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CRYSTAL STRUCTURE OF SODIUM GOLD(I) THIOSULPHATE DIHYDRATE, $Na_3Au(S_2O_3)_2 \cdot 2H_2O$

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Marjorie O. Faltens, and David H. Templeton

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Contribution from the Lawrence Berkeley Laboratory and Department of Chemistry, University of California, Berkeley, California 94720

Crystal Structure of Sodium Gold(I) Thiosulphate Dihydrate, $Na_3Au(S_2O_3)_2 \cdot 2H_2O$[1]

HELENA RUBEN, ALLAN ZALKIN, MARJORIE O. FALTENS, AND DAVID H. TEMPLETON

The crystal and molecular structure of $Na_3Au(S_2O_3)_2 \cdot 2H_2O$ has been determined from a single crystal X-ray diffraction study. The monoclinic space group is $P2_1/a$ with a = 18.206(2), b = 11.355(6), c = 5.436(4) Å, β = 97.87(5)°, Z = 4, d_x = 3.14 g cm^{-3}. The structure was refined by full-matrix least-squares to a conventional R factor of 0.055 using 1811 non-zero intensities. The gold atom is bonded to two sulfur atoms from two thiosulfate groups in a nearly linear arrangement (S-Au-S, 176.5°). The average Au-S, S-S, S-O bond distances are 2.28(1), 2.06(1), and 1.46(1) Å. The three sodium atoms are coordinated to irregular polyhedra of oxygen neighbors; two of the sodium atoms have six such neighbors and one has five. Hydrogen bond lengths are O-H...S = 3.24(1) and 3.33(1) Å, O-H...O = 2.75(1) Å.

Introduction

An extensive investigation of the Mössbauer spectra of gold compounds was undertaken by Faltens and Shirley[2] to seek systematic relationships of these spectra to chemical structure. It was learned that both the isomer shift and the magnitude of the quadrupole splitting varied over wide ranges for various compounds, and neither effect alone could be used even to distinguish between gold(I) and gold(III). However, systematic relations were discovered which permitted unique discrimination between linear sp complexes of Au(I) and square-planar dsp^2 complexes of Au(III) when both effects were considered, and a partial understanding was gained of the origin of the variation of the spectra. The present study of the structure of $Na_3Au(S_2O_3)_2 \cdot 2H_2O$ was done partly to verify its structure for the Mössbauer investigation and partly as a contribution to the crystal chemistry of salt hydrates.

Gold sodium thiosulfate, known clinically as Aurocidin, Crisalbine, or Sanocrysin,[3] has been used in some medical circles for the treatment of arthritis, tuberculosis, and leprosy.

Experimental

Sodium gold(I) thiosulfate was made from $HAuCl_4$ and $Na_2S_2O_3$ and recrystallized from water-alcohol solution as described by Brown.[4] A small crystal, approximately 0.17 x 0.06 x 0.02 mm was glued to a thin pyrex glass fiber with the long dimension of the crystal (the crystallographic *c* axis) parallel to the fiber axis.

The crystals were aligned on a G. E. XRD-5 manual diffractometer equipped with a quarter-circle Eulerian cradle, a scintillation counter and a pulse height discriminator. X-rays were produced by a molybdenum anode tube ($\lambda_{K\alpha_1}$ = 0.70926, $\lambda_{K\alpha_2}$ = 0.71354 and $\lambda_{K\beta}$ = 0.63225 Å) operated at 45 kv and 20 ma; a 3.0 mil Zr filter was located at the receiving collimator. The tube take-off angle was 2° for the cell dimension measurements and 4° for the intensity measurements.

Cell dimensions were determined from a series of measurements along the *h*00, 0*k*0, and 00*l* directions using peaks where the K_{α_1}, K_{α_2} and K_β were resolved. The β angle was measured directly from the angle between the *h*00 and 00*l* rows on the chi circle. All measurements were made at room temperature (≈23°). A total of 1966 independent reflections, all that are available in a quandrant of reciprocal space where $2\theta \leq 50°$, were counted for ten seconds each using the stationary-crystal stationary-counter technique. Background was plotted as a function of 2θ and applied routinely to the reflections with the exception of a minority for which background was measured individually because of streaking from lower orders. The most intense reflection (511) was 2303 counts per second after subtraction of the background.

The absorption parameter μ was calculated to be 145 cm^{-1}. An absorption correction was made with a program[5] that approximates the crystal with an array of grid points; it resulted in substantial improvement of the final agreement. The minimum and maximum corrections applied to the observed intensities were 1.35 and 2.62. An extinction correction was not indicated and not made. The usual Lorentz-polarization corrections were made.

