Table S1. The observed and the obs.-calc. frequencies (MHz) of measured rotational transitions in the ground state of the parent isotopomer of benzaldehyde.

Benzaldehyde, ground state of the parent isotopomer

Obs-calc differences are from fit of Watson's reduced Hamiltonian in ---> reduction-A, representation-I.r <---

TRANSITION	Observed	Obs-Calc	Error	Note	wt for blend
!					
! Merged FTMW measurements from Warsaw and Harvard					
!					
! aR_0,1-type lines					
!					
3, 1, 3 <- 2, 1, 2	7752.2719	-0.0002	0.0020		
3, 0, 3 <- 2, 0, 2	8207.0738	-0.0004	0.0020		
3, 2, 2 <- 2, 2, 1	8306.8593	0.0017	0.0020		
3, 2, 1 <- 2, 2, 0	8406.6476	0.0028	0.0020		
3, 1, 2 <- 2, 1, 1	8829.9395	-0.0001	0.0020		
4, 1, 4 <- 3, 1, 3	10309.4761	0.0002	0.0020		
4, 0, 4 <- 3, 0, 3	10831.4034	0.0000	0.0020		
4, 2, 3 <- 3, 2, 2	11056.2202	0.0007	0.0020		
4, 3, 2 <- 3, 3, 1	11122.9490	0.0007	0.0020		
4, 3, 1 <- 3, 3, 0	11129.5434	0.0011	0.0020		
4, 2, 2 <- 3, 2, 1	11300.5835	-0.0001	0.0020		
4, 1, 3 <- 3, 1, 2	11741.2378	-0.0001	0.0020		
5, 1, 5 <- 4, 1, 4	12846.7075	-0.0004	0.0020		
5, 0, 5 <- 4, 0, 4	13375.1072	0.0002	0.0020		
5, 2, 4 <- 4, 2, 3	13788.8803	-0.0007	0.0020		
5, 4, 2 <- 4, 4, 1	13900.5830	0.0022	0.0030		
5, 4, 1 <- 4, 4, 0	13900.9034	0.0027	0.0030		
5, 3, 3 <- 4, 3, 2	13920.1747	0.0000	0.0020		
5, 3, 2 <- 4, 3, 1	13943.0987	0.0007	0.0020		
5, 2, 3 <- 4, 2, 2	14258.2008	-0.0004	0.0020		
5, 1, 4 <- 4, 1, 3	14621.7220	0.0001	0.0020		
!					
6, 1, 6 <- 5, 1, 5	15362.4557	-0.0002	0.0020		
6, 0, 6 <- 5, 0, 5	15843.1646	0.0004	0.0020		
6, 2, 5 <- 5, 2, 4	16500.8965	0.0004	0.0020		
6, 4, 3 <- 5, 4, 2	16698.7032	0.0015	0.0020		
6, 4, 2 <- 5, 4, 1	16700.1348	-0.0013	0.0020		
6, 3, 4 <- 5, 3, 3	16723.1202	0.0008	0.0020		
6, 3, 3 <- 5, 3, 2	16783.4913	0.0016	0.0020		
6, 2, 4 <- 5, 2, 3	17269.9756	-0.0004	0.0020		
6, 1, 5 <- 5, 1, 4	17459.5886	-0.0007	0.0020		
!					
7, 1, 7 <- 6, 1, 6	17856.9156	-0.0002	0.0020		
7, 0, 7 <- 6, 0, 6	18255.4059	0.0005	0.0020		
7, 2, 6 <- 6, 2, 5	19188.6406	-0.0006	0.0020		
7, 5, 3 <- 6, 5, 2	19471.9805	0.0029	0.0030		
7, 5, 2 <- 6, 5, 1	19472.0498	0.0001	0.0030		
7, 4, 4 <- 6, 4, 3	19505.9502	0.0005	0.0020		
7, 4, 3 <- 6, 4, 2	19510.7022	0.0005	0.0020		
7, 3, 5 <- 6, 3, 4	19528.4336	-0.0005	0.0020		
7, 3, 4 <- 6, 3, 3	19661.4268	-0.0010	0.0020		
7, 1, 6 <- 6, 1, 5	20240.9395	-0.0003	0.0020		
7, 2, 5 <- 6, 2, 4	20311.0830	-0.0003	0.0020		
!					

```
   8,  1,  8 <-  7,  1,  7        20331.7354   -0.0001    0.0020
   8,  0,  8 <-  7,  0,  7        20637.3965   -0.0002    0.0020
   8,  2,  7 <-  7,  2,  6        21849.0264    0.0008    0.0020
   8,  5,  4 <-  7,  5,  3        22274.6455   -0.0010    0.0020
   8,  5,  3 <-  7,  5,  2        22274.9346    0.0007    0.0020
   8,  4,  5 <-  7,  4,  4        22322.8838    0.0003    0.0020
   8,  3,  6 <-  7,  3,  5        22331.3311   -0.0005    0.0020
   8,  4,  4 <-  7,  4,  3        22335.8272   -0.0006    0.0020
   8,  3,  5 <-  7,  3,  4        22588.5284    0.0005    0.0020
   8,  1,  7 <-  7,  1,  6        22950.9083   -0.0005    0.0020
   8,  2,  6 <-  7,  2,  5        23350.1915    0.0001    0.0020
!
   9,  1,  9 <-  8,  1,  8        22789.5635    0.0001    0.0020
   9,  0,  9 <-  8,  0,  8        23010.0547    0.0002    0.0020
   9,  2,  8 <-  8,  2,  7        24479.7373   -0.0007    0.0020
   9,  3,  6 <-  8,  3,  5        25574.2305   -0.0013    0.0020
   9,  1,  8 <-  8,  1,  7        25576.6846   -0.0008    0.0020
   9,  2,  7 <-  8,  2,  6        26359.4424   -0.0006    0.0020
!
  10,  1, 10 <-  9,  1,  9        25233.5244    0.0006    0.0020
  10,  0, 10 <-  9,  0,  9        25385.3106    0.0006    0.0020
  10,  2,  9 <-  9,  2,  8        27079.4902   -0.0008    0.0020
  10,  3,  8 <-  9,  3,  7        27906.5049   -0.0016    0.0020
!
! bR-type lines
!
   1,  1,  1 <-  0,  0,  0         6439.0467    0.0011    0.0030
!
   2,  1,  2 <-  1,  0,  1         8848.4092    0.0006    0.0020
   2,  2,  1 <-  1,  1,  0        16907.7569   -0.0019    0.0020
   2,  2,  0 <-  1,  1,  1        17292.4971   -0.0018    0.0020
!
   3,  1,  3 <-  2,  0,  2        11087.9190   -0.0002    0.0020
   3,  2,  2 <-  2,  1,  1        19317.1172    0.0013    0.0020
   3,  2,  1 <-  2,  1,  2        20520.8223   -0.0024    0.0020
   3,  3,  1 <-  2,  2,  0        27546.2871    0.0018    0.0020
   3,  3,  0 <-  2,  2,  1        27572.5371    0.0003    0.0020
!
   4,  1,  4 <-  3,  0,  3        13190.3209    0.0001    0.0020
   4,  0,  4 <-  3,  1,  3         7950.5584   -0.0001    0.0020
   4,  2,  3 <-  3,  1,  2        21543.3965    0.0007    0.0020
   4,  2,  2 <-  3,  1,  3        24069.1348   -0.0014    0.0020
   4,  4,  1 <-  3,  3,  0        38028.1231   -0.0011    0.0020
!
   5,  1,  5 <-  4,  0,  4        15205.6257    0.0004    0.0020
   5,  0,  5 <-  4,  1,  4        11016.1890   -0.0007    0.0020
   5,  2,  4 <-  4,  1,  3        23591.0401    0.0011    0.0020
   5,  4,  2 <-  4,  3,  1        40799.1641    0.0014    0.0020
   5,  4,  1 <-  4,  3,  2        40807.2188   -0.0004    0.0020
!
   6,  1,  6 <-  5,  0,  5        17192.9746    0.0004    0.0020
   6,  0,  6 <-  5,  1,  5        14012.6456   -0.0003    0.0020
   6,  2,  5 <-  5,  1,  4        25470.2139    0.0008    0.0020
   6,  3,  4 <-  5,  2,  3        35347.0996    0.0014    0.0020
   6,  3,  3 <-  5,  2,  4        36276.7090    0.0004    0.0020
!
   7,  1,  7 <-  6,  0,  6        19206.7266    0.0008    0.0020
   7,  0,  7 <-  6,  1,  6        16905.5948   -0.0006    0.0020
   7,  2,  6 <-  6,  1,  5        27199.2647   -0.0003    0.0020
   7,  3,  4 <-  6,  2,  5        39437.2403    0.0000    0.0020
!
   8,  1,  8 <-  7,  0,  7        21283.0567    0.0008    0.0020
   8,  0,  8 <-  7,  1,  7        19686.0752   -0.0011    0.0020
!
   9,  1,  9 <-  8,  0,  8        23435.2227    0.0001    0.0020
   9,  0,  9 <-  8,  1,  8        22364.3955    0.0002    0.0020
!
```

```
  10,  1, 10 <-  9,  0,  9        25658.6924    0.0005    0.0020
!
! bQ-type lines
!
   2,  2,  0 <-  2,  1,  1        11035.4050   -0.0023    0.0020
   2,  2,  1 <-  2,  1,  2        12089.0284   -0.0025    0.0020
!
   3,  2,  1 <-  3,  1,  2        10612.1121   -0.0004    0.0020
   3,  2,  2 <-  3,  1,  3        12643.6164    0.0000    0.0020
!
   4,  2,  2 <-  4,  1,  3        10171.4581   -0.0001    0.0020
   4,  2,  3 <-  4,  1,  4        13390.3609    0.0008    0.0020
   4,  3,  1 <-  4,  2,  2        18969.7022    0.0005    0.0020
   4,  3,  2 <-  4,  2,  3        19331.3047   -0.0008    0.0020
!
   5,  1,  4 <-  5,  0,  5         7193.7346    0.0001    0.0020
   5,  2,  3 <-  5,  1,  4         9807.9390    0.0015    0.0020
   5,  2,  4 <-  5,  1,  5        14332.5327   -0.0005    0.0020
   5,  3,  2 <-  5,  2,  3        18654.5987    0.0002    0.0020
   5,  3,  3 <-  5,  2,  4        19462.5977   -0.0015    0.0020
!
   6,  1,  5 <-  6,  0,  6         8810.1602    0.0006    0.0030
   6,  2,  4 <-  6,  1,  5         9618.3242    0.0001    0.0020
   6,  2,  5 <-  6,  1,  6        15470.9727   -0.0007    0.0020
   6,  3,  3 <-  6,  2,  4        18168.1124    0.0002    0.0020
   6,  3,  4 <-  6,  2,  5        19684.8223   -0.0003    0.0020
   6,  4,  2 <-  6,  3,  3        26773.0703   -0.0009    0.0020
!
   7,  1,  6 <-  7,  0,  7        10795.6934   -0.0007    0.0030
   7,  2,  5 <-  7,  1,  6         9688.4673   -0.0003    0.0020
   7,  2,  6 <-  7,  1,  7        16802.7002    0.0014    0.0020
   7,  3,  4 <-  7,  2,  5        17518.4571    0.0004    0.0020
   7,  3,  5 <-  7,  2,  6        20024.6163    0.0009    0.0020
   7,  4,  3 <-  7,  3,  4        26622.3457    0.0006    0.0020
!
   8,  2,  6 <-  8,  1,  7        10087.7495   -0.0006    0.0020
   8,  2,  7 <-  8,  1,  8        18319.9893    0.0004    0.0020
!
!-------------------------------------------------------------------------
!
!  Data from R.K.Kakar, E.A.Reinhart, C.R.Quade, T.Kojima, J.Chem.Phys.
!                                          52,3803-3813(1970)
!
!  Table I = Oklahoma spectrometer
!
   7,  6,  1 <-  6,  6,  0        19453.100     0.018     0.200          1.00E+00
   7,  6,  2 <-  6,  6,  1        19453.100     0.018     0.200          1.00E+00
   7,  5,  2 <-  6,  5,  1        19472.100     0.086     0.200          1.00E+00
   7,  5,  3 <-  6,  5,  2        19472.100     0.086     0.200          1.00E+00
   7,  4,  3 <-  6,  4,  2        19511.000     0.298     0.200
   7,  4,  4 <-  6,  4,  3        19506.100     0.150     0.200
   7,  3,  5 <-  6,  3,  4        19528.300    -0.134     0.200
   7,  2,  6 <-  6,  2,  5        19188.900     0.259     0.200
   7,  1,  6 <-  6,  1,  5        20240.900    -0.040     0.200
   7,  0,  7 <-  6,  0,  6        18255.700     0.295     0.200

   8,  7,  1 <-  7,  7,  0        22230.100     0.321     0.200          1.00E+00
   8,  7,  2 <-  7,  7,  1        22230.100     0.321     0.200          1.00E+00
   8,  6,  2 <-  7,  6,  1        22246.700     0.218     0.200          1.00E+00
   8,  6,  3 <-  7,  6,  2        22246.700     0.218     0.200          1.00E+00
   8,  4,  4 <-  7,  4,  3        22336.100     0.272     0.200
   8,  4,  5 <-  7,  4,  4        22322.900     0.016     0.200
   8,  3,  5 <-  7,  3,  4        22588.600     0.072     0.200
   8,  3,  6 <-  7,  3,  5        22331.500     0.168     0.200
   8,  2,  6 <-  7,  2,  5        23350.200     0.009     0.200
   8,  2,  7 <-  7,  2,  6        21849.200     0.174     0.200
   8,  1,  7 <-  7,  1,  6        22951.100     0.191     0.200
```

