.20
-3	17.7	18.23
-2	7.7	9.42
-1	13.8	13.78
0	6.2	7.64
3	67.7	73.42
5	55.0	52.86
6	14.7	13.99
7	8.5	9.42
12	81.2	79.49
13	26.7	26.69
14	37.4	36.13
	h=1,k=2	
15	42.0	42.47
14	31.1	30.31
12	24.4	23.53
11	113.9	112.39
10	31.2	31.30
9	14.7	14.82
8	55.5	54.56
7	5.2	4.46
6	16.5	15.10
5	80.9	79.19
4	105.0	101.95
3	147.5	157.06
2	91.0	97.43
1	219.0	286.59
0	143.2	178.10
-1	94.7	103.17
-2	38.8	39.99
-3	10.7	13.24
-4	99.3	90.68
-5	69.3	62.35
-6	10.8	9.23
-7	206.0	193.44
-8	30.5	28.70
-9	36.0	33.98
-10	68.4	65.00
-11	73.6	71.44
-12	5.3	4.79
-13	16.5	15.99
-14	44.9	44.82
-15	46.9	47.11
-16	19.5	18.85
	h=1,k=1	
-16	7.1	8.28
-15	42.2	43.87
-14	44.0	44.27
-13	39.6	39.74
-12	22.2	21.23
-11	115.9	113.83
-10	65.1	64.65
-9	26.4	26.17
-8	58.4	54.72
-7	23.7	25.19
-6	71.9	63.49
-5	67.1	65.35
-4	126.1	109.44
-2	67.8	62.68
-1	84.1	79.19
0	63.4	66.03
1	113.8	120.64
2	39.3	38.85
3	37.8	41.68
4	164.7	170.39
5	103.7	104.09
6	25.7	25.42
8	39.1	36.87
9	47.9	47.96
10	14.7	14.67
11	56.7	57.17
12	48.3	48.45
13	9.0	9.07
14	46.4	46.41
15	46.1	45.22
	h=1,k=0	
14	44.6	44.62
12	4.4	5.51
10	13.7	13.71
8	216.8	231.04
6	118.9	119.26
4	128.9	127.92
2	68.9	75.42
0	138.5	144.24
-2	155.5	138.39
-4	315.4	317.74
-6	85.8	76.73
-8	11.2	11.95
-10	130.5	130.52
-12	62.8	62.44
	h=2,k=0	
-12	32.4	33.11
-10	37.3	37.41
-8	60.6	61.39
-6	240.0	226.20
-4	186.1	187.61
-2	250.5	289.08
0	45.9	44.21
2	78.9	83.57
4	60.0	60.58
6	49.1	50.58
	h=2,k=1	
13	14.9	9.45
12	34.5	34.68
10	29.6	30.08
9	9.7	10.06
8	87.9	90.47
7	98.2	99.15
6	15.2	15.65
5	157.4	162.86
4	57.3	58.43
3	137.4	142.48
2	27.0	23.20
1	50.3	54.03
0	155.3	150.92
-1	35.8	30.52
-2	135.1	133.81
-3	262.7	255.44
-4	69.9	71.45
-5	18.9	19.01
-6	61.4	55.20
-7	207.5	190.91
-8	74.1	69.16
-10	86.5	82.16
-11	15.3	13.90
-12	4.4	3.22
-13	72.9	74.08
-14	24.2	25.09
-15	47.2	47.36
	h=2,k=2	
-16	16.2	13.50
-15	19.5	19.52
-14	32.4	31.93
-13	54.4	55.50
-12	28.4	23.53
-11	103.1	102.17
-10	23.7	23.43
-9	72.2	68.43
-8	65.4	61.58
-7	114.7	107.30
-6	107.5	99.19
-5	25.8	24.07
-4	66.8	64.85
-3	188.8	188.36
-2	70.2	68.91
-1	59.6	59.85
0	75.9	71.41
1	198.2	215.31
2	23.4	21.96
3	81.7	82.71
4	104.6	108.50
5	14.2	14.81
6	6.6	7.23
7	79.1	79.24
8	60.2	62.41
9	87.9	88.81
10	40.4	40.66
11	74.9	76.18
12	27.9	26.74
13	10.1	10.57
14	22.5	22.31
	h=2,k=3	
13	10.1	9.87
12	66.4	66.76
11	10.7	9.70
10	42.1	42.04
9	50.7	50.13
8	122.7	121.98
7	5.3	3.88
5	31.8	31.50
4	38.5	36.38
3	10.5	8.65
2	89.1	91.08
-8	58.4	54.72
-1	93.6	99.38
-2	108.8	113.68
-3	16.6	15.74
-4	43.0	41.98
-5	5.1	4.74
-6	45.9	43.66
-7	6.3	6.39
-8	39.8	37.91
-9	43.0	42.04
-10	161.2	153.92
-11	15.5	15.25
-12	38.0	36.96
-13	5.6	4.06
-14	15.1	14.11
-15	8.0	6.69
	h=2,k=4	
-14	27.9	28.22
-13	7.0	5.55
-12	31.0	31.05
-11	97.4	93.73
-10	6.9	4.80
-9	81.8	78.25
-8	19.3	18.14
-7	90.5	48.95
-6	95.3	93.39
-5	33.9	33.06
-4	81.6	82.32
-3	92.7	99.97
-2	10.4	9.27
-1	59.5	64.91
0	19.7	19.56
1	144.8	170.19
2	79.1	86.05
3	4.6	3.74
4	70.8	73.13
5	4.6	3.53
6	23.3	22.07
7	71.0	71.28
8	37.5	37.69
9	30.8	30.41
10	7.8	6.25
11	62.5	61.97
12	45.7	44.92
13	21.2	20.90
	h=2,k=5	
12	25.6	24.08
11	31.1	29.75
10	7.8	5.86
9	15.5	15.27
8	42.6	41.67
7	77.0	76.22
6	70.3	70.74
5	71.3	74.08
4	8.5	9.35
3	28.4	28.46
2	47.6	52.24
1	66.5	72.09
0	37.7	41.09
-1	8.9	6.08
-2	14.1	13.68
-3	113.1	121.64
-4	68.5	71.03
-5	9.4	8.32
-6	46.2	46.36
-7	95.8	92.29
-9	5.4	2.67
-10	28.1	26.05
-11	28.2	28.60
-12	13.2	12.40
-13	29.2	29.02
	h=2,k=6	
-12	15.8	15.69
-11	9.3	9.97
-10	1.7	1.80
-9	8.6	8.46
-8	29.7	27.92
-7	4.5	1.14
-6	76.1	77.99
-5	68.6	71.33
-4	52.2	54.94
-3	4.9	2.47
-2	34.6	34.52
0	12.2	12.86
1	8.6	9.65
2	5.4	.96
3	22.0	23.14
4	112.3	124.19
5	46.8	48.05
6	27.2	28.59
7	17.8	18.05
8	28.3	27.89
9	13.1	13.40
10	11.3	11.15
	h=2,k=7	
9	21.5	20.91
8	39.2	38.99
6	16.6	16.08
5	48.8	51.32
4	14.1	15.84
3	55.4	59.31
2	10.8	8.89
0	35.8	36.37
-1	36.2	36.81
-2	39.8	41.32
-3	48.1	50.34
-4	8.0	7.60
-5	8.8	8.81
-6	5.4	2.76
-7	54.6	54.01
-8	36.2	35.68
-9	18.0	17.89
-10	16.9	15.52
	h=2,k=8	
-8	32.6	30.53
-7	28.0	29.81
-6	14.4	14.39
-4	6.2	6.88
-3	39.2	40.69
-2	44.5	46.01
0	26.2	27.17
1	35.3	38.25
2	21.4	20.55
3	14.3	14.55
4	15.2	15.23
5	6.4	7.18
	h=2,k=9	
0	30.9	31.59
-1	31.0	30.56
-2	12.9	13.21
	h=3,k=0	
4	17.7	18.84
3	7.0	6.62
2	5.1	5.75
1	14.6	14.82
0	33.3	33.97
-1	5.5	4.91
-2	24.8	24.20
-3	39.5	40.31
-4	29.8	29.43
-5	18.9	19.15
-6	28.6	28.37
-7	12.3	12.12
	h=3,k=1	
-2	26.2	28.28
-1	34.6	35.37
0	33.3	35.04
1	53.8	55.86
2	26.9	27.35
3	12.2	12.51
-1	12.9	10.43
-2	35.3	33.30
-3	201.3	204.52
-4	3.7	1.19
-5	94.0	87.54
-6	70.9	66.52
-7	53.0	50.81
-8	22.0	21.43
-9	37.8	35.75
-10	102.8	97.07
-11	75.9	72.36
-12	56.8	56.49
-13	79.1	77.63
-14	25.4	25.78
-15	34.5	34.49
-16	9.6	10.80
	h=3,k=2	
-16	5.4	5.70
-15	11.6	10.35
-14	38.5	36.82
-13	27.1	26.89
-12	9.4	6.41
-11	65.8	63.88
-10	76.6	74.80
-9	91.0	86.63
-8	11.2	9.50
-7	182.2	173.68
-6	97.1	92.18
-5	65.7	61.92
-4	44.9	43.08
-3	11.7	9.57
-2	112.6	109.09
-1	5.8	4.68
0	119.3	116.45
1	236.7	243.41
2	43.3	40.74
3	57.9	57.59
4	102.4	103.71
5	105.5	107.35
6	55.3	55.80
8	60.7	57.44
9	36.5	36.97
10	24.5	25.06
11	81.0	83.53
12	31.8	34.52
13	33.8	34.74
	h=3,k=3	
12	93.7	97.38
10	102.5	104.90
8	38.3	40.01
6	4.2	3.25
4	172.4	182.05
2	175.8	180.46
0	194.5	201.15
-2	96.4	93.22
-4	12.2	11.04
-6	240.3	246.85
-8	78.7	77.45
-10	103.1	101.02
-12	26.4	25.90
-14	6.0	4.59
-16	53.1	54.57
	h=4,k=0	
-16	19.9	20.53
-14	41.7	41.52
-12	5.4	4.98
-10	199.0	196.08
-8	64.0	60.73
-6	4.5	4.16
-4	20.5	17.95
-2	146.2	150.62
0	139.3	142.40
2	116.1	118.11
4	34.8	33.87
6	36.8	37.57
8	125.0	130.60
10	37.4	38.56
12	43.0	46.25
	h=4,k=1	
12	48.5	50.03
11	43.0	45.30
10	8.1	9.42
9	86.7	88.85
8	47.8	50.15
7	42.6	42.73
6	13.4	13.50
5	25.3	23.96
4	61.3	62.64
3	11.7	10.96
2	103.4	101.93
1	192.8	197.71
0	13.2	13.62
-1	54.4	49.28
-2	73.7	70.84
-3	164.1	165.11
-4	84.4	82.33
-5	6.2	5.04
-6	103.7	100.90
-7	45.7	43.88
-8	22.5	21.63
-9	133.2	129.94
-10	26.9	26.85
-11	12.6	11.39
-12	32.0	31.66
-13	62.0	61.46
-14	50.9	52.28
-15	22.2	22.76
-16	29.2	30.70
	h=4,k=2	
-14	38.3	37.57
-13	57.7	56.97
-12	30.8	29.09
-11	7.6	9.27
-10	46.2	44.62
-9	37.1	35.62
-8	5.4	5.34
-7	119.1	112.55
-6	65.8	62.21
-5	122.0	118.45
-4	80.9	77.21
-3	133.9	129.36
-2	122.5	120.39
-1	91.4	90.83
0	91.4	90.83
1	64.4	62.22
2	15.6	13.47
3	6.6	6.46
5	143.8	145.71
6	7.2	6.34
7	35.6	34.93
8	61.4	61.87
10	4.5	3.23
11	15.0	15.53
12	30.5	31.20
	h=4,k=3	
10	8.2	7.21
9	14.4	14.48
8	26.6	27.48
7	20.9	22.25
6	78.7	77.13
5	26.8	25.75
4	141.8	142.96
3	37.5	35.79
2	71.9	72.83
1	15.3	11.99
0	23.8	24.04
-1	7.0	7.53
-2	8.4	7.90
-3	8.3	8.21
-4	31.3	29.92
-5	49.5	48.28
-6	194.3	192.63
-7	30.3	28.66
-8	36.2	34.65
-9	6.3	5.21
-10	17.1	15.09
-12	40.4	39.74
-14	66.1	65.37
-15	21.4	21.29
	h=4,k=4	
-14	19.0	16.43
-13	63.7	62.63
-12	34.5	33.59
-11	32.6	31.38
-10	41.8	40.44
-9	32.8	32.28
-8	36.0	34.40
-7	83.9	79.93
-6	36.0	35.12
-5	89.9	88.28
-4	12.9	14.26
-3	75.4	72.48
-2	106.3	105.24
-1	53.7	51.49
0	41.0	37.50
1	63.4	61.31
2	20.2	18.20
4	28.8	27.65
5	111.3	108.31
6	56.5	55.84
7	11.6	8.99
8	36.0	37.47
9	19.5	19.18
10	7.8	9.31
	h=4,k=5	
9	52.7	52.28
8	15.2	15.62
7	16.2	14.69
6	23.5	23.43
5	20.5	18.58
4	37.7	36.37
3	5.1	4.16
2	5.1	2.09
1	101.2	99.89
0	51.6	50.67
-1	45.5	44.77
-2	55.9	55.69
-3	94.9	94.32
-4	9.7	9.99
-6	49.2	47.99
	h=5,k=3	
-14	6.9	6.85
-13	8.6	8.12
-12	9.9	9.49
-11	27.0	25.27
-10	40.7	38.90
-9	73.6	69.07
-8	48.8	47.65
-7	36.5	35.57
-6	124.2	120.23
-5	4.2	5.35
-4	24.3	22.99
-3	29.8	28.57
-2	59.9	57.51
0	56.7	55.15
2	154.3	155.11
3	37.2	35.80
4	52.0	50.79
6	19.6	21.56
8	25.6	24.76
10	18.6	17.78
	h=5,k=2	
8	57.4	57.70
7	25.0	25.51
6	5.6	5.91
5	12.9	11.07
4	6.5	6.51
3	13.8	13.87
2	76.1	74.74
1	54.7	55.71
0	17.5	16.23
-1	24.0	22.29
-2	151.7	150.35
0	7.9	9.25
-1	62.6	62.01
-2	59.9	57.14
-3	15.7	14.11
-4	7.2	5.54
-5	74.4	71.47
-6	81.6	77.55
-7	72.6	71.66
-8	50.0	48.94
-9	129.0	125.80
-10	36.4	36.31
-11	26.0	24.93
-12	28.1	26.60
-13	6.7	5.28
-14	34.1	34.26
-15	39.7	41.70
	h=5,k=1	
-13	13.0	12.47
-14	14.1	14.23
-15	25.6	25.60
-12	5.2	4.82
-10	71.0	69.55
-9	43.2	41.53
-8	20.3	21.67
-7	53.7	52.71
-6	79.1	76.02
-7	87.9	86.27
-4	17.7	17.73
-3	159.5	157.94
-2	64.9	63.66
-1	94.2	90.84
0	94.2	94.75
1	37.1	35.03
2	78.0	77.16
4	53.0	53.28
5	107.2	107.7
6	15.6	16.43
7	12.6	13.20
8	24.2	24.43
9	63.0	63.38
10	26.5	25.96
	h=5,k=0	
8	88.8	92.79
6	15.5	15.40
4	5.4	5.23
2	18.9	21.07
0	23.8	25.00
-2	24.3	25.53
-4	12.5	13.81
-6	105.4	104.72
-10	57.5	58.67
-12	15.7	14.72
-14	63.1	63.86
	h=6,k=0	
4	37.1	38.02
-12	45.5	46.56
-10	26.4	25.70
-8	32.5	32.26
-6	180.0	190.08
-4	50.4	46.71
-2	50.1	48.97
0	7.8	7.75
2	107.1	108.61
4	70.4	72.29
6	81.5	84.02
8	19.3	19.75
	h=6,k=1	

