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Sulfaguanidine Monohydrate 의 結晶 및 分子構造

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The Crystal and Molecular Structure of Sulfaguanidine Monohydrate

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요 약. sulfaguanidine 1 수화물의 결정 및 분자구조를 X-선 회절법으로 규명하였다. 결정은 일사측제에 속하며 단위세포내에는 4 분자가 있으며 공간군은 P21/c 이다. 단위세포상수는 a==7.57 ±0.03, b=5.44±0.02, c=24.76±0.06Å, b=91.0±0.2° 이다. 결정구조는 패터슨 함수의 해석과 직접법을 병용하여 밝혀냈다. 원자좌표치는 최소자승법으로 정밀화하였으며 1542개의 독립적인 회 절반점에 대한 최종 R값은 0.14 이었다. sulfaguanidine 분자의 guanidyl 기의 질소원자는 2개의 다른 불분자를 매개로 하여 수소결합을 하므로써 다른 분자들의 sulfonyl 기의 산소원자에 연결되어 있다. 수소결합 형성에 있어서 물분자는 donor 와 acceptor 의 역할을 겸하고 있으며 수소결합은 사면체의 방향을 하고 있다. 2차원적인 수소결합에 의하여 연결된 분자들은 (001)면에 평행한 분자층 을 형성하고 있다.

Abstract. The crystal and molecular structure of sulfaguanidine monohydrate, $C_7H_{10}N_4O_2S \cdot H_2O_5$, was determined from visually estimated intensity data from Weissenberg photographs. The crystal data are monoclinic, space group $P2_1/c$ with four molecules in a unit cell of dimensions, a=7.57 ± 0.03 , $b=5.44\pm 0.02$, $c=24.76\pm 0.06$ Å, $\beta=91.0\pm 0.2^{\circ}$. The structure has been solved by an interpretation of a Patterson map and with a help of a direct procedure on a procejection. The parameters were refined isotropically by block-diagonal least-squares methods using 1542 observed independent reflections to give R=0.14. By hydrogen bonding a guanidyl nitrogen of a sulfaguanidine molecule is linked to the sulfonyl oxygens of the other molecules indirectly through two different water molecules. The role of water molecule is both a donor and an acceptor in hydrogen-bonding formation and these hydrogen bonds are tetrahedrally oriented. The hydrogen-bonding networks form infinite molecular layers parallel to (001) plane.

Introduction

As a part of a series of detailed structure

investigation on sulfur-containing compounds which have biologically active groups by X-ray single-crystal diffraction, the crystal and molecular structure of sulfaguanidine monohydrate

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has been determined in this work. Among the sulfa drugs containing sulfanilamido group, the crsytal structures of sulfanilamide^{1,2,3},

H₂N - SO₂NH₂, and sulfathiazole, ^{4,5}
H₂N - SO₂N =
$$\begin{bmatrix} H \\ N \\ N \\ S \end{bmatrix}$$
, have been stude

ied previously. In this experiment, we have investigated the conformation of the sulfaguanidine molecule, delocalization of π -electron on the guanidyl residue and the role of hydrated water molecule in the crystal-lattice environment.

Experimental

Prismatic crystals of sulfaguanidine monohydrate elongated along the b axis were obtained by slow evaporation from an aqueous acctone solution at room temperature.

Unit cell dimensions were determined by a least-squares refinement of 2θ values for 47 independent reflections measured on the h0l and 0kl Weissenberg photographs calibrated with superimposed Al-lines using Cu-K_a radiation.

The density of single crystals, measured by flotation method in a mixture of benzene and carbon tetrachloride, was found to be consistent with 4 formular units in a unit cell.

The existence of water molecule in the single crystal was confirmed by the strong O-H stretching peak at 3400 cm⁻¹ in IR-spectrum.

The crystal data are as follows; sulfaguanidine monohydrate,

$$H_2N - SO_2 - NH - C NH + H_2O,$$

monoclinic, $a=7.57\pm0.03, b=5.44\pm0.02, c=$

24. 76±0. 06 Å, $\beta=91.0\pm0.2^{\circ}$, V=1019.1Å³, M. W. =232. 26, Z=4, $D_m=1.50$, $D_c=1.51$ g cm⁻³, space group: $P2_1/c$ from systematic absence hol for l odd, 0k0 for k odd.

Intensity data were collected from equi-inclination Weissenberg photographs taken with Cu- K_a radiation by use of the multiple-film method. The layers from hol to h4l for the b axis and from 0kl to 1kl for the a axis were recorded. The approximate dimensions of the crystals used for gathering the intensity data for the a and b axes were $0.2 \times 0.2 \times 1.0$ and $0.2 \times 0.3 \times 0.7$ mm respectively. The relative intensities were measured by visual comparison with a calibrated intensity scale prepared from the same crystals with the X-ray beam under carefully controlled conditions.

The intensities were corrected for spot-shape, Lorentz and polarization effects, but not for extinction or absorption, and converted into the observed structure factors. Interlayer scaling constants were calculated from common equivalent reflections and the structure factors were placed approximately on an absolute scale following the Wilson method⁶. The final number of observed independent reflections was 1542. All calculations were performed on an IBM 1130 computer with a series of programs by Shiono (1968)⁷.

Structure Determination and Refinement

After the structure factors were converted into the normalized structure factors, E, a three-dimensional sharpened Patterson function was computed using 985 normalized structure factors greater than |E|=0.58. The sulfursulfur Harker peak was easily identified, and trial positional parameters for the six atoms of S, O(1), O(2), C(1), N(1) and N(2) whose numberings are shown in Fig. 1 were obtained from the inspection of the Patterson map.

Independently a Fourier projection of electron density on (0kl) plane with 78 reflections whose phases were determined manually by an application of the direct method⁸ showed the positions of some of fifteen atoms and these were consistent with those obtained from the Patterson map.

The initial R factor, $R = \sum ||F_a| - |F_c|| / \sum |F_b|$, based on these six atoms with all assumed $B=3.0 \text{ Å}^2$ was 0.56 for 1005 reflections. The R factor based on sulfur coordinates alone was 0.70. Two cycles of isotropic block-diagonal least-squares refinement⁹ reduced the R factor to 0.45. A three-dimensional Fourier synthesis, which was computed using the 1005 observed structure factors phased on the contribution of the six atoms, gave the clear picture of the molecule consistent with a chemically reasonable model. The refinement with isotropic temperature parameters was carried out successively and stopped after several cycles attaining a minimum R value of 0.14 for all the 1542 observed reflections. The function minimized was $w(|F_o|)$ $-|F_c|)^2$ and the weighting scheme w proposed by Cruickshank (1965)¹⁰ was used throughout the refinement. The form of the function, w, was $(A+B|F_o|+C|F_o|^2)^{-1}$, where $A=2|F_{\min}|$ $=3.14, B=1.00, C=2/|F_{max}|=0.0164.$ No allowance was made for the hydrogen atoms. Atomic scattering factor values were taken from the International Table for X-ray Crystallography¹¹. The final positional and thermal parameters for the atoms are given in Table 1 with their estimated standard deviations as calculated from the least-squres refinement. Observed and calculated structure factors based on the parameters in Table 1 are listed in Table 2. The atoms are numbered according to Fig. 1.

Table 1. Final atomic coordinates and isotropic thermal parameters in sulfaguanidine monohydrate. The estimated standard deviations given in parentheses refer to the last decimal positions

	x	у У	z	В
s	0. 2588(5)	0. 4835(7)	0.4041(1)	1. 68(6)
0(1)	0.366(2)	0.301(2)	0.4306(5)	2.6(2)
0(2)	0.323(2)	0.727(2)	0.4098(5)	2.9(2)
O (3)	0.349(2)	0.168(2)	0.5410(5)	2.7(2)
N (1)	0.266(3)	0.273(4)	0.1671(9)	5.1(4)
N (2)	0.055(2)	0.493(2)	0.4226(5)	2.7(2)
N (3)	0.022(2)	0.065(3)	0.4261(7)	3.2(3)
N (4)	-0.215(2)	0.324(3)	0.4386(7)	3.2(3)
C (1)	0.257(2)	0.412(3)	0. 3339(6)	2.4(3)
C (2)	0.338(3)	0.205(4)	0.3160(8)	2.8(3)
C (3)	0.338(3)	0.157(4)	0.2603(9)	3.5(4)
C (4)	0.261(3)	0.322(4)	0.2230(8)	3. 3(3)
C (5)	0.175(3)	0.519(4)	0.2433(9)	3.8(4)
C (6)	0.175(3)	0.573(4)	0.2983(8)	3. 0(3)
$\mathbf{C}(7)$	-0.038(2)	0.291(3)	0.4302(6)	2.2(3)

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Table 2. Observed and calculated structure factors. Columns are Index, $10|F_{o^{h_s}}|$ and $10|F_{cal}|$