The calculation of the estimated standard deviations of the intensities and the weighting scheme used in the least-squares are described elsewhere[6]. A value of p = 0.07 was found necessary to reduce the weights of the intense reflections so that their weighted residuals were comparable to those of lesser intensities. Our unpublished full-matrix least-squares program minimizes the function R_2^2 where $R_2^2 = \Sigma w(\Delta F)^2/\Sigma w F_0^2$. F_0 and F_c are observed and calculated structure factors, ΔF is the difference of their magnitudes, and w is the weighting factor. The program accommodates both the real and imaginary parts of the dispersion correction. Scattering factors for neutral Au, neutral S, neutral O and Na^+ were used[7] and modified for dispersion by adding -2.36, 0.1, 0.0 and 0.0 electrons[8] respectively. The imaginary dispersion terms are 8.89, 0.16, 0.0 and 0.04.

Results

Unit Cell and Space Group. The space group is $P2_1/a(C_{2h}^5)$ with cell dimensions at 23° of a = 18.206(6), b = 11.355(6), c = 5.436(4) Å and β = 97.87(5)°; the errors are subjective estimates.[9] With four formula units per unit cell and a cell volume of 1113 Å^3, the calculated density is 3.14 g cm^{-3}; a literature value[10] is 3.09 g cm^{-3}. The general position in $P2_1/a$ is $\pm(x,y,z;\ \frac{1}{2}+x,\frac{1}{2}-y,z)$.

Determination of the Structure. The gold atom location was deduced from a three-dimensional Patterson function. A Fourier synthesis phased by the gold atom revealed the locations of the sodium and sulfur atoms. A least-squares refinement of this partial structure using isotropic temperature factors resulted in $R = \Sigma|\Delta F|/\Sigma|F_0| = 0.21$. With anisotropic thermal parameters on the gold atom the R factor went to 0.19. A difference Fourier revealed the oxygen atom locations. Further refinement with all atoms anisotropic brought R to 0.078. After making the absorption correction the final agreement was R = 0.055 for 1811 non-zero intensities. R_2 was 0.063, and the standard deviation of an observation of unit weight was 1.19.[11]

Attempts to locate hydrogen atoms were unsuccessful. A difference Fourier showed 28 peaks larger than 1 Å^{-3}. The largest peak was 2 Å^{-3} and was near the gold atom as were several other large peaks. None of the top peaks appeared near reasonable locations for hydrogen atoms, and no attempt was made to include hydrogen in the calculations.

The final positional parameters with their standard deviations are listed in Table I. The anisotropic thermal parameters are listed in Table II.

Description of the Structure. The $Au(S_2O_3)_2^{3-}$ anion is shown in a schematic drawing in Fig. 1, and the crystal structure is shown in Fig. 2 and 3. Distances and angles are listed in Tables III and IV. The S-Au-S angle of 176.5(2)° is nearly linear, and there is no significant difference in the two Au-S bond lengths. The two S-S-Au angles are also essentially equal, and the dihedral angle of their planes is 67°. The two S-S distances are different from each other by little more than three standard deviations, but this difference is not considered to be significant. The average S-O bond distance of the six such bonds in this structure is 1.458 Å, and no individual value is more than its estimated standard deviation from this average. These dimensions of the thiosulfate group are in excellent agreement with those found by Baggio[12] for $[Co(NH_3)_5S_2O_3]Cl\cdot H_2O$, in which thiosulfate is complexed to cobalt through a sulfur atom as it is to gold in this crystal. As pointed out by Baggio,[12] the S-S bond is shorter in thiosulfates in which such complexing is absent, for example salts of Mg[13,14] and Na.[15,16]

Although the hydrogen atom positions were not determined directly from the diffraction data, one can identify three hydrogen bonds in the structure. The water molecule identified as O(7) has sulfur atom neighbors at 3.24(1) and 3.33(1) Å. These distances are within the range of O-H...S hydrogen bond distances which have been verified by nmr[17,18] and neutron diffraction[16] studies for sodium thiosulfate pentahydrate and magnesium thiosulfate hexahydrate, and the angles to these two sulfur neighbors and the two sodium neighbors are

roughly tetrahedral. On this basis we assign the two hydrogen atoms of O(7) to O-H...S bonds. Water molecule O(8) has a neighbor O(5) at 2.75(1) Å, a normal distance for an O-H...O hydrogen bond, and again the angles defined by this oxygen neighbor and the two sodium neighbors are reasonable for hydrogen bonding. The position of the other hydrogen on O(8) must complete a more or less tetrahedral coordination for this oxygen atom, but no neighbor is available close enough in a suitable direction for a hydrogen bond. Thus we conclude that O(8) makes only one hydrogen bond.