```
   8,  1,  8 <-  7,  1,  7        20331.900     0.164     0.200

   9,  8,  1 <-  8,  8,  0        25006.700     0.093     0.200           1.00E+00
   9,  8,  2 <-  8,  8,  1        25006.700     0.093     0.200           1.00E+00
   9,  7,  2 <-  8,  7,  1        25021.900     0.037     0.200           1.00E+00
   9,  7,  3 <-  8,  7,  2        25021.900     0.037     0.200           1.00E+00
   9,  6,  3 <-  8,  6,  2        25045.800     0.136     0.200           1.00E+00
   9,  6,  4 <-  8,  6,  3        25045.800     0.136     0.200           1.00E+00
   9,  5,  4 <-  8,  5,  3        25086.200     0.108     0.200           1.00E+00
   9,  5,  5 <-  8,  5,  4        25086.200     0.108     0.200           1.00E+00
   9,  4,  5 <-  8,  4,  4        25179.800    -0.106     0.200
   9,  4,  6 <-  8,  4,  5        25149.400     0.126     0.200
   9,  3,  6 <-  8,  3,  5        25574.100    -0.132     0.200
   9,  3,  7 <-  8,  3,  6        25126.100     0.035     0.200
   9,  2,  7 <-  8,  2,  6        26359.500     0.057     0.200
   9,  2,  8 <-  8,  2,  7        24479.800     0.062     0.200
   9,  1,  8 <-  8,  1,  7        25576.700     0.015     0.200
   9,  1,  9 <-  8,  1,  8        22789.600     0.037     0.200
!
!  Table I = Wyoming spectrometer
!
  10,  9,  1 <-  9,  9,  0        27783.550     0.049     0.100           1.00E+00
  10,  9,  2 <-  9,  9,  1        27783.550     0.049     0.100           1.00E+00
  10,  7,  3 <-  9,  7,  2        27818.660    -0.019     0.100           1.00E+00
  10,  7,  4 <-  9,  7,  3        27818.660    -0.019     0.100           1.00E+00
  10,  6,  4 <-  9,  6,  3        27851.350    -0.030     0.100           1.00E+00
  10,  6,  5 <-  9,  6,  4        27851.350    -0.030     0.100           1.00E+00
  10,  5,  5 <-  9,  5,  4        27908.400     0.011     0.100
  10,  4,  6 <-  9,  4,  5        28048.830    -0.078     0.100
  10,  4,  7 <-  9,  4,  6        27983.860     0.004     0.100
  10,  3,  7 <-  9,  3,  6        28620.830     0.123     0.100
  10,  3,  8 <-  9,  3,  7        27906.500    -0.007     0.100
  10,  2,  8 <-  9,  2,  7        29318.300    -0.063     0.100
  10,  2,  9 <-  9,  2,  8        27079.550     0.059     0.100
  10,  1,  9 <-  9,  1,  8        28112.570     0.056     0.100

  11, 10,  1 <- 10, 10,  0        30560.400    -0.025     0.100           1.00E+00
  11, 10,  2 <- 10, 10,  1        30560.400    -0.025     0.100           1.00E+00
  11,  6,  5 <- 10,  6,  4        30664.570     0.164     0.100           1.00E+00
  11,  6,  6 <- 10,  6,  5        30664.570     0.164     0.100           1.00E+00
  11,  5,  6 <- 10,  5,  5        30742.320     0.054     0.100
  11,  4,  7 <- 10,  4,  6        30950.660     0.001     0.100
  11,  4,  8 <- 10,  4,  7        30824.220     0.071     0.100
  11,  3,  8 <- 10,  3,  7        31717.830     0.124     0.100
  11,  3,  9 <- 10,  3,  8        30666.760     0.057     0.100
  11,  2,  9 <- 10,  2,  8        32212.090     0.046     0.100
  11,  2, 10 <- 10,  2,  9        29648.340     0.136     0.100
  11,  1, 10 <- 10,  1,  9        30564.900     0.221     0.100
  11,  1, 11 <- 10,  1, 10        27666.780     0.034     0.100
  11,  0, 11 <- 10,  0, 10        27767.700     0.070     0.100

  12,  6,  6 <- 11,  6,  5        33485.550    -0.001     0.100           1.00E+00
  12,  6,  7 <- 11,  6,  6        33485.550    -0.001     0.100           1.00E+00
  12,  5,  7 <- 11,  5,  6        33590.400    -0.200     0.100
  12,  5,  8 <- 11,  5,  7        33576.350     0.107     0.100
  12,  4,  8 <- 11,  4,  7        33894.750     0.099     0.100
  12,  4,  9 <- 11,  4,  8        33666.550     0.156     0.100
  12,  3,  9 <- 11,  3,  8        34841.480     0.007     0.100
  12,  3, 10 <- 11,  3,  9        33401.380     0.039     0.100
  12,  2, 10 <- 11,  2,  9        35028.020     0.027     0.100
  12,  2, 11 <- 11,  2, 10        32187.150     0.083     0.100
  12,  1, 11 <- 11,  1, 10        32952.470     0.089     0.100
  12,  1, 12 <- 11,  1, 11        30092.050     0.016     0.100
  12,  0, 12 <- 11,  0, 11        30157.450     0.124     0.100

  13, 12,  1 <- 12, 12,  0        36114.410     0.138     0.100           1.00E+00
  13, 12,  2 <- 12, 12,  1        36114.410     0.138     0.100           1.00E+00
```

```
  13, 11,  2 <- 12, 11,  1        36126.830     0.065     0.100            1.00E+00
  13, 11,  3 <- 12, 11,  2        36126.830     0.065     0.100            1.00E+00
  13, 10,  3 <- 12, 10,  2        36143.280     0.079     0.100            1.00E+00
  13, 10,  4 <- 12, 10,  3        36143.280     0.079     0.100            1.00E+00
  13,  9,  4 <- 12,  9,  3        36165.570     0.092     0.100            1.00E+00
  13,  9,  5 <- 12,  9,  4        36165.570     0.092     0.100            1.00E+00
  13,  7,  6 <- 12,  7,  5        36243.090     0.085     0.100            1.00E+00
  13,  7,  7 <- 12,  7,  6        36243.090     0.085     0.100            1.00E+00
  13,  6,  7 <- 12,  6,  6        36315.710     0.041     0.100            1.00E+00
  13,  6,  8 <- 12,  6,  7        36315.710     0.041     0.100            1.00E+00
  13,  5,  8 <- 12,  5,  7        36456.750     0.079     0.100
  13,  5,  9 <- 12,  5,  8        36426.840     0.095     0.100
  13,  4,  9 <- 12,  4,  8        36890.900     0.054     0.100
  13,  4, 10 <- 12,  4,  9        36505.710     0.033     0.100
  13,  3, 10 <- 12,  3,  9        37960.230     0.100     0.100
  13,  3, 11 <- 12,  3, 10        36106.170     0.092     0.100
  13,  2, 11 <- 12,  2, 10        37754.810     0.059     0.100
  13,  2, 12 <- 12,  2, 11        34698.460     0.025     0.100
  13,  1, 12 <- 12,  1, 11        35302.170     0.114     0.100
  13,  1, 13 <- 12,  1, 12        32511.750     0.056     0.100
  13,  0, 13 <- 12,  0, 12        32553.240     0.146     0.100

  14, 13,  1 <- 13, 13,  0        38891.340     0.175     0.100            1.00E+00
  14, 13,  2 <- 13, 13,  1        38891.340     0.175     0.100            1.00E+00
  14, 12,  2 <- 13, 12,  1        38903.360     0.057     0.100            1.00E+00
  14, 12,  3 <- 13, 12,  2        38903.360     0.057     0.100            1.00E+00
  14, 11,  3 <- 13, 11,  2        38918.910     0.019     0.100            1.00E+00
  14, 11,  4 <- 13, 11,  3        38918.910     0.019     0.100            1.00E+00
  14, 10,  4 <- 13, 10,  3        38939.600     0.187     0.100            1.00E+00
  14, 10,  5 <- 13, 10,  4        38939.600     0.187     0.100            1.00E+00
  14,  9,  5 <- 13,  9,  4        38967.380     0.133     0.100            1.00E+00
  14,  9,  6 <- 13,  9,  5        38967.380     0.133     0.100            1.00E+00
  14,  8,  6 <- 13,  8,  5        39006.490     0.059     0.100            1.00E+00
  14,  8,  7 <- 13,  8,  6        39006.490     0.059     0.100            1.00E+00
  14,  7,  7 <- 13,  7,  6        39064.320     0.012     0.100            1.00E+00
  14,  7,  8 <- 13,  7,  7        39064.320     0.012     0.100            1.00E+00
  14,  5,  9 <- 13,  5,  8        39344.920    -0.041     0.100
  14,  5, 10 <- 13,  5,  9        39286.740     0.161     0.100
  14,  4, 10 <- 13,  4,  9        39946.810    -0.047     0.100
  14,  4, 11 <- 13,  4, 10        39336.330     0.109     0.100
  14,  3, 12 <- 13,  3, 11        38777.800     0.017     0.100
  14,  2, 13 <- 13,  2, 12        37185.620     0.046     0.100
  14,  1, 13 <- 13,  1, 12        37638.480     0.100     0.100
  14,  1, 14 <- 13,  1, 13        34927.570     0.066     0.100
  14,  0, 14 <- 13,  0, 13        34953.500     0.168     0.100

  15,  1, 14 <- 14,  1, 13        39978.150     0.059     0.100
  15,  1, 15 <- 14,  1, 14        37340.880     0.119     0.100
  15,  0, 15 <- 14,  0, 14        37356.750     0.086     0.100

  16,  1, 16 <- 15,  1, 15        39752.520     0.150     0.100
  16,  0, 16 <- 15,  0, 15        39762.220     0.164     0.100
!
!  Table V = Q-branch b-dipole transitions
!
  12,  5,  8 <- 12,  4,  9        34172.590     0.124     0.150
  13,  4, 10 <- 13,  3, 11        27753.790    -0.200     0.150
  13,  5,  9 <- 13,  4, 10        34093.510    -0.023     0.150
  14,  5,  9 <- 14,  4, 10        32691.210     0.145     0.150
  16,  5, 12 <- 16,  4, 13        34126.320     0.126     0.150
  17,  5, 13 <- 17,  4, 14        34311.840    -0.046     0.150
  18,  5, 14 <- 18,  4, 15        34630.880     0.086     0.150
  19,  4, 16 <- 19,  3, 17        33964.550     0.084     0.150
  20,  5, 16 <- 20,  4, 17        35766.000     0.063     0.150
!
!-----------------------------------------------------------------------------
!
```

```
!  MMW measurements in Warsaw
!
! aR_0,1 type-II bandheads
!
  72,  0, 72 <- 71,  0, 71       174628.394     0.004     0.050
  71,  1, 70 <- 70,  1, 69       174627.671    -0.015     0.050
  70,  2, 68 <- 69,  2, 67       174630.202     0.001     0.050
  69,  3, 66 <- 68,  3, 65       174639.423    -0.019     0.050
  68,  4, 64 <- 67,  4, 63       174660.610    -0.032     0.050
  67,  5, 62 <- 66,  5, 61       174701.796    -0.053     0.050
  66,  6, 60 <- 65,  6, 59       174775.944    -0.062     0.050
  65,  7, 58 <- 64,  7, 57       174905.712     0.055     0.050
!
  73,  0, 73 <- 72,  0, 72       177035.640    -0.023     0.050
  72,  1, 71 <- 71,  1, 70       177034.915    -0.005     0.050
  71,  2, 69 <- 70,  2, 68       177037.288     0.006     0.050
  70,  3, 67 <- 69,  3, 66       177046.089    -0.036     0.050