b=0, k=0	16 366 208	9 65 72	15 166 152	8 195 202	5 62 55	8 556 505
4 324 319	18 285 219	10 451 490	16 271 268	9 268 225	6 53 54	9 56 12
6 1058 1548	20 72 56	11 597 612	17 336 303	10 330 285	8 58 43	10 623 599
8 1157 1098	22 191 130	12 357 326	19 292 215	11 62 52	9 85 79	11 198 211
10 114 36	26 163 148	13 47 65	21 107 112	12 199 170	h=0 k=2	12 119 100
12 835 815	28 111 99	14 234 168	22 190 170	13 220 231	0 95 124	13 86 93
16 158 154	h = 5 k = 0	15 79 67	23 87 81	15 249 206	2 57 61 ¦	14 177 192
18 77 100	2 200 188	16 191 121	25 136 141	16 238 224	3 444 499	15 160 172
22 64 62	4 444 406	17 347 267	27 216 193	17 189 132	4 212 192	16 403 365
26 107 107	6 332 273	18 415 417	28 108 123	21 79 68	5 197 167	17 153 130
30 180 173	8 158 169	19 209 186	30 53 57	22 80 58	.6 354 295	18 177 172
k=1 $k=0$	10 132 135	20 128 135	h=3 k=1	23 133 113	7 310 279	20 225 230
0 159 148	12 181 148	21 63 89	0 40 62	24 52 48	8 360 342	21 77 98
2 676 1037	14 417 350	22 107 110	1 428 412	25 87 97	9 65 43	22 81 91
4 94 115	16 107 107	23 59 42	2 310 287	26 45 34	10 356 377	23 79 88
6 747 800	18 381 282	24 139 127	3 120 134	h=6 k=1	11 143 146	25 56 59
8 1047 1076	20 196 159	25 133 138	4 272 236	1 341 307	12 163 152	26 82 123
10 558 496	22 119 131	26 115 127	5 91 109	2 90 90	13 106 98	28 43 58
12 746 699	24 98 96	27 154 175	6 328 291	4 162 143	14 104 119	29 49 57
14 774 710	26 123 119	28 70 78	7 367 328	5 307 259	16 275 228	h=3 k=2
16 419 324	h=6 k=0	29 204 31	9 258 254	7 210 162	17 245 212	0 439 423
18 325 255	2 77 86	31 121 150	10 130 120	11 181 179	18 198 140	1 53 41
20 286 289	4 184 152	$k = 1 \ k = 1$	13 266 221	12 67 69	19 163 120	2 236 212
24 130 143	6 74 50	0 567 659	14 158 117	13 151 118	20 303 227	4 71 87
26 98 107	8 75 75	1 645 876	15 192 167	15 261 197	26 122 130	6 219 166
28 182 131	10 239 208	2 520 624	17 288 241	17 127 108	28 74 69	7 289 253
30 108 77	12 258 188	3 369 380	18 360 322	18 244 190	29 31 34	3 282 242
h=2 k=0	16 405 324	4 298 239	19 143 142	21 170 142	30 51 67	10 58 87
0 960 1364	22 154 144	5 297 268	22 76 53	23 95 94	n = 1 = 2 0 499 565	11 241 198
2 191 186	<i>k</i> =7 <i>k</i> =0	6 336 278	23 171 150	h=7 k=1	1 183 171	12 303 219
6 210 168	0 103 83	7 339 357	24 88 86	0 149 139	2 506 534	13 109 120
8 352 299	2 275 190	8 76 93	25 166 157	1 85 96	3 122 129	14 134 119
10 104 80	4 297 247	9 367 340	26 107 92	3 270 236	4 124 103	15 110 103
12 98 87	6 199 156	10 297 204	29 162 148	4 59 54	5 117 133	16 113 102
14 325 271	8 180 151	11 93 59	<i>h</i> =4 <i>k</i> =1	6 131 121	6 348 263	17 88 114
16 438 400	10 209 163	12 191 104	0 287 243	7 194 160	7 418 379	18 302 268
18 433 359	14 205 135	13 383 314	1 264 208	9 237 189	8 389 281	21 41 40
20 298 275	18 156 130	14 242 211	2 147 156	10 132 114	9 52 36	22 79 124
22 419 357	20 73 46	15 236 167	3 298 255	12 184 131	10 51 45	23 35 26
26 373 313	$h=8 \ k=0$	16 59 54	4 92 90	13 276 231	11 252 222	24 196 199
28 267 231	0 177 140	17 374 281	5 237 224	14 60 72	12 315 297	25 33 31
30 111 72	2 138 120	18 267 179	6 139 113	15 217 175	14 262 196	27 35 30
h=3 k=0	6 69 66	19 292 260	8 191 154	16 53 74	16 87 80	28 101 128
0 854 959	10 114 71	20 132 103	10 166 138	17 123 80	18 526 406	h=4 k=2
2 456 345	12 149 114	22 88 89	11 207 196	18 85 70	19 31 16	0 394 391
4 382 339	14 80 63	23 99 109	12 191 137	19 90 85	21 73 66	1 249 251
6 56 87	16 171 134	25 184 187	15 260 226	20 83 54	24 231 203	2 282 294
8 270 254	h=9 k=0	29 149 147	17 212 195	h = 8 k = 1	25 36 29	3 81 115
10 131 63	0 97 71		18 181 161	1 173 176	26 68 65	4 159 114
12 181 155	2 226 134	0 589 735	19 119 106	4 117 102	27 38 30	5 13Z 105
14 195 182	4 153 130	1 251 260	20 49 26	5 147 125	28 76 101	0 313 319
18 389 279	8 121 101	2 260 254	21 129 141	7 81 71	29 32 32	1 60 60
24 196 165	10 184 147	J 302 249	23 96 100	8 71 69	30 86 102	6, 66, 0 10, 10, 10, 10, 10, 10, 10, 10, 10, 10,
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0 391 388	2 104 217	0 10 00		13 83 76	1 100 100	11 37 41 12 76 69
2 414 345	a 407 401	10 950 990	1 102 100	10 190 146	2 403 318 3 992 982	14 70 20
4 107 102	4 447 421 5 620 640	10 300 320	0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	10 93 82 t=0 t=1	0 000 200 1 1 150 200	14 26 26
10 JU7 248 10 070 099	0 012 002	12 106 111	2 379 340 3 365 100	V=A K=T	5 103 189	++
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24	107	120	- 11	145	153	15	288	285	3	495	505	7	157	174	19	31	42	70		
	101	54	19	57	65	17	156	144	4	51	93	8	56	52	20	102	123	10	82	65
25	11	- 24		100	100	19	94	40	5	197	207	9	19	4	21	101	110	11	25	I
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-	195	158	16	125	113	21	138	162	12	96	84	12	274	284	24	21	15	14	141	141
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13	83	96	23	168	156	0	- 54	60	19	101	101	44	25	30	5	161	168	1	28	24
14	53	64	24	93	86) 2	189	159	21	26	11	21		.,		101	200		79	87
15	43	24	25	43	55	3	309	330	22	57	32	22	121	96	6	40	34	-	1.5	154
10	40	000	20	95	82	4	63	51	23	87	86	23	141	120	7	135	152	3	191	104
16	200	200	00		27		49	37	1	i=6 k	=3	24	26	38	8	99	108	4	71	68
18	86	103	27	42	16		44	41		141	192	25	81	105	9	28	20	5	53	49
20	83	83	28	38	40	6	23	41		141	100	00	70	103	10	127	116	6	174	151
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1	161	136	²	164	110	10		100		160	149	2	291	289	14	36	33	· 10	144	120
2	97	94	3	107	125	11	179	162	9	100	143	1 1	020	304	15	129	194	11	45	46
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4		100		410	916	15	1.38	137	13	174	150	1 7	114	128	19	28	26	1 14	23	17
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h	-0 k -	=5	6	755	807	-1	590	702	-18	136	166	3	189	167	-11	33	26	-6	68	70
1	237	270	-8	579	547	-2	226	213	-19	140	165	4	64	81	-12	51	33	7	73	66
	93 82	91 110	-10	790	20 778		431	430	-20	97	104	-7	102	78	-13	101	106	8	394	394
4	109	120	-14	214	205		418	390	-23	98	139	-13	80	97 89	-15	263	206	-10	289	304
5	361	438	-16	189	196	-7	255	235	-25	76	85	-14	60	54	-16	41	210	-12	132	142
7	214	240	18	500	451	8	89	93	-27	72	107	-15	83	53	-17	136	143	-14	304	313
11	159	19 9	20	372	288	-9	276	291	-29	133	156	-17	165	166	-18	87	110	15	186	194
13	121	138	-22	319	233	-10	316	315	h	=4 k=	-1	18	123	116	-19	124	128	18	45	38
17	120	110	-24	230	185	-11	118	121	-1	358	355	21	63	80	-20	247	268	19	43	51
h:	-1 #-	-5	-26	198	139	-13	418	454	2	73	94	Å.	=8 k=	=1	-21	128	141	-20	44	49
3	143 218	173	-28	81 	-0 92	-14	132	132	3	184	432	-3	71	50	23	40	53 100	21	105	116
4	188	200	-2	310	296	-17	195	219	-5	254	405 235		90 80	70	-24	161	102	-23	99 98	(4 98
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9	153	169	8	327	270	20	120	125	-8	42	40	-12	62	47	-2	489	478	-2	200	176
10	109	122	10	516	501	23	55	72	-9	190	197	13	90	78	—3	88	92	4	122	124
12	107	125	-12	381	314	25	83	113	-10	160	153	-15	169	164	-4	189	149	-6	145	129
13	145	176	14	254	213	-26	56	60	11	250	225	h	=9 k=	-1	5	102	105	-7	127	103
15	142	136	-16	365	292	-27	81	74	13	67	74	1	215	165	6	34	12	-8	47	33
19	07 214	170	-20 -24	202	84	28	00 140	134	-14	125	118	3	113	90	-7	38	53	9	139	147
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5	96	101	h	=3 k=	0	-2	251	226	-19	76	100	h-	=1 k=	-2	-13	39	54	-13	59	66
6	93	125	-2	72	42	3	560	643	-21	228	273	1	227	244	-14	260	284	-14	148	181
8	102	113	4	164	161	-4	318	240	-25	59	111	-2	316	420	-15	111	117	-15	37	40
9	73	86	-6	188	132	-5	391	436	-27	93	148	3	242	193	-16	226	212	16	53.	93
12	138	128	8	109	90	-6	103	149	h.	=5 k=	1	-4	174	120	-18	124	122	-20	98	109
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14 15 15 2 4	62 -1 & 537 309	50 0 722 312	12 16 18 20	385 349 379 101	310 295 288 80	8 9 10 11	243 66 278 342	212 87 279 398		204 104 254 144	196 96 247 134	6 7 8	352 165 141 29	276 163 99 21	-22 -23 24 28	86 75 151 132	121 84 168 165		05 =7 k= 109 238 90	2 106 224 67
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14 15 -2 -4 -6 -8 -10 -12 -16 -18 -20	62 =1 k= 537 309 117 727 668 509 126 132 78	50 0 722 312 80 672 553 390 59 126 81	$ \begin{array}{r} -12 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ h \\ -4 \\ -10 \\ -16 \\ \end{array} $	385 349 379 101 227 176 56 	310 295 288 80 167 134 33 0 351 124 150	$ \begin{array}{r} -8 \\ -9 \\ -10 \\ -11 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \\ -19 \end{array} $	243 66 278 342 41 139 172 128 45 56 141	212 87 279 398 45 134 204 129 54 70 144	-3 -4 -5 -6 -7 -8 -9 -10 -11 -12 -13	203 204 104 254 144 237 59 153 50 121 48 332	196 96 247 134 249 68 152 42 127 29 358	$ \begin{array}{c c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ \end{array} $	352 165 141 29 74 200 34 139 129 42 131	276 163 99 21 67 209 16 156 119 59 123	$ \begin{array}{r} -22 \\ -23 \\ -24 \\ -28 \\ k^{*} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ \end{array} $	41 86 75 151 132 =4 k= 165 205 178 385 113 303	121 84 168 165 2 130 191 168 390 99 313	$ \begin{array}{c} -24 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -8 \\ -9 \\ -12 \\ -13 \\ \end{array} $	65 =7 k= 109 238 90 42 67 222 210 48 200 74	96 2 106 224 67 31 56 201 201 53 197 50
14 15 h -2 -4 -6 -8 -10 -12 -16 -18 -20 -22	62 =1 &== 537 309 117 727 668 509 126 132 78 416	50 0 722 312 80 672 553 390 59 126 81 300	$ \begin{array}{r} -12 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ h \\ -10 \\ -16 \\ -18 \\ \end{array} $	385 349 379 101 227 176 56 425 174 235 339	310 295 288 80 167 134 33 0 351 124 150 239	$ \begin{array}{r} -8 \\ -9 \\ -10 \\ -11 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \\ -19 \\ -20 \end{array} $	243 66 278 342 41 139 172 128 45 56 141 48	212 87 279 398 45 134 204 129 54 70 144 41	$ \begin{array}{r} -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \end{array} $	204 104 254 144 237 59 153 50 121 48 332 150	196 96 247 134 249 68 152 42 127 29 358 157	$ \begin{array}{c c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ \end{array} $	352 165 141 29 74 200 34 139 129 42 131 173	276 163 99 21 67 209 16 156 119 59 123 180	$ \begin{array}{r} -22 \\ -23 \\ -24 \\ -28 \\ k^{2} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ \end{array} $	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48	121 84 168 165 2 130 191 168 390 99 313 40	$ \begin{array}{c} -24 \\ -24 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -8 \\ -9 \\ -12 \\ -13 \\ -14 \\ \end{array} $	65 =7 k= 109 238 90 42 67 222 210 48 200 74 136	2 106 224 67 31 56 201 201 53 197 60 153
