546. *The Stereochemistry of Molecules Containing the* C=C=N *Group. Part III.*[1] *The Crystal and Molecular Structure of* N-*Ethyl*-2,2′-*dimethylsulphonylvinylideneamine.*

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A three-dimensional structural analysis of a third compound containing the >C=C=NR group is described. Crystals of *N*-ethyl-2,2′-dimethylsulphonylvinylideneamine are orthorhombic $P2_12_12_1$ with $a = 12{\cdot}02$, $b = 5{\cdot}85$, $c = 14{\cdot}47$ Å. The bond angle at the nitrogen atom is 144°, in sharp contrast to the value of 180° found in two other vinylideneamines in which R = Me. The bond lengths and other bond angles exhibit no unusual features, except the C=N bond which has the length of a normal C≡N bond. It is suggested that the linearity of the −N= system in *N*-methylvinylideneamines is due, at least partly, to hyperconjugation.

It has been shown in Parts I [2] and II [1] that the −N= system in two *N*-methylvinylideneamines is linear. In order for this system to achieve linearity the lone pair of electrons on the nitrogen atom must be absorbed into the rest of the molecule. In Part II it was suggested that hyperconjugation or electronegativity, or both, might account for the transfer of the lone pair. In an attempt to assess the relative importance of these two effects the crystal structure of *N*-ethyl-2,2′-dimethylsulphonylvinylideneamine, $(CH_3{\cdot}SO_2)_2C{=}C{=}NEt$, has been determined. Although some doubt has recently been cast on the importance of the part played by hyperconjugation in the ground state of molecules,[3,4] the results of the analysis show that hyperconjugation must play some part in the bonding of *N*-methylvinylideneamines.

Experimental.—The synthesis of *N*-ethyl-2,2′-dimethylsulphonylvinylideneamine will be described separately.[5]

Molecular formula: $C_6H_{11}O_4S_2N$, $M = 225{\cdot}29$. Orthorhombic, $a = 12{\cdot}02 \pm 0{\cdot}04$ Å, $b = 5{\cdot}85 \pm 0{\cdot}02$ Å, $c = 14{\cdot}47 \pm 0{\cdot}04$ Å. $U = 1017$ Å^3. $D_m = 1{\cdot}44$ g./c.c. (by flotation), $Z = 4$, $D_c = 1{\cdot}44$ g./c.c. $F(000) = 472$. Space group $P2_12_12_1$ (D_2^4 No. 41), origin at 1/4,1/4,1/4 from the Int. Tab. origin.[6] Cu-K_α radiation, $\mu = 44{\cdot}3$ cm.$^{-1}$, single crystal oscillation and Weissenberg photographs about the *a*, *b*, and *c* axes. No correction was made for absorption.

The relative intensities of 1120 reflections were estimated by eye from a calibration strip and were placed on the same scale by equating $\sum|F_o|$ in common reciprocal rows. An approximate scale and temperature factor were obtained from *h0l* intensities by the method of Howells, Phillips, and Rogers.[7] A further 176 reflections, the intensities of which were too low to be observed, were given a value of one-half of the minimum observable intensity and are included in the *R*-values quoted.

The *R*-value used in this paper is the residual $\dfrac{\sum\big||F_o| - |F_c|\big|}{\sum|F_o|}$.

Structure determination. The positions of the sulphur atoms were determined from the Patterson functions of the *a*- and *b*-axis projections. Fourier methods (in projection) were then used to obtain a complete set of co-ordinates. An attempt was made to refine these co-ordinates three-dimensionally by using 390 low-order planes. The least-squares method was used and the calculations were done on the Leeds University Pegasus computer. The programmes were devised by Dr. D. W. J. Cruickshank and Miss D. E. Pilling. The weights used were $w = 1$ throughout.

After four refinement cycles the residual fell slowly to 0·27, so the calculated phase angles were used to evaluate the electron density in three dimensions. This electron density showed that sites of the carbon atoms of the ethyl group (C5 and C6; see Fig. 1) were in error. New

[1] Part II, Bullough and Wheatley, *Acta Cryst.*, 1957, **10**, 233.
[2] Wheatley, *Acta Cryst.*, 1954, **7**, 68.
[3] Rao, *Nature*, 1960, **187**, 913.
[4] Dewar and Schmeising, *Tetrahedron*, 1959, **5**, 166.
[5] Challenger, unpublished work.
[6] "International Tables for X-Ray Crystallography," Vol. I, Kynoch Press, Birmingham, 1952.
[7] Howells, Phillips, and Rogers, *Acta Cryst.*, 1950, **3**, 210.

positions were found for these atoms and the trial structure thus produced was eventually refined by using all 1296 intensities. The final value of the residual (after anisotropic refinement of the temperature factors) was 0·10.

Results.—The results of the analysis are summarized in the Tables and Figures.

TABLE 1. *Final co-ordinates* (Å) *for* $(CH_3{\cdot}SO_2)_2C{=}C{=}N{\cdot}C_2H_5$.

	x	*y*	*z*		*x*	*y*	*z*
S 1	7·6002	1·2262	8·1286	C 1	7·1234	2·6999	8·9912
S 2	5·1042	0·9074	6·4295	C 2	4·9572	−0·8507	6·5980
O 1	9·0194	1·2757	7·8849	C 3	6·7959	1·2852	6·5881
O 2	4·7436	1·2630	5·0811	C 4	7·4642	1·5700	5·4431
O 3	7·0605	0·1090	8·8447	C 5	8·4458	2·6217	3·3902
O 4	4·4080	1·5310	7·5144	C 6	7·6254	3·9130	3·4620
N 1	8·0475	1·6813	4·4405				

FIG. 1. *Numbering of the atoms and the bond lengths in* $(CH_3{\cdot}SO_2)_2C{=}C{=}NEt$.

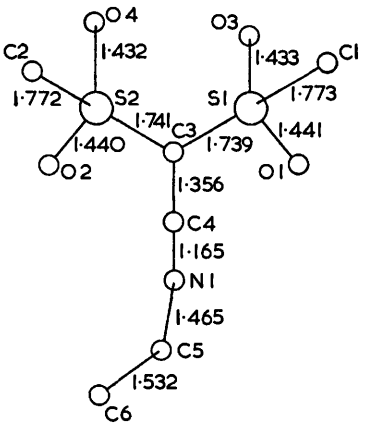

FIG. 2. *The unit cell contents viewed down the b-axis, and some of the more important van der Waals distances.*

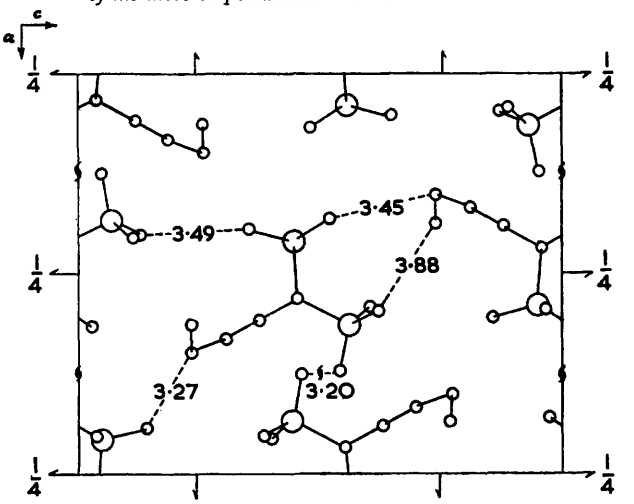

TABLE 2. U_{ij} (Å^2) *for* $(CH_3{\cdot}SO_2)_2C{=}C{=}N{\cdot}C_2H_5$.