The next nearest neighbor to the gold atom, besides S(3) and S(4), is another gold atom at 3.302(1) Å. The nearest neighbors of the sodium atoms are all oxygen atoms. Na(1) is coordinated by six oxygen atoms at distances of 2.36 to 2.63 Å; the two nearest neighbors are two oxygen atoms of water molecules. Na(2) is also coordinated by six oxygen atoms at distances from 2.35 to 2.54 Å, the nearest of which comes from a water molecule. Na(3) is coordinated to five oxygen atoms ranging in distance from 2.34 to 2.53 Å, the closest of which is from a water molecule. The stereogram in Fig 3 shows some of the coordination about the sodium ions.

Table I. Positional Parameters[a]

	x	y	z
Au	.07361(3)	.06692(5)	.1479(1)
S(1)	.0006(2)	.3396(3)	.2178(5)
S(2)	.2592(2)	.0358(3)	.1207(5)
S(3)	.1591(2)	-.0366(3)	-.0338(6)
S(4)	-.0063(2)	.1713(3)	.3497(6)
Na(1)	.2176(3)	.3252(5)	.3495(10)
Na(2)	.1254(3)	-.4095(4)	.3159(9)
Na(3)	.4129(3)	-.1027(5)	.2427(10)
O(1)	.0762(5)	.3809(8)	.2892(17)
O(2)	-.0521(5)	.4044(8)	.3419(16)
O(3)	-.0179(5)	.3425(10)	-.0505(15)
O(4)	.3108(5)	-.0169(9)	-.0319(16)
O(5)	.2543(5)	.1633(8)	.0916(16)
O(6)	.2783(5)	.0010(8)	.3803(15)
O(7)[b]	.1652(5)	-.2179(9)	.4284(19)
O(8)[b]	.3387(5)	-.2627(8)	.3212(17)

[a] Standard deviations in this and subsequent tables appear in parentheses.

[b] Water molecule oxygen atom.

Table II. Anisotropic Thermal Parameters[a]

	B_{11}	B_{22}	B_{33}	B_{12}	B_{13}	B_{23}
Au	2.02(2)	1.84(3)	2.58(3)	.18(2)	.31(2)	-.23(2)
S(1)	1.82(12)	1.71(12)	1.41(11)	.02(10)	.15(9)	.03(10)
S(2)	1.99(12)	1.73(12)	1.05(11)	.03(10)	.14(9)	.04(9)
S(3)	2.10(13)	2.15(14)	2.03(13)	.38(11)	.11(10)	-.52(11)
S(4)	2.57(14)	1.88(14)	3.01(15)	.19(11)	.93(11)	.02(12)
Na(1)	4.33(28)	2.62(25)	2.47(22)	1.05(21)	.19(19)	.35(19)
Na(2)	2.70(22)	2.13(22)	1.97(20)	.07(17)	.19(17)	-.09(17)
Na(3)	2.65(23)	2.90(24)	2.57(23)	-.25(19)	.30(18)	-.22(19)
O(1)	2.13(38)	2.48(41)	3.12(43)	-.45(31)	-.17(31)	.44(34)
O(2)	2.53(38)	2.29(39)	2.71(40)	.41(31)	.98(31)	-.10(31)
O(3)	2.95(42)	4.46(52)	1.56(36)	.02(39)	-.64(30)	-.19(35)
O(4)	2.66(40)	3.00(42)	2.03(37)	.67(34)	.31(30)	-.30(33)
O(5)	3.12(41)	1.95(37)	2.15(35)	-.52(32)	-.47(30)	.40(31)
O(6)	3.20(40)	1.92(39)	1.52(34)	.08(31)	-.21(29)	.07(29)
O(7)	2.51(40)	3.25(46)	3.91(46)	-.01(35)	.85(35)	.31(39)
O(8)	4.30(50)	2.09(40)	2.40(39)	-.34(35)	-.05(35)	-.43(32)

[a] The anisotropic temperature factor has the form

$$T = \exp[-0.25(B_{11}a_1^{*2}h^2 + \ldots \quad +2B_{12}a_1^*a_2^*hk + \ldots)]$$

Table III. Interatomic Distances[a]