14 15 h^{-2} -2 -4 -6 -8 -10 -12 -16 -20 -22 -24	62 =1 &= 537 309 117 727 668 509 126 132 78 416 131	50 0 722 312 80 672 553 390 59 126 81 300 118	$ \begin{array}{r} -12 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ h \\ -10 \\ -16 \\ -18 \\ -24 \\ \end{array} $	385 349 379 101 227 176 56 425 174 235 339 130	310 295 288 80 167 134 33 0 351 124 150 239 117	$ \begin{array}{c} -8 \\ -9 \\ -10 \\ -11 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \\ -19 \\ -20 \\ -21 \\ \end{array} $	243 66 278 342 41 139 172 128 45 56 141 48 156	212 87 279 398 45 134 204 129 54 70 144 41 180	$ \begin{array}{r} -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \end{array} $	204 104 254 144 237 59 153 50 121 48 332 150 179	196 96 247 134 249 68 152 42 127 29 358 157 212	$ \begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ \end{array} $	352 165 141 29 74 200 34 139 129 42 131 173 53	276 163 99 21 67 209 16 156 119 59 123 180 51	$ \begin{array}{r} -22 \\ -23 \\ -24 \\ -28 \\ -28 \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ \end{array} $	$ \begin{array}{r} & 86 \\ & 75 \\ & 151 \\ & 132 \\ & = 4 k = \\ & 165 \\ & 205 \\ & 178 \\ & 385 \\ & 113 \\ & 303 \\ & 48 \\ & 69 \\ \end{array} $	121 84 168 165 2 130 191 168 390 99 313 40 73	$ \begin{array}{c} -24 \\ -24 \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -8 \\ -9 \\ -12 \\ -13 \\ -14 \\ -16 \\ \end{array} $	63 =7 k= 109 238 90 42 67 222 210 48 200 74 136 104	56 22 106 224 67 31 56 201 201 53 197 60 153 122
14 15 h: -2 -4 -6 -8 -10 -12 -16 -12 -16 -20 -22 -24 -22	62 62 537 309 117 727 668 509 126 132 78 416 131 253	50 0 722 312 80 672 553 390 59 126 81 300 118 213	$ \begin{array}{r} -12 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ h^{-1} \\ -10 \\ -16 \\ -18 \\ -24 \\ h^{-1} \\ -24 \\ -24 \\ h^{-1} \\ -24 $	385 349 379 101 227 176 56 425 174 235 339 130 7 k=	310 295 288 80 167 134 33 351 124 150 239 117 0	$ \begin{array}{r} -8 \\ -9 \\ -10 \\ -11 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \\ -19 \\ -20 \\ -21 \\ -25 \\ \end{array} $	243 66 278 342 41 139 172 128 45 56 141 48 156 122	212 87 279 398 45 134 204 129 54 70 144 41 180 133	$ \begin{array}{r} -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \end{array} $	204 104 254 144 237 59 153 50 121 48 332 150 179 91	196 96 247 134 249 68 152 42 127 29 358 157 212 73	$ \begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ \end{array} $	352 165 141 29 74 200 34 139 129 42 131 173 53 81	276 163 99 21 67 209 16 156 119 59 123 180 51 90	$ \begin{array}{r} -22 \\ -23 \\ -24 \\ -28 \\ -28 \\ -28 \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ \end{array} $	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47	46 121 84 165 2 130 191 168 390 99 313 40 73 40	$\begin{array}{c} -24 \\ k \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -8 \\ -9 \\ -12 \\ -13 \\ -14 \\ -16 \\ -18 \end{array}$	63 =7 k = 109 238 90 42 67 222 210 48 200 74 136 104 126	56 22 106 224 67 31 56 201 201 53 197 60 153 122 153
14 15 h -2 -4 -6 -8 -10 -12 -16 -22 -22 -24 -22 -24 -22 -24 -22 -24 -22 -24 -22 -22 -22 -24 -22 -30 -	62 62 537 309 117 727 668 509 126 132 78 416 131 253 110	50 0 722 312 80 672 553 390 59 126 81 300 118 213 97	$ \begin{array}{c} -12 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ h \\ -4 \\ -10 \\ -16 \\ -18 \\ -24 \\ h \\ -4 \\ -4 \\ \end{array} $	385 349 379 101 227 176 56 425 174 235 339 130 	310 295 288 80 167 134 33 0 351 124 150 239 117 -0 88	$ \begin{array}{r} -8\\ -9\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -18\\ -19\\ -20\\ -21\\ -25\\ -26\end{array} $	243 66 278 342 41 139 172 128 45 56 141 48 156 122 78	212 87 279 398 45 134 204 129 54 70 144 41 180 133 60	$ \begin{array}{r} -3 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \end{array} $	204 104 254 144 237 59 153 50 121 48 332 150 179 91 95	196 96 247 134 249 68 152 42 127 29 358 157 212 73 113	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \end{array}$	352 165 141 29 74 200 34 139 129 42 131 173 53 81 70	276 163 99 21 67 209 16 156 119 59 123 180 51 90 45	$ \begin{array}{c} -23 \\ -23 \\ 24 \\ 28 \\ k: \\ -1 \\ -2 \\ -3 \\ -4 \\ 5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ \end{array} $	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47 242	121 84 165 2 130 191 168 390 99 313 40 73 40 261	$ \begin{array}{c} -24 \\ -24 \\ -11 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -8 \\ -9 \\ -12 \\ -13 \\ -14 \\ -16 \\ -18 \\ \hbar^{2} \end{array} $	65 =7 k = 109 238 90 42 67 222 210 48 200 74 136 104 126 =8 k =	36 2 106 224 67 31 56 201 53 197 60 153 122 153 122 153 2
13 15 h -2 -4 -6 -8 -10 -12 -12 -16 -12 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -20 -22 -24 -30 -	62 =1 &== 537 309 117 727 668 509 126 132 78 416 131 253 110 =2 &=	50 0 722 312 80 672 553 390 59 126 81 300 118 213 97 0	$ \begin{array}{r} -12 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ h^{2} \\ -4 \\ -10 \\ -16 \\ -18 \\ -24 \\ h^{2} \\ -4 \\ -6 \\ \end{array} $	385 349 379 101 227 176 56 425 174 235 339 130 7 <i>k</i> = 85 80	310 295 288 80 167 134 33 0 351 124 150 239 117 -0 88 77	$\begin{array}{c} -8\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -18\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\end{array}$	243 66 278 342 41 139 172 128 45 56 141 48 156 122 78 120	212 87 279 398 45 134 204 129 54 70 144 41 180 133 60 152	$\begin{array}{c} -1 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ \end{array}$	204 104 254 144 237 59 153 50 121 48 332 150 179 91 95 122	196 96 247 134 249 68 152 42 127 29 358 157 212 73 113 169	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ \end{array}$	352 165 141 29 74 200 34 139 129 42 131 173 53 81 70 46	276 163 99 21 67 209 16 156 119 123 180 51 90 45 55	$ \begin{array}{c} -23 \\ -23 \\ 24 \\ 28 \\ h^{2} \\ 1 \\ -2 \\ -3 \\ -4 \\ 5 \\ -6 \\ 7 \\ -8 \\ -9 \\ -10 \\ 12 \\ 12 \\ 12 $	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47 242 255 55	46 121 84 165 2 130 191 168 390 99 313 40 73 40 261 270	$\begin{array}{c} -24 \\ & A^{*} \\1 \\2 \\3 \\4 \\5 \\ -6 \\ -8 \\ -9 \\ -12 \\13 \\14 \\ -16 \\ -18 \\ A^{*} \\1 \end{array}$	65 =7 k = 109 238 90 42 67 222 210 48 200 74 136 104 125 =8 k = 145	36 2 106 224 67 31 56 201 53 197 60 153 122 153 2 153 2
13 15 3 -2 -4 -6 -8 -10 -12 -16 -12 -22 -24 -24 -	62 $=1 \ k =$ 537 309 117 727 668 509 126 132 78 416 131 253 110 $=2 \ k =$ 688 689 688 689 689 689 126 132 78 610 131 253 110 1000 100 100 100 100 100 100 100 100 100	50 0 722 312 80 672 553 390 59 126 81 300 118 213 97 0 812 304	$ \begin{array}{c} -10 \\ -112 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ -4 \\ -10 \\ -16 \\ -18 \\ -24 \\ -4 \\ -6 \\ -8 \\ -8 \\ -8 \\ -8 \\ -8 \\ -8 \\ -8 \\ -8$	385 349 379 101 227 176 56 56 425 174 235 339 130 	310 295 288 80 167 134 33 351 124 150 239 117 -0 88 77 81	$ \begin{array}{r} -8\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -18\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\\ -26\\ -26\\ -27\\ -26\\ -26\\ -26\\ -26\\ -26\\ -26\\ -26\\ -26$	243 66 278 342 41 139 172 128 45 56 141 48 156 122 78 120 = 3 29	212 87 279 398 45 134 204 129 54 70 144 41 180 133 60 152 152	$\begin{array}{c} -1 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -25 \end{array}$	204 104 254 144 237 59 153 50 121 48 332 150 179 91 95 122 58 22	196 96 247 134 249 68 152 42 127 29 358 157 212 73 113 169 82	$\begin{array}{c} -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \end{array}$	352 165 141 29 74 200 34 139 129 42 131 173 53 81 70 46 97	276 163 99 21 67 209 16 156 119 59 123 180 51 90 45 55 98	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -10 \\ -12 \\ -13 \\ -10 \\ -12 \\ -13 \\ -10 \\ -12 \\ -13 \\ -10 \\ -12 \\ -13 \\ -10 \\ -12 \\ -13 \\ -10 \\ -12 \\ -13 \\ -10 \\ -12 \\ -13 \\ -10 \\ -12 \\ -13 \\ -10 \\ -1$	$ \begin{array}{r} 41 \\ 86 \\ 75 \\ 151 \\ 132 \\ =4 \ k = \\ 165 \\ 205 \\ 178 \\ 385 \\ 113 \\ 303 \\ 48 \\ 69 \\ 47 \\ 242 \\ 255 \\ 82 \\ 106 $	46 121 84 168 165 22 130 191 168 390 99 313 40 73 40 261 270 78	$\begin{array}{c} -24\\ & & \\ & -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -113\\ & -14\\ & -16\\ & -18\\ & & \\ & & \\ & & \\ & -1\\ & & \\ & -2\\ & & \\ \end{array}$	63 =7 k = 109 238 90 42 67 222 210 48 200 74 136 104 126 =8 k = 145 67	36 2 106 224 67 31 56 201 53 197 60 153 122 153 2 121 79
$ \begin{array}{c} 14 \\ 15 \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -12 \\ -16 \\ -12 \\ -16 \\ -22 \\ -24 \\ -28 \\ -30 \\ -2 \\ -4 \\ -6 \\ -6 \\ -6 \\ -6 \\ -6 \\ -6 \\ -6 \\ -6$	62 62 537 309 117 727 668 509 126 132 78 416 131 253 110 -2 & 683 310 89	50 722 312 80 672 553 390 59 126 81 213 97 0 812 304 19	$ \begin{array}{c} -10 \\ -112 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ -4 \\ -10 \\ -16 \\ -18 \\ -24 \\ -4 \\ -6 \\8 \\8 \\10 \\12 \\10 \\12 \\10 \\10 \\12 \\10 \\$	385 349 379 101 227 176 56 425 174 235 339 130 -7 <i>k</i> = 85 80 133 118	310 295 288 80 167 134 33 351 124 150 2399 117 :0 88 877 81 97	$ \begin{array}{r} -8\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -18\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\\ -26\\ -27\\ -26\\ -27\\ -26\\ -27\\ -26\\ -27\\ -26\\ -27\\ -27\\ -26\\ -27\\ -27\\ -26\\ -27\\ -27\\ -26\\ -27\\ -27\\ -27\\ -26\\ -27\\ -27\\ -27\\ -26\\ -27\\ -27\\ -27\\ -26\\ -27\\ -27\\ -27\\ -27\\ -27\\ -27\\ -27\\ -27$	243 66 278 342 41 139 172 128 56 141 48 45 56 141 128 78 120 3 k= 282	212 87 279 398 45 134 204 129 54 70 144 41 180 133 60 152 =1 279 337	$\begin{array}{c} -3 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -26 \\ \end{array}$	203 204 254 144 237 59 153 50 121 153 332 150 121 121 179 91 58 58 73 73	342 196 96 247 134 249 68 152 42 127 2358 157 212 73 113 169 82 81	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \\ -27 \end{array}$	352 365 141 29 74 200 34 139 129 42 131 173 53 81 70 46 97 42	276 163 99 21 67 209 16 156 119 59 123 180 51 90 45 55 98 20 52	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ h^{2} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -14 \\ -15 \end{array}$	41 86 75 151 132 =4 k= 165 205 205 205 205 205 205 205 20	 101 121 84 165 2 130 191 158 390 99 313 40 251 261 270 78 203 104 	$\begin{array}{c} -24\\ & & \\ & -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -13\\ & -14\\ & -16\\ & -18\\ & & \\ & & \\ & & -1\\ & & \\ & & -2\\ & & \\ & & -4\\ & & \\ & & -1\\ & & \\ & & -2\\ & & \\ & & -4\\ & & \\ & & -1\\ & & \\ & & -2\\ & & \\ & & -4\\ & & \\ & & -1\\ & & \\ & & -2\\ & & \\ & & -4\\ & & \\ & & -1\\ & & \\ & & -2\\ & & \\ & & -4\\ & & \\ & & -1\\ & & \\ & & -2\\ & & \\ & & -4\\ & & \\ & & -1\\ & & \\ & & -2\\ & & \\ & & -4\\ & & \\ & & -1\\ & & \\ & & -2\\ & & \\ & & -2\\ & & \\ & & -2\\ & & \\ & & -2\\ & & \\ & & -2\\ & & \\ & & -2\\ & & \\ & & \\ & & -2\\ & & -2\\ & & \\ & & -2\\$	65 7 k= 109 238 90 42 67 222 210 48 200 48 200 48 104 126 -8 k= 145 -201 51 -20 -201 -	2 106 224 67 31 56 201 201 53 197 60 153 122 153 2 121 79 181 0