	U_{11}	U_{22}	U_{33}	$2U_{12}$	$2U_{23}$	$2U_{13}$
S 1	0·0375	0·0388	0·0404	0·0032	0·0075	−0·0037
S 2	0·0343	0·0698	0·0492	−0·0014	0·0164	−0·0085
O 1	0·0385	0·0602	0·0581	0·0089	0·0053	−0·0156
O 2	0·0563	0·1131	0·0659	−0·0247	0·0451	−0·0470
O 3	0·0670	0·0428	0·0466	−0·0111	0·0252	−0·0025
O 4	0·0437	0·0860	0·0678	0·0180	−0·0033	0·0201
N 1	0·0482	0·0734	0·0397	−0·0270	−0·0101	0·0064
C 1	0·0616	0·0485	0·0584	0·0052	−0·0118	−0·0042
C 2	0·0694	0·0567	0·0923	−0·0182	−0·0177	−0·0035
C 3	0·0466	0·0559	0·0423	−0·0069	0·0155	−0·0113
C 4	0·0435	0·0503	0·0511	0·0048	0·0078	−0·0069
C 5	0·0661	0·0800	0·0434	−0·0386	0·0160	0·0167
C 6	0·0691	0·0846	0·0821	−0·0161	0·0308	−0·0433

TABLE 3. *Standard deviations* (Å) *of the co-ordinates and* σ_{rms} *for* $(CH_3{\cdot}SO_2)_2C{=}C{=}N{\cdot}C_2H_5$.

	x	*y*	*z*	σ_{rms}		*x*	*y*	*z*	σ_{rms}
S 1	0·0023	0·0025	0·0025	0·0024	C 1	0·0121	0·0117	0·0117	0·0118
S 2	0·0025	0·0032	0·0029	0·0029	C 2	0·0129	0·0129	0·0143	0·0134
O 1	0·0064	0·0079	0·0070	0·0071	C 3	0·0096	0·0113	0·0095	0·0102
O 2	0·0079	0·0113	0·0081	0·0092	C 4	0·0098	0·0111	0·0101	0·0103
O 3	0·0081	0·0076	0·0071	0·0076	C 5	0·0125	0·0143	0·0111	0·0127
O 4	0·0071	0·0098	0·0078	0·0083	C 6	0·0132	0·0158	0·0136	0·0142
N 1	0·0084	0·0110	0·0080	0·0093					

TABLE 4. *Bond lengths and bond angles, and their standard deviations in* $(CH_3{\cdot}SO_2)_2C{=}C{=}N{\cdot}C_2H_5$.

Bond	Length (Å)	σ (Å)	Angle	Size	(σ)	Angle	Size	(σ)
S1–O1	1·441	0·0075	S1–C3–S2	121° 32′	0° 36′	O2–S2–C2	108° 15′	0° 35′
S2–O2	1·440	0·0097	S1–C3–C4	121° 49′	0° 48′	O2–S2–C3	105° 58′	0° 31′
S1–O3	1·433	0·0080	S2–C3–C4	116° 36′	0° 47′	O3–S1–C1	107° 41′	0° 31′
S2–O4	1·432	0·0088	N1–C4–C3	173° 21′	1° 08′	O3–S1–C3	107° 10′	0° 30′
S1–C1	1·773	0·0121	N1–C5–C6	111° 13′	1° 02′	O4–S2–C2	108° 38′	0° 34
S2–C2	1·772	0·0137	O1–S1–O3	118° 51′	0° 27′	O4–S2–C3	108° 00′	0° 30′
S1–C3	1·739	0·0104	O1–S1–C1	108° 35′	0° 30′	C1–S1–C3	106° 10′	0° 31′
S2–C3	1·741	0·0106	O1–S1–C3	107° 44′	0° 28′	C2–S2–C3	106° 42′	0° 34′
N1–C4	1·165	0·0139	O2–S2–O4	118° 41′	0° 32′	C4–N1–C5	144° 31′	1° 03′
N1–C5	1·465	0·0157						
C3–C4	1·356	0·0145						
C5–C6	1·532	0·0191						

TABLE 5. *Standard deviations* (Å²) *of the* U_{ij} *for* $(CH_3{\cdot}SO_2)_2C{=}C{=}N{\cdot}C_2H_5$.

	U_{11}	U_{22}	U_{33}	$2U_{12}$	$2U_{23}$	$2U_{13}$
S 1	0·0010	0·0011	0·0011	0·0019	0·0019	0·0018
S 2	0·0011	0·0017	0·0013	0·0025	0·0028	0·0021
O 1	0·0032	0·0044	0·0042	0·0070	0·0081	0·0066
O 2	0·0046	0·0079	0·0049	0·0120	0·0117	0·0084
O 3	0·0046	0·0039	0·0039	0·0079	0·0070	0·0074
O 4	0·0038	0·0062	0·0048	0·0093	0·0104	0·0075
N 1	0·0045	0·0064	0·0043	0·0104	0·0098	0·0077
C 1	0·0066	0·0060	0·0065	0·0121	0·0115	0·0118
C 2	0·0074	0·0071	0·0093	0·0134	0·0150	0·0148
C 3	0·0050	0·0059	0·0050	0·0101	0·0102	0·0088
C 4	0·0049	0·0057	0·0053	0·0103	0·0104	0·0090
C 5	0·0072	0·0096	0·0059	0·0151	0·0136	0·0114
C 6	0·0078	0·0100	0·0088	0·0160	0·0176	0·0146

TABLE 6. *Minimum van der Waals contacts* (Å) *in* $(CH_3{\cdot}SO_2)_2C{=}C{=}N{\cdot}C_2H_5$.

O–O	C–O	C–C	O–N	C–N
3·2	3·3	3·9	3·4	3·6

TABLE 7. *List of* $|F_o|$, $|F_c|$, A_c, *and* B_c (*for* N-*ethyl*-2,2′-*dimethylsulphonylvinylidene-amine*) *on ten times the absolute scale.*

h	k	l	$\|F_o\|$	$\|F_c\|$	A_c	B_c	h	k	l	$\|F_o\|$	$\|F_c\|$	A_c	B_c	h	k	l	$\|F_o\|$	$\|F_c\|$	A_c	B_c
0	0	2	1058	1138	1138	0	2	0	12	61	55	−55	0	4	0	14	81	80	−80	0
0	0	4	151	129	129	0	2	0	13	302	284	−284	0	4	0	15	92	73	−73	0
0	0	6	475	449	−449	0	2	0	14	25	3	3	0	4	0	16	139	130	−130	0
0	0	8	400	349	−349	0	2	0	15	107	108	−108	0	4	0	17	16	1	−1	0
0	0	10	503	502	−502	0	2	0	16	21	19	19	0	4	0	18	31	55	−55	0
0	0	12	305	279	−279	0	2	0	17	26	32	−32	0							
0	0	14	26	3	3	0	2	0	18	47	41	41	0	5	0	1	151	139	0	−139
0	0	16	226	197	197	0								5	0	2	55	47	0	−47
0	0	18	116	116	116	0	3	0	1	341	315	0	−315	5	0	3	118	105	0	105
							3	0	2	149	141	0	141	5	0	4	29	33	0	−33
1	0	1	412	458	0	458	3	0	3	530	485	0	−485	5	0	5	179	159	0	159
1	0	2	170	207	0	207	3	0	4	595	634	0	634	5	0	6	127	134	0	−134
1	0	3	180	158	0	−158	3	0	5	447	438	0	−438	5	0	7	389	368	0	368
1	0	4	63	30	0	30	3	0	6	396	389	0	389	5	0	8	183	173	0	−173
1	0	5	382	345	0	−345	3	0	7	20	39	0	39	5	0	9	308	297	0	297
1	0	6	289	293	0	293	3	0	8	6	14	0	14	5	0	10	26	21	0	−21
1	0	7	235	218	0	−218	3	0	9	355	365	0	365	5	0	11	26	34	0	34
1	0	8	323	304	0	304	3	0	10	34	27	0	−27	5	0	12	159	153	0	153
1	0	9	123	122	0	−122	3	0	11	135	102	0	102	5	0	13	26	38	0	−38
1	0	10	67	72	0	−72	3	0	12	213	191	0	−191	5	0	14	25	4	0	−4
1	0	11	25	16	0	16	3	0	13	26	23	0	−23	5	0	15	160	154	0	−154
1	0	12	93	83	0	−83	3	0	14	26	2	0	−2	5	0	16	46	44	0	44
1	0	13	166	151	0	−151	3	0	15	29	15	0	15	5	0	17	94	93	0	−93
1	0	14	106	88	0	−88	3	0	16	46	51	0	−51							
1	0	15	96	102	0	+102	3	0	17	47	45	0	45	6	0	0	630	664	−664	0
1	0	16	22	24	0	−24	3	0	18	48	46	0	46	6	0	1	173	151	−151	0
1	0	17	47	42	0	42								6	0	2	517	508	−508	0
1	0	18	56	65	0	−65	4	0	0	1340	1397	−1397	0	6	0	3	215	194	−194	0
							4	0	1	705	678	678	0	6	0	4	166	176	176	0
2	0	0	179	164	164	0	4	0	2	340	335	−335	0	6	0	5	116	111	−111	0
2	0	1	165	203	203	0	4	0	3	280	250	250	0	6	0	6	165	145	145	0
2	0	2	439	397	397	0	4	0	4	225	208	208	0	6	0	7	78	64	−64	0
2	0	3	1293	1337	1337	0	4	0	5	112	150	150	0	6	0	8	91	83	83	0
2	0	4	379	352	−352	0	4	0	6	129	126	126	0	6	0	9	75	66	66	0
2	0	5	222	211	211	0	4	0	7	21	39	39	0	6	0	10	260	247	247	0
2	0	6	79	55	−55	0	4	0	8	89	57	57	0	6	0	11	170	152	152	0
2	0	7	111	118	118	0	4	0	9	8	20	20	0	6	0	12	173	148	148	0
2	0	8	20	29	−29	0	4	0	10	151	142	142	0	6	0	13	105	104	104	0
2	0	9	103	97	−97	0	4	0	11	26	7	−7	0	6	0	14	68	58	58	0
2	0	10	48	69	−69	0	4	0	12	87	65	65	0	6	0	15	21	27	27	0
2	0	11	314	310	−310	0	4	0	13	56	62	−62	0	6	0	16	18	11	−11	0