Atoms	Dist (Å)	Atoms	Dist (Å)	Atoms	Dist (Å)
Au -Au	3.302(1)	S(4) -O(7)[b]	3.326(10)	O(1) -O(2)	2.408(12)
Au -S(3)	2.280(3)	Na(1)-O(1)	2.626(10)	O(1) -O(3)	2.381(12)
Au -S(4)	2.272(3)	Na(1)-O(4)	2.494(11)	O(1) -O(2)	3.224(13)
S(1) -O(1)	1.457(9)	Na(1)-O(5)	2.460(11)	O(1) -O(4)	2.885(12)
S(1) -O(2)	1.458(9)	Na(1)-O(6)	2.473(10)	O(1) -O(6)	3.286(12)
S(1) -O(3)	1.451(9)	Na(1)-O(7)[b]	2.355(11)	O(1) -O(8)[b]	2.940(12)
S(1) -S(4)	2.051(5)	Na(1)-O(8)[b]	2.399(11)	O(2) -O(2)	3.221(19)
S(1) -Na(3)	3.201(6)	Na(2)-O(1)	2.540(10)	O(2) -O(3)	2.408(12)
S(1) -Na(3)	3.203(6)	Na(2)-O(2)	2.433(10)	O(2) -O(4)	3.255(12)
S(2) -O(4)	1.463(9)	Na(2)-O(3)	2.392(10)	O(2) -O(6)	3.303(13)
S(2) -O(5)	1.458(9)	Na(2)-O(3)	2.392(10)	O(2) -O(7)[b]	3.315(13)
S(2) -O(6)	1.461(9)	Na(2)-O(4)	2.390(11)	O(2) -O(8)[b]	2.940(12)
S(2) -S(3)	2.069(5)	Na(2)-O(6)	2.459(10)	O(4) -O(6)	2.403(12)
S(2) -Na(3)	3.199(6)	Na(2)-O(7)[b]	2.348(11)	O(4) -O(5)	2.426(13)
S(3) -O(4)	2.770(10)	Na(3)-O(1)	2.532(11)	O(4) -O(6)	3.176(12)
S(3) -O(5)	2.881(9)	Na(3)-O(1)	2.931(11)	O(5) -O(6)	2.421(12)
S(3) -O(6)	2.936(9)	Na(3)-O(2)	2.381(11)	O(5) -O(7)[b]	3.118(13)
S(3) -O(7)[b]	3.239(11)	Na(3)-O(2)	3.157(11)	O(5) -O(8)[b]	2.751(12)
S(4) -O(1)	2.858(10)	Na(3)-O(3)	2.383(11)	O(6) -O(7)[b]	3.260(13)
S(4) -O(2)	2.774(10)	Na(3)-O(4)	2.423(10)	O(6) -O(8)[b]	3.222(13)
S(4) -O(3)	2.904(10)	Na(3)-O(6)	2.908(11)	O(7)[b]-O(8)[b]	3.328(14)

[a]Distances have not been corrected for thermal motion. The table lists all of the distances less than 3.35 Å.

[b]Water molecule oxygen atom.

Table IV. Selected Angles

S(3) - Au -S(4)	176.5(2)	O(2)-Na(2)-O(3)	88.0(4)
O(1) -S(1)-O(2)	112.0(7)	O(2)-Na(2)-O(4)	150.7(5)
O(1) -S(1)-O(3)	109.9(7)	O(3)-Na(2)-O(6)	170.6(5)
O(2) -S(1)-O(3)	112.3(7)	O(4)-Na(2)-O(6)	81.8(4)
O(4) -S(2)-O(5)	112.2(7)	O(4)-Na(2)-O(7)	118.8(5)
O(4) -S(2)-O(6)	110.5(7)	O(6)-Na(2)-O(7)	92.4(4)
O(5) -S(2)-O(6)	112.1(7)	O(1)-Na(3)-O(2)	82.0(4)
Au -S(3)-S(2)	103.6(1)	O(1)-Na(3)-O(3)	117.7(4)
Au -S(4)-S(1)	104.1(1)	O(1)-Na(3)-O(4)	126.6(5)
O(1)-Na(1)-O(4)	68.5(3)	O(1)-Na(3)-O(8)	74.1(4)
O(1)-Na(1)-O(5)	116.6(4)	O(2)-Na(3)-O(8)	146.8(5)
O(1)-Na(1)-O(6)	80.2(4)	O(3)-Na(3)-O(8)	113.3(5)
O(1)-Na(1)-O(7)	156.6(5)	S(3)- O(7)-S(4)	103.9(2)
O(1)-Na(1)-O(8)	71.5(4)	S(3)- O(7)-Na(1)	101.4(3)
O(4)-Na(1)-O(6)	79.5(4)	S(3)- O(7)-Na(2)	114.2(3)
O(5)-Na(1)-O(7)	80.7(4)	S(4)- O(7)-Na(1)	123.6(3)
O(5)-Na(1)-O(8)	107.0(4)	S(4)- O(7)-Na(2)	89.4(2)
O(7)-Na(1)-O(8)	88.9(4)	Na(1)-O(7)-Na(2)	123.5(4)
O(1)-Na(2)-O(2)	80.8(4)	Na(1)-O(8)-Na(3)	97.4(3)
O(1)-Na(2)-O(3)	90.6(4)	Na(1)-O(8)-O(5)	117.2(4)
O(1)-Na(2)-O(7)	167.7(5)	Na(3)-O(8)-O(5)	113.2(4)

(1) Research performed under the auspices of the U. S. Atomic Energy Commission.