Table 1. Continued.

l	Fo	Fc
8	20.3	21.27
7	14.7	14.90
6	36.6	36.36
5	101.5	104.21
4	7.1	6.61
3	51.5	50.73
2	48.7	48.40
1	60.4	61.63
0	15.2	13.96
-1	12.8	11.60
-2	98.0	97.23
-3	33.2	31.62
-4	4.7	2.87
-5	136.5	136.71
-6	28.1	27.05
-7	47.9	47.53
-8	13.6	12.44
-9	51.8	51.78
-10	24.0	23.57
-11	29.3	28.81
-12	49.7	50.96
-13	52.6	53.78
-14	37.0	36.64
h=6,k=2		
-13	60.8	59.67
-12	38.1	37.57
-11	8.6	9.16
-10	40.8	40.45
-9	99.4	99.71
-8	18.6	17.45
-7	17.1	18.41
-6	54.4	52.82
-5	42.2	41.39
-4	15.9	14.98
-3	61.3	59.91
-2	61.4	59.87
-1	100.3	97.83
0	46.5	44.76
1	95.8	94.05
2	48.1	46.72
3	11.6	11.38
4	16.7	15.96
5	32.1	31.00
6	32.6	32.51
7	14.0	13.89
8	8.7	8.64
9	63.2	64.23
h=6,k=3		
8	50.4	49.62
7	6.6	5.20
6	42.1	42.99
5	11.9	12.18
3	18.5	18.19
2	84.7	83.61
1	25.7	25.06
0	17.7	17.04
-1	46.9	45.32
-2	134.5	130.27
-3	10.2	3.81
-4	5.4	5.48
-5	13.2	12.72
-6	13.1	12.56
-7	8.3	7.49
-8	78.3	76.20
-10	88.5	86.29
-11	20.0	20.68
-12	52.2	52.08
-12	5.94	7.22
h=6,k=4		
-10	9.08	5.39
-9	75.0	72.04
-8	45.2	44.10
-7	26.2	24.79
-6	44.4	41.64
-5	7.4	7.48
-4	17.9	16.13
-3	69.4	65.72
-2	41.9	39.08
-1	70.8	68.11
0	11.9	11.02
1	56.2	53.59
2	68.2	65.86
3	34.5	34.08
4	8.5	5.63
5	17.7	13.51
6	20.9	20.17
7	10.6	8.26
h=6,k=5		
6	9.7	8.91
5	50.1	47.98
4	23.1	22.58
3	49.0	48.32
2	33.1	31.16
1	50.2	48.39
-1	9.0	3.69
-2	43.1	41.25
-3	46.3	43.01
-4	43.2	40.94
-5	65.8	63.24
-6	10.2	7.98
-7	12.7	10.89
-8	45.3	44.06
-9	48.5	46.49
-10	24.8	23.63
-11	9.6	8.88
h=6,k=6		
-8	6.7	5.74
-7	25.3	23.88
-6	80.8	76.12
-5	23.6	21.74
-4	41.6	40.82
-3	18.9	18.35
-2	18.8	17.30
-1	9.6	8.74
0	9.5	7.90
1	6.5	5.68
2	36.9	33.89
3	32.5	30.40
h=6,k=7		
-1	19.1	17.26
-2	32.3	29.47
-3	10.6	10.04
-4	24.2	23.90
h=7,k=6		
-1	10.4	9.22
-2	28.8	25.98
-4	18.6	17.75
-5	6.7	5.24
h=7,k=5		
-9	59.3	56.88