$\begin{array}{c} 13 \\ 15 \\ -2 \\ -2 \\4 \\ -6 \\ -8 \\ -10 \\ -12 \\ -16 \\ -10 \\ -12 \\ -22 \\ -24 \\ -28 \\ -30 \\ h \\ -2 \\ -4 \\ -6 \\ -8 \end{array}$	62 62 537 309 117 727 668 509 126 132 78 416 131 253 110 2 &= 683 310 9 429	50 0 722 312 80 672 553 390 59 126 81 300 118 213 97 0 812 304 19 401	$ \begin{array}{c} -10\\ -12\\ -16\\ -18\\ -20\\ -22\\ -24\\ -26\\ h_{1}\\ -26\\ -18\\ -16\\ -18\\ -24\\ -6\\ -8\\ -8\\ -10\\ -12\\ -16\\ -12\\ -16\\ \end{array} $	385 349 379 101 227 176 56 6 <i>k</i> = 425 174 235 339 130 =7 <i>k</i> = 85 80 133 118 156 324	310 310 2285 80 167 134 33 50 351 124 150 239 117 60 88 77 81 97 124 209	$\begin{array}{c} -8\\ -9\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -18\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\\ -26\\ -27\\ -2\\ -3\end{array}$	243 66 278 342 41 139 172 128 45 56 141 48 156 122 78 120 =3 k= 281 2382 201	212 87 229 338 45 134 2204 129 54 132 160 112 279 337 161	$\begin{array}{c} -3 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -26 \\ -1 \end{array}$	203 204 104 254 144 237 59 153 50 121 48 332 150 179 91 91 95 122 58 33 26 <i>k</i> = 73 73 73 242	342 196 96 247 134 249 68 152 42 127 29 358 157 212 73 113 1692 82 11 21 231	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \\ -27 \\ -28 \end{array}$	352 352 165 141 29 74 200 34 139 129 42 131 173 53 81 70 46 97 42 41 103	276 163 99 21 67 209 16 156 119 59 123 180 51 90 51 90 55 58 20 22 23	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ h^{2} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \end{array}$	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47 242 255 82 196 92 124	 121 84 165 130 191 168 390 99 313 40 73 40 261 270 78 203 104 169 	$\begin{array}{c} -24\\ & & & \\ -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -13\\ & -14\\ & -16\\ & -18\\ & & \\ & & \\ -1\\ & -2\\ & & \\ & -4\\ & -6\\ & -8\end{array}$	65 7 k= 109 238 90 42 67 222 210 48 200 74 136 104 126 =8 k= 145 61 136	2 106 224 67 31 56 201 201 53 197 60 153 122 153 2 153 2 153 2 153 122 153 2 153 139
$\begin{array}{c} 13 \\ 15 \\ -2 \\ -2 \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -12 \\ -16 \\ -20 \\ -22 \\ -24 \\ -28 \\ -30 \\ h \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \end{array}$	62 62 537 309 117 727 668 509 126 132 78 416 131 253 110 2 &= 683 310 89 429 509	50 0 722 312 80 672 553 390 59 126 81 213 97 -0 812 304 19 401 515	$ \begin{array}{c} -10\\ -116\\ -18\\ -20\\ -22\\ -24\\ -26\\ h_{1}\\ -26\\ -10\\ -16\\ -18\\ -24\\ -6\\ -8\\ -10\\ -12\\ -16\\ -18\\ -10\\ -12\\ -16\\ -18\\ -10\\ -12\\ -16\\ -18\\ -10\\ -12\\ -16\\ -18\\ -10\\ -18\\ -10\\ -12\\ -16\\ -18\\ -18\\ -10\\ -12\\ -16\\ -18\\ -18\\ -10\\ -12\\ -16\\ -18\\ -18\\ -10\\ -12\\ -16\\ -18\\ -18\\ -10\\ -12\\ -16\\ -18\\ -10\\ -12\\ -16\\ -18\\ -18\\ -10\\ -12\\ -16\\ -18\\ -18\\ -10\\ -12\\ -16\\ -18\\ -18\\ -10\\ -12\\ -16\\ -18\\ -18\\ -10\\ -12\\ -16\\ -18\\ -10\\ -12\\ -16\\ -18\\ -10\\ -12\\ -16\\ -18\\ -10\\ -12\\ -16\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -10\\ -18\\ -18\\ -10\\ -18\\ -10\\ -18\\ -18\\ -10\\ -18\\ -18\\ -18\\ -10\\ -18\\ -18\\ -18\\ -18\\ -10\\ -18\\ -18\\ -18\\ -18\\ -18\\ -18\\ -18\\ -18$	385 349 379 101 227 176 56 425 174 235 339 130 -7 k= 85 80 133 118 156 324 115	310 310 2285 80 167 134 33 351 124 150 239 117 60 88 87 77 81 77 124 209 97 4	$\begin{array}{c} -8\\ -9\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -18\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\\ -2\\ -2\\ -2\\ -2\\ -3\\ -4\end{array}$	243 66 278 342 41 139 172 128 45 56 141 48 156 122 78 120 3 k= 281 382 281 109 172	212 87 279 398 45 134 204 129 54 133 100 144 41 180 00 133 60 279 337 161 142	$\begin{array}{c} -3 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -26 \\ -1 \\ -3 \\ \end{array}$	203 204 104 254 144 237 59 153 50 121 48 332 150 179 91 91 91 95 58 33 22 58 73 73 73 73 242 242 104	342 196 96 247 134 249 68 152 42 127 29 358 157 212 73 113 169 82 81 21 231 109	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \\ -27 \\ -28 \\ -30 \\ \end{array}$	352 352 165 141 29 74 200 34 139 129 42 131 173 53 81 70 46 97 42 41 103 42	276 163 99 21 67 209 16 156 159 16 159 123 180 51 90 45 55 98 20 139 55	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ h^{2} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \end{array}$	41 86 75 151 132 -4 k= 165 205 178 385 113 303 48 69 47 242 255 82 255 82 196 92 124 90	 48 121 84 165 15 165 390 99 313 40 73 40 73 40 261 270 78 203 104 169 99 	$\begin{array}{c} -24\\ & & & \\ -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -13\\ & -14\\ & -16\\ & -18\\ & & \\ & & \\ -1\\ & -2\\ & & \\ & -4\\ & & \\ & -6\\ & & \\ & -8\\ & & \\ & -10\end{array}$	65 7 k= 109 238 90 42 67 222 210 48 200 74 136 104 126 61 136 61 136 240	350 22 106 224 67 31 56 201 201 53 197 60 153 122 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 239
$\begin{array}{c} 14 \\ 15 \\ h^{+} \\ -2 \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -12 \\ -16 \\ -12 \\ -22 \\ -24 \\ -28 \\ -30 \\ h \\ -2 \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -12 \end{array}$	62 62 537 309 117 727 668 509 126 132 78 416 131 253 110 =2 4= 683 310 89 429 509 420	50 0 722 312 80 672 553 390 59 126 81 213 97 -0 812 304 19 401 515 313	$ \begin{array}{c} -10\\ -116\\ -18\\ -20\\ -22\\ -24\\ -26\\ h_{1}\\ -26\\ -16\\ -18\\ -24\\ -6\\ -8\\ -10\\ -112\\ -16\\ -18\\ -20\\ \end{array} $	385 349 379 101 227 176 56 425 174 235 339 130 -7 <i>k</i> = 85 80 133 113 156 324 115 55	310 3195 288 80 167 134 33 351 124 150 239 117 60 88 77 81 97 124 209 94 53	$\begin{array}{c} -8\\ -9\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -18\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\\ -2\\ -2\\ -2\\ -3\\ -4\\ -5\end{array}$	243 66 278 342 41 139 172 128 45 56 124 141 48 156 122 78 120 = 3 k= 281 281 281 29 109 172 2358	212 212 279 398 45 134 204 129 54 45 134 204 129 54 41 180 133 60 162 162 279 337 161 142 242	$\begin{array}{c} -3 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -26 \\ -1 \\ -3 \\ -4 \\ \end{array}$	204 204 104 254 144 237 59 153 50 121 150 179 91 95 122 258 332 150 95 122 26 <i>k</i> - 26 <i>k</i> - 242 2104 140	342 196 247 134 249 68 152 42 127 29 358 157 212 73 113 169 82 81 231 109 144	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \\ -27 \\ -28 \\ -30 \\ \delta \end{array}$	352 165 167 167 167 169 74 200 34 139 129 42 131 131 53 81 70 46 97 42 41 131 103 42 42 41 131 20 42 42 42 42 42 42 42 42 42 42	276 163 99 21 67 209 16 156 119 59 123 180 51 90 45 55 98 20 139 55 22 23 23 23 23 23 23 23 23 23	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ h^{2} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \end{array}$	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47 242 255 82 255 82 196 92 124 90 106	121 121 84 168 165 2 130 191 168 390 99 313 40 73 40 261 270 78 203 104 169 99 112	$\begin{array}{c} -24\\ & & & \\ -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -13\\ & -14\\ & -16\\ & -18\\ & & \\ & & \\ -10\\ & -2\\ & -4\\ & -6\\ & -8\\ & -10\\ & -12\end{array}$	63 7 k= 109 238 90 42 67 222 210 48 200 74 136 104 136 104 126 61 136 61 136 240 240	350 22 106 224 67 31 56 201 201 53 197 60 153 122 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 139 239 40
$\begin{array}{c} 14 \\ 15 \\ 5 \\ -2 \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -12 \\ -16 \\ -12 \\ -22 \\ -24 \\ -28 \\ -30 \\ h \\ -2 \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -12 \\ -14 \end{array}$	62 62 537 309 117 727 668 509 126 132 78 416 131 253 110 83 310 89 429 509 420 732	50 0 722 312 80 672 553 390 59 126 81 213 97 -0 812 304 19 401 515 313 616	$ \begin{array}{c} -10\\ -12\\ -16\\ -18\\ -20\\ -22\\ -24\\ -26\\ h\\ -26\\ -10\\ -16\\ -18\\ -24\\ -6\\ -8\\ -10\\ -112\\ -16\\ -18\\ -20\\ \hbar\\ -20\\ \hbar \end{array} $	385 389 379 101 227 176 56 <i>s</i> =6 425 174 425 235 339 130 -7 <i>s</i> = 85 80 133 118 156 324 115 55 55 	310 3195 288 80 167 134 33 351 124 150 239 117 60 88 77 81 17 124 209 91 124 53 50 53 50	$\begin{array}{c} -8\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -18\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\\ -2\\ -3\\ -4\\ -5\\ -6\end{array}$	243 66 278 342 41 139 172 128 45 56 141 48 156 122 78 120 = 3 k= 281 281 281 29 109 172 2358 149	212 212 279 398 45 134 204 129 54 45 134 204 129 54 41 180 133 60 162 162 279 337 161 142 242 342 132	$\begin{array}{c} -3 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -26 \\ -1 \\ -3 \\ -4 \\ -5 \end{array}$	204 204 104 254 144 237 59 153 50 121 150 179 91 95 122 258 332 150 179 91 95 122 26 <i>k</i> 242 104 104 208	342 196 247 134 249 68 152 42 127 29 358 157 212 73 113 169 82 81 231 109 144 224	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \\ -27 \\ -28 \\ -30 \\ -1 \end{array}$	352 165 141 29 74 200 34 139 42 131 173 53 81 70 46 97 42 131 103 42 131 103 42 131 105 131 97 42 42 131 139 42 42 131 139 42 42 42 42 42 42 42 42 42 42	276 163 99 21 67 209 16 156 119 59 123 180 51 90 45 55 98 20 139 55 22 139 25 55 22 139	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ h^{2} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \\ -19 \end{array}$	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47 242 255 82 196 92 124 90 106 96	121 121 84 168 165 2 130 191 168 390 99 313 40 73 40 261 270 78 203 104 169 99 9112 103	$\begin{array}{c} -24\\ & & & \\ & -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -13\\ & -14\\ & -16\\ & -18\\ & & \\ & -11\\ & -2\\ & -4\\ & -6\\ & -8\\ & -8\\ & -10\\ & -12\\ & -14\end{array}$	65 7 k = 109 228 90 42 67 222 210 48 200 74 136 104 126 =8 k = 145 67 201 136 240 240 241 136 240 241 245 245 245 245 245 245 245 245	350 22 106 224 67 31 56 201 201 53 197 60 153 122 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 239 40 164