  69,  4, 65 <- 68,  4, 64       177066.428    -0.020     0.050
  68,  5, 63 <- 67,  5, 62       177105.854    -0.026     0.050
!
  87,  1, 86 <- 86,  1, 85       213136.236    -0.010     0.050
  85,  3, 82 <- 84,  3, 81       213141.579    -0.037     0.050
  84,  4, 80 <- 83,  4, 79       213152.948    -0.029     0.050
  83,  5, 78 <- 82,  5, 77       213174.921    -0.009     0.050
  82,  6, 76 <- 81,  6, 75       213212.987    -0.018     0.050
  81,  7, 74 <- 80,  7, 73       213275.328     0.009     0.050
  80,  8, 72 <- 79,  8, 71       213374.209    -0.047     0.050
!
  89,  0, 89 <- 88,  0, 88       215543.606    -0.080     0.050
  88,  1, 87 <- 87,  1, 86       215542.466    -0.018     0.050
  86,  3, 83 <- 85,  3, 82       215547.568    -0.013     0.050
  85,  4, 81 <- 84,  4, 80       215558.524    -0.012     0.050
  84,  5, 79 <- 83,  5, 78       215579.750     0.022     0.050
  83,  6, 77 <- 82,  6, 76       215616.411    -0.040     0.050
  82,  7, 75 <- 81,  7, 74       215676.414    -0.006     0.050
  81,  8, 73 <- 80,  8, 72       215771.255    -0.060     0.050
  80,  9, 71 <- 79,  9, 70       215919.720    -0.099     0.050
!
 105,  0,105 <-104,  0,104       254033.958     0.007     0.050             1.00E+00
 102,  3, 99 <-101,  3, 98       254033.958     0.007     0.050             1.00E+00
 104,  1,103 <-103,  1,102       254032.153    -0.005     0.050             1.00E+00
 103,  2,101 <-102,  2,100       254032.153    -0.005     0.050             1.00E+00
 101,  4, 97 <-100,  4, 96       254040.321    -0.032     0.050
 100,  5, 95 <- 99,  5, 94       254052.862    -0.008     0.050
  99,  6, 93 <- 98,  6, 92       254074.568    -0.002     0.050
  98,  7, 91 <- 97,  7, 90       254109.356     0.006     0.050
  97,  8, 89 <- 96,  8, 88       254162.581     0.015     0.050
  96,  9, 87 <- 95,  9, 86       254241.792     0.030     0.050
  95, 10, 85 <- 94, 10, 84       254358.028     0.098     0.050
  94, 11, 83 <- 93, 11, 82       254527.763    -0.067     0.050
!
 134,  0,134 <-133,  0,133       323741.014     0.073     0.050
 133,  1,132 <-132,  1,131       323739.213    -0.003     0.050
 132,  2,130 <-131,  2,129       323737.922    -0.040     0.050
 131,  3,128 <-130,  3,127       323737.922     0.031     0.050
 130,  4,126 <-129,  4,125       323739.911     0.040     0.050
 129,  5,124 <-128,  5,123       323745.041     0.077     0.050
 128,  6,122 <-127,  6,121       323754.518     0.037     0.050
 127,  7,120 <-126,  7,119       323770.029    -0.020     0.050
 126,  8,118 <-125,  8,117       323793.703    -0.009     0.050
 125,  9,116 <-124,  9,115       323828.018    -0.040     0.050
 124, 10,114 <-123, 10,113       323876.444     0.035     0.050
 121, 13,108 <-120, 13,107       324156.603    -0.001     0.050
!
! higher K-1 tails of aR_0,1-type lines for a given J
!
  59, 11, 48 <- 58, 11, 47       174609.773    -0.048     0.050
```

```
!
  60, 12, 48 <- 59, 12, 47       177091.255    -0.028     0.050
!
  61, 10, 51 <- 60, 10, 50       175143.398    -0.035     0.050
  61, 12, 50 <- 60, 12, 49       174783.597     0.011     0.050
  61, 14, 48 <- 60, 14, 47       174551.313    -0.002     0.050
  61, 14, 47 <- 60, 14, 46       175065.849    -0.003     0.050
!
  62, 11, 52 <- 61, 11, 51       175646.483     0.068     0.050
  62, 15, 47 <- 61, 15, 46       176931.974     0.002     0.050
  62, 15, 48 <- 61, 15, 47       176817.191    -0.035     0.050
  62, 17, 46 <- 61, 17, 45       175552.723    -0.037     0.050
  62, 17, 45 <- 61, 17, 44       175554.115    -0.028     0.050
  62, 18, 45 <- 61, 18, 44       175105.636     0.023     0.050
  62, 19, 44 <- 61, 19, 43       174741.960     0.013     0.050
!
  63,  9, 54 <- 62,  9, 53       175649.326     0.031     0.050
  63, 10, 54 <- 62, 10, 53       175452.378    -0.105     0.050
  63, 21, 42 <- 62, 21, 41       177074.326     0.023     0.050
  63, 22, 41 <- 62, 22, 40       176845.679     0.001     0.050
  63, 32, 31 <- 62, 32, 30       175647.896     0.079     0.050
  63, 33, 30 <- 62, 33, 29       175584.417     0.076     0.050
  63, 34, 29 <- 62, 34, 28       175526.289     0.013     0.050
  63, 35, 28 <- 62, 35, 27       175473.014     0.022     0.050
  63, 36, 27 <- 62, 36, 26       175423.981     0.035     0.050
  63, 37, 26 <- 62, 37, 25       175378.772     0.101     0.050
  63, 38, 25 <- 62, 38, 24       175336.839     0.076     0.050
  63, 40, 23 <- 62, 40, 22       175261.755     0.070     0.050
  63, 41, 22 <- 62, 41, 21       175228.046     0.107     0.050
  63, 42, 21 <- 62, 42, 20       175196.481     0.086     0.050
  63, 43, 20 <- 62, 43, 19       175166.877     0.034     0.050
  63, 45, 18 <- 62, 45, 17       175113.047     0.052     0.050
!
  72, 14, 58 <- 71, 14, 57       213377.336     0.015     0.050
!
  73, 13, 60 <- 72, 13, 59       214035.104     0.019     0.050
  73, 15, 58 <- 72, 15, 57       214016.152     0.056     0.050
!
  74, 15, 60 <- 73, 15, 59       212972.587    -0.013     0.050
  74, 16, 58 <- 73, 16, 57       214136.743    -0.039     0.050
  74, 16, 59 <- 73, 16, 58       212685.817    -0.011     0.050
!
  75, 12, 63 <- 74, 12, 62       213087.463    -0.024     0.050
  75, 17, 58 <- 74, 17, 57       215294.433     0.011     0.050
  75, 18, 57 <- 74, 18, 56       214053.024    -0.040     0.050
  75, 18, 58 <- 74, 18, 57       213994.386    -0.010     0.050
  75, 19, 57 <- 74, 19, 56       213227.393    -0.034     0.050
  75, 19, 56 <- 74, 19, 55       213234.574    -0.005     0.050
!
  76, 12, 64 <- 75, 12, 63       215244.984     0.021     0.050
  76, 20, 57 <- 75, 20, 56       215592.095     0.011     0.050
  76, 24, 52 <- 75, 24, 51       213927.644    -0.006     0.050
  76, 25, 51 <- 75, 25, 50       213651.317    -0.001     0.050
  76, 27, 49 <- 75, 27, 48       213196.994     0.018     0.050
  76, 28, 48 <- 75, 28, 47       213008.210    -0.014     0.050
  76, 29, 47 <- 75, 29, 46       212839.826     0.023     0.050
  76, 30, 46 <- 75, 30, 45       212688.718    -0.006     0.050
  76, 31, 45 <- 75, 31, 44       212552.557     0.001     0.050
  76, 32, 44 <- 75, 32, 43       212429.312     0.018     0.050
!
  77, 25, 52 <- 76, 25, 51       216545.307    -0.005     0.050
  77, 30, 47 <- 76, 30, 46       215541.009    -0.035     0.050
  77, 31, 46 <- 76, 31, 45       215399.199    -0.008     0.050
  77, 32, 45 <- 76, 32, 44       215270.840    -0.014     0.050
  77, 48, 29 <- 76, 48, 28       214178.060    -0.022     0.050
  77, 49, 28 <- 76, 49, 27       214141.509     0.081     0.050
  77, 50, 27 <- 76, 50, 26       214106.696    -0.030     0.050
```

```
  77, 51, 26 <- 76, 51, 25       214073.789    -0.028     0.050
  77, 53, 24 <- 76, 53, 23       214012.786    -0.038     0.050
  77, 58, 19 <- 76, 58, 18       213883.151    -0.072     0.050
  77, 59, 18 <- 76, 59, 17       213860.469    -0.025     0.050
  77, 60, 17 <- 76, 60, 16       213838.637    -0.019     0.050
  77, 61, 16 <- 76, 61, 15       213817.584    -0.061     0.050
  77, 62, 15 <- 76, 62, 14       213797.400    -0.006     0.050
  77, 65, 12 <- 76, 65, 11       213740.781    -0.026     0.050
!
  78, 10, 68 <- 77, 10, 67       213779.198    -0.015     0.050
  78, 11, 68 <- 77, 11, 67       213773.986     0.005     0.050
  78, 11, 67 <- 77, 11, 66       216558.348    -0.047     0.050
!
 112, 22, 91 <-111, 22, 90       323756.648     0.055     0.050
 114, 29, 85 <-113, 29, 84       323734.138    -0.025     0.050
 115, 34, 81 <-114, 34, 80       324197.840    -0.042     0.050
 115, 35, 80 <-114, 35, 79       323844.185    -0.030     0.050
!
!
! Miscellaneous bR-type lines
!
  18, 17,  2 <- 17, 16,  1       176881.628    -0.053     0.050
  20, 16,  5 <- 19, 15,  4       174744.437    -0.010     0.050
  23, 15,  9 <- 22, 14,  8       175370.727    -0.065     0.050
  23, 20,  3 <- 22, 19,  4       213828.656    -0.030     0.050
  24, 20,  4 <- 23, 19,  5       216604.454    -0.015     0.050
  31, 17, 15 <- 30, 16, 14       212905.923     0.002     0.050
  37, 15, 23 <- 36, 14, 22       213830.609     0.012     0.050
  40, 14, 27 <- 39, 13, 26       213979.369     0.028     0.050            1.00E+00
  40, 14, 26 <- 39, 13, 27       213979.369     0.028     0.050            1.00E+00
  73, 13, 61 <- 72, 12, 60       213181.347     0.010     0.050
  76, 12, 64 <- 75, 13, 63       213014.850    -0.012     0.050
  77, 11, 66 <- 76, 12, 65       214003.992     0.026     0.050
  78, 10, 68 <- 77, 11, 67       213763.904     0.003     0.050
  78, 11, 68 <- 77, 10, 67       213789.235    -0.058     0.050
  49, 25, 25 <- 48, 24, 24       324264.844    -0.012     0.050
!
! bQ-type lines
!
  68, 24, 44 <- 68, 23, 45       177026.249     0.023     0.050
  69, 24, 45 <- 69, 23, 46       176796.306     0.065     0.050
  74, 24, 50 <- 74, 23, 51       175468.378     0.062     0.050
  71, 29, 42 <- 71, 28, 43       216728.223    -0.027     0.050
  72, 29, 43 <- 72, 28, 44       216586.219    -0.004     0.050
  78, 29, 49 <- 78, 28, 50       215594.697     0.031     0.050
  79, 29, 50 <- 79, 28, 51       215404.024    -0.040     0.050
  80, 29, 51 <- 80, 28, 52       215205.645     0.069     0.050
  85, 29, 56 <- 85, 28, 57       214085.424     0.043     0.050
  86, 29, 57 <- 86, 28, 58       213833.671    -0.094     0.050
  89, 29, 60 <- 89, 28, 61       213018.084     0.025     0.050
  90, 29, 61 <- 90, 28, 62       212724.670    -0.038     0.050
  91, 29, 62 <- 91, 28, 63       212419.996    -0.020     0.050
 104,  8, 96 <-104,  7, 97       212761.677     0.074     0.050
```