l	Fo	Fc
-8	31.2	28.99
-6	38.8	37.21
-5	28.7	27.05
-4	7.7	6.50
-3	9.8	10.19
-2	24.3	23.13
-1	54.5	51.84
0	30.2	29.03
1	33.5	31.96
3	32.7	31.96
h=7,k=4		
4	22.7	21.18
2	38.2	35.53
1	5.1	3.26
0	30.5	30.35
-1	64.7	63.79
-2	33.8	32.29
-3	30.8	28.93
-4	6.4	5.50
-5	69.0	66.68
-6	41.2	39.90
-7	26.9	25.19
-8	29.9	29.92
-9	10.4	9.44
-10	24.4	22.13
-11	39.0	38.98
h=7,k=3		
-12	80.6	79.72
-11	21.8	23.08
-7	8.7	9.53
-6	56.9	54.87
-4	43.3	42.29
-3	30.7	29.23
-2	107.3	103.94
-1	21.9	22.16
0	30.2	29.26
2	24.3	24.35
4	5.2	5.04
6	65.7	64.76
h=7,k=2		
6	19.1	20.48
5	53.9	53.18
4	10.6	10.40
3	30.7	30.89
2	41.0	41.30
1	11.8	11.02
0	16.3	15.16
-1	76.7	75.06
-2	51.4	50.98
-3	51.8	52.34
-4	44.0	43.18
-5	101.2	99.69
-6	44.5	44.21
-7	25.9	25.40
-8	35.9	34.49
-9	18.8	18.43
-10	33.7	33.48
-11	22.3	22.34
-12	5.4	4.43
-13	59.7	60.91
h=7,k=1		
-13	19.5	20.24
-12	32.4	32.99
-10	27.8	27.38
-9	102.7	103.28
-8	13.0	12.37
-7	6.2	6.68
-6	54.7	54.54
-5	71.4	70.86
-4	56.8	56.37
-3	21.5	22.04
-2	51.0	49.20
-1	65.3	64.90
0	23.6	24.56
1	74.4	76.26
3	61.0	59.98
4	6.4	5.49
5	31.3	32.00
6	52.4	53.85
7	7.0	5.26
h=7,k=0		
6	7.5	8.30
2	132.9	136.51
0	12.1	13.16
-2	31.3	31.60
-4	8.9	8.40
-6	61.2	62.55
-8	94.4	96.15
-10	77.5	80.86
-12	26.2	26.60
h=8,k=0		
-8	69.8	70.16
-6	18.3	18.86
-4	31.8	31.90
-2	106.6	111.02
0	63.6	64.54
2	44.0	45.57
4	12.2	11.15
h=8,k=1		
4	9.4	9.25
3	21.0	21.06
2	47.9	49.07
1	28.4	29.27
0	5.0	3.47
-1	85.8	87.22
-2	16.0	15.08
-3	17.5	18.47
-4	24.1	23.60
-5	30.3	31.30
-6	23.3	24.63
-7	23.4	23.58
-8	49.6	51.03
-9	62.4	64.26
-10	19.1	16.76
-11	32.5	33.17
h=8,k=2		
-11	19.6	19.63
-9	51.2	50.95
-8	30.3	29.88
-7	18.3	18.25
-6	17.3	18.10
-5	82.6	80.34
-3	9.1	6.99
-2	34.7	34.65
-1	5.7	7.06
0	6.1	6.45
1	24.9	25.22
2	32.8	33.66
3	62.1	62.65
h=8,k=3		

l	Fo	Fc
2	58.6	58.29
1	9.4	9.44
0	13.1	12.51
-1	6.9	6.07
-2	10.1	8.90
-3	18.8	18.37
-4	68.6	67.35
-5	15.2	15.16
-6	64.7	63.11
-7	20.9	20.93
-8	48.8	49.21
-10	18.2	17.34
h=8,k=4		
-8	7.9	8.64
-7	13.7	13.67
-5	75.6	73.08
-4	31.5	30.18
-3	19.6	18.65
-2	28.7	26.85
-1	6.8	1.07
0	12.5	13.11
h=9,k=2		
-2	18.0	18.57
-4	35.5	35.63
-5	29.8	29.61
-6	32.6	33.90
h=9,k=1		
-6	18.9	18.96
-5	64.8	65.51
-4	9.7	9.16
-3	10.8	9.99
-2	16.2	16.50
-1	45.1	45.02
h=9,k=0		
0	23.8	23.81
-2	52.8	55.73
-4	64.1	64.63
-6	30.6	31.76
-8	15.2	15.63
18	49.3	47.61
h=0,k=1		
20	8.1	7.84
19	25.8	25.08
18	21.8	22.62
17	23.7	23.67
h=0,k=2		
16	10.4	12.29
17	18.2	18.30
18	15.5	16.05
19	7.6	9.12
h=0,k=3		
19	8.1	9.88
18	11.4	11.88
17	11.5	9.70
16	26.4	26.30
h=0,k=4		
15	56.1	52.11
16	12.4	12.32
17	7.9	6.67
18	16.4	15.35
19	14.1	14.33
h=0,k=5		
19	11.8	11.64
18	14.5	12.20
17	10.4	7.76
15	8.6	7.91
14	25.6	23.80
h=0,k=6		
13	11.3	10.23
14	5.6	4.27
15	6.7	7.00
16	7.8	7.69
18	23.3	21.57
h=0,k=7		
17	20.2	19.08
15	6.6	8.02
14	11.3	9.32
13	13.3	13.46
12	25.3	23.46
11	41.7	38.27
h=0,k=8		
9	6.8	7.00
10	16.3	14.12
11	19.2	18.78
12	16.8	15.44
14	6.0	4.38
15	16.7	15.03
h=0,k=9		
14	16.0	14.57
13	6.5	7.91
12	6.4	2.33
10	9.4	10.98
9	9.1	7.74
8	22.4	20.20
7	8.8	8.33
6	11.6	12.52
5	36.5	36.08
4	30.0	31.72
h=0,k=10		
0	7.8	10.06
1	11.4	12.69
2	9.4	7.97
3	21.2	22.03
5	8.7	7.61
6	8.7	7.27
7	11.6	9.54
8	12.1	10.96
9	16.4	14.82
h=0,k=11		
8	10.8	10.46
7	10.5	9.86
4	9.8	8.85
3	9.5	9.58
2	20.5	19.03
h=0,k=12		
0	11.2	10.69
1	7.0	6.82
2	7.0	6.25
-1	9.9	9.04
h=1,k=11		
-8	8.3	7.15
-6	13.4	13.18
-3	8.2	8.34
-2	17.7	16.17
-1	6.4	6.58
0	9.0	9.40
3	8.2	7.91
4	6.3	7.56
5	9.9	10.00
6	11.4	11.49

l	Fo	Fc
8	7.4	5.79
h=1,k=10		
11	15.1	13.67
9	14.1	12.65
8	7.2	7.04
6	6.7	5.94
5	20.1	19.99
4	9.5	9.73
3	10.2	10.14
2	10.6	9.84
1	11.9	12.26
0	9.3	9.54
-1	23.3	23.30
-3	8.9	8.55
-5	16.5	15.06
-6	9.5	7.50
-7	20.7	19.85
-8	9.9	8.71
-9	5.6	6.84
h=1,k=9		
-14	8.9	8.37
-12	9.9	9.82
-10	9.1	8.50
-9	21.6	20.91
-8	21.4	22.98
-6	24.9	25.76
-5	15.3	16.14
4	31.7	34.36
5	10.1	10.14
6	15.9	16.55
8	7.6	7.98
9	5.3	4.15
10	13.1	13.20
12	10.9	11.76
13	16.7	16.07
h=1,k=8		
15	10.6	8.70
14	12.3	12.31
12	17.4	17.33
11	23.3	23.03
10	7.3	8.47
9	10.3	10.04
8	17.4	17.83
-9	9.7	8.71
-10	24.2	23.18
-11	18.6	17.26
-13	7.9	9.14
-14	13.9	13.12
-15	14.3	13.40
-16	14.3	14.49
h=1,k=7		
-16	13.6	14.22
-15	17.6	16.63
-14	12.3	13.47
-13	13.3	12.61
-12	11.4	10.40
-11	32.3	32.29
11	8.0	8.07
12	20.5	20.56
13	18.6	18.75
14	8.4	5.98
15	14.7	12.93
h=1,k=6		
17	12.4	10.19
16	14.8	13.38
14	24.6	23.15
13	7.2	5.78
12	5.8	6.13
-13	28.2	28.25
-14	35.1	33.15
-15	8.8	7.66
-16	10.0	9.84
h=1,k=5		
-19	11.7	12.31
-18	11.3	9.48
-17	7.5	7.20
-16	7.4	7.31
-15	29.8	29.71
-14	23.4	23.61
14	12.1	11.41
15	30.0	29.60
16	21.2	21.21
17	6.5	5.24
18	11.5	10.01
h=1,k=4		
19	9.1	6.70
18	13.5	14.18
17	8.0	9.02
16	19.4	20.46
15	29.2	28.40
14	15.5	15.01
-15	11.1	10.49
-17	34.2	34.15
-18	11.2	9.97
-19	21.5	21.54
h=1,k=3		
-18	48.8	49.28
-17	8.5	9.55
-16	11.2	10.76
15	8.4	7.52
16	12.3	12.11
18	6.6	4.73
h=1,k=2		
20	6.5	6.45
19	12.2	12.82
18	21.4	20.69
17	8.2	6.96
16	12.7	11.91
-17	37.7	38.34
-18	5.6	5.42
-19	18.7	19.49
-20	9.2	9.65
-21	12.1	11.32
h=1,k=1		
-21	21.9	23.31
-18	18.2	19.54
16	12.4	11.68
17	17.9	17.80
18	11.1	11.94
19	24.1	24.07
20	11.2	10.33
h=1,k=0		
20	11.0	10.75
18	36.3	38.41
16	24.9	24.60
-20	17.0	17.96
-18	24.4	26.48
16	46.6	47.80
18	10.6	12.24
h=1,k=1		