TABLE 7. (*Continued.*)

h	k	l	$\|F_o\|$	$\|F_c\|$	A_c	B_c	h	k	l	$\|F_o\|$	$\|F_c\|$	A_c	B_c	h	k	l	$\|F_o\|$	$\|F_c\|$	A_c	B_c
6	0	17	11	6	6	0	13	0	1	46	36	0	−36	3	1	10	60	39	−36	−16
							13	0	2	67	55	0	55	3	1	11	275	281	52	276
7	0	1	156	127	0	−127	13	0	3	27	25	0	−25	3	1	12	75	84	−77	−34
7	0	2	268	262	0	−262	13	0	4	148	140	0	140	3	1	13	154	155	−49	147
7	0	3	60	68	0	−68	13	0	5	55	54	0	54	3	1	14	89	84	−44	72
7	0	4	591	607	0	−607	13	0	6	168	167	0	167	3	1	15	46	42	18	38
7	0	5	230	234	0	234	13	0	7	55	47	0	47	3	1	16	83	101	−49	89
7	0	6	535	562	0	−562	13	0	8	38	36	0	36	3	1	17	16	31	−23	−21
7	0	7	115	97	0	97	13	0	9	103	94	0	94	3	1	18	11	16	9	13
7	0	8	227	221	0	−221														
7	0	9	71	49	0	−49	14	0	0	142	128	128	0	4	1	0	80	68	0	−68
7	0	10	176	164	0	164	14	0	1	45	25	25	0	4	1	1	163	180	−179	20
7	0	11	36	56	0	56	14	0	2	129	118	118	0	4	1	2	366	337	−141	−306
7	0	12	80	215	0	215	14	0	3	8	2	2	0	4	1	3	147	147	146	−20
7	0	13	24	4	0	4	14	0	4	66	55	55	0	4	1	4	380	348	348	19
7	0	14	129	111	0	111	14	0	5	7	15	−15	0	4	1	5	36	27	−23	−14
7	0	15	19	17	0	−17	14	0	6	92	85	−85	0	4	1	6	213	181	180	−18
7	0	16	38	39	0	39	14	0	7	9	19	−19	0	4	1	7	235	216	199	−84
														4	1	8	196	183	−33	180
8	0	0	212	192	192	0	15	0	1	98	90	0	−90	4	1	9	245	214	203	−69
8	0	1	240	215	−215	0	15	0	2	6	19	0	−19	4	1	10	193	183	−131	128
8	0	2	136	135	135	0	15	0	3	53	51	0	−51	4	1	11	25	27	27	5
8	0	3	159	174	−174	0	15	0	4	4	11	0	11	4	1	12	85	54	−40	36
8	0	4	45	16	−16	0								4	1	13	25	13	−12	3
8	0	5	9	26	26	0	0	1	1	266	264	0	−264	4	1	14	59	40	−5	−40
8	0	6	11	5	−5	0	0	1	2	190	241	241	0	4	1	15	87	77	−77	−4
8	0	7	83	69	−69	0	0	1	3	448	354	0	354	4	1	16	47	47	30	−36
8	0	8	63	63	63	0	0	1	4	635	652	−652	0	4	1	17	36	41	−40	6
8	0	9	39	34	−34	0	0	1	5	40	11	0	−11							
8	0	10	78	45	−45	0	0	1	6	276	270	−270	0	5	1	0	611	589	0	589
8	0	11	72	53	−53	0	0	1	7	305	283	0	283	5	1	1	297	285	−281	−47
8	0	12	24	22	22	0	0	1	8	21	19	−19	0	5	1	2	462	431	74	425
8	0	13	21	20	20	0	0	1	9	6	27	0	27	5	1	3	102	101	−89	−49
8	0	14	60	51	51	0	0	1	10	48	23	23	0	5	1	4	205	206	102	−179
8	0	15	15	29	29	0	0	1	11	34	55	0	−55	5	1	5	120	112	92	63
							0	1	12	26	24	24	0	5	1	6	76	67	46	−49
9	0	1	374	344	0	344	0	1	13	94	77	0	−77	5	1	7	175	170	95	−140
9	0	2	128	113	0	−113	0	1	14	35	48	−48	0	5	1	8	125	117	3	−117
9	0	3	180	182	0	182	0	1	15	25	6	0	−6	5	1	9	153	144	138	−43
9	0	4	193	182	0	−182	0	1	16	22	21	−21	0	5	1	10	218	209	12	−209
9	0	5	72	62	0	−62	0	1	17	19	5	0	−5	5	1	11	182	155	18	−154
9	0	6	225	230	0	−230	0	1	18	47	51	−51	0	5	1	12	92	80	−19	−77
9	0	7	312	301	0	−301								5	1	13	56	58	39	−44
9	0	8	45	12	0	12	1	1	0	1352	1437	0	−1437	5	1	14	65	60	−7	60
9	0	9	343	336	0	−336	1	1	1	523	559	534	−167	5	1	15	21	27	−26	9
9	0	10	80	70	0	70	1	1	2	694	744	274	−692	5	1	16	65	63	−24	58
9	0	11	58	168	0	−168	1	1	3	529	540	157	−516	5	1	17	14	17	−17	2
9	0	12	22	33	0	33	1	1	4	389	338	306	143							
9	0	13	20	31	0	31	1	1	5	443	431	−111	−416	6	1	0	95	119	0	−119
9	0	14	68	65	0	65	1	1	6	98	107	96	45	6	1	1	192	211	−175	−118
9	0	15	72	68	0	68	1	1	7	248	237	−216	−98	6	1	2	152	141	−129	−57
							1	1	8	323	322	92	309	6	1	3	80	90	−18	−88
10	0	0	58	57	−57	0	1	1	9	146	137	−135	−21	6	1	4	262	258	−249	−65
10	0	1	29	41	−41	0	1	1	10	272	269	−85	255	6	1	5	279	272	211	−171
10	0	2	149	149	−149	0	1	1	11	307	304	−100	287	6	1	6	134	109	−58	92
10	0	3	76	65	−65	0	1	1	12	139	132	0	132	6	1	7	348	335	304	−142
10	0	4	139	137	−137	0	1	1	13	188	175	−1	175	6	1	8	119	120	−97	70
10	0	5	59	47	−47	0	1	1	14	114	106	−82	−67	6	1	9	162	139	138	−13
10	0	6	11	32	32	0	1	1	15	71	60	4	60	6	1	10	91	85	72	46
10	0	7	56	32	32	0	1	1	16	114	114	−2	−113	6	1	11	135	120	90	79
10	0	8	154	138	138	0	1	1	17	51	61	56	26	6	1	12	160	145	145	−9
10	0	9	122	113	−113	0	1	1	18	69	68	−15	−66	6	1	13	82	92	−36	85
10	0	10	41	37	37	0								6	1	14	63	46	42	−19
10	0	11	48	49	−49	0	2	1	0	437	387	0	−387	6	1	15	99	93	−88	30
10	0	12	19	15	15	0	2	1	1	366	341	231	251	6	1	16	32	39	31	−24
10	0	13	16	29	−29	0	2	1	2	322	332	306	129	6	1	17	65	97	−97	−2
10	0	14	11	17	−17	0	2	1	3	129	151	−145	40							
							2	1	4	514	535	506	−173	7	1	0	71	70	0	−70
11	0	1	83	74	0	74	2	1	5	315	331	−181	277	7	1	1	319	291	−175	232
11	0	2	133	136	0	136	2	1	6	339	305	304	15	7	1	2	161	157	−101	−121
11	0	3	52	62	0	62	2	1	7	392	370	−354	107	7	1	3	148	137	−36	133
11	0	4	187	188	0	188	2	1	8	205	191	187	38	7	1	4	101	102	−98	28
11	0	5	111	97	0	−97	2	1	9	95	78	−60	51	7	1	5	134	136	19	134
11	0	6	279	254	0	254	2	1	10	133	124	3	124	7	1	6	95	88	−64	−60
11	0	7	94	86	0	−86	2	1	11	88	81	−29	−76	7	1	7	168	170	72	154
11	0	8	96	78	0	78	2	1	12	136	130	−128	24	7	1	8	25	15	−15	−3
11	0	9	61	66	0	−66	2	1	13	92	77	−53	−56	7	1	9	54	38	34	−17
11	0	10	99	99	0	−99	2	1	14	48	59	−58	−10	7	1	10	88	78	−10	−78
11	0	11	26	14	0	−14	2	1	15	94	95	90	−31	7	1	11	119	102	43	−92
11	0	12	118	103	0	−103	2	1	16	63	64	−62	−14	7	1	12	25	39	30	−25
							2	1	17	74	68	68	−3	7	1	13	72	75	19	−73
12	0	0	56	46	46	0	2	1	18	12	18	−18	4	7	1	14	65	57	0	−57
12	0	1	14	14	−14	0								7	1	15	35	39	11	−37
12	0	2	53	41	41	0	3	1	0	702	747	0	747	7	1	16	41	43	26	−35
12	0	3	78	84	−84	0	3	1	1	356	317	−35	−315							
12	0	4	8	6	−6	0	3	1	2	423	427	355	238	8	1	0	115	122	0	122
12	0	5	123	116	−116	0	3	1	3	47	74	−2	−74	8	1	1	152	155	151	−34
12	0	6	11	28	28	0	3	1	4	301	293	284	−71	8	1	2	263	269	−259	72
12	0	7	38	32	−32	0	3	1	5	39	30	−9	−29	8	1	3	175	173	−55	−164
12	0	8	78	71	−71	0	3	1	6	156	160	147	64	8	1	4	452	467	−464	46
12	0	9	72	67	67	0	3	1	7	79	92	74	55	8	1	5	160	162	22	−161
12	0	10	18	5	−5	0	3	1	8	151	151	−11	−151	8	1	6	456	465	−464	−29
12	0	11	79	68	68	0	3	1	9	112	120	−9	119	8	1	7	176	165	−134	−95