Table S2. The observed and the obs.-calc. frequencies (MHz) of measured rotational transitions in the first excited torsional state of benzaldehyde.

---

benzaldehyde, v=1

Obs-calc differences are from fit of Watson's reduced Hamiltonian in ---> reduction-A, representation-I.r <---

TRANSITION	Observed	Obs-Calc	Error	Note	wt for blend
!					
! Data from R.K.Kakar, E.A.Reinhart, C.R.Quade, T.Kojima, J.Chem.Phys. 52,3803-3813(1970)					
!					
! Table I = Oklahoma spectrometer					
!					
7, 6, 1 <- 6, 6, 0	19470.200	-0.118	0.200		1.0000
7, 6, 2 <- 6, 6, 1	19470.200	-0.118	0.200		1.0000
7, 5, 2 <- 6, 5, 1	19489.300	0.062	0.200		1.0000
7, 5, 3 <- 6, 5, 2	19489.300	0.062	0.200		1.0000
7, 4, 3 <- 6, 4, 2	19528.300	0.384	0.200		
7, 4, 4 <- 6, 4, 3	19523.300	0.158	0.200		
7, 3, 4 <- 6, 3, 3	19679.100	0.394	0.200		
7, 3, 5 <- 6, 3, 4	19545.200	-0.275	0.200		
7, 2, 5 <- 6, 2, 4	20326.990	0.250	0.200		
7, 2, 6 <- 6, 2, 5	19206.400	0.356	0.200		
7, 1, 6 <- 6, 1, 5	20254.200	-0.038	0.200		
7, 0, 7 <- 6, 0, 6	18274.300	-0.106	0.200		
8, 7, 1 <- 7, 7, 0	22249.600	0.121	0.200		1.0000
8, 7, 1 <- 7, 7, 0	22249.600	0.121	0.200		1.0000
8, 6, 2 <- 7, 6, 1	22266.300	0.128	0.200		1.0000
8, 6, 3 <- 7, 6, 2	22266.300	0.128	0.200		1.0000
8, 5, 3 <- 7, 5, 2	22294.600	0.137	0.200		1.0000
8, 5, 4 <- 7, 5, 3	22294.600	0.137	0.200		1.0000
8, 4, 4 <- 7, 4, 3	22355.500	0.000	0.200		
8, 4, 5 <- 7, 4, 4	22342.500	0.003	0.200		
8, 3, 5 <- 7, 3, 4	22608.400	0.096	0.200		
8, 3, 6 <- 7, 3, 5	22350.600	-0.097	0.200		
8, 2, 6 <- 7, 2, 5	23367.300	0.009	0.200		
8, 2, 7 <- 7, 2, 6	21869.200	0.222	0.200		
8, 1, 7 <- 7, 1, 6	22966.300	0.187	0.200		
8, 1, 8 <- 7, 1, 7	20356.800	0.196	0.200		
8, 0, 8 <- 7, 0, 7	20660.200	0.265	0.200		
9, 8, 1 <- 8, 8, 0	25028.800	0.029	0.200		1.0000
9, 8, 2 <- 8, 8, 1	25028.800	0.029	0.200		1.0000
9, 7, 2 <- 8, 7, 1	25044.000	-0.017	0.200		1.0000
9, 7, 3 <- 8, 7, 2	25044.000	-0.017	0.200		1.0000
9, 6, 3 <- 8, 6, 2	25067.800	-0.003	0.200		1.0000
9, 6, 4 <- 8, 6, 3	25067.800	-0.003	0.200		1.0000
9, 5, 4 <- 8, 5, 3	25108.300	0.092	0.200		1.0000
9, 5, 5 <- 8, 5, 4	25108.300	0.092	0.200		1.0000
9, 4, 5 <- 8, 4, 4	25202.400	0.347	0.200		
9, 4, 6 <- 8, 4, 5	25171.000	-0.284	0.200		
9, 3, 7 <- 8, 3, 6	25147.600	-0.109	0.200		
9, 2, 7 <- 8, 2, 6	26377.800	0.007	0.200		
9, 2, 8 <- 8, 2, 7	24502.300	0.024	0.200		
9, 1, 8 <- 8, 1, 7	25593.900	0.048	0.200		
9, 1, 9 <- 8, 1, 8	22817.800	0.069	0.200		
9, 0, 9 <- 8, 0, 8	23036.400	0.083	0.200		
!					

```
!  Table I = Wyoming spectrometer
!
  10,  9,  1 <-  9,  9,  0        27808.020    -0.110     0.100           1.0000
  10,  9,  2 <-  9,  9,  1        27808.020    -0.110     0.100           1.0000
  10,  8,  2 <-  9,  8,  1        27822.330    -0.035     0.100           1.0000
  10,  8,  3 <-  9,  8,  2        27822.330    -0.035     0.100           1.0000
  10,  7,  3 <-  9,  7,  2        27843.250    -0.034     0.100           1.0000
  10,  7,  4 <-  9,  7,  3        27843.250    -0.034     0.100           1.0000
  10,  6,  4 <-  9,  6,  3        27875.930    -0.035     0.100           1.0000
  10,  6,  5 <-  9,  6,  4        27875.930    -0.035     0.100           1.0000
  10,  5,  5 <-  9,  5,  4        27932.860    -0.092     0.100
  10,  4,  7 <-  9,  4,  6        28008.210    -0.015     0.100
  10,  2,  8 <-  9,  2,  7        29337.740    -0.123     0.100
  10,  2,  9 <-  9,  2,  8        27104.830     0.165     0.100

  11,  7,  4 <- 10,  7,  3        30647.900     0.078     0.100           1.0000
  11,  7,  5 <- 10,  7,  4        30647.900     0.078     0.100           1.0000
  11,  6,  5 <- 10,  6,  4        30691.670     0.238     0.100           1.0000
  11,  6,  6 <- 10,  6,  5        30691.670     0.238     0.100           1.0000
  11,  5,  6 <- 10,  5,  5        30769.480     0.204     0.100
  11,  4,  7 <- 10,  4,  6        30977.690    -0.175     0.100
  11,  4,  8 <- 10,  4,  7        30850.740    -0.085     0.100
  11,  3,  9 <- 10,  3,  8        30692.800     0.023     0.100
  11,  2,  9 <- 10,  2,  8        32232.760     0.112     0.100
  11,  2, 10 <- 10,  2,  9        29676.230     0.151     0.100
  11,  1, 11 <- 10,  1, 10        27701.680    -0.017     0.100
  11,  0, 11 <- 10,  0, 10        27801.680     0.167     0.100

  12,  6,  6 <- 11,  6,  5        33515.000    -0.014     0.100           1.0000
  12,  6,  7 <- 11,  6,  6        33515.000    -0.014     0.100           1.0000
  12,  5,  7 <- 11,  5,  6        33620.150     0.081     0.100
  12,  5,  8 <- 11,  5,  7        33605.700     0.089     0.100
  12,  4,  8 <- 11,  4,  7        33924.550     0.077     0.100
  12,  3,  9 <- 11,  3,  8        34869.480     0.015     0.100
  12,  3, 10 <- 11,  3,  9        33429.600     0.010     0.100
  12,  2, 10 <- 11,  2,  9        35049.710     0.029     0.100
  12,  2, 11 <- 11,  2, 10        32217.770     0.046     0.100
  12,  1, 11 <- 11,  1, 10        32977.170     0.089     0.100
  12,  1, 12 <- 11,  1, 11        30130.480     0.023     0.100
  12,  0, 12 <- 11,  0, 11        30195.110     0.111     0.100

  13, 12,  1 <- 12, 12,  0        36146.280    -0.013     0.100           1.0000
  13, 12,  2 <- 12, 12,  1        36146.280    -0.013     0.100           1.0000
  13, 11,  2 <- 12, 11,  1        36158.840     0.064     0.100           1.0000
  13, 11,  3 <- 12, 11,  2        36158.840     0.064     0.100           1.0000
  13, 10,  3 <- 12, 10,  2        36175.060    -0.139     0.100           1.0000
  13, 10,  4 <- 12, 10,  3        36175.060    -0.139     0.100           1.0000
  13,  8,  5 <- 12,  8,  4        36228.900     0.130     0.100           1.0000
  13,  8,  6 <- 12,  8,  5        36228.900     0.130     0.100           1.0000
  13,  7,  6 <- 12,  7,  5        36274.880    -0.060     0.100           1.0000
  13,  7,  7 <- 12,  7,  6        36274.880    -0.060     0.100           1.0000
  13,  5,  8 <- 12,  5,  7        36488.700     0.077     0.100
  13,  5,  9 <- 12,  5,  8        36458.650     0.160     0.100
  13,  4,  9 <- 12,  4,  8        36923.440     0.114     0.100
  13,  3, 10 <- 12,  3,  9        37988.950    -0.170     0.100
  13,  3, 11 <- 12,  3, 10        36136.680     0.179     0.100
  13,  2, 11 <- 12,  2, 10        37777.600     0.072     0.100
  13,  2, 12 <- 12,  2, 11        34731.910    -0.061     0.100
  13,  1, 12 <- 12,  1, 11        35330.280     0.099     0.100
  13,  1, 13 <- 12,  1, 12        32553.500    -0.132     0.100
  13,  0, 13 <- 12,  0, 12        32594.630     0.111     0.100

  14, 13,  1 <- 13, 13,  0        38925.680     0.029     0.100           1.0000
  14, 13,  2 <- 13, 13,  1        38925.680     0.029     0.100           1.0000
  14, 12,  2 <- 13, 12,  1        38937.900     0.122     0.100           1.0000
  14, 12,  3 <- 13, 12,  2        38937.900     0.122     0.100           1.0000
  14, 11,  3 <- 13, 11,  2        38953.350    -0.004     0.100           1.0000
```

```
  14, 11,  4 <- 13, 11,  3        38953.350    -0.004     0.100           1.0000
  14, 10,  4 <- 13, 10,  3        38973.830    -0.031     0.100           1.0000
  14, 10,  5 <- 13, 10,  4        38973.830    -0.031     0.100           1.0000
  14,  9,  5 <- 13,  9,  4        39001.690     0.013     0.100           1.0000
  14,  9,  6 <- 13,  9,  5        39001.690     0.013     0.100           1.0000
  14,  8,  6 <- 13,  8,  5        39040.930     0.095     0.100           1.0000
  14,  8,  7 <- 13,  8,  6        39040.930     0.095     0.100           1.0000
  14,  7,  7 <- 13,  7,  6        39098.630    -0.050     0.100           1.0000
  14,  7,  8 <- 13,  7,  7        39098.630    -0.050     0.100           1.0000
  14,  5,  9 <- 13,  5,  8        39379.420    -0.021     0.100
  14,  4, 10 <- 13,  4,  9        39981.910    -0.053     0.100
  14,  4, 11 <- 13,  4, 10        39369.440     0.060     0.100
  14,  3, 12 <- 13,  3, 11        38810.400     0.001     0.100
  14,  2, 13 <- 13,  2, 12        37222.000    -0.094     0.100
  14,  1, 13 <- 13,  1, 12        37670.370     0.061     0.100
  14,  1, 14 <- 13,  1, 13        34973.120     0.131     0.100