l	Fo	Fc
19	6.4	6.18
18	10.9	11.28
16	13.7	14.04
15	58.2	58.25
-17	35.6	37.07
-18	22.7	24.59
-19	13.4	14.83
-20	6.9	7.66
-21	9.7	10.46
h=2,k=2		
-21	21.9	22.81
-20	13.7	14.44
-19	10.2	10.61
-18	29.0	30.48
-17	27.4	28.33
15	10.7	10.34
16	12.7	12.05
19	25.3	26.88
h=2,k=3		
18	28.8	26.99
16	7.3	7.21
15	5.9	4.47
14	7.1	6.88
-16	16.2	16.07
-18	28.6	29.65
-19	8.4	9.78
-20	15.8	17.15
h=2,k=4		
-18	15.2	16.32
-17	25.4	25.40
-16	10.7	11.07
-15	16.6	16.55
14	7.8	6.09
15	7.7	6.61
16	12.0	12.60
17	12.7	12.43
h=2,k=5		
17	6.5	1.95
16	10.5	10.37
15	31.4	30.16
14	10.6	10.52
13	20.8	20.38
-14	22.7	23.23
-15	25.5	26.32
-16	29.1	29.85
-17	22.1	22.31
-18	13.9	13.27
-19	10.4	9.09
h=2,k=6		
-18	17.4	16.44
-17	8.3	9.12
-15	20.7	20.27
-14	49.3	48.53
-13	6.5	7.37
12	23.3	21.97
13	15.5	14.40
14	26.4	24.32
16	19.3	18.71
17	9.7	9.36
h=2,k=7		
15	23.0	21.14
14	10.9	9.57
13	9.6	9.29
11	16.2	16.32
-11	15.1	14.83
-12	8.3	6.94
-13	34.7	34.05
-14	8.9	7.51
-15	19.8	19.73
-17	8.2	7.99
h=2,k=8		
-15	5.5	2.75
-14	11.7	11.30
-13	10.2	7.87
-12	19.2	20.34
-11	18.6	18.58
-10	9.0	9.65
-9	10.1	10.69
7	19.8	20.21
8	21.7	20.99
9	21.5	22.74
10	26.9	26.49
11	18.0	17.50
14	10.2	9.86
h=2,k=9		
12	17.3	16.00
11	5.8	6.15
10	6.2	6.32
9	19.5	18.56
8	18.5	18.83
6	9.5	9.68
3	10.8	10.99
2	20.4	22.34
1	11.4	11.35
-3	6.3	5.33
-4	20.1	22.05
-5	6.2	4.93
-6	14.0	13.82
-7	8.7	8.04
-9	20.0	20.10
-10	25.9	26.28
-11	10.4	9.62
-12	11.0	12.15
-13	7.6	6.36
h=2,k=10		
-12	6.3	5.90
-11	15.6	15.62
-9	15.4	15.74
-6	8.8	8.88
-5	20.6	20.79
-4	8.8	9.03
-3	8.7	9.15
-1	14.6	14.77
1	22.7	23.64
2	17.3	17.42
3	10.9	10.49
5	10.3	10.04
7	13.7	15.09
h=2,k=11		
6	16.6	15.30
2	13.7	13.88
0	9.8	8.88
-3	8.2	8.59
-4	12.6	13.11
-6	11.5	10.37
-7	8.7	9.42
-8	8.9	7.13
h=3,k=11		
-8	15.1	14.01
-4	10.8	10.86

l	Fo	Fc
-3	5.3	.36
-2	13.1	11.99
0	6.9	5.19
1	10.2	11.00
2	14.2	13.58
4	8.9	9.00
h=3,k=10		
9	15.8	15.17
7	13.0	12.79
6	8.3	6.98
4	5.6	5.93
3	15.8	16.68
2	7.4	7.51
1	8.0	5.35
-1	16.4	17.24
-2	13.4	13.22
-3	21.3	22.44
-4	10.6	9.81
-5	8.5	8.89
-7	10.1	9.79
-9	15.6	15.00
-10	5.6	4.15
-12	8.3	5.87
h=3,k=9		
-14	18.6	17.11
-13	12.6	11.53
-12	7.4	7.04
-11	15.9	15.91
-10	14.4	14.28
-8	14.0	14.94
-7	7.9	8.38
-6	12.0	10.91
-5	15.0	14.24
-4	19.9	19.23
-3	6.0	3.16
-2	27.8	29.83
-1	22.9	24.12
0	7.1	6.17
2	15.3	16.48
4	21.7	21.67
5	12.3	12.29
7	17.7	17.35
8	21.0	20.57
9	11.6	12.03
10	10.6	9.39
h=3,k=8		
12	7.0	5.90
10	15.3	15.25
9	10.0	11.38
7	9.9	8.96
6	34.0	34.19
5	37.8	40.43
-8	4.8	4.11
-9	10.9	12.28
-10	16.3	15.64
-11	20.4	20.10
-12	23.7	23.17
-13	19.2	18.89
-16	15.8	14.50
h=3,k=7		
-17	24.4	23.43
-16	6.6	6.64
-15	14.9	13.91
-14	10.1	11.30
-13	12.5	13.00
-12	7.1	6.46
-11	28.5	27.10
8	21.7	21.76
9	5.8	4.97
10	11.6	9.70
11	31.5	29.50
12	6.4	5.58
13	8.6	18.09
h=3,k=6		
13	20.5	19.67
12	45.2	44.24
11	8.5	6.87
10	15.8	13.75
-13	6.3	3.30
-14	9.3	8.73
-15	11.3	12.04
-16	22.9	23.12
-18	21.3	20.75
h=3,k=5		
-18	11.6	12.87
-17	32.1	32.15
-16	6.9	8.31
-15	13.2	14.74
-14	24.4	23.12
12	12.3	13.02
13	13.0	13.76
14	20.1	20.84
15	19.8	19.50
16	8.5	8.75
h=3,k=4		
15	30.9	29.48
14	12.6	11.28
13	9.2	10.36
-15	30.0	29.90
-17	10.0	8.96
-18	12.1	10.92
-19	19.6	19.95
h=3,k=3		
-20	22.7	23.96
14	16.7	15.12
16	30.1	29.71
17	12.6	12.12
18	17.8	17.24
h=3,k=2		
18	7.4	7.26
17	6.7	7.13
16	13.3	13.83
15	35.9	36.28
14	6.4	6.59
-17	6.3	5.54
-18	9.6	8.98
-19	5.4	4.47
-20	10.8	10.42
-21	19.4	20.21
h=3,k=1		
-21	14.7	15.76
-20	12.1	12.95
-19	11.8	11.43
-17	57.8	60.37
14	9.3	8.22
15	24.9	26.80
16	20.5	21.51
18	10.4	9.43
h=3,k=0		
18	9.7	8.64

l	Fo	Fc
16	6.3	9.27
14	17.4	18.46
-18	56.7	59.78
-20	8.6	9.58
h=4,k=0		
-20	31.0	34.58
-18	7.2	9.23
16	14.6	15.37
18	12.6	12.51
h=4,k=1		
16	1.3	12.56
15	7.7	8.33
14	5.8	5.54
13	18.5	20.40
-17	25.9	27.52
-18	18.6	20.15
-19	13.1	14.16
-21	27.1	29.49
h=4,k=2		
-20	17.4	18.24
-19	11.4	10.83
-18	9.9	8.42
-17	47.0	48.77
-16	27.7	28.63
13	34.3	36.17
14	11.0	10.93
15	32.6	33.25
16	8.5	8.89
17	7.1	8.83
h=4,k=3		
16	32.1	33.26
15	6.0	3.62
14	13.1	12.37
13	9.9	8.81
12	40.6	40.10
-16	35.3	35.89
-18	31.2	32.81
h=4,k=4		
-19	8.6	8.01
-18	13.1	14.61
-17	26.2	26.71
-16	6.9	7.77
-15	12.9	11.78
11	26.1	25.45
12	18.7	19.91
13	18.2	19.01
15	21.8	20.74
16	16.5	15.55
h=4,k=5		
16	10.1	8.33
15	7.0	8.19
13	8.2	9.58
12	16.0	15.74
11	25.4	24.30
10	28.7	28.94
-14	13.1	12.26
-15	14.0	14.24
-16	13.5	13.61
-17	20.6	21.81
-18	18.5	18.76
h=4,k=6		
-17	7.6	8.21
-16	11.5	12.42
-15	9.8	9.08
-14	19.5	17.58
9	25.4	24.05
10	9.6	11.63
11	16.4	16.21
12	21.4	19.81
13	11.8	12.99
h=4,k=7		
13	9.8	10.76
12	16.0	14.88
11	7.4	7.55
10	14.7	13.81
9	27.9	28.15
8	10.6	9.62
7	31.4	29.89
-11	24.8	25.01
-12	8.2	7.39
-13	10.9	8.12
-14	14.4	14.95
-15	14.5	15.31
-16	5.8	5.55
-17	9.5	8.57
h=4,k=8		
-16	15.7	15.17
-14	19.3	19.18
-13	13.2	13.39
-12	5.6	2.51
-10	18.4	17.13
-9	15.3	14.15
-8	20.3	20.00
-7	21.3	20.84
4	13.5	15.62
5	27.2	27.02
6	23.1	22.29
7	7.3	6.39
8	17.6	16.79
11	7.6	8.30
12	8.9	8.13
h=4,k=9		
9	8.7	6.02
7	9.8	10.98
6	15.4	16.89
5	9.0	7.50
4	18.5	18.58
3	18.6	18.40
2	13.2	14.78
0	17.1	16.78
-1	6.2	4.97
-2	19.6	21.14
-3	11.7	11.78
-5	28.9	27.53
-6	27.9	27.45
-8	10.2	9.24
-10	5.2	3.59
-12	14.8	14.89
-14	9.8	10.08
h=4,k=10		
-11	12.1	10.57
-8	7.5	6.46
-7	20.1	19.86
-5	16.3	16.43
-4	8.1	6.62
-3	6.1	4.31
-2	11.7	12.27
-1	18.5	18.39
1	7.3	8.40
3	12.5	11.48