TABLE 7. (*Continued.*)

h	k	l	$\|F_o\|$	$\|F_c\|$	A_c	B_c
8	1	8	160	138	−128	−52
8	1	9	163	140	−139	11
8	1	10	155	162	128	−99
8	1	11	98	96	−64	71
8	1	12	127	110	109	−15
8	1	13	80	68	13	67
8	1	14	109	114	114	13
8	1	15	46	48	28	38
9	1	0	32	23	0	−23
9	1	1	75	56	−29	−49
9	1	2	85	55	−55	−1
9	1	3	52	51	−32	−40
9	1	4	133	129	−66	110
9	1	5	161	160	22	−158
9	1	6	11	21	−21	−4
9	1	7	38	21	6	−20
9	1	8	103	90	−14	−89
9	1	9	69	48	14	46
9	1	10	35	26	19	19
9	1	11	92	75	16	74
9	1	12	21	20	−5	−19
9	1	13	38	46	9	45
9	1	14	31	43	18	−39
9	1	15	26	24	−9	22
10	1	0	145	127	0	127
10	1	1	192	184	184	7
10	1	2	149	153	99	117
10	1	3	86	91	81	42
10	1	4	115	105	101	29
10	1	5	158	155	−155	5
10	1	6	67	56	40	−39
10	1	7	207	192	−186	51
10	1	8	143	143	73	−123
10	1	9	169	162	−162	8
10	1	10	72	72	−24	−67
10	1	11	89	87	−87	−3
10	1	12	45	48	−41	−25
10	1	13	36	43	38	−21
10	1	14	16	34	−19	28
11	1	0	98	95	0	95
11	1	1	38	31	30	7
11	1	2	165	163	−55	154
11	1	3	98	92	12	92
11	1	4	58	58	−54	19
11	1	5	100	97	−2	97
11	1	6	78	80	−59	−54
11	1	7	13	25	2	25
11	1	8	46	43	2	−43
11	1	9	31	30	−1	30
11	1	10	20	29	4	−29
11	1	11	19	3	−3	0
11	1	12	14	26	26	−1
12	1	0	13	5	0	5
12	1	1	47	45	−8	45
12	1	2	95	98	96	16
12	1	3	63	60	26	54
12	1	4	120	126	126	7
12	1	5	91	92	15	91
12	1	6	160	167	166	−16
12	1	7	39	35	19	29
12	1	8	27	25	15	−20
12	1	9	42	36	34	13
12	1	10	86	85	−85	−3
12	1	11	40	58	28	−51
13	1	0	47	48	0	−48
13	1	1	83	75	67	33
13	1	2	56	56	−10	−56
13	1	3	56	50	36	35
13	1	4	45	54	−29	−46
13	1	5	60	72	−21	69
13	1	6	33	33	1	33
13	1	7	27	31	−31	1
13	1	8	74	74	−1	74
13	1	9	78	91	−38	−82
14	1	0	34	36	0	−36
14	1	1	81	80	−80	3
14	1	2	42	40	0	−40
14	1	3	55	49	−29	40
14	1	4	59	63	63	−3
14	1	5	58	59	46	37
14	1	6	56	52	52	−2
14	1	7	48	60	51	32
15	1	0	118	127	0	−127
15	1	1	39	48	46	−16
15	1	2	108	133	18	−132
0	2	0	1199	1183	−1183	0
0	2	1	457	497	0	−497
0	2	2	671	681	−681	0
0	2	3	252	239	0	−239
0	2	4	352	325	−325	0
0	2	5	149	147	0	147
0	2	6	78	67	−67	0
0	2	7	288	278	0	278
0	2	8	475	458	458	0
0	2	9	356	333	0	333
0	2	10	343	336	336	0
0	2	11	183	178	0	178
0	2	12	133	105	105	0
0	2	13	28	33	0	−33
0	2	14	87	74	−74	0
0	2	15	80	55	0	−55
0	2	16	103	91	−91	0
0	2	17	87	90	0	−90
1	2	0	55	39	39	0
1	2	1	207	212	104	−185
1	2	2	587	512	195	−473
1	2	3	115	110	83	−72
1	2	4	147	148	100	109
1	2	5	140	121	108	−53
1	2	6	243	241	−62	233
1	2	7	350	352	63	346
1	2	8	31	25	−22	−11
1	2	9	216	211	54	204
1	2	10	160	159	−38	−155
1	2	11	109	108	−71	82
1	2	12	63	66	1	−66
1	2	13	52	39	−39	4
1	2	14	35	52	−2	52
1	2	15	56	66	−28	−60
1	2	16	49	58	3	58
1	2	17	39	56	−22	−51
2	2	0	143	150	150	0
2	2	1	187	165	133	−97
2	2	2	353	370	88	−359
2	2	3	119	102	−101	7
2	2	4	281	242	−95	−222
2	2	5	303	281	−279	31
2	2	6	206	233	21	−222
2	2	7	210	210	−200	66
2	2	8	55	39	−12	−37
2	2	9	141	128	89	92
2	2	10	142	139	82	112
2	2	11	336	347	347	11
2	2	12	153	145	−32	141
2	2	13	159	161	160	13
2	2	14	91	105	−14	104
2	2	15	22	39	−12	−37
2	2	16	19	28	16	22
2	2	17	15	19	3	−19
3	2	0	73	116	−116	0
3	2	1	400	367	98	354
3	2	2	154	145	−145	6
3	2	3	207	185	166	−82
3	2	4	479	460	122	−444
3	2	5	169	145	145	5
3	2	6	246	240	−25	−238
3	2	7	189	170	98	139
3	2	8	167	147	117	−88
3	2	9	127	118	−67	97
3	2	10	120	120	78	91
3	2	11	39	64	−47	−44
3	2	12	119	109	47	99
3	2	13	87	94	−76	−55
3	2	14	26	27	−13	24
3	2	15	81	78	−39	−67
3	2	16	20	18	−17	5
3	2	17	15	11	8	−8
4	2	0	594	518	518	0
4	2	1	129	104	50	91
4	2	2	183	157	117	−104
4	2	3	156	132	125	43
4	2	4	168	173	87	−149
4	2	5	132	115	33	−110
4	2	6	100	79	−27	−74
4	2	7	235	223	−204	−90
4	2	8	114	115	−113	−23
4	2	9	189	173	53	−164
4	2	10	159	147	−142	39
4	2	11	32	42	31	−29
4	2	12	96	96	21	94
4	2	13	29	30	6	30
4	2	14	105	98	94	28
4	2	15	25	60	−25	54
4	2	16	21	88	82	33
5	2	0	202	221	−221	0
5	2	1	185	176	−152	88
5	2	2	313	274	−118	248
5	2	3	189	187	10	−187
5	2	4	58	66	−64	14
5	2	5	201	193	−76	−177
5	2	6	356	327	172	−278
5	2	7	210	193	−38	−189
5	2	8	138	131	124	42
5	2	9	108	98	−35	−91
5	2	10	190	170	133	106
5	2	11	31	46	46	−3
5	2	12	29	38	−13	36
5	2	13	28	27	25	10
5	2	14	73	62	−48	−38
5	2	15	116	98	22	96
5	2	16	52	66	−58	−33
6	2	0	11	8	8	0
6	2	1	214	218	−10	218
6	2	2	180	170	145	89
6	2	3	179	190	186	37
6	2	4	139	135	133	23
6	2	5	103	94	92	19
6	2	6	51	58	14	56
6	2	7	196	188	143	−122
6	2	8	85	95	−95	−9
6	2	9	181	163	−103	−126
6	2	10	189	182	−181	9
6	2	11	183	155	−139	−69
6	2	12	31	61	−31	−53
6	2	13	122	105	−103	−20
6	2	14	26	5	−5	2
6	2	15	22	23	−12	20
6	2	16	18	41	−40	−10
7	2	0	95	94	−94	0
7	2	1	78	70	−70	3
7	2	2	282	284	−42	280
7	2	3	353	347	−304	166
7	2	4	329	355	−85	345
7	2	5	215	197	−186	−64
7	2	6	208	236	43	232
7	2	7	216	194	−168	−96
7	2	8	93	82	52	64
7	2	9	133	109	56	−93
8	2	0	300	317	−317	0
8	2	1	32	49	18	46
8	2	2	125	137	−135	23
8	2	3	118	131	−129	24
8	2	4	63	64	11	63
8	2	5	9	9	9	−2
8	2	6	106	89	89	−2
8	2	7	99	101	98	−24
8	2	8	39	51	−22	46
8	2	9	22	22	22	4
9	2	0	308	275	275	0
9	2	1	270	262	−18	−262
9	2	2	145	127	127	6
9	2	3	116	110	−107	−25
9	2	4	103	110	60	93
9	2	5	220	206	−126	162
9	2	6	227	215	−113	183
9	2	7	142	123	−30	120
9	2	8	143	134	−133	14
9	2	9	147	138	6	138
10	2	0	201	195	195	0
10	2	1	93	85	−60	61
10	2	2	108	107	104	26
10	2	3	103	100	−76	65
10	2	4	56	62	57	25
10	2	5	46	47	−31	−35
10	2	6	91	88	−73	49
10	2	7	32	24	−11	−21
10	2	8	42	40	−39	9
10	2	9	106	88	77	−42
11	2	0	91	91	91	0
11	2	1	39	34	34	−5
11	2	2	183	181	78	−163
11	2	3	113	113	113	−1
11	2	4	120	123	16	−122
11	2	5	80	86	68	53
11	2	6	112	109	−47	−99
11	2	7	71	57	47	33
11	2	8	98	95	−94	−10
11	2	9	9	17	−10	−13