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(11) Supplementary Material Available. A listing of structure factor amplitudes will appear following these pages in the microfilm edition of this volume of the journal. Photocopies of the supplementary material from this paper only or microfiche (105 x

148 mm, 20x reduction, negatives) containing all of the supplementary material for the papers in this issue may be obtained from the Journals Department, American Chemical Society, 1155 16th St., N.W., Washington, D.C. 20036. Remit check or money order for 0.00 for photocopy or $0.00 for microfiche, referring to code number INORG-00000.

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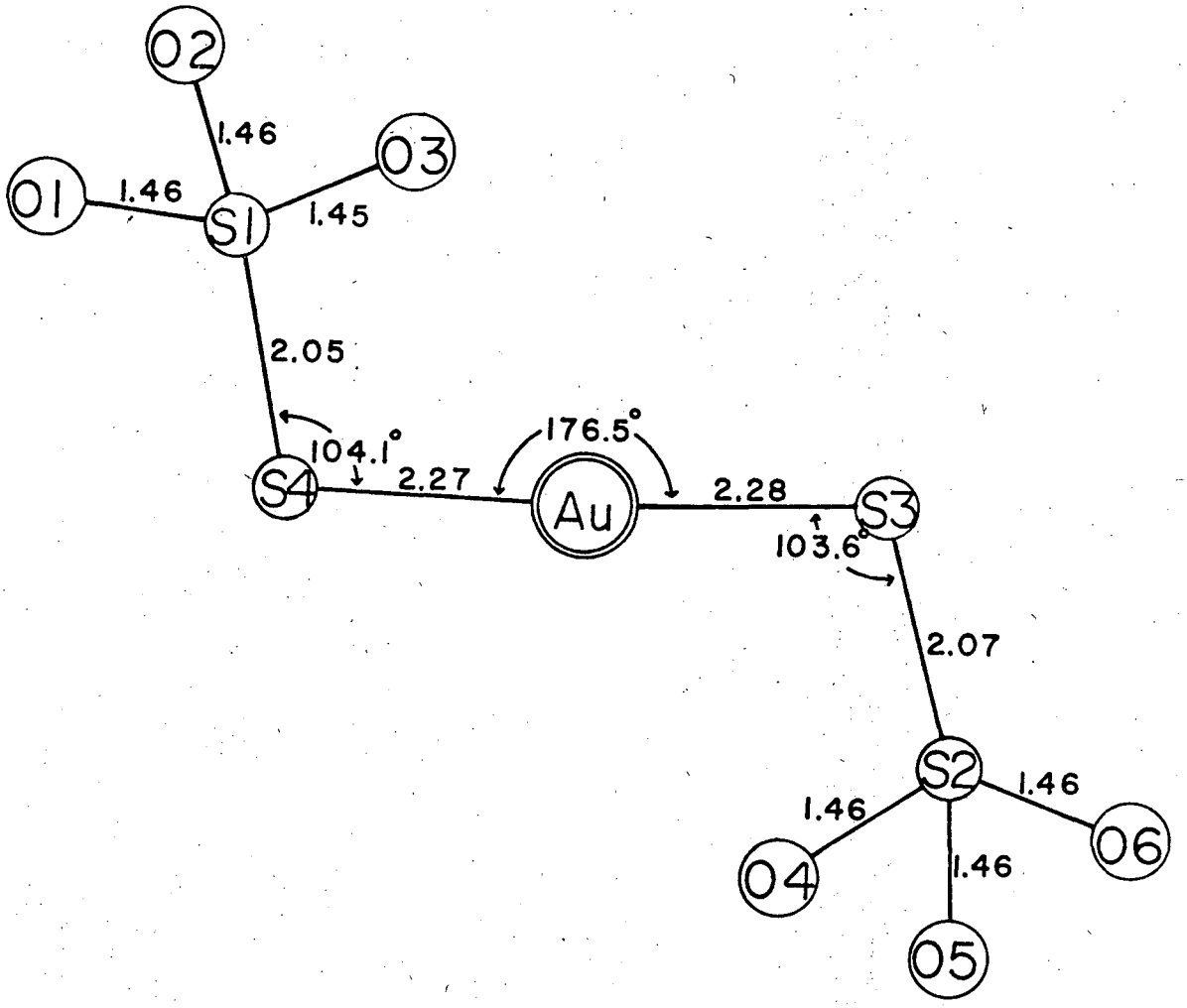
(18) Z.M. El Saffar, *Acta Crystallogr.*, B**25**, 1708 (1969).