$\begin{array}{c} 14 \\ 15 \\ h^{*} \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -12 \\ -16 \\ -10 \\ -22 \\ -24 \\ -28 \\ -30 \\ h \\ -10 \\ -12 \\ -14 \\ -16 \end{array}$	62 62 537 309 117 727 668 509 126 132 78 416 131 253 110 83 310 89 429 509 420 732 465	50 0 722 312 80 672 553 390 59 126 81 213 97 -0 812 304 19 401 515 313 616 368	$ \begin{array}{c} -10\\ -12\\ -16\\ -18\\ -20\\ -22\\ -24\\ -26\\ h_{-}\\ -26\\ -10\\ -16\\ -18\\ -24\\ -6\\ -8\\ -10\\ -12\\ -16\\ -18\\ -20\\ -4\\ -4\\ -6\\ -7\\ -4\\ -6\\ -7\\ -4\\ -6\\ -7\\ -4\\ -6\\ -7\\ -4\\ -6\\ -7\\ -4\\ -6\\ -7\\ -7\\ -4\\ -6\\ -7\\ -7\\ -7\\ -7\\ -7\\ -7\\ -7\\ -7\\ -7\\ -7$	385 389 379 101 227 176 56 425 235 339 130 -7 85 80 133 119 156 324 115 55 55 -8 4 -8 4	310 295 288 80 167 134 33 351 124 150 239 117 150 88 877 81 17 97 81 97 124 53 94 53 90 231	$\begin{array}{c} -8\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\\ -2\\ -3\\ -4\\ -5\\ -6\\ -7\end{array}$	243 66 278 342 41 139 172 128 45 56 124 122 78 120 = 3 k= 281 281 281 281 29 172 2358 149 214	313 212 212 212 87 279 398 45 134 204 129 54 70 144 180 133 60 162 161 142 342 342 132 190	$\begin{array}{c} -1 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -26 \\ -1 \\ -3 \\ -4 \\ -5 \\ -7 \end{array}$	203 204 104 254 144 237 59 153 50 121 150 179 91 95 122 258 33 2 57 3 73 73 73 73 73 73 73 73 73 73 73 73	342 196 247 134 249 68 152 42 127 29 358 157 212 73 113 169 82 81 231 109 144 255	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \\ -27 \\ -28 \\ -30 \\ -1 \\ -2 \\ -2 \\ -2 \\ -2 \\ -28 \\ -30 \\ -1 \\ -2 \end{array}$	352 165 167 167 167 167 167 169 74 200 34 139 42 131 173 53 81 70 46 97 42 41 103 42 41 103 42 41 103 42 42 41 105 105 105 105 105 105 105 10	276 163 99 21 67 209 16 156 119 59 123 180 51 90 45 55 98 20 139 55 22 177 361	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ h^{2} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \\ -19 \\ -20 \end{array}$	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47 242 255 82 196 92 124 90 106 96 118	 48 121 84 165 2 130 191 168 390 99 313 40 73 40 261 203 104 169 99 112 103 114 	$\begin{array}{c} -24\\ & & & \\ & -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -13\\ & -14\\ & -16\\ & -18\\ & & \\ & -11\\ & -2\\ & -4\\ & -6\\ & -8\\ & -8\\ & -10\\ & -12\\ & -14\\ & -15\end{array}$	65 7 k = 109 228 90 42 67 222 210 48 200 74 136 104 126 =8 k = 145 67 201 136 240 24 240 240 59 40 40 45 45 45 45 45 45 45 45 45 45	3% 22 106 224 67 31 56 201 201 53 197 60 153 122 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 239 40 164 51
$\begin{array}{c} 14 \\ 15 \\ 5 \\ h^{*} \end{array}$ $\begin{array}{c} -2 \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -12 \\ -16 \\ -12 \\ -22 \\ -24 \\ -28 \\ -30 \\ h \\ -12 \\ -4 \\ -6 \\ -8 \\ -10 \\ -112 \\ -14 \\ -16$	62 62 537 309 117 727 668 509 126 132 78 416 131 253 110 89 429 509 429 509 420 732 465 91	50 0 722 312 80 672 553 390 59 126 81 213 97 -0 812 304 19 401 515 313 616 368 100 200 200 200 200 200 200 200	$ \begin{array}{c} -10\\ -12\\ -16\\ -18\\ -20\\ -22\\ -24\\ -26\\ h_{-}\\ -26\\ -10\\ -16\\ -18\\ -24\\ -6\\ -8\\ -10\\ -12\\ -16\\ -18\\ -20\\ -4\\ -4\\ -6\\ -10\\ -12\\ -16\\ -18\\ -20\\ -4\\ -10\\ -12\\ -16\\ -18\\ -20\\ -4\\ -10\\ -10\\ -12\\ -16\\ -18\\ -20\\ -4\\ -10\\ -10\\ -10\\ -10\\ -10\\ -10\\ -10\\ -10$	385 389 379 101 227 176 56 425 174 425 339 130 133 158 155 55 55 55 55 324 118 118 155 55 55 174 155 55 55 56 80 133 118 156 156 156 156 156 157 176 176 176 176 176 176 176 17	310 3195 288 80 167 134 33 351 124 150 239 117 60 88 77 81 17 209 94 53 60 231 74 74 209 231 74 231 245 245 245 245 245 245 245 245	$\begin{array}{c} -8\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\\ -2\\ -3\\ -4\\ -5\\ -6\\ -7\\ -8\\ \end{array}$	243 66 278 342 41 139 172 128 45 56 124 141 48 156 122 78 120 = 3 k= 281 20 109 172 358 149 172	212 212 279 398 45 134 204 129 54 41 129 54 41 180 133 60 162 152 279 337 161 162 279 337 161 142 242 132	$\begin{array}{c} -3 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -26 \\ -1 \\ -3 \\ -4 \\ -5 \\ -7 \\ -9 \\ -9 \\ -9 \\ -9 \\ -1 \\ -3 \\ -4 \\ -5 \\ -7 \\ -9 \\ -9 \\ -9 \\ -9 \\ -9 \\ -1 \\ -1 \\ -1$	204 204 104 254 144 237 59 153 50 121 48 332 150 179 91 95 122 58 332 242 242 242 244 104 104 208 78 87 78	196 196 247 134 249 68 152 42 29 358 157 212 73 113 169 82 11 231 109 144 255 97	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \\ -27 \\ -28 \\ -30 \\ -1 \\ -2 \\ -3 \\ -3 \\ -1 \\ -2 \\ -3 \\ -3 \\ -3 \\ -3 \\ -3 \\ -3 \\ -3$	352 165 167 167 167 167 167 167 169 129 42 131 173 53 81 70 46 97 42 41 103 42 41 103 42 41 103 42 41 103 42 42 42 41 105 129 42 42 42 42 42 42 42 42 42 42	276 163 99 21 67 209 16 7209 16 156 119 59 123 180 51 90 45 55 98 20 21 139 55 20 139 21 23 23 23 23 23 23 23 23 23 23	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ h^{2} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \\ -19 \\ -20 \\ -22 \\ \end{array}$	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47 242 255 82 196 92 124 90 106 96 118 148 148 164 178 113 113 113 113 113 113 113 11	 48 121 84 165 2 130 191 168 390 99 313 40 73 40 264 270 78 203 104 169 99 112 103 114 175 	$\begin{array}{c} -24\\ & & & \\ & -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -13\\ & -14\\ & -16\\ & -18\\ & & \\ & -12\\ & -14\\ & -6\\ & -8\\ & -10\\ & -12\\ & -14\\ & -15\\ & & \\ & & \\ & & \\ \end{array}$	65 7 k = 109 228 90 42 67 222 210 48 200 74 136 104 126 -8 k = 145 67 201 1.36 240 240 1.36 240 240 -8 k = 1.09 -8 k = -8	350 22 106 224 67 31 56 201 201 203 197 60 153 122 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 164 51 -2 -164 51
$\begin{array}{c} 14 \\ 15 \\ h^{*} \\ -2 \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -112 \\ -16 \\ -10 \\ -22 \\ -24 \\ -28 \\ -30 \\ h \\ -2 \\ -24 \\ -28 \\ -30 \\ h \\ -10 \\ -112 \\ -14 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -8 \\ -10 \\ -12 \\ -14 \\ -16 \\ -8 \\ -20 \\ -22 \\ -24 \\ -28 \\ -30 \\ h \\ -2 \\ -28 \\ -30 \\ -2 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -28 \\ -30 \\ -30 \\ -28 \\ -30 \\ -30 \\ -28 \\ -30 \\ -30 \\ -28 \\ -30 \\ -$	62 62 537 309 117 727 668 509 126 132 78 416 131 253 110 83 429 509 429 509 429 732 465 91 368	50 0 722 312 80 672 553 390 59 126 81 213 97 70 812 304 19 401 515 313 616 3688 100 319 72 72 72 72 72 72 72 72 72 72	$ \begin{array}{c} -10 \\ -12 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ h \\ -26 \\ -4 \\ -10 \\ -16 \\ -18 \\ -24 \\ -6 \\ -8 \\ -10 \\ -12 \\ -16 \\ -18 \\ -20 \\ -4 \\ -10 \\ -12 \\ -16 \\ -18 \\ -20 \\ -4 \\ -10 \\ -14 \\ -14 \\$	385 389 379 101 227 176 56 425 174 425 133 130 -7 85 80 133 118 155 55 -8 4	310 3195 2888 80 167 1344 33 351 124 150 239 117 60 88 77 81 97 124 53 97 209 94 53 50 231 74 57 231 205 231 235 235 235 235 235 235 235 235	$\begin{array}{c} -8\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -18\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\\ -2\\ -3\\ -4\\ -5\\ -6\\ -7\\ -8\\ -9\\ -9\\ -2\\ -3\\ -4\\ -5\\ -6\\ -7\\ -8\\ -9\\ -9\\ -2\\ -2\\ -3\\ -4\\ -5\\ -6\\ -7\\ -8\\ -9\\ -9\\ -2\\ -2\\ -2\\ -2\\ -2\\ -2\\ -2\\ -2\\ -2\\ -2$	243 66 278 342 41 139 172 128 45 56 141 48 156 122 78 120 	212 212 279 398 45 134 204 129 54 41 180 133 60 152 279 337 161 152 279 337 161 142 242 132 190 207 100	$\begin{array}{c} -3 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -26 \\ -1 \\ -3 \\ -4 \\ -5 \\ -7 \\ -91 \\ -11 \\ $	204 204 104 254 144 237 59 153 50 121 150 179 91 95 122 242 242 208 73 73 73 74 242 244 208 78 87 181	196 196 247 134 249 68 152 42 127 29 358 157 212 73 113 169 82 81 231 109 144 255 97 178 61	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \\ -27 \\ -28 \\ -30 \\ -1 \\ -2 \\ -3 \\ -1 \\ -2 \\ -3 \\ -1 \\ -2 \\ -3 \\ -5 \\ -5 \\ -5 \\ -5 \\ -5 \\ -5 \\ -5$	352 365 141 29 74 200 34 139 129 42 131 173 53 81 173 53 81 173 53 81 103 97 42 41 103 42 129 42 139 42 42 42 42 42 42 42 42 42 42	276 163 99 21 67 209 16 156 119 59 123 180 51 90 45 55 98 20 139 55 22 177 361 218 91 21 20 21 20 20 21 20 21 20 20 21 20 20 20 20 20 20 20 20 20 20	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ h^{2} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \\ -19 \\ -20 \\ -22 \\ $	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47 242 255 82 196 92 124 90 106 96 118 144 66 96 118 124 106 113 113 113 113 113 113 113 11	101 121 84 165 2 130 191 168 390 313 40 264 203 104 169 99 112 103 114 175 82	$\begin{array}{c} -24\\ & & & \\ & -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -13\\ & -14\\ & -16\\ & -18\\ & & \\ & -11\\ & -12\\ & -4\\ & -6\\ & -8\\ & -10\\ & -12\\ & -14\\ & -15\\ & & \\$	65 7 k = 109 228 90 42 67 222 210 48 200 74 136 104 126 =8 k = 145 67 201 136 240 24 240 24 136 136 136 240 24 245 208 208 208 208 208 208 208 208	3% 22 106 224 67 31 56 201 201 203 197 60 153 122 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 239 40 164 51 -2 146
$\begin{array}{c} 14 \\ 15 \\ h^{*} \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -112 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -28 \\ -30 \\ h \\ -12 \\ -14 \\ -16 \\ -18 \\ -10 \\ -112 \\ -14 \\ -16 \\ -18 \\ -20 \\ -22 \\ -26 \\ -22 \\ -26 \\ -22 \\ -26 \\ -26 \\ -22 \\ -26$	62 62 537 309 117 727 668 509 126 132 78 416 131 253 110 89 429 509 429 509 429 732 465 91 368 110 257	50 0 722 312 80 672 553 390 59 126 81 213 97 70 812 304 19 401 515 313 616 3688 100 319 97 72 210 80 97 210 80 80 81 80 80 81 80 80 81 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 81 80 81 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 80 81 80 81 80 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 81 80 80 80 80 80 80 80 80 80 80	$\begin{array}{c} -10 \\ -12 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ h \\ -26 \\ h \\ -10 \\ -16 \\ -18 \\ -24 \\ h \\ -6 \\ -8 \\ -10 \\ -112 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -14 \\ -10 \\ -14 \\ -10 \\ h \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -14 \\ -10 \\ -14 \\ -16 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -14 \\ -10 \\ -14 \\ -16 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -14 \\ -10 \\ -14 \\ -16 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -14 \\ -10 \\ -14 \\ -10 \\ -14 \\ -16 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -14 \\ -10 \\ -14 \\ -16 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -14 \\ -16 \\ -16 \\ -18 \\ -20 \\ h \\ -10 \\ -14 \\ -10 \\ -16 \\ -16 \\ -18 \\ -20 $	385 387 389 379 101 227 176 56 425 174 425 339 130 133 156 80 133 118 155 55 55 55 55 117 118 56 81 118 118 115 55 55 117 118 118 118 118 119 59 42 119 119 119 119 119 119 119 11	310 319 2285 288 80 167 134 33 351 124 150 239 117 50 88 77 81 17 209 94 53 -0 231 74 57 136 136 136 136 137 124 150 124 150 239 117 124 117 124 150 239 117 124 150 239 117 124 150 124 150 124 117 124 150 124 117 124 117 124 117 124 150 124 117 124 150 124 117 124 150 124 117 124 150 124 117 124 150 124 150 124 150 124 150 124 150 124 150 124 150 124 150 124 150 124 150 124 150 124 150 150 150 150 150 150 150 150	$\begin{array}{c} -8\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -18\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\\ -2\\ -3\\ -4\\ -5\\ -6\\ -7\\ -8\\ -9\\ -10\\ -11\\ \end{array}$	243 66 278 342 41 139 172 128 45 56 124 141 48 156 122 78 120 = 3 k= 281 20 109 172 358 149 214 212 256 248	212 212 279 398 45 134 204 129 54 41 129 54 41 180 133 60 162 152 279 337 161 162 142 242 132 190 207 100 213 212	