l	Fo	Fc
5	18.3	18.26
6	11.8	11.29
7	8.1	8.90
h=4,k=11		
2	6.6	5.95
0	11.5	10.95
-1	6.1	7.17
-2	7.9	8.01
-3	10.2	9.23
-6	8.9	7.61
h=5,k=11		
-4	15.6	13.77
0	10.3	11.52
h=5,k=10		
6	8.7	9.47
5	8.0	4.97
3	6.4	9.22
2	9.8	7.53
1	17.0	17.78
0	7.5	6.52
-1	9.5	9.08
-3	10.4	11.35
-4	6.4	7.59
-5	18.5	18.18
-6	5.6	6.07
-8	7.7	5.42
-9	13.0	11.91
h=5,k=9		
-12	5.7	6.32
-11	5.8	6.33
-10	19.6	19.53
-9	12.9	11.71
-8	9.9	10.71
-7	19.6	19.91
-6	18.9	19.11
-5	6.9	6.40
-4	15.6	15.82
-2	12.8	12.47
-1	8.4	6.99
0	10.6	10.89
2	23.9	22.90
3	24.8	23.47
4	6.5	4.29
6	6.7	8.93
8	11.1	11.19
h=5,k=8		
10	13.2	13.07
9	20.1	19.27
8	6.4	6.89
5	13.0	12.08
4	27.8	26.71
3	9.1	8.79
2	24.5	23.02
1	27.4	25.86
0	18.5	17.58
-1	17.8	18.34
-2	18.3	18.06
-3	5.1	4.21
-4	5.5	2.95
-5	14.4	14.93
-6	25.0	23.99
-7	14.6	12.97
-8	37.5	36.95
-9	27.6	26.49
-11	10.4	9.74
-12	11.9	11.25
-14	8.4	8.72
h=5,k=7		
-17	8.8	8.71
-16	6.0	6.20
-15	10.4	11.77
-14	5.6	6.70
-13	39.3	37.43
-12	10.9	9.31
-11	15.6	15.08
-10	14.3	13.13
-9	19.6	19.47
4	12.1	11.72
5	29.7	29.26
6	10.4	9.02
7	6.5	4.38
8	8.3	7.75
9	21.0	20.23
10	19.3	19.52
11	9.7	10.11
12	7.9	8.14
h=5,k=6		
10	16.5	17.02
9	7.5	8.55
8	36.2	33.96
7	33.0	31.79
-11	27.7	27.59
-12	41.8	39.84
-13	6.9	7.26
-14	40.1	38.88
-15	12.8	12.80
-16	6.7	4.67
-18	10.7	10.36
h=5,k=5		
-19	10.3	10.81
-17	16.3	14.18
-16	22.7	22.28
-14	6.3	8.35
-13	50.2	49.23
9	35.9	36.06
12	12.6	11.71
13	14.9	13.36
h=5,k=4		
14	12.2	11.54
13	29.1	28.78
12	5.7	3.91
11	28.0	27.59
-14	14.6	14.25
-15	31.9	31.73
-16	14.2	14.44
-17	40.7	40.75
-18	6.0	4.31
h=5,k=3		
-20	16.0	15.99
-19	9.7	10.98
-18	27.5	27.64
-16	56.0	56.42
11	6.9	8.77
12	59.2	59.94
13	11.3	11.82
14	7.6	10.20
h=5,k=2		
16	10.0	8.88
15	13.6	13.21
13	29.1	30.80

l	Fo	Fc
12	9.4	6.24
11	16.6	17.62
-16	10.3	9.51
-17	39.8	41.57
-18	12.2	13.81
-19	11.5	11.99
-20	10.3	10.73
-21	14.7	15.98
h=5,k=1		
-21	14.5	15.55
-20	21.3	22.87
-19	17.8	19.81
-18	7.7	7.27
-17	24.0	25.36
-16	9.8	10.03
12	22.6	23.74
13	13.8	13.76
15	19.3	20.28
16	7.6	6.73
h=5,k=0		
16	36.7	36.74
-16	18.1	19.62
-18	13.3	13.98
-20	29.3	32.02
h=6,k=0		
-20	12.8	12.44
-18	14.1	14.70
-16	61.4	64.05
10	13.4	14.43
12	34.6	35.42
14	8.4	9.77
h=6,k=1		
15	11.2	12.41
13	33.5	34.22
12	19.0	20.21
11	7.8	7.88
10	21.3	22.61
-15	24.3	23.83
-16	20.0	20.66
-17	44.2	46.27
-20	7.2	7.99
h=6,k=2		
-19	21.8	23.50
-18	17.2	18.55
-17	19.5	21.07
-16	29.4	30.42
-15	14.3	13.18
10	6.3	5.74
11	14.0	13.73
12	17.2	16.57
13	12.4	12.85
15	6.7	4.27
h=6,k=3		
12	16.4	16.96
11	12.5	12.02
10	37.5	39.31
-14	28.0	27.73
-15	5.8	6.27
-18	14.9	16.77
-20	35.1	37.70
h=6,k=4		
-19	22.1	22.86
-18	7.2	6.16
-16	22.2	23.27
-15	18.9	19.50
-14	17.6	17.69
-13	40.4	38.82
8	17.1	16.95
9	49.2	49.00
10	17.0	16.94
12	11.8	11.23
13	7.5	8.76
14	7.8	8.28
h=6,k=5		
13	23.1	23.37
12	6.9	6.80
10	6.9	6.51
8	19.5	19.15
-12	22.0	21.03
-13	29.9	29.98
-14	29.1	29.65
-15	15.9	15.27
-16	13.4	15.01
-17	26.6	26.52
h=6,k=6		
-16	26.8	25.06
-15	18.6	19.46
-13	6.6	7.59
-12	25.8	25.86
-10	24.2	23.14
4	29.4	26.93
6	37.7	38.15
7	18.8	18.51
10	7.4	3.14
12	18.7	17.19
h=6,k=7		
11	11.1	10.69
10	7.5	8.69
7	15.8	17.57
6	23.7	22.66
5	34.2	32.28
4	12.2	12.82
2	6.7	6.09
1	25.9	25.02
-5	49.8	48.93
-7	30.2	29.26
-9	10.3	9.06
-10	15.9	15.30
-11	23.1	23.08
-12	15.2	16.55
-13	19.0	17.79
-15	13.0	11.38
h=6,k=8		
-13	18.5	17.40
-12	21.5	20.29
-11	6.6	4.99
-10	20.3	19.52
-9	16.8	17.09
-8	6.5	6.11
-6	15.9	14.50
-4	14.5	12.63
-3	17.1	15.72
-2	16.0	14.86
-1	26.5	27.13
0	35.7	34.84
1	16.6	15.96
2	8.9	9.02
4	8.8	7.77
5	12.0	11.03
6	15.5	13.22

Table 1. Continued.