Figure Captions

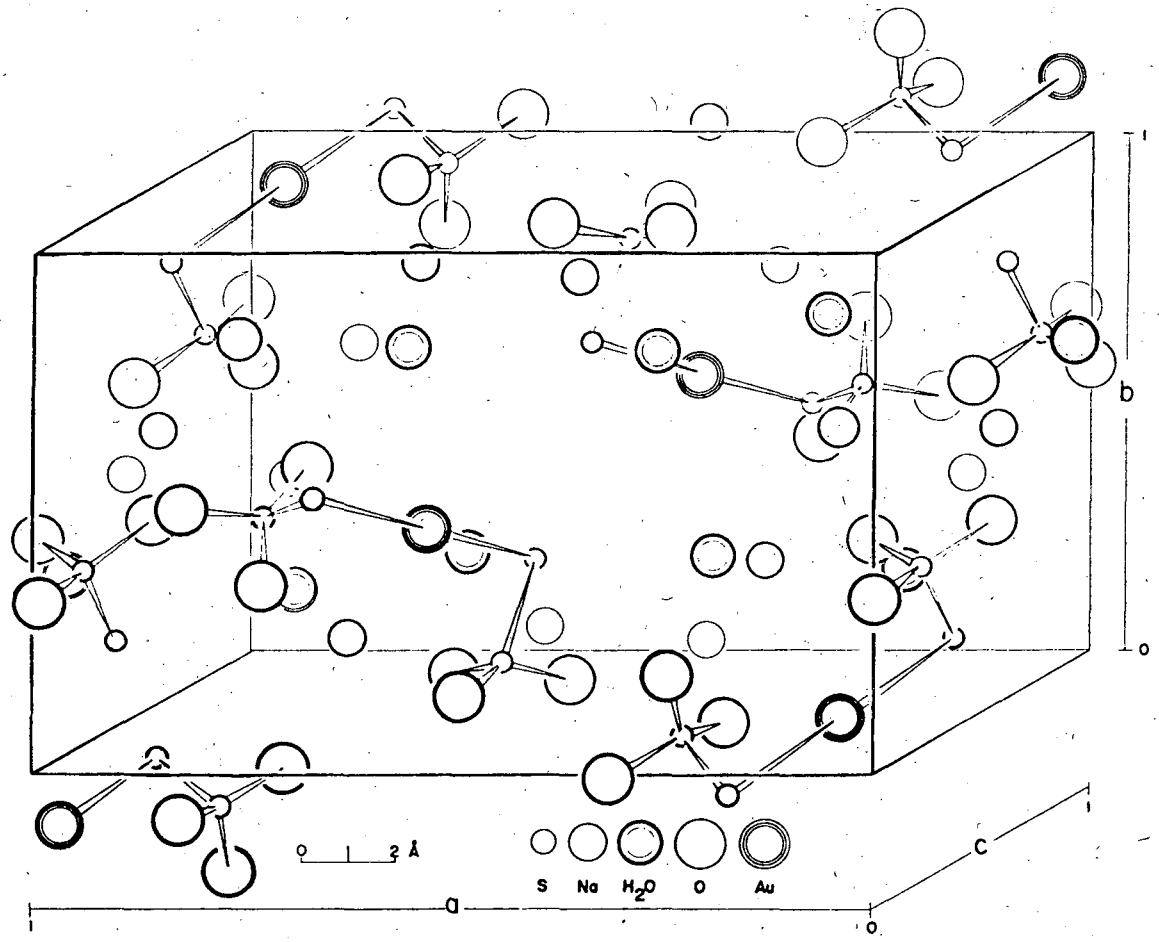
Figure 1. Schematic drawing of the $Au(S_2O_3)_2^{3-}$ anion.

Figure 2. Perspective view of the unit cell contents.

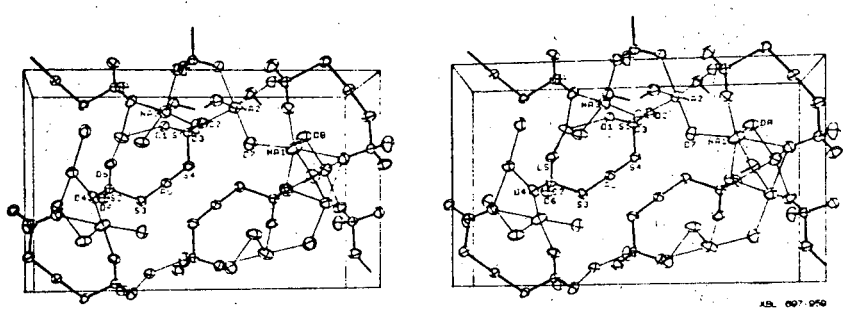
Figure 3. Stereogram showing the coordination to sodium atoms.