  14,  0, 14 <- 13,  0, 13        34998.580     0.105     0.100

  15,  1, 14 <- 14,  1, 13        40013.980    -0.054     0.100
  15,  1, 15 <- 14,  1, 14        37389.900     0.083     0.100
  15,  0, 15 <- 14,  0, 14        37405.580     0.085     0.100

  16,  1, 16 <- 15,  1, 15        39805.200     0.185     0.100
  16,  0, 16 <- 15,  0, 15        39814.700     0.145     0.100
!
!-------------------------------------------------------------------------
!
!  MMW measurements in Warsaw
!
!  aR_0,1 bandheads
!
  72,  0, 72 <- 71,  0, 71       174882.592    -0.047     0.050
  71,  1, 70 <- 70,  1, 69       174867.568     0.091     0.050
  70,  2, 68 <- 69,  2, 67       174855.559     0.031     0.050
  68,  4, 64 <- 67,  4, 63       174856.917     0.026     0.050
  67,  5, 62 <- 66,  5, 61       174883.386     0.023     0.050
  66,  6, 60 <- 65,  6, 59       174942.507     0.006     0.050
  65,  7, 58 <- 64,  7, 57       175056.710     0.119     0.050
  65,  8, 58 <- 64,  8, 57       175055.719    -0.014     0.050
  64,  9, 56 <- 63,  9, 55       175258.371     0.016     0.050
  64,  8, 56 <- 63,  8, 55       175273.484     0.002     0.050

  61, 12, 50 <- 60, 12, 49       174903.350    -0.067     0.050

  88,  0, 88 <- 87,  0, 87       213448.560    -0.011     0.050
  87,  1, 86 <- 86,  1, 85       213432.890    -0.003     0.050
  86,  2, 84 <- 85,  2, 83       213419.107    -0.018     0.050
  84,  4, 80 <- 83,  4, 79       213406.088    -0.022     0.050
  83,  5, 78 <- 82,  5, 77       213413.452    -0.015     0.050
  82,  6, 76 <- 81,  6, 75       213436.829    -0.001     0.050
  81,  7, 74 <- 80,  7, 73       213484.229    -0.005     0.050
  80,  8, 72 <- 79,  8, 71       213567.930    -0.004     0.050
  79,  9, 70 <- 78,  9, 69       213707.669    -0.099     0.050

  89,  0, 89 <- 88,  0, 88       215858.349    -0.023     0.050
  88,  1, 87 <- 87,  1, 86       215842.626    -0.043     0.050
  87,  2, 85 <- 86,  2, 84       215828.798    -0.022     0.050
  86,  3, 83 <- 85,  3, 82       215818.774    -0.003     0.050
  85,  4, 81 <- 84,  4, 80       215815.174    -0.031     0.050
  84,  5, 79 <- 83,  5, 78       215821.797    -0.010     0.050
  83,  6, 77 <- 82,  6, 76       215843.854     0.028     0.050
  82,  7, 75 <- 81,  7, 74       215888.886    -0.020     0.050
  80,  9, 71 <- 79,  9, 70       216101.289     0.001     0.050           1.0000
  80, 10, 71 <- 79, 10, 70       216101.289     0.001     0.050           1.0000
  79, 10, 69 <- 78, 10, 68       216320.140    -0.028     0.050
  79, 11, 69 <- 78, 11, 68       216316.904     0.047     0.050
  78, 11, 67 <- 77, 11, 66       216703.861     0.041     0.050
```

```
  78, 12, 67 <- 77, 12, 66       216658.675    -0.010     0.050

 105,  0,105 <-104,  0,104       254404.804    -0.051     0.050
 104,  1,103 <-103,  1,102       254388.839     0.001     0.050
 103,  2,101 <-102,  2,100       254373.953    -0.038     0.050
 102,  3, 99 <-101,  3, 98       254361.580    -0.011     0.050
 101,  4, 97 <-100,  4, 96       254353.305     0.016     0.050
 100,  5, 95 <- 99,  5, 94       254351.241    -0.010     0.050
  98,  7, 91 <- 97,  7, 90       254378.411    -0.022     0.050
  97,  8, 89 <- 96,  8, 88       254416.818     0.002     0.050
  96,  9, 87 <- 95,  9, 86       254480.945    -0.017     0.050
  95, 10, 85 <- 94, 10, 84       254581.774     0.030     0.050

 134,  0,134 <-133,  0,133       324212.717    -0.014     0.050
 133,  1,132 <-132,  1,131       324196.308    -0.022     0.050
 132,  2,130 <-131,  2,129       324180.453     0.018     0.050
 131,  3,128 <-130,  3,127       324165.834     0.082     0.050
 130,  4,126 <-129,  4,125       324153.145     0.007     0.050
 129,  5,124 <-128,  5,123       324143.656     0.009     0.050
 128,  6,122 <-127,  6,121       324138.604     0.027     0.050
 127,  7,120 <-126,  7,119       324139.568     0.026     0.050
 126,  8,118 <-125,  8,117       324148.578     0.013     0.050
 125,  9,116 <-124,  9,115       324168.186    -0.023     0.050
 124, 10,114 <-123, 10,113       324201.750    -0.014     0.050
 123, 11,112 <-122, 11,111       324253.535     0.014     0.050
!
!  higher K-1 tails of aR_0,1-type lines for J+1<-J sequences
!
  59, 11, 48 <- 58, 11, 47       174698.962    -0.028     0.050
  61, 14, 48 <- 60, 14, 47       174697.254    -0.059     0.050

  62, 15, 48 <- 61, 15, 47       176967.883     0.038     0.050
  62, 15, 47 <- 61, 15, 46       177085.782    -0.070     0.050
  62, 18, 45 <- 61, 18, 44       175256.007     0.026     0.050

  63, 37, 26 <- 62, 37, 25       175532.826     0.035     0.050
  63, 36, 27 <- 62, 36, 26       175577.997     0.002     0.050
  63, 35, 28 <- 62, 35, 27       175626.951    -0.018     0.050
  63, 34, 29 <- 62, 34, 28       175680.204     0.023     0.050

  73, 13, 60 <- 72, 13, 59       214092.676     0.040     0.050

  74, 15, 60 <- 73, 15, 59       213126.157     0.032     0.050
  74, 16, 59 <- 73, 16, 58       212857.811     0.019     0.050

  75, 12, 63 <- 74, 12, 62       213185.591    -0.043     0.050
  75, 18, 58 <- 74, 18, 57       214176.735    -0.014     0.050
  75, 20, 55 <- 74, 20, 54       212791.475     0.051     0.050
  75, 20, 56 <- 74, 20, 55       212790.672     0.018     0.050

  76, 24, 52 <- 75, 24, 51       214111.120     0.011     0.050
  76, 28, 48 <- 75, 28, 47       213192.056    -0.009     0.050
  76, 29, 47 <- 75, 29, 46       213023.772     0.019     0.050
  76, 30, 46 <- 75, 30, 45       212872.833     0.048     0.050
  76, 31, 45 <- 75, 31, 44       212736.705    -0.020     0.050
  76, 32, 44 <- 75, 32, 43       212613.555    -0.015     0.050
  76, 33, 43 <- 75, 33, 42       212501.631    -0.023     0.050

  77, 25, 52 <- 76, 25, 51       216731.169     0.001     0.050
  77, 26, 51 <- 76, 26, 50       216478.899    -0.063     0.050
  77, 30, 47 <- 76, 30, 46       215727.351    -0.072     0.050
  77, 31, 46 <- 76, 31, 45       215585.667    -0.029     0.050
  77, 32, 45 <- 76, 32, 44       215457.438    -0.015     0.050

 115, 18, 97 <-114, 18, 96       323825.750    -0.081     0.050
 115, 19, 97 <-114, 19, 96       323752.550     0.009     0.050
 115, 36, 79 <-114, 36, 78       323796.870     0.034     0.050
```

```
!
!  bQ-type lines
!
  64, 24, 40 <- 64, 23, 41       176795.193    -0.109     0.050
  63, 24, 39 <- 63, 23, 40       176975.300     0.024     0.050
  62, 24, 38 <- 62, 23, 39       177146.492     0.092     0.050

  72, 29, 43 <- 72, 28, 44       215318.978     0.002     0.050
  70, 29, 41 <- 70, 28, 42       215596.774     0.024     0.050
  66, 29, 37 <- 66, 28, 38       216083.271    -0.022     0.050
  65, 29, 36 <- 65, 28, 37       216191.608    -0.028     0.050
  64, 29, 35 <- 64, 28, 36       216295.015     0.001     0.050
  63, 29, 34 <- 63, 28, 35       216393.592    -0.001     0.050
!
!  bR-type lines
!
  21, 16,  6 <- 20, 15,  5       176877.282    -0.082     0.050
  23, 20,  4 <- 22, 19,  3       213013.470     0.001     0.050
  24, 20,  5 <- 23, 19,  4       215791.718    -0.001     0.050
  26, 19,  8 <- 25, 18,  7       213700.903     0.039     0.050
  27, 19,  8 <- 26, 18,  9       216476.458    -0.042     0.050
  34, 16, 19 <- 33, 15, 18       212822.292     0.018     0.050
  35, 16, 20 <- 34, 15, 19       215564.786     0.049     0.050
  37, 15, 23 <- 36, 14, 22       213272.636     0.038     0.050
  38, 15, 23 <- 37, 14, 24       215980.952    -0.075     0.050          1.0000
  38, 15, 24 <- 37, 14, 23       215980.952    -0.075     0.050          1.0000
  38, 20, 19 <- 37, 19, 18       254585.425     0.001     0.050
  41, 14, 28 <- 40, 13, 27       216119.250    -0.043     0.050
  41, 28, 14 <- 40, 27, 13       324135.089     0.012     0.050
  49, 12, 38 <- 48, 11, 37       216384.396     0.011     0.050
  52, 24, 28 <- 51, 23, 29       323849.506     0.020     0.050
  67, 19, 48 <- 66, 18, 49       323904.122     0.049     0.050
  69, 13, 57 <- 68, 12, 56       214064.268     0.005     0.050
  77, 12, 65 <- 76, 13, 64       216030.211     0.082     0.050
  79, 10, 69 <- 78, 11, 68       216310.536    -0.029     0.050
```