l	Fo	Fc
7	6.9	8.44
8	6.5	5.72
9	14.0	14.38
h=6,k=9		
6	6.5	6.89
4	10.6	10.69
3	8.5	8.15
2	15.6	15.28
1	9.7	9.97
0	6.0	3.20
-1	20.8	19.96
-2	29.5	25.05
-4	8.4	8.80
-8	15.1	14.90
-10	12.3	10.86
-11	14.1	14.42
-12	12.1	11.73
h=6,k=10		
-8	9.7	8.27
-7	15.3	13.94
-3	12.5	12.11
-1	10.3	9.30
0	9.0	6.26
2	10.2	10.33
3	16.3	14.47
h=7,k=9		
-8	6.5	7.58
-6	19.0	17.62
-5	14.8	12.66
-4	10.5	10.44
-3	19.3	17.81
-2	14.9	13.74
-1	8.8	7.96
0	10.8	10.82
4	7.6	5.20
h=7,k=8		
7	8.0	9.88
6	11.5	9.29
5	18.4	15.55
4	12.1	8.78
3	10.8	9.52
2	12.7	11.11
0	10.0	10.99
-1	11.9	13.40
-2	10.1	10.29
-4	26.5	24.87
-5	30.4	28.53
-7	6.1	4.93
-8	14.9	13.17
-10	18.4	17.76
-12	6.4	5.29
-13	11.0	10.23
h=7,k=7		
-15	21.0	21.60
-13	13.5	13.33
-12	6.6	6.66
-11	8.2	9.85
-10	8.9	9.17
-9	36.1	34.65
-8	15.3	14.47
-7	9.9	11.31
-6	8.7	6.88
-5	23.5	21.87
-4	20.2	19.26
-3	24.7	23.59
-2	16.2	14.98
-1	6.2	5.58
0	9.5	6.51
1	31.5	30.59
2	13.2	11.77
3	27.8	27.13
4	11.7	10.27
6	19.6	19.25
7	12.3	11.15
9	13.1	11.94
h=7,k=6		
10	16.2	16.14
8	12.2	10.75
5	5.7	.78
4	6.8	7.95
3	25.8	24.65
2	58.0	57.01
1	14.5	14.73
-6	26.9	25.00
-7	31.9	32.35
-8	39.5	38.89
-9	6.6	6.40
-10	28.8	26.73
-11	10.6	11.29
-12	7.1	5.57
-14	15.7	16.21
-16	27.7	27.66
h=7,k=5		
-18	20.8	21.01
-17	11.7	-11.21
-16	16.5	15.59
-15	9.3	-10.67
-14	9.6	-10.50
-12	21.5	-20.79
	4	30.1	28.52
	9	24.0	24.05
	7	18.0	-17.89
	5	53.7	-52.91
-12	12.0	12.67
-13	51.5	50.30
-14	6.1	-9.46
-15	5.8	-5.53
-16	10.8	10.54
-17	19.3	-19.33
-18	6.5	-3.60
h=7,k=3		
-18	13.1	13.88
-16	21.2	21.48
-14	34.3	-34.39
	7	26.2	25.40
	8	22.5	-21.98
	10	14.8	-17.03
	13	15.5	13.69
h=7,k=2		
	12	13.3	12.99
	10	14.2	-15.66
	9	35.4	36.42
	8	18.7	-19.33
	7	22.1	-21.40
-14	7.0	-7.72
-15	20.9	-21.35
-16	19.2	-19.60
-17	24.3	-23.85
-18	9.8	10.29
-19	9.0	-10.08
-20	21.1	22.88
h=7,k=1		
-20	13.6	14.87
-19	21.1	-23.01
-18	16.0	16.39
-17	21.7	23.11
-16	30.5	31.76
-15	35.1	36.02
	8	13.2	-14.05
	9	35.6	36.05
13	24.6	-25.53
14	11.2	-11.52
h=7,k=0		
14	12.4	14.85
12	26.4	30.60
10	35.4	-35.62
8	7.3	-9.01
-14	31.0	-32.06
-16	54.4	-58.82
-18	9.5	-9.39
-20	24.1	26.25
h=8,k=0		
-18	27.0	27.98
-16	6.9	6.84
-14	11.0	-10.89
-12	54.4	-57.33
	6	29.5	29.26
	8	20.6	-20.97
	10	40.7	-41.69
	12	8.9	9.9
h=8,k=1		
	10	6.5	6.3
	9	35.4	35.3
	8	8.1	8.9
	7	22.8	23.8
	6	20.1	19.9
	5	17.0	17.9
-12	19.2	19.1
-13	45.3	46.8
-14	11.0	10.0
-16	16.6	17.4
-17	9.5	9.7
-19	29.3	31.4
h=8,k=2		
-19	7.2	7.8
-18	5.9	6.8
-17	25.3	27.2
-16	7.3	7.0
-15	34.6	36.0
-14	26.8	27.4
-13	19.4	20.1
-12	41.1	43.9
	4	24.5	24.8
	5	41.2	41.6
	6	22.1	22.3
	7	14.2	15.6
	10	14.3	11.7
	11	8.3	9.4
h=8,k=3		
	10	14.1	13.2
	7	9.8	11.0
	6	47.9	49.0
	3	11.8	11.2
-12	16.9	16.6
-13	11.7	13.5
-14	17.2	17.8
-15	13.2	16.4
-16	45.9	47.6
h=8,k=4		
-17	20.8	21.8
-15	34.3	35.3
-14	10.3	8.5
-13	10.3	9.7
-12	31.0	32.2
-11	25.7	26.6
-10	5.6	3.1
-9	34.4	34.5
1	23.5	23.4
2	22.8	24.5
3	29.5	30.0
4	9.1	6.9
5	27.4	26.6
6	24.1	24.0
7	18.4	18.6
10	12.9	12.9
h=8,k=5		
9	21.0	20.1
8	9.0	8.3
7	22.5	23.4
6	13.2	13.0
5	5.9	8.3
2	23.3	22.5
1	16.3	14.0
0	33.4	32.7
-1	52.5	52.3
-4	25.9	24.8
-5	18.0	17.2
-6	16.7	15.4
-8	10.7	10.7
-9	31.8	31.3
-10	34.6	35.1
-11	29.1	28.9
-12	19.2	18.72
-13	33.0	32.52
-14	8.4	8.85
-15	7.6	4.92
-16	15.0	13.89
h=8,k=6		
-15	5.9	3.80
-14	7.4	7.70
-12	29.6	29.65
-11	20.5	20.12
-10	13.0	11.90
-8	34.2	34.19
-7	5.6	4.77
-6	16.4	15.06
-5	7.5	3.97
-4	11.5	11.81
-3	9.0	8.08
-2	44.1	41.80
-1	19.7	19.32
0	21.4	21.73
1	19.2	18.71
2	18.8	18.10
3	8.4	7.55
4	8.1	7.77
6	21.0	19.84
7	6.4	6.46
8	6.3	5.76
h=8,k=7		
5	11.2	10.97
4	11.0	11.56
3	14.9	13.30
2	17.9	15.91
0	16.3	15.13
-1	29.0	28.18
-2	6.4	7.43
-3	20.3	19.22
-5	7.5	7.06
-7	27.8	26.97
-8	19.2	18.30
-9	23.5	21.94
-11	8.4	6.97
-13	19.3	18.37
h=8,k=8		
-9	17.5	15.30
-8	16.5	14.66
-6	14.7	13.88
-5	18.8	17.89
-4	12.4	10.27
-2	17.0	15.43
0	12.0	9.13
1	7.6	6.52
2	6.8	6.28
3	19.1	19.29
4	16.1	14.87
h=8,k=9		
-17	9.1	9.42
-16	17.5	19.32
-14	15.4	13.21
-13	7.9	8.69
-12	25.9	26.13
-10	20.0	20.32
-8	87.1	69.67
-7	18.1	16.51
-6	7.2	6.63
h=9,k=7		
-5	6.0	2.68
-4	9.5	9.76
-11	25.0	25.49
-2	25.7	26.49
-1	8.2	8.01
0	26.9	26.65
1	18.3	17.09
2	52.9	53.27
3	9.8	9.49
4	15.3	15.51
h=9,k=2		
9	19.1	19.00
7	8.4	6.81
6	17.1	17.70
5	13.3	13.85
3	36.6	40.00
2	9.0	8.21
0	21.8	22.01
-1	55.1	50.91
-7	26.1	28.33
-8	12.5	12.34
-9	56.4	51.89
-10	6.9	6.90
-11	20.2	21.26
-12	16.8	17.01
-13	19.2	19.28
-14	12.7	12.97
-15	24.7	26.44
-16	23.5	25.46
h=9,k=1		
-18	12.9	14.46
-17	19.0	19.07
-16	7.7	10.57
-15	24.2	25.65
-14	11.9	10.10
-13	31.4	33.89
-12	18.3	18.21
-11	39.1	40.57
-10	5.8	5.38
-8	31.9	32.92
-7	29.7	29.66
-6	24.2	25.25
-5	22.8	23.64
-4	23.1	23.80
-3	6.5	3.51
-2	29.2	28.95
-1	14.3	14.98
0	14.7	16.46
h=9,k=0		
10	8.1	9.06
6	53.2	55.83
4	10.7	12.26
2	13.8	14.83
-10	29.0	29.93
-12	71.0	76.23
-14	30.6	32.12
-16	35.0	38.77
-17	37.2	39.90
-9	41.5	42.30
-12	6.0	6.65
-8	41.1	44.56
-6	18.8	20.65
-4	59.5	61.98
-2	6.9	6.13
0	16.9	18.42
2	30.7	32.54
4	23.4	23.23
6	25.4	27.07
h=10,k=1		
7	12.1	11.70
6	14.9	14.69
5	7.9	8.68
3	37.9	39.87
2	16.1	16.82
1	6.2	5.95
0	16.1	16.08
-2	12.0	12.99
-3	20.3	21.17
-4	23.1	23.88
-5	45.1	47.88
-7	27.2	29.03
-8	12.9	13.61
-9	32.2	34.26
-10	8.9	9.21
-11	6.5	6.60
-12	26.9	28.73
-13	8.0	7.05
-15	30.2	32.78
h=10,k=2		
-16	9.9	10.18
-15	7.4	11.41
-13	15.5	16.61
-12	16.2	16.91
-11	38.1	41.07
-10	24.0	26.24
-9	17.2	18.76
-8	32.8	33.24
-7	18.3	19.26
-5	25.4	26.70
-4	16.8	16.98
-3	15.5	15.32
-1	49.4	51.39
0	20.7	20.83
1	10.4	9.69
0	39.0	39.98
-2	26.9	27.07
-3	9.6	7.28
-4	27.9	28.75
-6	14.2	14.32
-8	27.6	28.25
-10	10.7	10.40
-11	13.5	13.07
-12	37.6	39.20
h=10,k=4		
-14	7.0	.98
-13	21.9	22.84
-12	6.8	8.18
-11	30.4	32.13
-9	10.7	9.79
-8	30.6	31.90
-7	20.4	20.49
-5	18.7	18.51
-4	10.4	9.57
-2	8.8	8.25
-1	37.7	38.00
0	15.8	16.17
1	10.7	9.61
2	12.0	11.09
3	6.6	7.17
4	12.0	11.67
h=10,k=5		
3	24.8	23.92
0	14.2	14.01
-1	6.2	5.93
-2	14.3	13.59
-3	6.0	6.63
-5	19.2	19.25
-6	17.0	17.71
-7	23.9	24.65
-8	15.4	15.28
-9	21.0	20.38
-12	15.4	15.78
h=10,k=6		
-10	6.4	7.19
-8	20.2	19.23
-7	16.0	15.95
-6	7.3	5.93
-4	25.7	25.17
-3	11.6	11.87
-2	8.3	8.25
0	7.5	6.02
h=11,k=5		
-15	15.5	17.47
-6	12.9	13.13
-4	15.7	15.11
h=11,k=4		
1	9.3	8.96
0	20.4	21.11
-1	12.5	11.05
-3	18.4	18.15
-5	23.6	25.20
-6	6.7	5.16
-8	7.5	8.24
-9	15.7	17.19
-10	12.4	14.09
h=11,k=3		
-12	21.1	22.25
-10	16.7	20.03
-8	14.7	15.37
-7	6.0	4.47
-6	5.8	4.52
-4	39.6	40.89
-3	11.7	12.53
-2	6.0	4.90
0	13.8	13.42
2	6.0	7.13
h=11,k=2		
3	18.7	18.95
2	6.0	6.80
0	15.4	15.93
-1	16.9	17.69
-2	17.1	18.15
-3	22.1	23.06
-4	12.5	13.51
-5	25.4	27.16
-7	16.0	16.18
-8	10.8	11.93
-10	9.5	10.71
-11	20.2	20.73
-12	20.8	22.98
h=11,k=1		
8	61.0	60.86
-11	39.6	37.76
h=11,k=3		
0	133.7	176.08
1	120.0	149.02
4	175.7	197.10
8	30.3	31.16
9	5.3	4.56
10	59.9	58.61
11	8.1	7.65
h=11,k=2		
-3	314.2	287.37
-1	168.7	191.62
h=11,k=0		
-14	6.4	7.09
h=12,k=4		
-8	5.6	6.79
-6	5.7	3.85
-4	18.9	15.73
-2	7.1	8.39
h=12,k=1		
-3	16.7	17.06
-4	12.9	12.99
-5	17.9	18.25
-6	6.3	6.98
-7	11.2	10.24
-8	14.9	16.52
-9	8.0	4.07
h=12,k=2		
-7	26.4	30.02
-6	12.5	13.57
-5	8.5	8.76
-4	14.6	16.05
h=0,k=2		
10	165.4	163.95
14	16.6	19.14
h=0,k=1		
14	18.6	17.02
12	40.4	40.81
11	120.0	119.43
9	15.4	12.78
h=0,k=2		
1	62.1	98.84
2	52.6	55.34
3	197.7	218.89
h=1,k=9		
-1	11.6	10.77
h=1,k=8		
-8	25.9	25.14
h=1,k=7		
-10	24.2	24.07
-9	36.3	36.14
-8	16.1	19.04
-7	12.6	12.55
-6	5.9	7.42
-5	37.0	37.95
-4	38.8	40.15
-3	71.4	76.52
-2	26.0	27.95
0	35.3	38.18
1	36.8	40.74
2	16.1	18.00
3	31.8	34.79
4	21.8	23.63
5	8.2	7.64
6	35.6	36.14
7	61.6	62.96
8	13.6	13.38
9	30.1	29.53
h=1,k=1		
11	7.8	8.03
10	8.1	7.00
-5	29.8	30.17
h=1,k=5		
-11	62.8	60.49
-10	30.8	31.30
h=1,k=4		
8	61.0	60.86
-11	39.6	37.76
h=1,k=3		
0	133.7	176.08
1	120.0	149.02
4	175.7	197.10
8	30.3	31.16
9	5.3	4.56
10	59.9	58.61
11	8.1	7.65
h=1,k=2		
-3	314.2	287.37
-1	168.7	191.62
h=1,k=0		
-14	6.4	7.09
h=2,k=9		
-16	11.6	12.30
h=2,k=8		
-16	6.1	1.92
-14	113.1	116.63
-2	127.1	126.26
0	34.7	36.06
2	65.4	63.72
11	38.9	38.51