TABLE 7. (*Continued.*)

h	*k*	*l*	$\lvert F_o\rvert$	$\lvert F_c\rvert$	A_c	B_c
12	2	0	73	68	68	0
12	2	1	49	48	17	−45
12	2	2	76	73	47	56
12	2	3	95	79	69	−38
12	2	4	107	103	−65	79
12	2	5	39	39	39	7
12	2	6	73	77	−42	64
12	2	7	11	4	−4	2
12	2	8	33	36	36	−5
12	2	9	67	52	−44	28
13	2	0	39	41	−41	0
13	2	1	86	72	31	65
13	2	2	16	26	−6	−25
13	2	3	89	82	81	−10
13	2	4	63	68	2	−67
13	2	5	69	115	85	−78
13	2	6	63	62	6	−62
13	2	7	49	54	38	−38
13	2	8	26	28	23	−17
14	2	0	68	50	−50	0
14	2	1	100	89	28	−84
14	2	2	78	71	−70	9
14	2	3	51	50	30	−40
14	2	4	33	37	−28	24
14	2	5	31	33	25	21
14	2	6	28	38	37	−8
0	3	1	11	5	0	−5
0	3	2	96	98	−98	0
0	3	3	178	180	0	−180
0	3	4	415	415	415	0
0	3	5	53	37	0	−37
0	3	6	314	317	317	0
0	3	7	15	47	0	−47
0	3	8	48	54	54	0
0	3	9	79	73	0	−73
0	3	10	170	169	−169	0
0	3	11	14	8	0	−8
9	3	12	114	98	−98	0
0	3	13	14	17	0	−17
0	3	14	13	11	−11	0
0	3	15	12	16	0	16
0	3	16	9	20	20	0
0	3	17	35	58	0	58
1	3	0	316	324	0	324
1	3	1	426	394	−393	32
1	3	2	320	344	−287	190
1	3	3	235	239	−224	83
1	3	4	249	247	−240	58
1	3	5	236	223	22	222
1	3	6	274	270	−267	38
1	3	7	216	222	218	45
1	3	8	142	150	−106	−106
1	3	9	310	309	309	−2
1	3	10	88	97	59	−77
1	3	11	218	232	174	−154
1	3	12	128	115	112	−25
1	3	13	40	62	−20	−59
1	3	14	113	93	91	22
1	3	15	54	59	−59	−2
1	3	16	9	24	17	17
1	3	17	32	62	−60	−16
2	3	0	259	247	0	247
2	3	1	182	182	−121	−136
2	3	2	133	129	−107	−72
2	3	3	279	267	−30	−266
2	3	4	167	169	−164	−39
2	3	5	214	212	71	−199
2	3	6	34	29	−25	14
2	3	7	342	337	298	−158
2	3	8	99	103	−99	31
2	3	9	183	170	170	−2
2	3	10	54	38	−38	8
2	3	11	41	54	47	26
2	3	12	14	37	−3	−37
2	3	13	59	68	−14	67
2	3	14	53	63	30	−56
2	3	15	82	89	−71	54
2	3	16	9	26	16	−20
3	3	0	345	341	0	−341
3	3	1	142	116	69	−93
3	3	2	303	272	−214	−167
3	3	3	120	118	63	−99
3	3	4	242	244	−242	31
3	3	5	82	103	−77	−69
3	3	6	194	202	−202	−3
3	3	7	72	62	−60	17
3	3	8	151	164	−120	12
3	3	9	138	136	−84	−106
3	3	10	107	101	98	25
3	3	11	96	113	−42	−105
3	3	12	131	146	145	17
3	3	13	42	41	40	−9
3	3	14	88	106	94	−48
3	3	15	45	48	26	41
3	3	16	45	63	5	−63
4	3	0	334	313	0	313
4	3	1	79	70	14	−68
4	3	2	248	221	68	211
4	3	3	174	174	−165	54
4	3	4	166	167	−167	−1
4	3	5	79	65	3	−65
4	3	6	236	236	−193	−137
4	3	7	46	60	2	60
4	3	8	103	107	30	−102
4	3	9	91	76	48	59
4	3	10	167	168	107	−130
4	3	11	67	68	2	68
4	3	12	94	75	70	−25
4	3	13	26	46	0	46
4	3	14	12	16	−16	3
4	3	15	9	20	7	19
4	3	16	7	32	−11	30
5	3	0	194	185	0	−185
5	3	1	263	259	259	−10
5	3	2	115	99	42	−90
5	3	3	159	170	92	−143
5	3	4	81	64	−43	−48
5	3	5	103	100	33	−94
5	3	6	39	54	−48	25
5	3	7	160	159	−158	12
5	3	8	31	47	−46	11
5	3	9	207	207	−199	55
5	3	10	91	92	−26	88
5	3	11	156	165	−119	114
5	3	12	14	15	15	1
5	3	13	71	80	15	79
5	3	14	12	28	−14	−24
5	3	15	38	45	40	21
6	3	0	122	100	0	100
6	3	1	163	168	35	165
6	3	2	216	215	61	206
6	3	3	212	214	2	214
6	3	4	49	32	32	−3
6	3	5	201	218	−131	174
6	3	6	113	109	−88	−65
6	3	7	159	155	−100	118
6	3	8	170	169	21	−168
6	3	9	88	84	−80	−25
6	3	10	65	70	43	−55
6	3	11	14	53	−5	−53
6	3	12	13	21	−12	−18
6	3	13	61	73	7	−72
6	3	14	42	50	0	50
6	3	15	58	84	59	−60
7	3	0	269	283	0	283
7	3	1	108	110	97	−51
7	3	2	138	147	103	105
7	3	3	81	83	63	−55
7	3	4	134	145	145	−6
7	3	5	94	101	43	−92
7	3	6	119	114	77	−84
7	3	7	120	115	−8	−115
7	3	8	86	98	91	−36
7	3	9	88	85	−78	32
7	3	10	34	38	−9	−37
7	3	11	49	71	−39	59
7	3	12	41	57	−57	−7
7	3	13	45	53	−45	27
7	3	14	39	57	−33	46
8	3	0	154	157	0	−157
8	3	1	159	154	−60	142
8	3	2	163	180	93	−154
8	3	3	242	238	52	232
8	3	4	146	151	149	18
8	3	5	260	280	29	278
8	3	6	189	203	197	49
8	3	7	89	82	2	82
8	3	8	92	86	38	77
8	3	9	34	34	32	12
8	3	10	67	80	−26	76
8	3	11	74	102	10	−102
8	3	12	35	38	−29	24
8	3	13	68	99	20	−97
8	3	14	45	22	−22	−1
9	3	0	47	49	0	49
9	3	1	60	60	20	57
9	3	2	55	58	−13	56
9	3	3	165	171	53	162
9	3	4	53	64	−25	−58
9	3	5	108	119	−18	118
9	3	6	44	42	36	−21
9	3	7	58	51	−6	51
9	3	8	11	21	21	−2
9	3	9	53	44	−3	−44
9	3	10	12	33	17	−28
9	3	11	59	67	−18	−65
9	3	12	9	19	−18	4
9	3	13	28	73	−10	−73
10	3	0	203	220	0	−220
10	3	1	67	75	−30	−68
10	3	2	185	195	−95	−170
10	3	3	32	42	17	−39
10	3	4	47	64	−61	−18
10	3	5	96	108	87	−64
10	3	6	88	85	−30	79
10	3	7	53	47	32	−35
10	3	8	140	163	−17	162
10	3	9	28	17	12	−11
10	3	10	56	79	−12	78
10	3	11	8	27	21	18
11	3	0	113	125	0	−125
11	3	1	38	40	13	38
11	3	2	94	108	30	−103
11	3	3	9	9	−8	4
11	3	4	59	58	58	7
11	3	5	34	37	−21	31
11	3	6	86	91	75	53
11	3	7	45	55	−54	10
11	3	8	38	36	22	29
11	3	9	7	19	−1	−19
11	3	10	8	41	−23	34
12	3	0	11	7	0	−7
12	3	1	76	75	56	−49
12	3	2	21	30	−28	10
12	3	3	106	126	14	−126
12	3	4	28	33	−29	−16
12	3	5	96	115	−34	−109
12	3	6	42	40	−36	16
12	3	7	40	51	−38	−35
12	3	8	47	3	0	−3
13	3	0	9	44	0	−44
13	3	1	71	76	−71	−27
13	3	2	33	32	25	−19
13	3	3	35	48	−47	−10
13	3	4	52	69	60	33
13	3	5	5	19	1	−19
13	3	6	6	15	11	10
14	3	0	51	85	0	5
14	3	1	36	49	47	−13
14	3	2	45	63	38	50
14	3	3	28	43	0	−43
0	4	0	342	340	340	0
0	4	1	347	337	0	337
0	4	2	101	86	86	0
0	4	3	209	197	0	197
0	4	4	99	118	−118	0
0	4	5	108	118	0	−118
0	4	6	9	4	4	0
0	4	7	170	167	0	−167
0	4	8	78	72	72	0
0	4	9	297	290	0	−290
0	4	10	28	34	34	0
0	4	11	141	126	0	−126
0	4	12	26	40	−40	0
0	4	13	24	3	0	3
0	4	14	20	21	−21	0
0	4	15	56	62	0	62
1	4	0	179	182	−182	0
1	4	1	269	257	−16	257
1	4	2	192	204	−117	−167
1	4	3	49	46	43	14
1	4	4	252	273	−110	−250
1	4	5	87	98	−19	−96
1	4	6	148	164	127	−104
1	4	7	132	148	−13	−147
1	4	8	51	53	51	14
1	4	9	116	113	−68	−90
1	4	10	111	110	98	49
1	4	11	25	62	−5	−62
1	4	12	47	46	−4	46