$\begin{array}{c} -3 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -26 \\ -1 \\ -3 \\ -4 \\ -5 \\ -7 \\ -9 \\ -11 \\ -15 \\ -15 \\ -15 \\ -7 \\ -9 \\ -11 \\ -15 \\ -15 \\ -7 \\ -9 \\ -11 \\ -15 \\ -15 \\ -7 \\ -9 \\ -11 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -7 \\ -9 \\ -15 \\ -7 \\ -9 \\ -7 \\ -7$	204 204 104 254 144 237 59 153 50 121 153 50 121 150 179 91 95 122 258 332 215 26 <i>k</i> = 242 2104 104 140 208 87 181 9204	342 196 247 134 249 68 152 42 229 358 157 212 73 113 169 82 113 109 231 109 144 255 97 178 238	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \\ -27 \\ -28 \\ -30 \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \end{array}$	352 365 141 29 74 200 34 139 129 42 131 173 53 81 173 53 81 173 53 81 103 97 42 41 103 97 42 42 139 129 42 139 129 42 139 129 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 131 109 42 200 42 42 131 109 42 200 42 42 42 42 42 42 42 42 42 42	276 163 99 21 67 209 16 156 119 59 123 180 51 90 45 55 98 20 139 55 22 177 361 218 91 91 90 91 90 99 99 10 10 10 10 10 10 10 10 10 10	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ h^{2} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \\ -19 \\ -20 \\ -22 \\ -26 \\ -27 \\ z^{\prime} \end{array}$	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47 242 255 82 196 92 124 90 106 96 118 144 66 55 5 5 5 5 5 5 5 5 5 5 5 5	 40 121 84 165 2 130 191 168 390 99 313 40 73 40 264 270 78 203 404 203 104 169 99 112 103 114 175 82 59 20 	$\begin{array}{c} -24\\ & & & \\ & -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -13\\ & -14\\ & -16\\ & -18\\ & & \\ & -12\\ & -14\\ & -16\\ & -18\\ & & \\ & -12\\ & -4\\ & -6\\ & -8\\ & -10\\ & -12\\ & -14\\ & -15\\ & & \\ & -2\\ & -3\\ & -4\\ & & \\ & -2\\ & -3\\ & -4\\ & & \\ & -2\\ & -3\\ & -4\\ & & \\ & -2\\ & -3\\ & -4\\ & & \\ & -2\\ & -3\\ & -4\\ & & \\ & -2\\ & -3\\ & -4\\ & & \\ & -2\\ & -3\\ & -4\\ & & \\ & -2\\ & -3\\ & -4\\ & & \\ & -2\\ & -3\\ & -4\\ & & \\ & -2\\ & -2\\ & -3\\ & -4\\ & & \\ & -2\\ & -2\\ & -4\\ & & \\ & -2\\ & -2\\ & -4\\ & & \\ & -2\\ & -2\\ & -4\\ & & \\ & -2\\ & -2\\ & -4\\ & & \\ & -2\\ & $	65 7 k = 109 228 90 42 67 222 210 48 200 74 136 104 126 -8 k = 145 67 201 1.36 240 240 240 -8 k = 1.36 240 245 245 245 206 -8 k = 245 207 -8 k = 245 207 -8 k = 245 207 -8 k = 207 -8 k = 207 -9 k =	3% 22 106 224 67 31 56 201 201 203 197 60 153 122 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 164 51 52 146 31
$\begin{array}{c} 14 \\ 15 \\ h^{*} \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -112 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -28 \\ -30 \\ h \\ -12 \\ -14 \\ -16 \\ -18 \\ -10 \\ -112 \\ -14 \\ -16 \\ -18 \\ -20 \\ -22 \\ -26 \\ -30 \end{array}$	62 62 537 309 117 727 668 509 126 132 78 416 131 253 110 253 110 89 429 509 429 732 465 91 368 110 267 264	50 0 722 312 80 553 390 59 126 81 213 97 70 812 304 19 401 515 313 616 6368 100 319 97 72 10 80 118 118 10 118 10 10 10 10 10 10 10 10 10 10	$\begin{array}{c} -10 \\ -12 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ h \\ -22 \\ -24 \\ -6 \\ -10 \\ -16 \\ -18 \\ -24 \\ -6 \\ -8 \\ -10 \\ -112 \\ -16 \\ -18 \\ -20 \\ -4 \\ -10 \\ -14 \\ -10 \\ -14 \\ -10 \\ -14 \\ -2 \\ -2 \\ -2 \\ -2 \\ -2 \\ -2 \\ -2 \\ -$	385 387 388 379 101 227 176 56 425 174 425 339 130 133 156 80 133 118 155 55 55 55 55 117 118 58 415 55 55 55 55 55 56 117 176 176 176 176 176 176 17	310 295 288 80 167 134 33 351 124 150 239 117 88 77 81 97 124 209 94 53 97 124 53 97 124 209 94 53 51 124	$\begin{array}{c} -8\\ -9\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -19\\ -20\\ -21\\ -25\\ -26\\ -27\\ k\\ -1\\ -2\\ -3\\ -4\\ -5\\ -6\\ -7\\ -8\\ -9\\ -10\\ -11\\ -12\end{array}$	243 66 278 342 41 139 172 128 45 56 141 141 48 156 122 78 129 128 141 139 281 109 172 281 109 281 281 281 281 281 281 281 281	212 212 279 398 45 134 204 129 54 47 70 144 180 133 60 152 279 337 161 152 279 337 161 142 242 132 190 207 100 213 212 188	$\begin{array}{c} -3 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -26 \\ -1 \\ -3 \\ -4 \\ -5 \\ -7 \\ -9 \\ -111 \\ -12 \\ -15 \\ -18 \\ $	204 204 104 254 144 237 59 153 50 121 153 50 121 159 91 95 122 258 332 159 91 95 122 268 87 181 93 204 86	196 196 247 134 249 68 152 42 29 358 157 212 73 113 169 82 113 169 82 231 109 144 255 97 178 81 238 84	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \\ -27 \\ -28 \\ -30 \\ -30 \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \end{array}$	352 365 141 29 74 200 34 139 129 42 131 173 53 81 173 53 81 173 53 81 103 97 42 41 103 42 139 42 42 42 42 42 42 42 42 42 42	276 163 99 211 67 209 16 156 119 59 123 180 51 19 05 55 98 20 52 139 55 55 98 20 52 139 55 55 98 20 163 199 119 156 119 159 123 180 51 199 123 180 55 98 20 180 55 98 20 199 199 109 119 59 123 180 55 55 98 20 109 109 109 109 109 109 109 10	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ h^{2} \\ -2 \\ -28 \\ h^{2} \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \\ -19 \\ -20 \\ -22 \\ -26 \\ -27 \\ h^{2} \\ -1 \\ \end{array}$	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47 242 255 82 196 92 124 90 106 96 118 144 66 5 5 k= 42	101 121 84 165 2 130 191 168 390 313 40 264 200 104 169 99 112 103 114 175 82 59 2 31	$\begin{array}{c} -24\\ & & & \\ & -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -13\\ & -14\\ & -16\\ & -18\\ & & \\ & -12\\ & -14\\ & -16\\ & -18\\ & & \\ & -12\\ & -4\\ & -6\\ & -8\\ & -10\\ & -12\\ & -14\\ & -15\\ & & \\ & -2\\ & -3\\ & -4\\ & -7\end{array}$	65 7 109 228 90 42 67 222 210 48 200 74 136 104 126 -8 201 136 104 126 -8 201 136 201 136 201 1376 202 203 -8 -145 67 201 240 24 159 40 -9 218	30 22 106 224 67 31 56 201 201 203 197 60 153 122 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 153 2 164 51 -2 146 31 41 120
$\begin{array}{c} 14 \\ 15 \\ h^{*} \\ -2 \\ -4 \\ -6 \\ -8 \\ -10 \\ -112 \\ -16 \\ -10 \\ -22 \\ -24 \\ -28 \\ -30 \\ h \\ -2 \\ -24 \\ -28 \\ -30 \\ h \\ -10 \\ -112 \\ -14 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -5 \\ -30 \\ h \\ -2 \\ -26 \\ -30 \\ h \\ -22 \\ -26 \\ -30 \\ -22 \\ -26 \\ -30 \\ h \\ -26 \\ -30 \\ h \\ -26 \\ -30 \\ -22 \\ -26 \\ -30 \\ h \\ -26 \\ -30 \\ -26 \\ -30 \\ h \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -26 \\ -30 \\ -30 \\ -26 \\ -30 \\ -30 \\ -26 \\ -30 $	62 62 537 309 117 727 668 509 126 131 132 78 416 132 78 416 132 78 416 133 310 89 429 509 429 509 429 509 420 772 78 420 79 20 420 79 20 420 70 20 420 70 20 420 20 420 20 420 20 420 20 420 20 420 20 420 20 420 20 420 20 420 20 420 20 420 4	50 0 722 312 80 672 553 390 59 126 81 213 97 70 812 304 19 401 515 313 304 19 401 515 313 304 19 401 515 313 304 19 97 210 812 80 97 210 812 81 81 81 81 81 81 81 81 81 81	$\begin{array}{c} 10 \\ -12 \\ -16 \\ -18 \\ -20 \\ -22 \\ -24 \\ -26 \\ -4 \\ -10 \\ -16 \\ -18 \\ -24 \\ -6 \\ -8 \\ -10 \\ -12 \\ -16 \\ -18 \\ -20 \\ -4 \\ -10 \\ -12 \\ -16 \\ -18 \\ -20 \\ -4 \\ -10 \\ -14 \\ -16 \\ -8 \\ -20 \\ -4 \\ -6 \\ -8 \\ -20 \\ -4 \\ -6 \\ -8 \\ -20 \\ -4 \\ -6 \\ -8 \\ -20 \\ -4 \\ -10 \\ -14 \\ -16 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -10 \\ -14 \\ -16 \\ -16 \\ -18 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -10 \\ -14 \\ -16 \\ -18 \\ -2 \\ -4 \\ -10 \\ -14 \\ -16 \\ -16 \\ -18 \\ -2 \\ -4 \\ -10 \\ -14 \\ -16 \\ -16 \\ -18 \\ -2 \\ -4 \\ -10 \\ -14 \\ -16 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -10 \\ -14 \\ -6 \\ -8 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -8 \\ -2 \\ -4 \\ -6 \\ -8 \\ -8 \\ -8 \\ -8 \\ -8 \\ -8 \\ -8$	2.545 349 379 101 227 176 56 56 425 174 235 339 130 7 k= 85 80 133 118 156 324 115 55 55 	310 295 288 80 167 134 50 331 124 150 239 117 81 97 124 209 94 209 94 53 97 124 53 50 231 74 55 136 124 130 231 124	$\begin{array}{c} -8\\ -9\\ -9\\ -10\\ -11\\ -13\\ -14\\ -15\\ -16\\ -17\\ -18\\ -19\\ -20\\ -21\\ -22\\ -27\\ -22\\ -27\\ -27\\ -2\\ -3\\ -4\\ -5\\ -6\\ -7\\ -8\\ -9\\ -10\\ -11\\ -112\\ -13\\ \end{array}$	243 66 278 342 41 139 172 128 45 56 122 78 120 	313 212 279 398 45 134 129 54 70 144 1180 60 152 279 337 161 142 190 207 100 213 212 188 466	$\begin{array}{c} -3 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -16 \\ -17 \\ -18 \\ -19 \\ -23 \\ -25 \\ -26 \\ -1 \\ -3 \\ -4 \\ -5 \\ -7 \\ -9 \\ -111 \\ -12 \\ -18 \\ -21 \\ -18 \\ -21 \\ \end{array}$	204 204 104 254 144 237 59 153 50 121 48 332 150 179 91 52 159 122 58 73 242 104 140 208 78 87 181 93 204 87 57	136 96 247 134 249 68 152 42 127 29 358 157 212 73 113 169 82 81 109 82 81 109 124 224 55 97 178 81 238 84 115 115	$\begin{array}{c} -3 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -11 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -18 \\ -19 \\ -20 \\ -21 \\ -23 \\ -24 \\ -25 \\ -27 \\ -28 \\ -30 \\ -1 \\ -2 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ \end{array}$	352 165 141 29 74 200 34 139 129 42 131 173 53 81 173 53 81 173 53 81 173 53 81 109 42 42 131 173 53 81 109 42 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 139 42 131 173 53 81 42 42 42 42 42 42 42 42 42 42	276 163 99 211 67 209 16 156 119 59 123 180 51 19 123 180 51 19 55 98 20 55 98 20 55 98 20 139 55 98 20 163 199 119 59 123 180 55 98 20 180 55 98 20 180 55 98 20 180 55 98 20 180 55 98 20 180 55 98 20 180 55 98 20 180 55 98 20 180 55 98 20 19 19 19 19 19 59 123 180 55 98 20 19 19 55 98 20 19 19 55 98 20 10 10 10 55 98 20 10 10 10 55 98 20 10 10 55 98 20 10 10 55 98 20 10 10 55 98 20 10 10 55 98 20 10 10 10 55 98 10 10 55 10 10 10 55 98 10 10 55 10 10 55 10 10 55 10 10 10 55 10 10 10 55 10 10 10 10 10 10 10 10 10 10	$\begin{array}{c} -22 \\ -23 \\ -24 \\ -28 \\ h^2 \\ -28 \\ h^2 \\ -1 \\ -22 \\ -3 \\ -4 \\ -5 \\ -6 \\ -7 \\ -8 \\ -9 \\ -10 \\ -12 \\ -13 \\ -14 \\ -15 \\ -16 \\ -17 \\ -18 \\ -19 \\ -20 \\ -22 \\ -26 \\ -27 \\ h^2 \\ -27 \\ h^2 \\ -22 \\ -26 \\ -27 \\ h^2 \\ -22 \\ h^2 \\ -27 \\ h^2 \\ -22 \\ h^2 \\ -27 \\ -27 \\ h^2 \\ -27 \\ -27 \\ h^2 \\ -27 \\ -$	41 86 75 151 132 =4 k= 165 205 178 385 113 303 48 69 47 242 255 82 196 92 124 90 106 95 118 144 66 35 15 14 25 15 178 138 113 113 113 113 113 113 11	 43 121 124 168 165 12 130 191 168 390 99 313 40 73 203 114 175 82 59 22 31 645 	$\begin{array}{c} -24\\ & & & \\ & -1\\ & -2\\ & -3\\ & -4\\ & -5\\ & -6\\ & -8\\ & -9\\ & -12\\ & -13\\ & -14\\ & -16\\ & -18\\ & -11\\ & -18\\ & -11\\ & -18\\ & -14\\ & -6\\ & -8\\ & -10\\ & -12\\ & -4\\ & -6\\ & -8\\ & -10\\ & -12\\ & -14\\ & -15\\ & -2\\ & -3\\ & -7\\ & -8\\ & -7\\ & -8\\ \end{array}$	05 =7 k= 109 238 90 42 67 222 210 48 200 74 136 104 136 104 136 67 201 67 201 61 136 240 24 159 40 -9 -9 4- 128 34	30 22 106 224 67 31 56 201 203 197 60 153 197 60 153 122 153 2 121 79 181 83 139 40 164 51 22 146 31 40 41 120 46