O2
1.46
O3
O1
1.46
S1
1.45
2.05
104.1°
176.5°
S4
2.27
Au
2.28
S3
103.6°
2.07
S2
1.46
1.46
O6
O4
1.46
O5

XBL 707-1307



MUB-11229


XBL 807-950

OBSERVED AND CALCULATED STRUCTURE FACTORS OF SODIUM GOLD THIOSULFATE DIHYDRATE
FCA(0,0,0) = 1896

H	FOB	FCA
K,L= 0, 0		
2	354	324
4	113	113
6	365	360
8	225	210
10	18	20
12	524	491
14	414	387
16	177	164
18	25	40
20	149	137
K,L= 1, 0		
1	309	292
2	200	191
3	74	73
4	206	208
5	365	350
6	91	90
6	90	90
7	213	228
8	246	253
9	382	381
10	225	239
11	170	171
12	123	123
13	284	283
14	0	22*
15	176	179
16	114	116
17	59	53
18	36	40
19	74	76
20	0	12*
21	207	192
K,L= 2, 0		
0	450	473
1	170	174
2	15	26
3	339	333
4	176	176
5	358	339
6	492	473
7	88	94
8	299	297
9	100	103
10	130	128
11	241	241
12	136	144
13	90	92
14	131	136
15	22	21
16	57	61
17	196	201
18	107	102
19	74	73
20	125	125
21	49	38
K,L= 3, 0		
1	264	271
2	331	333
3	92	80
4	569	565
5	148	138
6	192	185
7	108	107
8	299	321
9	70	75
10	450	468
11	58	55
12	156	146
13	138	138
14	73	75
15	88	88
16	256	267
17	35	27
18	163	168
19	0	9*
20	0	14*
K,L= 4, 0		
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1	282	283
2	363	363
3	422	419
4	46	40
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6	139	130
7	87	94
8	35	44
9	258	269
10	130	124
11	245	257
12	36	34
13	116	120
14	105	107
15	125	122
16	64	64
17	257	247
18	26	18
19	147	140
20	0	16*
K,L= 5, 0		
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2	226	231
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5	38	22
6	129	143
7	247	285
8	126	129
9	0	14*
10	272	284
11	21	20
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13	118	116
14	23	6
15	79	84
16	229	222
17	26	1
18	174	163
19	95	90
K,L= 6, 0		
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2	218	215
3	219	220
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5	302	318
6	385	398
7	158	153
8	388	370
9	183	176
10	120	124
11	176	188
12	81	87
13	23	21
14	125	131
15	43	56
16	26	27
17	163	166
18	119	120
19	28	39
K,L= 7, 0		
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3	34	52
4	53	56
5	131	135
6	35	41
7	310	322
8	64	65
9	123	141
10	22	11
11	121	134
12	33	22
13	279	291
14	141	137
15	150	159
16	26	32
17	0	10*
18	0	19*
K,L= 8, 0		
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2	203	217
3	138	137
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6	227	235
7	86	84
8	175	158
9	52	49
10	0	11*
11	0	10*
12	185	182
13	72	61
14	206	219
15	76	57
16	95	101
17	0	9*
K,L= 9, 0		
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2	138	141
3	40	40
4	132	136
5	208	216
6	74	79
7	217	236
8	99	104
9	138	138
10	132	142
11	67	76
12	107	112
13	161	147
14	81	97
15	130	131
K,L= 10, 0		
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2	80	77
3	269	296
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5	86	101
6	50	46
7	0	10*
8	26	28
9	177	195
10	53	34
11	173	191
12	133	141
13	39	42
14	90	91
K,L= 11, 0		
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2	203	198
3	93	88
4	160	174
5	27	25
6	71	73
7	27	22
8	81	92
9	47	56
10	215	187
11	28	27
12	146	153
K,L= 12, 0		
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2	118	113
3	183	204
4	40	44
5	114	127
6	0	17*
7	0	8*
8	0	13*
9	164	171
K,L= 13, 0		
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2	114	107
3	0	15*
4	136	148
5	91	115
K,L= 0, 1		
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-8	420	396
-6	221	202
-4	383	358
-2	447	429
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2	263	252
4	410	402
6	350	345
8	24	21
10	196	179
12	430	399
14	163	145
16	156	136
18	275	248
20	134	111
K,L= 1, 1		
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-18	44	40
-17	122	123
-16	41	48
-15	191	208
-14	177	193
-13	148	165
-12	158	176
-11	285	317
-10	171	199
-9	441	472
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-3	251	242
-2	122	117
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1	49	51
2	345	325
3	377	368
4	404	387