Table S3. The observed and the obs.-calc. frequencies (MHz) of measured rotational transitions in the ground state of the parent isotopomer of anisole.

Anisole, ground state

Obs-calc differences are from fit of Watson's reduced Hamiltonian in ---> reduction-A, representation-I.r <---

TRANSITION	Observed	Obs-Calc	Error	Note	wt for blend
!					
! FTMW, IFPAN					
!					
! a-type					
!					
3, 1, 3 <- 2, 1, 2	7763.867	0.000	0.002		
3, 0, 3 <- 2, 0, 2	8217.876	0.000	0.002		
3, 1, 2 <- 2, 1, 1	8853.205	0.001	0.002		
!					
4, 1, 4 <- 3, 1, 3	10322.950	0.000	0.002		
4, 0, 4 <- 3, 0, 3	10837.722	0.000	0.002		
4, 2, 3 <- 3, 2, 2	11079.582	0.000	0.002		
4, 2, 2 <- 3, 2, 1	11342.554	0.000	0.002		
4, 1, 3 <- 3, 1, 2	11769.481	0.000	0.002		
!					
5, 1, 5 <- 4, 1, 4	12860.811	-0.001	0.002		
5, 0, 5 <- 4, 0, 4	13373.038	0.000	0.002		
5, 1, 4 <- 4, 1, 3	14652.044	0.000	0.002		
!					
6, 0, 6 <- 5, 0, 5	15831.915	-0.001	0.002		
!					
! b-type					
!					
3, 1, 3 <- 2, 0, 2	10886.979	0.000	0.002		
4, 0, 4 <- 3, 1, 3	8168.619	-0.001	0.002		
5, 0, 5 <- 4, 1, 4	11218.708	0.000	0.002		
6, 1, 6 <- 5, 0, 5	17018.206	-0.001	0.002		
!					
4, 2, 2 <- 4, 1, 3	9558.716	-0.001	0.002		
5, 2, 3 <- 5, 1, 4	9224.728	0.000	0.002		
5, 2, 4 <- 5, 1, 5	13742.126	-0.001	0.002		
6, 2, 4 <- 6, 1, 5	9083.646	-0.001	0.002		
7, 2, 5 <- 7, 1, 6	9221.867	0.000	0.002		
!					
! MMW IFPAN					
!					
! a+b-type, type-II bandheads					
!					
70, 0, 70 <- 69, 0, 69	169954.686	0.008	0.050		
69, 1, 68 <- 68, 1, 67	169901.865	-0.010	0.050		
68, 2, 66 <- 67, 2, 65	169852.263	0.025	0.050		
67, 3, 64 <- 66, 3, 63	169809.146	-0.005	0.050		
66, 4, 62 <- 65, 4, 61	169777.664	-0.012	0.050		
65, 5, 60 <- 64, 5, 59	169765.636	0.016	0.050		
64, 6, 58 <- 63, 6, 57	169785.627	0.056	0.050		
63, 8, 56 <- 62, 8, 55	169858.654	-0.008	0.050		
63, 7, 56 <- 62, 7, 55	169859.511	0.021	0.050		
62, 8, 54 <- 61, 8, 53	170034.606	-0.086	0.050		
72, 0, 72 <- 71, 0, 71	174774.483	0.008	0.050		
71, 1, 70 <- 70, 1, 69	174721.567	-0.024	0.050		
70, 2, 68 <- 69, 2, 67	174671.680	0.023	0.050		

```
  69,  3, 66 <- 68,  3, 65       174627.777    -0.023     0.050
  68,  4, 64 <- 67,  4, 63       174594.617    -0.014     0.050
  67,  5, 62 <- 66,  5, 61       174579.160     0.006     0.050
  66,  6, 60 <- 65,  6, 59       174592.412    -0.013     0.050

  89,  0, 89 <- 88,  0, 88       215734.186    -0.026     0.050
  88,  1, 87 <- 87,  1, 86       215680.811    -0.016     0.050
  87,  2, 85 <- 86,  2, 84       215629.165    -0.008     0.050
  86,  3, 83 <- 85,  3, 82       215581.004    -0.004     0.050
  85,  4, 81 <- 84,  4, 80       215538.712    -0.007     0.050
  84,  5, 79 <- 83,  5, 78       215505.590    -0.001     0.050
  83,  6, 77 <- 82,  6, 76       215486.227    -0.009     0.050
  82,  7, 75 <- 81,  7, 74       215487.283     0.000     0.050
  81,  8, 73 <- 80,  8, 72       215518.552    -0.037     0.050
  80,  9, 71 <- 79,  9, 70       215595.478    -0.035     0.050

 105,  0,105 <-104,  0,104       254268.455    -0.019     0.050
 104,  1,103 <-103,  1,102       254214.774    -0.018     0.050
 103,  2,101 <-102,  2,100       254162.256     0.027     0.050
 102,  3, 99 <-101,  3, 98       254111.921    -0.016     0.050
 101,  4, 97 <-100,  4, 96       254065.436     0.037     0.050
 100,  5, 95 <- 99,  5, 94       254024.583     0.034     0.050
  99,  6, 93 <- 98,  6, 92       253991.939     0.011     0.050
  98,  7, 91 <- 97,  7, 90       253970.883    -0.044     0.050
  97,  8, 89 <- 96,  8, 88       253966.162     0.006     0.050
  95, 10, 85 <- 94, 10, 84       254033.695     0.002     0.050
  94, 11, 83 <- 93, 11, 82       254128.798    -0.016     0.050

 135,  0,135 <-134,  0,134       326468.196    -0.055     0.050
 134,  1,133 <-133,  1,132       326414.138    -0.042     0.050
 133,  2,131 <-132,  2,130       326360.628     0.000     0.050
 132,  3,129 <-131,  3,128       326308.194    -0.025     0.050
 131,  4,127 <-130,  4,126       326257.700    -0.012     0.050
 130,  5,125 <-129,  5,124       326209.990    -0.041     0.050
 129,  6,123 <-128,  6,122       326166.387     0.077     0.050
 128,  7,121 <-127,  7,120       326128.036     0.090     0.050
 126,  9,117 <-125,  9,116       326074.678    -0.015     0.050
 125, 10,115 <-124, 10,114       326064.696    -0.063     0.050
 124, 11,113 <-123, 11,112       326070.384    -0.060     0.050
 122, 13,109 <-121, 13,108       326148.791    -0.101     0.050
 121, 14,107 <-120, 14,106       326236.385    -0.007     0.050
 120, 15,105 <-119, 15,104       326370.836    -0.077     0.050
 119, 16,103 <-118, 16,102       326570.218     0.125     0.050

 136,  1,135 <-135,  1,134       331224.589    -0.127     0.050
 135,  2,133 <-134,  2,132       331171.200     0.086     0.050
 134,  3,131 <-133,  3,130       331118.734     0.126     0.050

 133,  5,128 <-132,  5,127       333424.860     0.067     0.050
 132,  6,126 <-131,  6,125       333380.417     0.011     0.050
 130,  8,122 <-129,  8,121       333308.333     0.029     0.050
 128, 10,118 <-127, 10,117       333271.318     0.049     0.050
 127, 11,116 <-126, 11,115       333272.737     0.012     0.050
 125, 13,112 <-124, 13,111       333336.851    -0.006     0.050
 124, 14,110 <-123, 14,109       333412.428    -0.043     0.050
!
! lines down the band split into a+b quartets
!
  79, 10, 69 <- 78, 11, 68       215740.852    -0.002     0.050
  79, 11, 69 <- 78, 11, 68       215742.841     0.002     0.050
  79, 10, 69 <- 78, 10, 68       215743.973     0.018     0.050
  79, 11, 69 <- 78, 10, 68       215745.938    -0.002     0.050

  78, 11, 67 <- 77, 12, 66       215967.680    -0.009     0.050
  78, 12, 67 <- 77, 12, 66       216000.420    -0.010     0.050
  78, 12, 67 <- 77, 11, 66       216050.028     0.008     0.050
!
```

```
! high K-1
!
  75, 15, 61 <- 74, 15, 60       216108.654     0.011     0.050
  75, 17, 59 <- 74, 17, 58       216030.534    -0.054     0.050
  76, 22, 55 <- 75, 22, 54       215493.056     0.053     0.050
  77, 31, 46 <- 76, 31, 45       216101.785     0.016     0.050
  77, 32, 45 <- 76, 32, 44       215963.973     0.079     0.050
  77, 33, 44 <- 76, 33, 43       215838.877    -0.010     0.050
  77, 34, 43 <- 76, 34, 42       215725.148     0.015     0.050
  77, 35, 42 <- 76, 35, 41       215621.287     0.014     0.050
  77, 36, 41 <- 76, 36, 40       215526.167     0.014     0.050
  77, 37, 40 <- 76, 37, 39       215438.792     0.007     0.050
  77, 38, 39 <- 76, 38, 38       215358.323    -0.001     0.050

  93, 12, 81 <- 92, 13, 80       254289.947    -0.114     0.050
  93, 13, 81 <- 92, 13, 80       254290.702    -0.030     0.050           1.0000
  93, 12, 81 <- 92, 12, 80       254290.702    -0.030     0.050           1.0000
  93, 13, 81 <- 92, 12, 80       254291.408     0.006     0.050

  88, 19, 70 <- 87, 19, 69       254454.938     0.022     0.050
  90, 28, 62 <- 89, 28, 61       254369.840    -0.019     0.050
  90, 29, 61 <- 89, 29, 60       254049.445    -0.013     0.050

 111, 24, 87 <-110, 24, 86       323115.993     0.018     0.050
 113, 28, 85 <-112, 28, 84       323085.333    -0.052     0.050
 115, 43, 72 <-114, 43, 71       323063.597     0.002     0.050
!
!  b-type lines
!
  18, 17,  2 <- 17, 16,  1       170111.797     0.003     0.050
  24, 21,  4 <- 23, 20,  3       215894.662    -0.032     0.050
  30, 24,  6 <- 29, 23,  7       254400.054    -0.008     0.050
  35, 32,  4 <- 34, 31,  3       326447.160    -0.034     0.050
  37, 16, 22 <- 36, 15, 21       215397.088     0.039     0.050
  39, 30, 10 <- 38, 29,  9       323044.868    -0.020     0.050
  40, 15, 26 <- 39, 14, 25       216090.776     0.036     0.050
  40, 31, 10 <- 39, 30,  9       333092.675    -0.024     0.050
  46, 13, 34 <- 45, 12, 33       215806.558     0.011     0.050
  46, 18, 29 <- 45, 17, 28       254662.098     0.067     0.050
  48, 27, 22 <- 47, 26, 21       326204.159     0.036     0.050
  49, 11, 38 <- 48, 10, 39       215906.017     0.019     0.050
  55, 15, 41 <- 54, 14, 40       254294.898     0.083     0.050
  56, 24, 33 <- 55, 23, 32       326258.081    -0.022     0.050
  62,  8, 54 <- 61,  9, 53       169992.369     0.000     0.050
  62,  9, 54 <- 61,  8, 53       170062.194     0.028     0.050
  63,  7, 56 <- 62,  8, 55       169857.285     0.015     0.050
  63,  8, 56 <- 62,  7, 55       169860.901     0.020     0.050
  63, 13, 51 <- 62, 12, 50       215815.972     0.031     0.050
  71, 19, 53 <- 70, 18, 52       326501.902     0.104     0.050
  71, 19, 52 <- 70, 18, 53       326558.742     0.107     0.050
  88, 16, 73 <- 87, 15, 72       254157.481    -0.001     0.050
  92, 13, 79 <- 91, 14, 78       254544.686     0.037     0.050
!
!  bQ-type
!
 113, 11,102 <-113, 10,103       215374.271    -0.014     0.050

  45, 24, 21 <- 45, 23, 22       170176.630    -0.018     0.050
  46, 24, 22 <- 46, 23, 23       170107.187     0.005     0.050
  47, 24, 23 <- 47, 23, 25       170032.941    -0.062     0.050
  48, 24, 24 <- 48, 23, 25       169953.841    -0.047     0.050
  49, 24, 25 <- 49, 23, 26       169869.631     0.022     0.050
  50, 24, 26 <- 50, 23, 27       169779.931     0.001     0.050

  92, 31, 61 <- 92, 30, 62       215670.835    -0.039     0.050
  93, 31, 62 <- 93, 30, 63       215387.405     0.027     0.050
```

113, 32, 81 <-113, 31, 82	215766.361	-0.057	0.050
91, 36, 55 <- 91, 35, 56	254485.704	-0.027	0.050
92, 36, 56 <- 92, 35, 57	254319.917	0.000	0.050
56, 45, 11 <- 56, 44, 12	323218.329	0.009	0.050
57, 45, 12 <- 57, 44, 13	323198.623	0.045	0.050
58, 45, 13 <- 58, 44, 14	323177.785	-0.014	0.050
59, 45, 14 <- 59, 44, 15	323155.950	0.001	0.050
60, 45, 15 <- 60, 44, 16	323133.014	0.024	0.050
61, 45, 16 <- 61, 44, 17	323108.897	0.013	0.050
63, 45, 18 <- 63, 44, 19	323057.050	-0.028	0.050
64, 45, 19 <- 64, 44, 20	323029.304	0.004	0.050

Table S4. The observed and the obs.-calc. frequencies (MHz) of measured rotational transitions in the first excited torsional state of anisole.

Anisole, v=1

Obs-calc differences are from fit of Watson's reduced Hamiltonian in ---> reduction-A, representation-I.r <---