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Sulfaguanidine Monohydrate 의 結晶 및 分子構造

			· · · · · · · · · · · · · · · · · · ·			
A-1 A-3		-18 56 73	-20 69 66	-22 76 100	-21 24 26	h=6 k=4
1 074 076	91 128 140	-19 68 65	-21 23 67	23 66 73	-22 96 134	-1 52 42
-1 254 256	-21 120 140	-90 40 51	h=7 k=3	-24 114 131	-23 25 44	2 38 51
-2 196 208	-23 27 24	20 40 01	_1 53 28	25 23 38	-24 70 102	-3 72 69
-3 122 149		-21 95 119	0 196 145	-26 55 67		-4 84 96
-4 154 126	-25 96 130	-22 45 43		-20 33 0/	h=4 k=4	-6 79 78
-5 140 179	-27 38 57	-23 24 25	-3 188 165	n=2 c=4	1 81 82	0 70 90
-6 137 167	h=3 k=3	-24 37 42	-5 131 117	-1 93 79	-2 39 27	
-7 312 331	-1 191 155	-25 56 98	-6 61 58	-2 22 56	-3 26 41	
-8 73 87	-2 109 87	h=5 k=3	7 162 194	3 107 107	_4 101 199	-11 30 51
9 256 222	-3 334 313	-1 221 195	8 42 31	-4 213 238	_F 01 79	-13 26 54
-11 104 192	-4 167 113	-2 73 62	9 51 54	-5 72 80		14 66 88
	-5 189 203	-3 489 498	-10 91 102	-6 178 186	-6 145 157	-16 64 91
	7 190 169	-4 68 62	-11 135 151	7 91 89	-7 35 62	h = 7 k = 4
-13 212 196	-1 100 100	_5 71 89	-13 149 162		-8 129 132	-2 72 81
<i>⊷</i> 14 54 60	-8 154 132		_14 01 92	-9 64 80	-9 31 13	-3 157 147
-15 90 107	-9 135 123		-14 21 22	-10 919 941	-10 183 184	-4 74 67
-16 143 145	-10 71 79	7 216 223	13 40 51			-4 /4 00
-17 72 70	-13 88 83	-8 53 76	-16 21 28	-11 27 37	-13 23 41	
18 69 79	-14 136 140	9 244 249	-17 80 131		-14 131 153	-5 58 63
19 166 168	-15 147 172		h=8 k=3	-13 52 47	-15 37 41	-8 74 94
-20 32 43	-16 95 125	-12 39 43	-1 51 66	-14 144 165	16 141 167	9 47 52
-21 143 163		-13 141 151	-2 70 89	-15 43 59	17 119 122	-10 28 32
-22 39 52	-18 99 115	-15 93 96	-3 146 144	-16 147 169		-11 43 53
-23 110 150	-19 179 200	-16 31 28	-4 82 69	-17 39 53		-12 19 48
-25 64 71	-20 68 81	-17 25 31	-5 123 130		-20 120 107	$k = 1 \ k = 5$
-00 60 04	-21 174 195	-18 32 35		-19 26 42	21 40 07	-2 101 105
-20 69 94		-19 98 134	9 132 140	-20 165 200	-22 10 50	-3 184 210
-2/ 13 50	-93 134 173	-22 40 48	-11 22 50	-21 71 76	k=5 k=4	-4 72 119
$\mu = 2 \kappa = 3$			h=1 k=4	-22 105 119	—I 38 11	-+ 13 112
-1 300 303	06 09 179	-1 182 169	-1 123 137	-23 63 76	-2 157 141	-5 116 139
-2 77 94	-20 50 120	-2 50 71	2 238 252	-24 57 67	-3 162 163	-/ 149 193
3 207 157	n=4 k-3	0 70 101		-25 34 49	-4 38 27	-8 89 97
-4 14 7	1 141 124	- 4 190 112	_1 249 286	<i>b</i> =3 <i>k</i> =4	-5 157 161	-12 60 42
-5 207 188	-2 241 245		-4 249 200	-1 44 65	-6 134 114	-13 71 121
6 131 130	-3 115 138	5 -5 214 214		_9 101 911	-7 55 78	-15 60 75
-7 34 34	-4 44 53	36 35 30		-2 151 241	-8 219 250	—17 79 119
-8 123 119	5 206 182	2 7 95 105	-7 27 37	-3 70 60		-18 74 95
-9 159 172	-6 131 133	78 43 38	-8 224 285	-0 190 191	-10 65 99	-19 53 83
-10 39 32	-7 32 40	69 136 143	-9 99 90	-8 285 308	11 110 195	-20 46 20
-11 24 31	-8 62 58	8	-10 32 7	9 33 35	-11 110 123	k=1 k=6
-12 35 34	9 286 32	3 -12 55 53	-11 100 91	-10 144 157	-12 86 108	
-13 146 199	-10 52 6	4 -13 38 50	-12 171 161	-12 191 212	-13 85 93	-4 /9 104
- 10 140 100	-11 213 22	8 -14 37 29	-14 82 93	-14 137 163	-14 97 100	5 82 124
-14 107 144	-12 90 8	9 15 89 104		-15 31 42		-6 68 115
-15 201 219	_14 118 12	2 -16 52 64	-17 56 49		-17 56 67	8 109 134
-16 230 222	10 10 10	0 -17 59 45	-18 138 167	-18 132 170	-18 77 102	-12 51 64
-17 83 92	-15 162 20	-18 97 41			19 37 61	14 46 76-
-18 96 126	-16 63 3	0 _10 _01 11		-20 56 89)	-15 43 52
—19 84 8 ₉	-17 177 17	813 20 11	-21 00 10			l
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Description and Discussion of the Structure