TABLE 7. (*Continued.*)

h	k	l	$\|F_o\|$	$\|F_c\|$	A_c	B_c
1	4	13	21	47	−24	41
1	4	14	19	44	−18	40
1	4	15	14	35	6	35
2	4	0	114	109	−109	0
2	4	1	31	31	24	21
2	4	2	281	278	−83	265
2	4	3	54	43	20	−38
2	4	4	174	183	−77	166
2	4	5	58	87	−82	27
2	4	6	179	173	27	171
2	4	7	121	141	−136	−38
2	4	8	111	106	104	20
2	4	9	69	56	−55	−10
2	4	10	80	88	62	−63
2	4	11	25	39	−32	−23
2	4	12	119	125	6	−125
2	4	13	20	9	−3	8
2	4	14	78	78	−15	−77
2	4	15	13	14	14	1
3	4	0	28	37	37	0
3	4	1	267	255	−55	249
3	4	2	29	27	−19	19
3	4	3	181	189	−188	18
3	4	4	128	137	−18	136
3	4	5	174	183	−94	−157
3	4	6	87	81	31	75
3	4	7	165	159	−97	−126
3	4	8	33	28	21	20
3	4	9	93	89	71	−53
3	4	10	27	28	−16	−23
3	4	11	82	73	40	−61
3	4	12	25	37	−23	−29
3	4	13	74	86	80	−31
3	4	14	19	27	−27	4
3	4	15	42	53	43	30
4	4	0	182	176	−176	0
4	4	1	138	125	17	−124
4	4	2	49	48	−48	4
4	4	3	132	117	−111	−37
4	4	4	121	122	110	53
4	4	5	182	197	−180	80
4	4	6	29	30	17	−24
4	4	7	165	162	−116	113
4	4	8	52	59	−38	45
4	4	9	145	131	−5	131
4	4	10	29	33	−25	−21
4	4	11	96	94	79	51
4	4	12	26	18	4	−17
4	4	13	71	84	70	−47
4	4	14	19	29	−27	−11
5	4	0	131	122	122	0
5	4	1	107	105	0	105
5	4	2	175	169	103	133
5	4	3	107	101	−15	100
5	4	4	143	157	16	156
5	4	5	28	36	−25	25
5	4	6	158	150	−114	99
5	4	7	51	55	52	18
5	4	8	95	91	−91	−8
5	4	9	22	19	14	−14
5	4	10	98	88	−81	−36
5	4	11	26	38	15	−35
5	4	12	24	16	14	−7
5	4	13	21	33	13	−31
5	4	14	16	58	43	−39
6	4	0	56	47	47	0
6	4	1	142	130	−67	−112
6	4	2	146	140	52	−130
6	4	3	86	83	−83	−7
6	4	4	151	151	83	−126
6	4	5	120	110	−102	−43
6	4	6	73	58	−23	−53
6	4	7	53	51	−27	43
6	4	8	121	120	−116	−29
6	4	9	86	69	39	56
6	4	10	91	62	−49	38
6	4	11	26	91	78	48
6	4	12	24	53	16	51
6	4	13	20	53	46	27
7	4	0	88	91	91	0
7	4	1	161	148	103	−107
7	4	2	82	80	60	54
7	4	3	192	180	178	−26
7	4	4	35	36	36	−4
7	4	5	179	188	171	77
7	4	6	61	57	−33	−46
7	4	7	119	100	92	39
7	4	8	73	69	−63	−28
7	4	9	11	23	−22	5
8	4	0	182	176	176	0
8	4	1	66	60	−5	59
8	4	2	143	151	150	−5
8	4	3	58	44	44	3
8	4	4	8	13	−13	−2
8	4	5	99	108	107	−15
8	4	6	63	59	−55	22
8	4	7	89	87	60	−64
8	4	8	72	75	−73	−19
8	4	9	54	41	−4	−41
9	4	0	176	171	−171	0
9	4	1	51	45	0	−45
9	4	2	159	129	−129	4
9	4	3	105	102	101	−17
9	4	4	34	30	−29	−7
9	4	5	66	62	62	−6
9	4	6	83	75	68	−32
9	4	7	32	26	17	−19
9	4	8	115	117	117	0
9	4	9	8	18	−6	−17
10	4	0	48	34	−34	0
10	4	1	95	70	45	−54
10	4	2	88	84	−73	41
10	4	3	91	89	70	−55
10	4	4	80	81	−54	61
10	4	5	125	118	118	10
10	4	6	8	7	−1	7
10	4	7	78	82	68	46
10	4	8	54	57	57	−4
10	4	9	60	49	−36	32
11	4	0	56	59	−59	0
11	4	1	92	71	−28	−65
11	4	2	8	13	−9	−9
11	4	3	100	105	−98	−37
11	4	4	16	20	−20	4
11	4	5	75	72	−71	−10
11	4	6	51	47	35	32
11	4	7	54	56	−41	38
11	4	8	29	37	36	7
12	4	0	71	69	−69	0
12	4	1	9	2	−2	0
12	4	2	61	65	−61	−21
12	4	3	27	27	25	10
12	4	4	51	51	−7	−51
12	4	5	5	12	3	12
12	4	6	45	50	29	−41
13	4	0	67	65	65	0
13	4	1	6	15	−10	−10
13	4	2	42	46	16	−43
0	5	1	140	120	0	−120
0	5	2	143	128	−128	0
0	5	3	167	158	0	−158
0	5	4	153	149	−149	0
0	5	5	141	132	0	−132
0	5	6	95	89	−89	0
0	5	7	13	4	0	−4
0	5	8	11	18	18	0
0	5	9	59	66	0	66
0	5	10	26	13	13	0
0	5	11	32	50	0	50
0	5	12	56	68	68	0
0	5	13	52	54	0	54
1	5	0	102	113	0	−113
1	5	1	160	152	149	26
1	5	2	163	160	160	13
1	5	3	89	83	56	−62
1	5	4	125	128	120	45
1	5	5	52	65	−57	31
1	5	6	93	97	96	−13
1	5	7	67	84	−83	8
1	5	8	93	93	17	−92
1	5	9	94	90	−88	17
1	5	10	24	39	−29	−26
1	5	11	21	50	−50	1
1	5	12	19	46	−41	21
1	5	13	14	13	3	12
2	5	0	154	139	0	139
2	5	1	153	145	142	28
2	5	2	170	178	−35	174
2	5	3	47	46	−23	40
2	5	4	22	13	−13	2
2	5	5	129	129	−113	63
2	5	6	112	119	24	−116
2	5	7	87	94	−59	73
2	5	8	120	116	9	−115
2	5	9	69	62	−59	19
2	5	10	44	62	21	−58
2	5	11	20	39	−38	−9
2	5	12	18	3	1	−2
2	5	13	14	39	−33	−21
3	5	0	140	145	0	145
3	5	1	100	83	−83	−10
3	5	2	101	99	97	17
3	5	3	74	73	−56	47
3	5	4	56	60	58	12
3	5	5	121	123	61	106
3	5	6	49	36	−11	−34
3	5	7	115	118	77	90
3	5	8	11	16	−15	−5
3	5	9	91	80	79	8
3	5	10	24	33	−26	−20
3	5	11	21	44	33	−29
3	5	12	18	23	−23	−1
3	5	13	13	44	−37	−24
4	5	0	18	21	0	21
4	5	1	141	139	114	80
4	5	2	44	42	37	−21
4	5	3	116	105	22	103
4	5	4	52	56	55	−9
4	5	5	100	101	−35	94
4	5	6	33	35	35	−3
4	5	7	31	36	−34	−11
4	5	8	9	16	−13	9
4	5	9	69	55	−11	−54
4	5	10	25	40	−21	34
4	5	11	44	80	−58	−56
4	5	12	56	17	−16	−4
5	5	0	41	48	0	48
5	5	1	113	100	−100	−5
5	5	2	89	85	−83	19
5	5	3	62	63	−9	63
5	5	4	105	101	−74	−69
5	5	5	87	77	20	74
5	5	6	78	65	−63	17
5	5	7	83	74	62	41
5	5	8	69	71	1	71
5	5	9	79	64	54	−34
5	5	10	72	58	54	21
5	5	11	20	65	21	−61
5	5	12	15	53	48	−22
6	5	0	116	95	0	−95
6	5	1	34	31	4	−31
6	5	2	136	126	73	−103
6	5	3	29	37	36	5
6	5	4	53	54	51	−19
6	5	5	39	52	44	−28
6	5	6	61	61	8	61
6	5	7	55	49	10	−48
6	5	8	102	98	5	98
6	5	9	25	14	−14	3
6	5	10	22	43	−8	43
6	5	11	18	23	4	23
7	5	0	99	89	0	−89
7	5	1	59	51	39	32
7	5	2	138	108	−74	−78
7	5	3	40	41	34	23
7	5	4	89	90	−87	−23
7	5	5	151	30	−28	12
7	5	6	51	46	−27	37
7	5	7	63	71	−70	7
7	5	8	71	76	−21	73
7	5	9	69	56	−56	−2
8	5	0	12	13	0	13
8	5	1	73	61	−41	−46
8	5	2	34	43	22	37
8	5	3	138	121	4	−121
8	5	4	7	4	−2	4
8	5	5	91	125	23	−123
8	5	6	27	34	−34	−3
8	5	7	60	55	−7	−55
8	5	8	31	23	−11	−20
8	5	9	7	10	−10	−3
9	5	0	53	57	0	−57
9	5	1	12	21	15	−15
9	5	2	66	63	61	−18
9	5	3	58	48	−16	−45
9	5	4	96	98	96	20