TRANSITION	Observed	Obs-Calc	Error	Note	wt for blend
!					
! MMW IFPAN					
!					
! a+b-type, type-II bandheads					
!					
70, 0, 70 <- 69, 0, 69	170171.759	-0.019	0.050		
69, 1, 68 <- 68, 1, 67	170106.111	-0.007	0.050		
68, 2, 66 <- 67, 2, 65	170043.641	0.023	0.050		
67, 3, 64 <- 66, 3, 63	169987.644	-0.005	0.050		
66, 4, 62 <- 65, 4, 61	169943.260	0.002	0.050		
65, 5, 60 <- 64, 5, 59	169918.243	0.018	0.050		
64, 7, 58 <- 63, 7, 57	169925.086	0.001	0.050		1.0000
64, 6, 58 <- 63, 6, 57	169925.086	0.001	0.050		1.0000
72, 0, 72 <- 71, 0, 71	174997.829	-0.013	0.050		
71, 1, 70 <- 70, 1, 69	174932.079	-0.022	0.050		
70, 2, 68 <- 69, 2, 67	174869.336	0.031	0.050		
69, 3, 66 <- 68, 3, 65	174812.562	-0.007	0.050		
68, 4, 64 <- 67, 4, 63	174766.499	0.009	0.050		
67, 5, 62 <- 66, 5, 61	174738.167	0.009	0.050		1.0000
66, 6, 60 <- 65, 6, 59	174738.167	0.009	0.050		1.0000
89, 0, 89 <- 88, 0, 88	216010.545	-0.043	0.050		
88, 1, 87 <- 87, 1, 86	215944.258	-0.090	0.050		
87, 2, 85 <- 86, 2, 84	215879.819	-0.019	0.050		
86, 3, 83 <- 85, 3, 82	215818.805	-0.006	0.050		
85, 4, 81 <- 84, 4, 80	215763.628	-0.017	0.050		
84, 5, 79 <- 83, 5, 78	215717.603	-0.013	0.050		
83, 6, 77 <- 82, 6, 76	215685.316	-0.002	0.050		
82, 7, 75 <- 81, 7, 74	215673.376	0.015	0.050		
81, 8, 73 <- 80, 8, 72	215691.584	0.011	0.050		
80, 9, 71 <- 79, 9, 70	215755.201	-0.040	0.050		1.0000
80, 10, 71 <- 79, 10, 70	215755.201	-0.040	0.050		1.0000
105, 0,105 <-104, 0,104	254594.229	-0.009	0.050		
104, 1,103 <-103, 1,102	254527.695	-0.007	0.050		
102, 3, 99 <-101, 3, 98	254399.090	-0.052	0.050		
101, 4, 97 <-100, 4, 96	254339.749	0.003	0.050		
100, 5, 95 <- 99, 5, 94	254286.012	-0.011	0.050		
99, 6, 93 <- 98, 6, 92	254240.558	0.051	0.050		
98, 7, 91 <- 97, 7, 90	254206.615	0.035	0.050		
97, 8, 89 <- 96, 8, 88	254188.805	-0.030	0.050		
96, 9, 87 <- 95, 9, 86	254193.636	-0.024	0.050		
95, 10, 85 <- 94, 10, 84	254230.183	-0.016	0.050		
94, 11, 83 <- 93, 11, 82	254312.100	0.043	0.050		
130, 5,125 <-129, 5,124	326562.584	0.032	0.050		
129, 6,123 <-128, 6,122	326506.018	0.036	0.050		
127, 8,119 <-126, 8,118	326410.536	-0.083	0.050		
126, 9,117 <-125, 9,116	326375.745	0.010	0.050		
125, 10,115 <-124, 10,114	326352.837	-0.033	0.050		

```
 124, 11,113 <-123, 11,112       326345.618     0.037     0.050
 123, 12,111 <-122, 12,110       326358.501    -0.019     0.050
 122, 13,109 <-121, 13,108       326397.906     0.012     0.050
 121, 14,107 <-120, 14,106       326472.155    -0.036     0.050
 120, 15,105 <-119, 15,104       326593.348    -0.035     0.050

 127, 10,117 <-126, 10,116       331163.321     0.101     0.050

!
! lines down the band split into a+b quartets
!
  63,  7, 56 <- 62,  8, 55       169983.609     0.047     0.050
  63,  8, 56 <- 62,  8, 55       169985.019     0.059     0.050
  63,  7, 56 <- 62,  7, 55       169985.835     0.045     0.050

  62,  8, 54 <- 61,  9, 53       170105.033    -0.029     0.050
  62,  9, 54 <- 61,  9, 53       170132.639     0.008     0.050
  62,  9, 54 <- 61,  8, 53       170175.087    -0.002     0.050

  65,  7, 58 <- 64,  8, 57       174784.764    -0.062     0.050
  65,  8, 58 <- 64,  8, 57       174785.506    -0.029     0.050           1.0000
  65,  7, 58 <- 64,  7, 57       174785.506    -0.029     0.050           1.0000
  65,  8, 58 <- 64,  7, 57       174786.280     0.036     0.050

  64,  9, 56 <- 63,  9, 55       174910.286     0.031     0.050
  64,  8, 56 <- 63,  8, 55       174916.562    -0.049     0.050
  64,  9, 56 <- 63,  8, 55       174928.050    -0.018     0.050

  79, 10, 69 <- 78, 10, 68       215890.320     0.029     0.050
  79, 11, 69 <- 78, 10, 68       215892.303     0.016     0.050

  78, 11, 67 <- 77, 12, 66       216100.093    -0.013     0.050

  93, 13, 81 <- 92, 12, 80       254461.283     0.076     0.050
!
! high K-1
!
  59, 10, 49 <- 58, 10, 48       169968.097     0.019     0.050
  59, 11, 48 <- 58, 11, 47       174346.770    -0.007     0.050
  59, 12, 47 <- 58, 12, 46       175092.421    -0.003     0.050
  59, 13, 47 <- 58, 13, 46       169899.447     0.031     0.050
  60, 18, 42 <- 59, 18, 41       169981.341     0.065     0.050           1.0000
  60, 18, 43 <- 59, 18, 42       169981.341     0.065     0.050           1.0000
  60, 19, 42 <- 59, 19, 41       169627.590     0.005     0.050
  61, 10, 51 <- 60, 11, 50       170088.968    -0.036     0.050
  61, 15, 47 <- 60, 15, 46       174700.256     0.000     0.050
  62, 21, 41 <- 61, 21, 40       174887.281    -0.042     0.050
  62, 23, 39 <- 61, 23, 38       174450.638    -0.014     0.050

  75, 17, 59 <- 74, 17, 58       216109.793     0.043     0.050
  76, 21, 55 <- 75, 21, 54       216076.718     0.000     0.050           1.0000
  76, 21, 56 <- 75, 21, 55       216076.718     0.000     0.050           1.0000
  76, 22, 55 <- 75, 22, 54       215589.740    -0.006     0.050
  77, 32, 45 <- 76, 32, 44       216073.708    -0.024     0.050
  77, 33, 44 <- 76, 33, 43       215949.414     0.069     0.050
  77, 34, 43 <- 76, 34, 42       215836.151    -0.006     0.050
  77, 35, 42 <- 76, 35, 41       215732.813     0.001     0.050
  77, 36, 41 <- 76, 36, 40       215638.160    -0.003     0.050

  90, 29, 61 <- 89, 29, 60       254168.513    -0.014     0.050
  91, 47, 44 <- 90, 47, 43       254438.265    -0.055     0.050

 115, 32, 83 <-114, 32, 82       326587.476    -0.013     0.050
!
!  b-type lines
!
  18, 17,  2 <- 17, 16,  1       169681.247    -0.044     0.050
```

```
  24, 21,  3 <- 23, 20,  4       215362.396    -0.039     0.050
  26, 14, 13 <- 25, 13, 12       170160.174    -0.067     0.050
  32, 18, 15 <- 31, 17, 14       215851.945    -0.025     0.050
  33, 12, 21 <- 32, 11, 22       174529.085     0.002     0.050
  33, 12, 22 <- 32, 11, 21       174527.736     0.021     0.050
  36, 11, 25 <- 35, 10, 26       174669.434     0.012     0.050
  36, 11, 26 <- 35, 10, 25       174543.645     0.053     0.050
  40, 15, 26 <- 39, 14, 25       215753.428    -0.014     0.050
  43, 14, 29 <- 42, 13, 30       216105.631    -0.026     0.050
  43, 14, 30 <- 42, 13, 29       216102.310    -0.008     0.050
  43, 29, 14 <- 42, 28, 15       326169.318     0.069     0.050
  46, 13, 34 <- 45, 12, 33       215546.976    -0.046     0.050
  46, 18, 29 <- 45, 17, 28       254250.276    -0.028     0.050
  49, 12, 37 <- 48, 11, 38       216013.323    -0.011     0.050
  51, 26, 25 <- 50, 25, 26       326570.215     0.068     0.050
  55, 15, 40 <- 54, 14, 41       254190.038    -0.026     0.050
  58, 24, 34 <- 57, 23, 35       331147.555    -0.039     0.050
  59, 23, 37 <- 58, 22, 36       326473.618     0.011     0.050
  63,  9, 54 <- 62, 10, 53       174967.567    -0.003     0.050
  71, 19, 53 <- 70, 18, 52       326126.040     0.093     0.050
  75, 18, 57 <- 74, 17, 58       326556.966     0.059     0.050
  77, 12, 65 <- 76, 13, 64       216095.013    -0.005     0.050
  77, 18, 60 <- 76, 17, 59       326367.166    -0.008     0.050
  71, 19, 52 <- 70, 18, 53       326182.064    -0.004     0.050
  80, 18, 63 <- 79, 17, 62       326438.420    -0.014     0.050
!
!  bQ-type
!
  29, 24,  5 <- 29, 23,  6       170150.881     0.026     0.050
  30, 24,  6 <- 30, 23,  7       170131.820     0.039     0.050
  32, 24,  8 <- 32, 23,  9       170087.559    -0.050     0.050
  35, 24, 11 <- 35, 23, 12       170004.223    -0.012     0.050
  36, 24, 12 <- 36, 23, 13       169971.292    -0.008     0.050
  37, 24, 13 <- 37, 23, 14       169935.484    -0.051     0.050
  38, 24, 14 <- 38, 23, 15       169896.711    -0.071     0.050
  39, 24, 15 <- 39, 23, 16       169854.884     0.012     0.050
  42, 24, 18 <- 42, 23, 19       169708.415    -0.049     0.050
  43, 24, 19 <- 43, 23, 20       169652.176     0.017     0.050

  63, 25, 38 <- 63, 24, 39       174958.013    -0.002     0.050
  64, 25, 39 <- 64, 24, 40       174788.342    -0.010     0.050
  65, 25, 40 <- 65, 24, 41       174609.960    -0.054     0.050
  66, 25, 41 <- 66, 24, 42       174422.690     0.033     0.050
  82, 25, 57 <- 82, 24, 58       169847.329     0.007     0.050

  90, 26, 64 <- 90, 25, 65       174713.892    -0.049     0.050          1.0000
  90, 26, 65 <- 90, 25, 66       174713.892    -0.049     0.050          1.0000

  87, 31, 56 <- 87, 30, 57       216117.383     0.048     0.050
  88, 31, 57 <- 88, 30, 58       215883.212    -0.008     0.050
  89, 31, 58 <- 89, 30, 59       215640.074    -0.046     0.050

  84, 36, 48 <- 84, 35, 49       254522.973     0.087     0.050
  85, 36, 49 <- 85, 35, 50       254394.403     0.061     0.050
  86, 36, 50 <- 86, 35, 51       254261.049     0.041     0.050
  87, 36, 51 <- 87, 35, 52       254122.765     0.015     0.050
  88, 36, 52 <- 88, 35, 53       253979.510     0.078     0.050

 104, 46, 58 <-104, 45, 59       326544.035    -0.081     0.050
 105, 46, 59 <-105, 45, 60       326429.362    -0.038     0.050

 105,  7, 98 <-105,  6, 99       216070.774    -0.029     0.050
```

Table S5. The observed and the obs.-calc. frequencies (MHz) of measured rotational transitions in the ground states of singly substituted 13C and the 18O isotopomer of benzaldehyde.

	13C.1		13C.2		13C.3	
4 1 4 <- 3 1 3	10207.0399	0.0002	10302.8168	0.0000	10287.6934	-0.0006
4 0 4 <- 3 0 3	10722.8232	0.0011	10824.3830	0.0001	10809.7067	-0.0017
4 2 2 <- 3 2 1	11175.3236	-0.0012				
4 1 3 <- 3 1 2	11612.8160	0.0009	11733.3779	0.0000	11732.7160	0.0009
5 1 5 <- 4 1 4	12720.0129	0.0007	12838.4319	0.0000	12818.2265	0.0007
5 0 5 <- 4 0 4	13244.6188	-0.0016	13366.5205	-0.0001	13343.2909	0.0009
5 1 4 <- 4 1 3	14463.3376	0.0002	14611.9694	0.0000	14608.9044	-0.0005

	13C.4		13C.5		13C.6	
4 1 4 <- 3 1 3	10229.3531	0.0005	10274.5385	-0.0012	10177.0198	-0.0014
4 0 4 <- 3 0 3	10748.4216	-0.0020	10796.2694	0.0009	10690.6294	0.0003
4 2 2 <- 3 2 1	11231.0619	-0.0001				
4 1 3 <- 3 1 2	11666.3871	0.0003	11723.4585	0.0006	11571.1254	-0.0003
5 1 5 <- 4 1 4	12745.5170	0.0000	12801.3579	0.0008	12683.2066	0.0012
5 0 5 <- 4 0 4	13267.5796	0.0010	13324.9137	-0.0004	13207.1440	-0.0003
5 1 4 <- 4 1 3	14526.2893	0.0002	14596.5836	-0.0006	14412.3860	0.0003

	13C.7		18O	
4 1 4 <- 3 1 3	10206.7155	-0.0002	9913.0113	-0.0002
4 0 4 <- 3 0 3	10723.5845	0.0018	10409.5806	0.0001
4 2 3 <- 3 2 2			10599.5062	0.0009
4 2 2 <- 3 2 1	11189.9086	-0.0001	10805.8645	0.0015
4 1 3 <- 3 1 2			11234.1555	-0.0020
5 1 5 <- 4 1 4	12718.5137	0.0000	12357.0344	-0.0015
5 0 5 <- 4 0 4	13241.4216	-0.0009	12870.9060	0.0013
5 1 4 <- 4 1 3	14477.9552	-0.0002		

Table S6. The observed and the obs.-calc. frequencies (MHz) of measured rotational transitions in the ground states of singly substituted 13C and the 18O isotopomer of anisole.