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6	141	119
7	145	140
8	64	60
9	72	69
10	55	61
11	328	318
12	109	104
13	205	201
14	153	150
15	23	3
16	152	149
17	202	187
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19	85	80
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K,L=	2,	1
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-15	23	28
-14	77	87
-13	115	124
-12	123	128
-11	229	253
-10	300	302
-9	18	14
-8	214	222
-7	124	133
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-3	109	114
-2	207	210
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2	240	223
3	155	144
4	396	375
5	25	25
6	65	66
7	253	246
8	125	124
9	224	220
10	165	155
11	28	28
12	254	256
13	143	138
14	100	104
15	203	199
16	108	113
17	87	91
18	132	125
19	54	57
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K,L=	3,	1
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-16	49	61
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-14	223	249
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-9	93	87
-8	152	162
-7	56	57
-6	384	392
-5	205	209
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-2	23	19
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1	68	66
2	391	403
3	20	31
4	305	303
5	161	156
6	192	190
7	127	115
8	465	463
9	0	6*
10	298	290
11	106	106
12	149	138
13	152	146
14	150	152
15	33	36
16	221	217
17	72	72
18	52	51
19	27	29
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K,L=	4,	1
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-14	42	48
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-12	39	55
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-10	37	47
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-8	28	35
-7	203	200
-6	19	30
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-4	26	25
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1	373	401
2	44	39
3	207	211
4	63	59
5	17	7
6	30	30
7	404	405
8	18	21
9	332	320
10	28	6
11	72	75
12	71	68
13	174	172
14	0	8*
15	215	215
16	0	19*
17	131	129
18	38	40
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K,L=	5,	1
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-12	154	166
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1	156	145
2	242	236
3	162	165
4	98	100
5	221	218
6	245	249
7	105	118
8	427	417
9	121	118
10	248	255
11	170	166
12	39	37
13	80	75
14	63	64
15	25	22
16	142	149
17	26	32
18	27	17
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K,L=	6,	1
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-16	143	154
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-13	133	146
-12	25	28
-11	169	158
-10	115	106
-9	41	38
-8	242	239
-7	72	66
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-5	296	296
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-3	30	24
-2	327	328
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0	135	129
1	252	243
2	62	64
3	192	187
4	252	248
5	58	66
6	203	217
7	219	211
8	41	37
9	166	175
10	175	179
11	23	16
12	169	184
13	89	88
14	134	133
15	125	122
16	102	103
17	90	93
18	152	156
K,L=	7,	1
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1	143	124
2	20	26
3	223	222
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7	162	161
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13	134	141
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K,L=	8,	1
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-4	144	142
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2	131	144
3	80	83
4	259	264
5	128	135
6	283	281
7	32	15
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11	25	30

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15	77	76
16	88	82
K,L=	9,	1
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-7	144	169
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1	24	16
2	230	240
3	243	231
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5	206	219
6	59	68
7	87	90
8	155	163
9	25	19
10	67	76
11	181	191
12	26	34
13	108	111
14	149	150
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-9	28	33
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-7	188	177
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2	51	54
3	154	160
4	124	132
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6	114	125
7	134	118
8	37	39
9	160	176
10	54	51
11	27	34
12	92	101
13	109	107
K,L=	11,	1
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-5	29	20
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2	315	286
3	38	29
4	108	129
5	27	17
6	72	81
7	55	57
8	135	140
9	48	53
10	69	64
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K,L=	12,	1
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1	178	163
2	29	8
3	126	146
4	50	51
5	41	38
6	41	47
7	136	165
8	0	13*
K,L=	13,	1
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3	53	74
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K,L=	0,	2
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8	141	139
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12	38	34
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K,L=	1,	2
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-12	94	98
-11	310	313
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-8	131	136
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1	230	226
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3	405	402
4	188	181
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6	194	191
7	308	307
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9	238	230
10	20	8
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12	154	147
13	39	40
14	24	26
15	150	141
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K,L=	2,	2
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2	320	309
3	61	62
4	197	201
5	170	167
6	30	33
7	424	411
8	243	231
9	220	213
10	228	221
11	112	113
12	126	124
13	228	217
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15	112	109
16	152	153
17	64	64
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3	198	193
4	251	240
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6	414	404
7	109	110
8	99	92
9	0	16*
10	110	112
11	53	53
12	238	238
13	40	36
14	127	130
15	61	57
16	36	38
17	109	100
18	106	109
K,L=	4,	2
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8	28	23
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13	280	269
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16	26	21
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7	20	12
8	198	202
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11	108	105
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K,L=	6,	2
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13	79	81
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15	27	36
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K,L=	7,	2
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3	351	362
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8	87	92
9	213	226
10	103	111
11	113	128
12	108	114
13	0	21*
14	0	17*
15	157	163
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K,L=	8,	2
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1	124	113
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3	84	75
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5	23	22
6	24	20
7	34	36
8	124	131
9	25	36
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12	70	76
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K,L=	9,	2
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3	145	143
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6	160	159
7	127	134
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9	140	150
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5	185	203
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7	227	209
8	38	35
9	27	25
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K,L=	11,	2
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0	177	172
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2	79	87
3	30	32
4	30	49
5	157	149
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K,L=	0,	3
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