18°C. Three test-reflections were measured for every 100 reflections. Two test-reflections varied less than 10 %, while the third one varied up to 25 %, probably due to a slight change in crystal position in the capillary. The crystal position was readjusted once during experiment. Precision unit cell data were determined by least squares methods,[4] using 48 reflections measured with a manual diffractometer and Cu$K\alpha$-radiation. The intensities were corrected for effects due to absorption [4-6] and secondary extinction.[4,7]

Oscillation and Weissenberg photographs were taken of the molecular complex formed by equimolecular amounts of carbon tetrachloride and the present addition compound, sealed off in glass-capillaries containing a trace of free carbon tetrachloride.

CRYSTAL DATA

The crystals of MTCL:I_2 (1:1) are monoclinic. The unit cell dimensions are (estimated standard deviations in parenthesis)

$a = 8.777$ (4) Å, $b = 8.724$ (4) Å
$c = 15.595$ (3) Å and $\beta = 104.64$ (1)°.

The space group is $P2_1/c$. The unit cell contains four molecules of the addition compound (formula $C_7H_{13}NSI_2$). The experimental density is 2.27 ± 0.02 g/cm³, the calculated value is 2.283 g/cm³.

Crystalline MTCL:I_2:CCl_4 (1:1:1) is also monoclinic. The space group is $P2_1/c$ and the unit cell dimensions are $a = 13.6$ Å, $b = 8.0$ Å, $c = 16.4$ Å and $\beta = 104°$. The figures are probably correct to about 0.5 %. There are four molecules of the adduct per unit cell. Experimental density was obtained as *ca.* 2.2 g/cm³. The theoretical value is 2.11 g/cm³.

Table 2. Atomic parameters. E.s.d. in parenthesis. The β_{ij}-values are computed according to the formula exp $[-(\beta_{11}h^2+\beta_{22}k^2+\beta_{33}l^2+\beta_{12}hk+\beta_{13}hl+\beta_{23}kl)]$. The left-hand index on H denotes the attached carbon atom.

Atom	x	y	z	β_{11}	β_{22}	β_{33}	β_{12}	β_{13}	β_{23}
I_1	−0.09216(5)	−0.10107(6)	0.31189(3)	0.01293(7)	0.02133(9)	0.00487(2)	0.0005(1)	0.00193(6)	−0.00081(7)
I_2	0.17056(4)	0.07421(4)	0.41263(2)	0.01111(6)	0.01458(6)	0.00369(2)	0.0055(1)	0.00573(5)	0.00228(5)
S	0.4154(2)	0.2262(2)	0.5159(1)	0.0135(2)	0.0180(3)	0.00392(7)	−0.0026(4)	0.0081(2)	−0.0034(2)
N	0.6930(5)	0.3141(6)	0.5016(3)	0.0115(7)	0.0136(7)	0.0030(2)	0.000(1)	0.0042(5)	−0.0003(6)
C_1	0.5645(6)	0.2411(6)	0.4633(3)	0.0117(8)	0.0113(7)	0.0032(2)	0.001(1)	0.0051(7)	−0.0011(7)
C_2	0.5490(7)	0.1729(9)	0.3737(4)	0.0134(9)	0.021(1)	0.0044(3)	−0.008(2)	0.0086(9)	−0.0084(9)
C_3	0.5458(9)	0.292(1)	0.3020(4)	0.015(1)	0.031(2)	0.0033(3)	0.010(2)	0.0031(9)	−0.003(1)
C_4	0.7042(9)	0.3513(9)	0.2966(4)	0.019(1)	0.021(1)	0.0042(3)	0.004(2)	0.010(1)	0.004(1)
C_5	0.7985(9)	0.4274(8)	0.3811(5)	0.020(1)	0.017(1)	0.0057(4)	−0.003(2)	0.013(1)	0.000(1)
C_6	0.8284(7)	0.3231(8)	0.4607(4)	0.0102(8)	0.020(1)	0.0043(3)	−0.003(2)	0.0047(8)	−0.0001(9)
C_7	0.7182(8)	0.3877(8)	0.5893(4)	0.018(1)	0.018(1)	0.0031(3)	−0.000(2)	0.0043(8)	−0.0035(8)
H_{21}	0.649	0.096	0.377						
H_{22}	0.439	0.107	0.356						
H_{31}	0.489	0.240	0.238						
H_{32}	0.476	0.390	0.314						
H_{41}	0.773	0.255	0.282						
H_{42}	0.686	0.435	0.243						
H_{51}	0.912	0.464	0.371						
H_{52}	0.733	0.528	0.394						
H_{61}	0.853	0.208	0.440						
H_{62}	0.931	0.366	0.510						
H_{71}	0.711	0.301	0.639						
H_{72}	0.628	0.475	0.587						
H_{73}	0.835	0.441	0.607						

DETERMINATION OF THE STRUCTURE OF MTCL:I_2 (1:1).

Approximate coordinates of the two crystallographically nonequivalent iodine atoms were derived from the three-dimensional Patterson map. The electron density map showed peaks of reasonable peak heights and positions for all atoms, except hydrogen, corresponding to an ordered structure. The R-value, computed on the basis of these positions and an overall isotropic B-value of 4 Å^2, was obtained as 0.195. The structure was refined further, using least squares techniques and anisotropic temperature factors, yielding an R-value of 5.84 % and a weighted R-value of 6.43 %.

The experimental intensity data were then corrected for effects of absorption [4-6] and extinction.[4,7] As a result the atomic parameter values changed, however, only insignificantly.

The positions of the hydrogen atoms of the ring system were now calculated on the basis of a C–H bond length of 1.09 Å [8] and tetrahedral bond angles. The positional parameters thus derived were then included as fixed parameters together with an overall isotropic temperature factor of 4 Å^2 in a least squares refinement of the previous parameters. While most of the intermolecular parameters remained practically unchanged, two of the C–C bond lengths changed by as much as 0.02–0.03 Å by this procedure. The final R-value is 4.87 % and the weighted R-value 5.34 %. Observed and calculated structure factors are given in Table 1 and atomic parameters in Table 2. However, neither the positions of the hydrogen atoms of the cyclic group nor those of the methyl hydrogen atoms showed up distinctly in the three-dimensional electron density difference map.

The matrices of the β_{ij}-values given in Table 2 are all positive definite. The corresponding root-mean square amplitudes of vibration are presented in Table 3. These values served as basis of a least squares analysis of the librational motion of the organic molecule taken as a rigid body.[9,10] The result is given in Table 4.[4]

DISCUSSION OF THE STRUCTURE

Values of intramolecular parameters are given in Table 5 and Fig. 1. The C_1–N and C_1–S bond lengths are not significantly different from

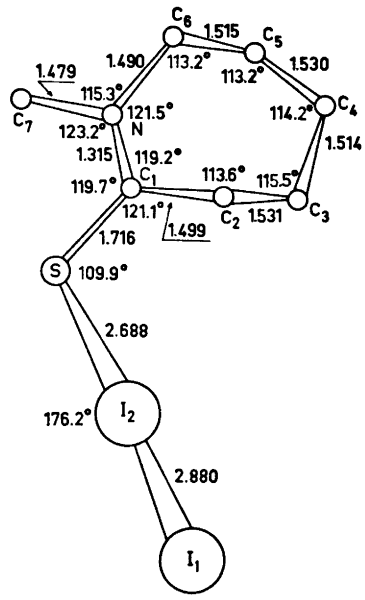

Fig. 1. The molecular complex as view along the [010]-direction of the unit cell.

the corresponding values reported for other thioamides [11] and related compounds.[12-14] The results are consistent with the assumption that the lone electron pair on nitrogen is considerably delocalized over the N, C_1 (and S) centres in the compound.

The bond angles centred at C_1 are close to 120°. The methyl group seems to be somewhat bent away from the sulfur atom, probably for sterical reasons.

The positions of C_1, C_2, C_6, C_7, N, and S deviate slightly, although probably significantly, from a planar arrangement, while C_1, C_2, C_6 and N are found to be co-planar within experimental error.

The cyclic group has the chair conformation, with C_1 and N at the basis and C_4 at the top. The observed C–C bond lengths are near to the corresponding covalent single bond distances.[8] The angles between single bond directions within the ring are found to be somewhat larger than tetrahedral, possibly because of sterical repulsions between non-bonded next-neighbour carbon atoms. The N–C_6 and N–C_7 bond lengths are found to be significantly larger than 1.44 Å observed in 1,3-dimethyl-2(3H)-imidazolethione [13] and quoted for amides.[15] A

Table 3. R.m.s. amplitudes of vibration along principal axes of vibration defined by the components of the unit vector in fractional coordinates.