TABLE 7. (*Continued.*)

h	*k*	*l*	$\|F_o\|$	$\|F_c\|$	A_c	B_c	*h*	*k*	*l*	$\|F_o\|$	$\|F_c\|$	A_c	B_c	*h*	*k*	*l*	$\|F_o\|$	$\|F_c\|$	A_c	B_c
9	5	5	42	89	−5	−88	2	6	9	28	25	21	−14	7	6	7	33	39	−39	−2
9	5	6	78	75	70	26	2	6	10	14	41	−15	38							
9	5	7	54	59	−26	−53								8	6	0	60	51	−51	0
9	5	8	18	18	17	−5	3	6	0	80	70	70	0	8	6	1	85	78	−30	−73
							3	6	1	81	74	59	−45	8	6	2	61	49	−42	−26
10	5	0	65	73	0	73	3	6	2	46	54	51	16	8	6	3	47	49	−35	−34
10	5	1	66	55	−50	22	3	6	3	69	75	67	−32	8	6	4	29	26	9	−24
10	5	2	56	49	10	48	3	6	4	20	20	19	−6	8	6	5	31	36	−31	18
10	5	3	32	36	−23	28	3	6	5	74	78	77	−11							
10	5	4	35	34	23	26	3	6	6	55	65	−64	−13	9	6	0	72	70	70	0
10	5	5	5	34	−11	32	3	6	7	32	45	31	33	9	6	1	6	25	2	25
10	5	6	33	37	25	−28	3	6	8	65	67	−67	−2	9	6	2	51	48	42	−23
10	5	7	18	36	15	32	3	6	9	60	55	−26	48							
							3	6	10	14	34	−33	6	0	7	1	53	49	0	49
11	5	1	60	49	−49	−2								0	7	2	44	40	−40	0
11	5	2	42	38	12	36	4	6	0	16	27	−27	0	0	7	3	102	93	0	93
11	5	3	45	43	−27	−33	4	6	1	92	72	32	64	0	7	4	6	2	−2	0
11	5	4	19	27	16	22	4	6	2	47	35	−2	35	0	7	5	98	103	0	103
							4	6	3	86	83	74	36	0	7	6	35	38	38	0
0	6	0	56	49	49	0	4	6	4	49	45	−20	40							
0	6	1	142	128	0	−128	4	6	5	66	66	57	−33	1	7	0	40	38	0	−38
0	6	2	20	28	28	0	4	6	6	41	49	25	42	1	7	1	19	23	−7	−22
0	6	3	94	80	0	−80	4	6	7	53	76	19	−73	1	7	2	44	43	−40	−17
0	6	4	8	5	5	0	4	6	8	7	18	15	10	1	7	3	48	40	−20	−34
0	6	5	20	27	0	27	4	6	9	68	70	6	−69	1	7	4	45	40	−36	18
0	6	6	44	46	−46	0								1	7	5	5	6	0	−6
0	6	7	92	91	0	91	5	6	0	15	18	−18	0	1	7	6	32	37	−33	16
0	6	8	33	30	−30	0	5	6	1	79	47	2	−47							
0	6	9	103	92	0	92	5	6	2	36	36	−35	6	2	7	0	83	91	0	−91
0	6	10	18	18	−18	0	5	6	3	81	82	65	−50	2	7	1	22	24	20	−14
0	6	11	39	59	0	59	5	6	4	20	31	−4	−31	2	7	2	93	96	−10	−95
							5	6	5	42	41	36	−19	2	7	3	6	7	−5	−5
1	6	0	94	90	90	0	5	6	6	63	71	38	−60	2	7	4	34	38	1	−38
1	6	1	29	55	−43	−34	5	6	7	59	13	3	13	2	7	5	12	13	−12	−6
1	6	2	133	120	120	0	5	6	8	44	48	36	−31	2	7	6	40	58	17	56
1	6	3	71	73	−72	−14	5	6	9	52	46	−26	38							
1	6	4	54	52	19	48								3	7	0	34	4	0	4
1	6	5	59	62	−62	2	6	6	0	62	65	−65	0	3	7	1	62	60	45	−40
1	6	6	85	95	−61	72	6	6	1	16	10	10	0	3	7	2	7	10	9	5
1	6	7	86	24	−5	24	6	6	2	94	99	−48	86	3	7	3	61	57	31	−48
1	6	8	94	93	−87	31	6	6	3	7	9	−8	−5	3	7	4	15	20	20	4
1	6	9	55	57	43	38	6	6	4	94	100	7	100	3	7	5	32	30	−25	−15
1	6	10	15	66	−60	−27	6	6	5	41	41	31	28							
1	6	11	9	46	41	22	6	6	6	73	70	21	66	4	7	0	27	28	0	−28
							6	6	7	15	21	10	19	4	7	1	29	26	−4	−25
2	6	0	60	64	64	0	6	6	8	22	29	27	11	4	7	2	45	34	30	−17
2	6	1	52	46	38	26								4	7	3	66	70	−26	−65
2	6	2	87	101	34	−95	7	6	0	52	51	−51	0	4	7	4	26	26	12	−23
2	6	3	91	93	90	21	7	6	1	32	30	−29	6	4	7	5	33	56	−17	−54
2	6	4	86	80	4	−80	7	6	2	31	31	−26	−18							
2	6	5	36	38	37	−9	7	6	3	80	69	−69	8	5	7	0	26	27	0	27
2	6	6	66	55	−23	−51	7	6	4	40	36	−28	−22	5	7	1	7	2	−2	0
2	6	7	27	35	29	−21	7	6	5	58	52	−52	2	5	7	2	59	71	69	19
2	6	8	26	25	−24	−7	7	6	6	33	39	26	−29							