Bond lengths and angles. The intramolecular bond lengths and angles are given in *Table* 3 and *Fig.* 1.

The benzene ring is slightly distorted from regular hexagon. Ring C-C bond lengths vary from 1.35 to 1.41 Å with the average 1.39 Å which is in agreement with the C-C bond length in crystalline benzene of 1.392 \pm 0.010 Å (Cox, Cruickshank & Smith, 1958)¹². It has been pointed out by many authors, namely, Klug (1970)¹³, Arona & Sundaralingam (1917) ¹⁴, Kruger & Gafner (1971)⁴ etc., that there is a tendency of stabilization of p-substituted benzene ring with the distortion from regular hexagonal form. It was found that the internal valence angles (range $117\sim122^{\circ}$) at the substituted benzene ring show significant deviations from the ideal hexagonal value of 120° .

The C(4)-N(1) bond length is 1.41 Å which

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Table 3. Bond lengths and angles in sulfaguanidine monohydrate.

 Con Hatted	arandaru	ucviations	Briett .	111	parentneses	refer	to	tпę	last	decimal	positions	ot	respective	value	:8

i	j	$D_{ij}(\text{\AA})$	i	j	k	∠ <i>ii</i> ≰(°)
S	C (1)	1.78(2)	O(1)	s	O(2)	114.3(7)
S	O (1)	1.43(1)	O(1)	S	C (1)	106.8(7)
S	O (2)	1.42(1)	O(1)	S	N (2)	115. 2(7)
S	N (2)	1.62(1)	O (2)	S	C(1)	107. 4(8)
C (1)	C (2)	1.36(3)	O (2)	S	N (2)	105.8(7)
C (2)	C (3)	1.40(3)	j C (1)	S	N (2)	107.0(7)
C (3)	C (4)	1.41(3)	C (2)	C (1)	S	120(1)
C (4)	C (5)	1.35(3)	C (6)	C (I)	s	119(1)
C (5)	C (6)	1.39(3)	C (1)	C (2)	C (3)	119(2)
-C (6)	C (1)	1.38(3)	C (2)	C (3)	C (4)	121(2)
C (4)	N (1)	1.41(3)	C (3)	C (4)	C (5)	117(2)
N (2)	C (7)	1.32(2)	C (4)	C (5)	C (6)	122(2)
C (7)	N (3)	1.32(2)	C (5)	C (6)	C (1)	119(2)
C (7)	N (4)	1.37(2)	C (6)	C (I)	C (2)	121(2)
			N(1)	C (4)	C (3)	120(2)
			N(I)	C (4)	C (5)	122(2)
			S	N (2)	C (7)	122(I)
			N (2)	C (7)	N (3)	125(2)
			N (2)	C (7)	N(4)	116(1)
			N (3)	C (7)	N(4)	118(2)



Fig. 1. Bond lengths (Å) and angles(°) in sulfaguanidine monohydrate

is shorter than the C-N single bond of 1.47 Å

(Pauling, 1950)¹⁵.

Bonding around the sulfur atom is distorted from the ideal tetrahedral. The maximum and minimum values for O-S-N are 115° and 106° respectively as in sulfathiazole II4. Distorfrom the tetrahedral symmetry is the tion general property of the derivatives of sulfanilamide. Reference to Table 4 shows a comparison of bond lengths and angles around the tetrahedral sulfur atom obtained from the compounds containing sulfonyl group with their average values. The S-C(1) bond length 1.78 Å is longer than the theoretical $S-C(sp^2)$ value 1.75 Å calculated from the atomic radii and electronegativities given by Truter (1962)¹⁹. The S–O(1) and S–N(2) bond lengths are in good agreement with the results of the other related compounds. The S-O(2) bond length 1.42 Å is insignificantly shorter than the S-O

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Table 4. A comparison of bond lengths and angles around sulfur atom in sulfaguanidine monohydrate and related compounds.

	<u> </u>	Bond ler	ngth (Å)		Pange of angle(°)
Compound	S	s-0		S-C(1)	Kange of angle()
Sulfaguanidine monohydrate α-Sulfanilamide ¹ β-Sulfanilamide ² γ-Sulfanilamide ³ Sulfanilamide monohydrate ¹⁴ Sulfathiazole II ⁴ Sulfathiazole I & III ⁵ Methanesulfonanilide ¹⁷ 2'-Hydroxymethanesulfonanilide ¹³ C. C. Dichard N. a. tolylaulfonyl sulfilimine ¹⁸	1. 42(1) $1. 41$ $1. 448(2)$ $1. 448(2)$ $1. 435(2)$ $1. 435(2)$ $1. 434(10)$ $1. 425(2)$ $1. 435(6)$ $1. 430(8)$	$\begin{array}{c} \mathbf{1.43(1)}\\ 1.47\\ \mathbf{1.454(2)}\\ \mathbf{1.45(1)}\\ \mathbf{1.463(9)}\\ \mathbf{1.444(2)}\\ \mathbf{1.444(2)}\\ \mathbf{1.440(6)}\\ \mathbf{1.443(2)}\\ \mathbf{1.447(6)}\\ \mathbf{1.435(8)} \end{array}$	1.62(1) 1.61 1.620(2) 1.67(2) 1.620(11) 1.539(3) 1.606(9) 1.633(2) 1.617(6) 1.598(8)	1.78(2) 1.74 1.750(2) 1.74(1) 1.748(12) 1.759(3) 1.754(9) 1.746(2) 1.736(6) 1.756(8)	$105.8 \sim 115.2$ $105.7 \sim 119.0$ $105.5 \sim 118.2$ $106.4 \sim 117.3$ $106.0 \sim 118.3$ $104.7 \sim 116.6$ $104.8 \sim 117.1$ $105.3 \sim 118.6$ $106.5 \sim 119.2$ $104.8 \sim 118.1$
Average	1.4	44	1.62	1.75	109. 5

average value 1.44 Å (see Table 4).

In the guanidyl residue the C--N bond lengths range from 1.32 Å in C(7)--N(2) to 1.37 Å in C(7)--N(4), angles from 116° in N(2)--C(7) --N(4) to 125° in N(2)--C(7)--N(3). There is a considerable deviation from the dimensions of free guanidium ion, $(C(NH_2)_3)^+$ (Haas, Harris & Mills, 1965)²⁰ of the average values 1.323 Å and 120°. This fact indicates that there is a flexibility in the geometry of guanidyl residue, subjecting to the crystal lattice environment.

By Wheatley's order/length curve for C--N bonds²¹, the C--N bond length of 1.319 Å corresponds to 50 % double bond character. Thus the bond lengths and the good planarity of guanidyl residue (see *Table* 5) suggest that the guanidyl residue has a delocalized π -electron system.

Planarity and molecular conformation The least-squares planes are listed in *Table* 5. The benzene ring is planar within the errors of the structure determination. The sulfur atom and the nitrogen atom N(1) are displaced 0.07 and 0.05 Å respectively from the ring plane. The

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Fig. 2. Newman projection down the C(1)-S, S-N
(2) and N(2)-C(7) bonds in sulfaguanidine monohydrate showing the conformation angles in degrees.

small distortion may result from the crystal packing forces. The guanidyl residue is planar within the experimental error and the sulfur atom is displaced -0.18 Å from the guanidyl plane which makes an angle of 96° with the benzene ring plane.

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Atoms in plane	Atoms out of plane	Distance in Å from best plane	Constant
A. Benzene ring			····
C (1)		0. 01	A=0.857
C (2)		0.00	B = 0.511
C (3)		-0.01	C = -0.073
C (4)		0.03	D=2.072
C (5)		-0.02	
C (6)		0.00	
	S	0.07	
	N (1)	0.05	
	O (1)	0.19	
B. Guanidyl residue			
N (2)		0. 01	A=0, 182
N (3)		-0.01	B=0.018
N (4)		-0.01	C=0.983
C (7)		0. 03	D=10.383
	S	-0.18	
	O (1)	0.60	
	O (2)	0.07	

Table 5. Least-squares planes in sulfaguanidine monohydrate. Equation for plane: Ax+By+Cz=D, where x, y, z are in Å



Fig. 3. Projection of the crystal structure of sulfaguanidine monohydrate along the b axis. Dashed lines are hydrogen bonds; arrows indicate donor direction

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Fig. 4. Projection of the crystal structure of sulfaguanidine monohydrate along the *c* axis. Dashed lines are hydrogen bonds;)arrows indicate donor direction. Dotted lines are probable hydrogen bonds.

The conformation angles about the C(1)-S, S-N(2) and N(2)-C(7) bonds are given in Fig. 2. The molecule does not take a symmetrical form. The benzene ring makes a torsional angle of 62° rather than 90° to the S-N(2) bond while in the case of sulfanilamide monohydrate¹⁶ it is 88°.

Hydrogen bonding and crystal packing. The structure of sulfaguanidine monohydrate projected along the b and c axis is shown in Fig. 3 and Fig. 4. The molecules related by the symmetry centers are joined indirectly by two N—H(guanidyl)…O—H(water)…O(sulfonyl) hydrogen bonds. As shown in Fig. 5, the water molecule plays an important role in the hydrogen-bonding scheme by forming a distorted tetrahedral configuration and water oxygen O(3) is involved in two donor O(3)—H ... O hydrogen bonds to sulfonyl oxygen O(1) and O(2j) and two acceptor N—H...O(3) hydrogen bonds from N(4f) and N(4g). The close inter- and intramolecular approaches less than 3.5 Å are listed in *Table* 6. Though the positions of the hydrogen atoms can not be located under the present accuracy it may be possible to suppose the hydrogen location and then consider the possi-

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i	j	$D_{ij}(\text{\AA})$	i	Ĵ	k	$\angle_{ijk}(\circ)$
O (3)	O (2j)	2.80(2)*	0(1)	0 (3)	N(4f)	115(1)
O (3)	0(1)	2.83(2)*	O (1)	O (3)	N (4g)	87(1)
O (3)	N(4f)	2.91(2)*	O (1)	O (3)	O(2i)	108(1)
O (3)	N (4 g)	3.00(2)*	O(2j)	O (3)	N(4f)	115(1)
N (3)	O (1)	2.90**	O(2j)	O (3)	N(4g)	92(1)
N (3)	O (2a)	2.97**	N(4f)	O (3)	N(4g)	134(1)
N (3)	N (2a)	3.13**				
				Symme	etry code :	
0(1)	O (2a)	3. 18		x	۰ ۷	2
N (4)	O(1c)	3. 18	a	x	-1+v	2
O (3)	N(3f)	3. 20	Ь	x	1+v	Σ
O (3)	N (1 <i>h</i>)	3. 21	c	-1+x	V V	z
0(1)	O (3i)	3.40	d	1+ <i>x</i>	y y	2
O (2)	C (2b)	3.49	e	1+x	1+v	ź
			f	-x	y	1-2
			g	-x	1-y	1 <i>z</i>
			h	x	1/2 - y	1/2+z
			i	1-x	-y	1 - z
			j	1-x	1y	1-z
			k	1-x	2-y	1-2

Table 6. Intermolecular distances and angles in sulfaguanidine monohydrate

*hydrogen bonds, **probable hydrogen bonds



Fig. 5. Environment around water oxygen atom O(3) in sulfaguanidine monohydrate

bilities of forming the hydrogen bonds. Analogy in this way may lead to the conclusion that N(3) can be involved in the three hydrogen bonds, one acceptor N(3)...H.-N(2a) (intermolecular), two donor N(3)...H...O(1) (intramolecular) and N(3)...H...O(2a) (intermolecular).. These are shown as the dotted lines in Fig. 4. But as there is only one hydrogen atom available to N(3), the latter two donor hydrogen bonds may exist as bifurcated hydrogen bond O(1)

in the form of N(3)-H $\begin{pmatrix} O(1)\\ (2a) \end{pmatrix}$

These hydrogen-bonding networks form infinite molecular layers parallel to (001) plane and the interlayer force appears to be van der Waals in character.

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