	R.m.s. ampl.(Å)	Components of unit vectors of principal axes		
S	.276	.0421	−.0983	.0297
	.231	.0924	.0583	.0374
	.164	−.0597	.0178	.0459
C_1	.214	.0911	−.0451	.0448
	.213	.0580	.0993	.0031
	.170	−.0470	.0352	.0487
C_2	.318	.0439	−.0933	.0348
	.210	.0967	.0538	.0299
	.163	−.0510	.0312	.0478
C_3	.359	.0298	.1090	−.0071
	.227	.1137	−.0297	.0156
	.190	.0065	.0192	.0640
C_4	.302	.0627	.0893	.0302
	.258	.0926	−.0688	.0227
	.181	.0380	.0208	−.0545
C_5	.299	.0985	−.0191	.0475
	.261	.0046	.1110	.0164
	.191	.0644	.0212	−.0432
C_6	.280	.0153	−.1137	.0016
	.223	.0493	.0054	.0652
	.183	.1058	.0139	−.0120
N	.229	.0145	.1136	−.0017
	.207	.1123	−.0117	.0342
	.177	−.0325	.0104	.0567
C_7	.275	.0000	−.1101	.0178
	.250	.1173	−.0027	.0114
	.176	.0101	.0317	.0628
I_2	.253	.0560	.0939	.0285
	.201	.0601	−.0634	.0508
	.174	−.0844	.0172	.0316
I_1	.288	.0063	.1132	−.0085
	.248	−.0428	.0178	.0528
	.212	.1095	.0005	.0391

simple, non-trivial explanation of this has not been found.

The I_1-I_2-S group is almost linear, the deviation from linearity is believed to be due to intermolecular interactions in the lattice. The observed iodine-iodine bond length of nearly 2.88 Å is about 0.22 Å longer than in the free molecule and also significantly greater than

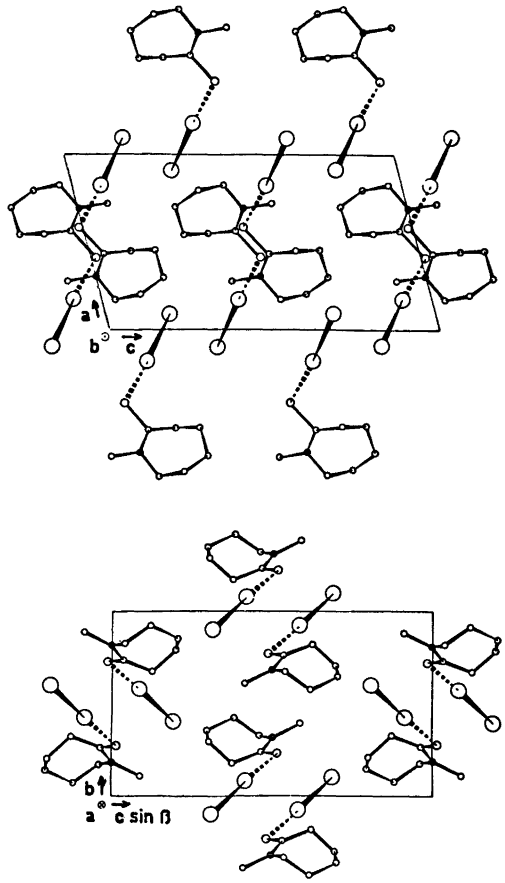

Fig. 2. The [010]- and [100]-projection of the unit cell, respectively.

observed in iodine complexes with thioethers, ethers, and amines.[1] On the other hand, the iodine-sulfur distance of 2.69 Å is significantly shorter than observed in iodine complexes with thioethers. These findings are consistent with the view that the present charge-transfer bond is somewhat stronger than that observed in complexes between iodine and thioethers.[1a–1d] The sulfur atom, no doubt, acts as an electron donor towards the electrophilic iodine molecule. The delocalization of the lone electron pair on nitrogen leads to an accumulation of negative charge on the thioamide sulfur atom in excess of that existing in thioethers. Hence the former sulfur should be able to donate electrons more strongly to the acceptor than the sulfur atom in thioethers, giving rise to a stronger charge-transfer bond.[2]

Table 4. The results of the rigid body analysis. Mass centre of MTCL in (0.612, 0.292, 0.446). $\Delta u_{rms} = 0.0032$ Å.[10]

$$\widetilde{T} = \begin{Bmatrix} 37(2) & 5(2) & 4(1) \\ & 47(3) & -2(2) \\ & & 33(2) \end{Bmatrix} \times 10^{-4} \text{ Å}^2$$

Principal axes of $\widetilde{T}$:

Eigenvalue (Å²)	R.M.S.(Å)	Direction cosines I_1	I_2	I_3
0.0496	0.223	0.3861	0.9223	0.0165
0.0385	0.196	0.6914	−0.3010	0.6567
0.0286	0.169	0.6106	−0.2421	−0.7540

$$\widetilde{L} = \begin{Bmatrix} 9(1) & -2(1) & -8(1) \\ & 3(1) & 1(1) \\ & & 11(1) \end{Bmatrix} \times 10^{-4} \text{rad.}^2$$

Principal axes of $\widetilde{L}$:

Eigenvalue (rad²)	R.M.S. (rad.)	Direction cosines I_1	I_2	I_3
0.0190	0.138	−0.6571	0.1219	0.7439
0.0036	0.060	−0.2684	0.8843	−0.3821
0.0014	0.038	−0.7044	−0.4507	−0.5484

$$\widetilde{S} = \begin{Bmatrix} -7(13) & 67(6) & -23(8) \\ 1(5) & -11(11) & -12(5) \\ -29(8) & -38(6) & 18(40) \end{Bmatrix} \times 10^{-4} \text{ rad. Å}$$

Trace of S has been set = 0

Principal axes of $\widetilde{S}$.

Eigenvalue (rad.Å)	Direction cosines I_1	I_2	I_3
0.0038	−0.5097	−0.4140	0.7542
0.0002	0.2043	0.7933	0.5735
0.0040	−0.8358	0.4464	−0.3197

Displacement of libration axes (Å) ($^{j}\varrho_i$ = displ. of L_j along L_i) $^{1}\varrho_2 = -0.159$, $^{2}\varrho_1 = 0.068$, $^{3}\varrho_1 = 2.038$, $^{1}\varrho_3 = -0.373$, $^{2}\varrho_3 = 0.531$, and $^{3}\varrho_2 = -0.508$. Effective cross translation parallel with the L_1, L_2, and L_3-axes (Å): 0.016, −0.019, and −0.029, respectively.

The C_1-S-I_2 bond angle is close to 110° and the iodine molecule is nearly co-planar with the $S-C_1-C_2$ group. Because of the double bond character of the C_1-S bond, a strong electron density due to one of the lone pairs on the sulfur atom should be set up in directions close to the $S-I_2$ direction. This orbital is therefore believed to be the main source of charge-transfer on complex formation. If sp^2-hybridized orbitals are not formed on sulfur, as has been claimed,[16] the lone pair orbitals are pure p-orbitals, the deviation of $\angle C_1-S-I_2$ from 90° originating in sterical interactions.

A reasonable description of bond and symmetry properties of the $S-I_2-I_1$ group in the ground state is provided by a model, wherein the four σ-electrons of the $S-I_2-I_1$ group are assumed axially delocalized in the complex giving rise to three-centre bonds, centred at the S, I_2 and I_1 sites. This principle is frequently used to describe related molecular complex structures.[17]

Table 5. Intramolecular parameters.

a. Bond lengths (Å) and angles (°).

	Observed	Corrected		Observed
I_1-I_2	2.880(1)		I_1-I_2-S	176.21(04)
I_2-S	2.688(2)		I_2-S-C_1	109.9 (2)
$S-C_1$	1.716(5)	1.721	$S-C_1-C_2$	121.1 (4)
C_1-C_2	1.493(7)	1.499	$C_1-C_2-C_3$	113.3 (6)
C_2-C_3	1.523(10)	1.531	$C_2-C_3-C_4$	115.5 (6)
C_3-C_4	1.506(11)	1.514	$C_3-C_4-C_5$	114.2 (5)
C_4-C_5	1.520(10)	1.530	$C_4-C_5-C_6$	113.2 (6)
C_5-C_6	1.508(9)	1.515	C_5-C_6-N	113.2 (5)
C_6-N	1.487(7)	1.490	C_6-N-C_7	115.3 (5)
$N-C_7$	1.476(7)	1.479	C_6-N-C_1	121.5 (4)
$N-C_1$	1.302(7)	1.315	$N-C_1-C_2$	119.2 (4)
			C_1-N-C_7	123.2 (5)
			$N-C_1-S$	119.7 (4)

b. Atomic deviations (Å) from least squares plane through C_1, C_2, C_6, C_7, N, and S.

C_1	C_2	C_6	C_7	N	S	I_1	I_2
−0.07	−0.024	0.039	−0.023	−0.015	0.012	0.051	−0.039

The atoms execute pronounced vibrational motion in the solid. The outer halogen atom vibrates more strongly than the central one, as is to be expected (Table 3).

Results of the rigid body motion analysis for the organic molecule are shown in Table 4. The root mean square deviation of the Δu(ij)'s[10] is relatively close to the values reported for other cyclic systems.[18] The magnitude of the figure indicates that the rigid-body motion approximation is acceptable, although not a good one,[10] as to be expected. It appears that the rotational oscillation is quite marked, especially about one of the principal axes. This gave rise to bond length corrections [4,10] ranging from 0.004 Å to 0.013 Å (Table 5).

The methyl group is likely to reorientate about the $N-C_7$ bond. The shortest S−H separation is 2.38 Å, assuming a C−H bond length of 1.09 Å and tetrahedral H−C−C bond angles. This is considerably shorter than expected for a normal S−H van der Waals contact distance.[14] The region of minimum potential energy corresponds most likely to the maximum S−H separation of 2.89 Å, which occurs when each of two hydrogen atoms are equally distant from the sulfur atom. None of the H−H separations or other types of separation involving the methyl group in this position, are less than the corresponding normal contact distance. These interactions can therefore hardly displace the methyl group appreciably from the predicted equilibrium orientation.

Other short intramolecular distances between "non-bonded" atoms of particular interest are $S-C_2=2.80$ Å, $S-N=2.62$ Å, $S-C_7=2.97$ Å, $I_2-C_1=3.65$ Å, and $I_2-C_2=3.63$ Å.

The structure viewed along the [010]- and [100]-directions of the unit cell is shown in Fig. 2. Intermolecular distances are of the van der Waals type.

The crystallographic computer program used is quoted in Ref. 4.

Acknowledgement. Thanks are due to mag. scient. E. Augdahl for helpful discussions and the gift of a sample of MTCL.

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Received September 17, 1973.