Table 2 gives the anisotropic thermal parameters (in Å²). These are the components of Cruickshank's anisotropic thermal motion tensor.[8]

Table 3 gives the standard deviations and σ_{rms} for the atomic co-ordinates. The values were found from the standard least-squares formulæ.[9]

For Table 4 the deviations were calculated from the formulæ given by Cruickshank and Robertson.[10]

Table 5 gives the standard deviations of the anisotropic thermal parameters.

Table 6 gives some of the smaller Van der Waals contacts.

Table 7 gives $|F_o|$, $|F_c|$, A_c, and B_c.

Discussion.—In Table 8 the bond lengths obtained in the present analysis are compared with standard lengths given in " Interatomic Distances." [11] It will be seen that the agreement is everywhere good except for the C=N bond, which is not significantly different from a carbon–nitrogen triple bond. The same feature was observed in the two previous studies on vinylideneamines.[1,2]

The angle C3=C4=N1 differs significantly from 180° ($\Delta/\sigma = 6{\cdot}0$) but the direction of displacement of the nitrogen atom is perpendicular to the plane S1–C4–S2. Such a

[8] Cruickshank, *Acta Cryst.*, 1956, **9**, 747.
[9] Whittaker and Robinson, " Calculus of Observations," Chapter IX, Blackie and Sons, London, 1940.
[10] Cruickshank and Robertson, *Acta Cryst.*, 1953, **6**, 698.
[11] Interatomic Distances " (L. E. Sutton), *Chem. Soc. Special Publ.* No. 11, London, 1958.

distortion preserves the *mm* symmetry of the π-bond in C4=N1, and thus might occur particularly easily.

In Table 9 the present molecular dimensions are compared with the analogous ones obtained by Wheatley for $(CH_3{\cdot}SO_2)_2C{=}C{=}NMe$. Some differences in bond angles occur but these, with the exception of C4=N1–C5, are small and are probably attributable to

TABLE 8. *Bond lengths* (Å) *and angles in* $(CH_3{\cdot}SO_2)_2C{=}C{=}N{\cdot}C_2H_5$ *compared with standard values taken from "Interatomic Distances."* [10]

Bond	Length in $(CH_3{\cdot}SO_2)_2C{=}C{=}NEt$	Length in standard	Standard source
S1–O1	1·441	1·43	Sulphones
S2–O2	1·440	1·43	Sulphones
S1–O3	1·433	1·43	Sulphones
S2–O4	1·432	1·43	Sulphones
S1–C1	1·773	1·80	S-methyl
S2–C2	1·772	1·80	S-methyl
S1–C3	1·739	1·73	S–C heterocyclic
S2–C3	1·741	1·73	S–C heterocyclic
N1–C4	1·165	—	Not enough data
N1–C5	1·465	1·475	N–C normal
C3–C4	1·356	1·337	C–C normal
C5–C6	1·532	1·541	C–C normal

Angle	Size in $(CH_3{\cdot}SO_2)_2C{=}C{=}N{\cdot}C_2H_5$	Size in standard
S1–C3–S2	121·5°	120·0°
S1–C3–C4	121·8	120·0
S2–C3–C4	116·6	120·0
N1–C4–C3	173·4	180·0
N1–C5–C6	111·2	109·5
O1–S1–O3	118·8	109·5
O1–S1–C1	108·6	109·5
O1–S1–C3	107·7	109·5
O2–S2–O4	118·7	109·5
O2–S2–C2	108·2	109·5
O2–S2–C3	106·0	109·5
O3–S1–C1	107·7	109·5
O3–S1–C3	107·2	109·5
O4–S2–C2	108·6	109·5
O4–S2–C3	108·0	109·5
C1–S1–C3	106·2	109·5
C2–S2–C3	106·7	109·5
C4–N1–C5	144·5	120·0

TABLE 9. *Comparison of the bond lengths* (Å) *and angles in* $(CH_3{\cdot}SO_2)_2C{=}C{=}N{\cdot}C_2H_5$ *with corresponding values in* $(CH_3{\cdot}SO_2)_2C{=}C{=}N{\cdot}CH_3$.

Bond	Length in NEt compound	Length in NMe compound	Δ/σ
S1–O1	1·441	1·431	1·4
S2–O2	1·440	1·431	0·9
S1–O3	1·433	1·435	0·3
S2–O4	1·432	1·435	0·3
S1–C1	1·773	1·770	0·3
S2–C2	1·772	1·770	0·1
S1–C3	1·739	1·726	1·3
S2–C3	1·741	1·726	1·4
N1–C4	1·165	1·154	0·8
N1–C5	1·465	1·426	2·4
C3–C4	1·356	1·342	0·9
C5–C6	1 532	—	—
Distance			
S1–S2	3·03	3·03	—

Angle	Size in NEt compound	Size in NMe compound	Δ/σ
S1–C3–S2	121·5°	122·6°	0·7
S1–C3–C4	121·8	118·7	3·9
S2–C3–C4	116·6	118·7	2·6
N1–C4–C3	173·4	180·0	6·0
N1–C5–C6	111·2	—	—
O1–S1–O3	118·8	118·4	0·8
O1–S1–C1	108·6	108·7	0·2
O1–S1–C3	107·7	107·3	0·8
O2–S2–O4	118·7	118·4	0·6
O2–S2–C2	108·2	108·7	0·8
O2–S2–C3	106·0	107·3	2·6
O3–S1–C1	107·7	106·7	2·0
O3–S1–C3	107·2	108·3	2·2
O4–S2–C2	108·6	106·7	3·2
O4–S2–C3	108·0	108·3	0·6
C1–S1–C3	106·2	106·7	1·0
C2–S2–C3	106·7	106·7	—
C4–N1–C5	144·5	180·0	31·0

packing requirements. The lengths of the C3=C4, C4=N1, and N1–C5 bonds are all found to be slightly, though not significantly, greater than those found by Wheatley (Δ/σ values are 0·9, 0·8, and 2·4 respectively). It is interesting that the N1–C5 bond is significantly longer than was found by Bullough and Wheatley for $CH_3{\cdot}SO_2{\cdot}C(SO_2{\cdot}Ph){=}C{=}NMe$ (Δ/σ = 5·2).

There is no significant difference between the length of the N1–C5 bond found here (1·465 Å) and the standard single bond value (1·475 Å). Thus the length of the N1–C5 bond is consistent with the absence of hyperconjugation.

The most noticeable feature of the present analysis is the value found for the angle C4=N1–C5. The angle between the valencies of the nitrogen atom is no longer 180°,

nor has the angle dropped to 120°, but an intermediate value of 144° is adopted. These values indicate that the methyl group, attached to the nitrogen atom in *N*-methylvinylideneamines, brings about complete delocalisation of the lone pair of electrons on the nitrogen, while a similarly substituted ethyl group cannot do so. By analogy, a carbanion of the type RR'C=C=$\bar{C}$–Me ought to have a linear C=C–Me angle.

In view of these results it seems reasonable to classify the *N*-methylvinylideneamines investigated [1,2] as examples of hyperconjugation in molecules in the ground state. However, it cannot be assumed that hyperconjugation is the only factor affecting linearity. In the *N*-ethylvinylideneamine the shortness of the N1=C4 bond, and the intermediate value adopted by the C4=N1–C5 angle could be ascribed either to the reduced hyperconjugation of the ethyl group or to the electron-attracting power of the sulphonyl groups. A more thorough examination of the effects of electronegativity is required to establish the relative importance of hyperconjugation and electronegativity.

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