	13C.1		13C.2		13C.3	
4 0 4 <- 3 0 3	10830.7806	0.0004	10815.9382	-0.0009	10754.6579	-0.0005
3 1 3 <- 2 0 2	10880.6805	-0.0007	10811.3795	0.0000	10748.1459	0.0008
5 0 5 <- 4 1 4	11210.8645	0.0010	11248.6423	0.0009	11186.8848	0.0000
4 1 4 <- 3 0 3	12984.4481	0.0013	12908.4415	0.0006	12833.0858	-0.0013
6 0 6 <- 5 1 5	14180.1652	-0.0011	14205.4110	-0.0004	14126.5658	0.0003
5 1 5 <- 4 0 4	15006.2622	-0.0006	14925.1076	-0.0002	14838.1446	0.0007

	13C.4		13C.5		13C.6	
4 1 4 <- 3 1 3	10188.3256	-0.0006				
4 0 4 <- 3 0 3	10695.2972	-0.0009	10728.8312	0.0013	10801.2425	-0.0014
3 1 3 <- 2 0 2	10813.1407	0.0001	10777.7742	-0.0001	10777.4340	-0.0004
5 0 5 <- 4 1 4	11003.5150	-0.0007	11105.8849	0.0013	11252.1420	0.0011
4 1 4 <- 3 0 3			12861.5481	-0.0004	12870.2747	0.0018
6 0 6 <- 5 1 5	13945.8011	0.0013	14047.1295	-0.0018	14201.8051	-0.0004
5 1 5 <- 4 0 4	14894.6293	0.0002	14864.1549	0.0003	14883.3620	-0.0007

	13C.7		18O	
4 1 4 <- 3 1 3			10145.5483	-0.0009
4 0 4 <- 3 0 3	10626.0223	0.0023	10650.6656	0.0017
3 1 3 <- 2 0 2	10775.2314	-0.0006	10755.9582	-0.0009
5 0 5 <- 4 1 4	10900.7071	-0.0011	10969.4554	-0.0001
4 1 4 <- 3 0 3	12845.9398	-0.0004	12828.4180	0.0005
6 0 6 <- 5 1 5	13828.4912	0.0000	13897.7040	-0.0003
5 1 5 <- 4 0 4	14833.8478	-0.0001		

Table S7. The observed and the obs.-calc. frequencies (MHz) of measured Stark components of rotational transitions in the parent isopomer of benzaldehyde.

TRANSITIONS (F and MF in units of 1/2):

	J	K -1	K +1	<- J	K -1	K +1	F	MF	<- F	MF	Volts	Obs	Obs-Calc
1.	4	1	4	3	1	3	8	0	6	0	0.0	10309.47560	-0.00023
2.	4	1	4	3	1	3	8	0	6	0	4197.6	10309.33200	-0.00157
3.	4	1	4	3	1	3	8	0	6	0	5930.8	10309.18950	-0.00287
4.	4	1	4	3	1	3	8	0	6	0	7268.8	10309.04750	-0.00335
5.	4	1	4	3	1	3	8	0	6	0	8394.6	10308.90570	-0.00439
6.	4	1	4	3	1	3	8	0	6	0	9384.7	10308.76800	-0.00211
!													
7.	4	1	4	3	1	3	8	4	6	4	4197.6	10310.13300	0.00076
8.	4	1	4	3	1	3	8	4	6	4	5930.8	10310.78780	0.00208
9.	4	1	4	3	1	3	8	4	6	4	7268.8	10311.44290	0.00024
10.	4	1	4	3	1	3	8	4	6	4	8394.6	10312.09680	-0.00129
11.	4	1	4	3	1	3	8	4	6	4	9384.7	10312.74890	-0.00299
!													
12.	4	0	4	3	0	3	8	0	6	0	0.0	10831.40350	0.00019
13.	4	0	4	3	0	3	8	0	6	0	4197.6	10831.18880	-0.00197
14.	4	0	4	3	0	3	8	0	6	0	5930.8	10830.97700	-0.00258
15.	4	0	4	3	0	3	8	0	6	0	7268.8	10830.76360	-0.00409
16.	4	0	4	3	0	3	8	0	6	0	8394.8	10830.55690	0.00024
17.	4	0	4	3	0	3	8	0	6	0	9384.7	10830.34690	0.00026
18.	4	0	4	3	0	3	8	0	6	0	10279.1	10830.13640	-0.00095
!													
19.	4	0	4	3	0	3	8	4	6	4	4197.6	10831.63840	0.00150
20.	4	0	4	3	0	3	8	4	6	4	5930.8	10831.87030	0.00060
21.	4	0	4	3	0	3	8	4	6	4	7268.8	10832.10430	0.00030
22.	4	0	4	3	0	3	8	4	6	4	8394.8	10832.33730	-0.00075
23.	4	0	4	3	0	3	8	4	6	4	9384.7	10832.57090	-0.00079
24.	4	0	4	3	0	3	8	4	6	4	10279.1	10832.80320	-0.00204
!													
25.	4	0	4	3	0	3	8	6	6	6	4197.6	10832.19430	-0.00057
26.	4	0	4	3	0	3	8	6	6	6	5930.8	10832.98290	-0.00065
27.	4	0	4	3	0	3	8	6	6	6	7268.8	10833.77550	-0.00158
28.	4	0	4	3	0	3	8	6	6	6	8394.8	10834.56770	-0.00190
29.	4	0	4	3	0	3	8	6	6	6	9384.7	10835.36130	0.00081
!													
30.	4	1	4	3	0	3	8	0	6	0	0.0	13190.32100	0.00041
31.	4	1	4	3	0	3	8	2	6	2	4197.6	13190.07000	-0.00212
32.	4	1	4	3	0	3	8	2	6	2	5930.8	13189.82320	-0.00177
33.	4	1	4	3	0	3	8	2	6	2	7268.8	13189.57380	-0.00292
34.	4	1	4	3	0	3	8	2	6	2	8394.6	13189.32850	-0.00076
35.	4	1	4	3	0	3	8	2	6	2	9384.7	13189.08070	-0.00192
36.	4	1	4	3	0	3	8	2	6	2	10279.1	13188.83440	-0.00221

Standard deviation = 0.001961

FINAL RESULTS OF LEAST SQUARES FITTING PROCEDURE

FITTED CONSTANTS:

A	/MHz	5234.364013047011212	1:Xab /MHz	0.
B	/MHz	1564.274370862518026	1:XJ.a/kHz	0.
C	/MHz	1204.681906530923015	1:XK.a/kHz	0.
DJ	/kHz	0.068822722	1:XJbc/kHz	0.

DJK	/kHz	0.149368377	1:Ma	/MHz	0.
DK	/kHz	0.748944906000000	1:Mb-c	/MHz	0.
dJ	/kHz	0.017954735	1:Tr	/MHz	0.
dK	/kHz	0.289229385	1:Xd	/kHz	0.
HJ	/ Hz	0.			
HJK	/ Hz	0.			
HKJ	/ Hz	0.			
HK	/ Hz	0.			
hJ	/ Hz	0.			
hJK	/ Hz	0.	Mu.a	/D	2.90611(56)
hK	/ Hz	0.	Mu.b	/D	1.18827(47)
LKKJ	/mHz	0.	Mu.c	/D	0.
1:Xa	/MHz	0.	d	/cm	26.930
1:Xb-c	/MHz	0.	k	/cm	0.

CORRELATION COEFFICIENTS:

	Mu.a	Mu.b
Mu.a	1.0000	
Mu.b	-0.6082	1.0000

Table S8. The observed and the obs.-calc. frequencies (MHz) of measured Stark components of rotational transitions in the parent isopomer of anisole.

TRANSITIONS (F and MF in units of 1/2):

	J	K -1	K +1	<- J	K -1	K +1	F	MF	<- F	MF	Volts	Obs	Obs-Calc
1.	3	1	3	2	0	2	6	0	4	0	0.0	10886.97940	-0.00012
2.	3	1	3	2	0	2	6	0	4	0	3778.5	10886.79810	-0.00112
3.	3	1	3	2	0	2	6	0	4	0	6544.6	10886.43760	-0.00121
4.	3	1	3	2	0	2	6	0	4	0	7560.2	10886.25800	-0.00010
5.	3	1	3	2	0	2	6	0	4	0	8452.8	10886.07740	-0.00045
6.	3	1	3	2	0	2	6	0	4	0	9259.7	10885.89680	-0.00089
7.	3	1	3	2	0	2	6	0	4	0	10000.9	10885.71850	0.00071
8.	3	1	3	2	0	2	6	0	4	0	11002.1	10885.45430	0.00139
!													
9.	3	1	3	2	0	2	6	4	4	4	3778.5	10887.38210	0.00030
10.	3	1	3	2	0	2	6	4	4	4	6544.6	10888.18710	0.00090
11.	3	1	3	2	0	2	6	4	4	4	7560.2	10888.58870	-0.00095
12.	3	1	3	2	0	2	6	4	4	4	8452.8	10888.99290	0.00076
13.	3	1	3	2	0	2	6	4	4	4	9259.7	10889.39460	0.00005
14.	3	1	3	2	0	2	6	4	4	4	10000.9	10889.79720	0.00076
15.	3	1	3	2	0	2	6	4	4	4	11002.1	10890.38680	-0.00151
!													
16.	4	0	4	3	0	3	8	0	6	0	0.0	10837.72240	-0.00023
17.	4	0	4	3	0	3	8	4	6	4	3778.5	10837.82330	0.00233
18.	4	0	4	3	0	3	8	4	6	4	6544.6	10838.01880	0.00116
19.	4	0	4	3	0	3	8	4	6	4	7560.2	10838.11690	0.00062
20.	4	0	4	3	0	3	8	4	6	4	8452.8	10838.21570	0.00099
21.	4	0	4	3	0	3	8	4	6	4	9259.7	10838.31360	0.00048
22.	4	0	4	3	0	3	8	4	6	4	10000.9	10838.41070	-0.00071
23.	4	0	4	3	0	3	8	4	6	4	11002.1	10838.55660	0.00042
!													
24.	4	0	4	3	0	3	8	6	6	6	3778.5	10837.97940	0.00058
25.	4	0	4	3	0	3	8	6	6	6	6544.6	10838.49080	-0.00031
26.	4	0	4	3	0	3	8	6	6	6	7560.2	10838.74900	0.00095
27.	4	0	4	3	0	3	8	6	6	6	8452.8	10839.00510	0.00072
28.	4	0	4	3	0	3	8	6	6	6	9259.7	10839.26010	-0.00055

Standard deviation = 0.000934

FINAL RESULTS OF LEAST SQUARES FITTING PROCEDURE

FITTED CONSTANTS:

A	/MHz	5028.844084915160237	1:Xab /MHz	0.
B	/MHz	1569.364393868730076	1:XJ.a/kHz	0.
C	/MHz	1205.825529159249981	1:XK.a/kHz	0.
DJ	/kHz	0.06150031	1:XJbc/kHz	0.
DJK	/kHz	0.03925811	1:Ma /MHz	0.
DK	/kHz	0.78470595	1:Mb-c/MHz	0.
dJ	/kHz	0.01648814	1:Tr /MHz	0.
dK	/kHz	0.1964627	1:Xd /kHz	0.
HJ	/ Hz	0.		
HJK	/ Hz	0.		
HKJ	/ Hz	0.		
HK	/ Hz	0.		
hJ	/ Hz	0.		
hJK	/ Hz	0.	Mu.a /D	0.6937(10)

hK	/ Hz	0.	Mu.b	/D	1.05470(10)
LKKJ	/mHz	0.	Mu.c	/D	0.
1:Xa	/MHz	0.	d	/cm	26.929999999999999
1:Xb-c	/MHz	0.	k	/cm	0.

CORRELATION COEFFICIENTS:

	Mu.a	Mu.b
Mu.a	1.0000	
Mu.b	-0.7075	1.0000

Table S9. Substitution coordinates (Angstr) for anisole and benzaldehyde.

ANISOLE	a	da	b	db	R
C1	0.4638	-0.0032	-0.2294	0.0065	0.5170(50)
C2	0.0819i		1.0831	0.0014	1.0799(24)
C3	-1.3379	0.0011	1.3431	0.0011	1.8960(14)
C4	-2.2649	0.0007	0.2997	0.0050	2.2846(11)
C5	-1.8055	-0.0008	-1.0162	0.0015	2.0718(13)
C6	-0.4347	-0.0035	-1.2908	0.0012	1.3623(19)
C7	2.7651	-0.0005	0.4143	0.0036	2.7959(9)
O	1.7898	0.0008	-0.6151	0.0024	1.8924(14)

BENZALDEHYDE	a	da	b	db	R
C1	0.4710	0.0032	-0.2172	0.0069	0.5181(50)
C2	0.0918i		1.0928	0.0014	1.0891(24)
C3	-1.3499	0.0011	1.3345	0.0011	1.8983(14)
C4	-2.2522	0.0007	0.2583	0.0058	2.2668(12)
C5	-1.7833	0.0008	-1.0494	0.0014	2.0692(13)
C6	-0.4004	0.0038	-1.2923	0.0012	1.3532(19)
C7	1.9416	0.0008	-0.5020	0.0030	2.0053(13)
O	2.8034	0.0005	0.3507	0.0043	2.8252(9)
D	2.2167	0.0007	-1.5784	0.0011	2.7197(10)