Skeletal Molecular Structure of Monocarbahexaborane from Microwave Spectral Studies

Gary L. McKown,[1a] Betsey P. Don,[1a] Robert A. Beaudet,* [1a] Phillip J. Vergamini,[1b] and Llewellyn H. Jones[1b]

Contribution from the Department of Chemistry, University of Southern California, University Park, Los Angeles, California 90007, and the Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico 87544. *Received February 13, 1976*

Abstract: The microwave spectra of ten isotopic species of CB_5H_7 have been assigned. The skeletal molecular structure is a distorted octahedron with one face containing three long B–B bonds. There is further evidence that the bridge hydrogen is located above this face. The bond distances are: r(C–B(2)) = 1.60 ± 0.01; r(C–B(5)) = 1.63 ± 0.01; r(B(2)–B(3)) = 1.87 ± 0.01; r(B(2)–B(6)) = 1.89 ± 0.01; r(B(4)–B(5)) = 1.72 ± 0.01; r(B(5)–B(6)) = 1.70 ± 0.01; r(B(2)–B(5)) = 1.70 ± 0.01 Å. The dipole moment was measured to be 1.43 ± 0.04 D.

The structure of the carborane CB_5H_7 is of interest because it represents the first one-carbon carborane to be synthesized and it is the only known closo carborane with a bridge hydrogen. Onak, Drake, and Dunks[2] initially prepared the compound from 1-methylpentaborane by using a silent electrical discharge. From the ^{11}B and ^{1}H NMR and the infrared analyses, they inferred that the molecule is a distorted octahedron. Prince and Schaeffer[3] showed chemically the presence of a single bridge hydrogen and substantiated the octahedral form.

The temperature-dependence study of the ^{11}B NMR spectrum showed that the 2,3-borons and 4,5-borons are different equivalent sets at −30 °C, but their peaks coalesce at higher temperatures into one B–H terminal doublet.[4] Hence the molecule has a plane of symmetry which bisects the B(2)–B(3) and the B(4)–B(5) bonds and which contains both the B(6) and C atoms.

However, CB_5H_7 is unique insofar as the bridge hydrogen has no obvious location as in a nido or arachno carborane, where an open face can easily accept it. The position of the bridge hydrogen is unconfirmed spectroscopically and it could conceivably be located (a) in an equatorial position or (b) above an octahedral face. Since four-centered hydrogen bonds have been found in metal cluster compounds, choice b is not to be ruled out. It is possible also to explain the NMR evidence in terms of tautomerization of the bridge hydrogen between boron 2, 3, 4, and 5. Since the structure will be "frozen" on a gigahertz (GHz) time scale, the microwave structural determination can differentiate unambiguously between the possible configurations.

The microwave spectrum of the isomers present in the normally abundant CB_5H_7 and in a ^{13}C enriched sample were obtained. It has not been possible to prepare the bridge deuterated isotopic species, but the skeletal structure of all the heavy atoms has been determined from the data. All the ambiguities in the signs of the atom coordinates were resolved by using the data from both the singly and doubly substituted isotopic species. Our results confirm that CB_5H_7 has an octahedral shape with an enlarged trigonal B–B–B face. These results are interpreted as a strong indication that the bridge hydrogen lies above a face and is involved in four-center bonding. The dipole moment was determined to be 1.43 D. A preliminary report of this work has been published.[5]

Experimental Section

The samples of both the normal and ^{13}C enriched CB_5H_7 were provided by T. Onak and were used without further purification except for the removal of some noncondensable gas, which presumably was hydrogen. The samples were stored in Pyrex tubes at liquid nitrogen temperature, where there was no apparent decomposition. However, the compound was very hygroscopic and underwent slow decomposition in the copper wave guide. At 0 °C the decomposition rate was intolerable, while at dry ice temperature the vapor pressure was negligible. The difficulty was alleviated by using finely ground dry ice and avoiding direct contact of the coolant with the wave guide. The microwave spectrum was observed in the region 8–38 GHz using a conventional 100-kHz Stark-modulated spectrometer.

Results

Data and Analysis. Since naturally abundant boron contains ^{11}B and ^{10}B in a 4:1 ratio, many isotopically substituted isomers produce observable spectra in CB_5H_7. Assuming that the molecule contains a mirror plane, B(2) and B(3) would be equivalent; likewise B(4) and B(5). Then the intensity of lines originating from the normal, 2-^{10}B, 4-^{10}B, and 6-^{10}B species should be in the ratio 4:2:2:1. Furthermore, the disubstituted species 2,6-^{10}B, 4,6-^{10}B, and 2,4-^{10}B are expected to have absorption intensities one-eighth that of the parent species.

Intense high J Q-branch lines with spacings of 5–10 MHz were present in nearly all spectral regions. By using low Stark modulation voltages, the R-branch lines were identified from their Stark effects. Only b type lines were observed.

Three sets of strong lines with approximate intensity ratios of 4:2:2 were first observed and assigned to the parent, 2-^{10}B, and 4-^{10}B species, respectively. Predictions then led to identification of weaker 6-^{10}B species lines, which were about one-quarter as intense as those of the parent species. This distribution is consistent with both the NMR data and the assumed molecular symmetry.

Successive isotopic forms could be found with greater ease as rotational constants were progressively more accurately predicted. In this manner absorption lines due to the 2,6-^{10}B and 4,6-^{10}B species were identified. The 2,4-^{10}B species could not be assigned because the transitions were weak and obscured by nearby stronger lines. Assignment of a ^{13}C-enriched sample proceeded in like manner, although transitions of doubly substituted ^{10}B species were too weak to be observed. The assigned transitions are listed in Table I and the rotational constants and the moments of inertia in Table II.

Stark Effect and Dipole Moment. Dipole moment measurements were hampered by first-order contributions to the Stark effect of many lines and by the dense spectrum. For example, the Stark effect of the 3_{03}–2_{12} line is approximately first order up to fields of several hundred volts per centimeter, at which point interference from the 3_{13}–2_{02} line is observed. Stark measurements on some transitions in uncluttered regions with second-order character are shown in Table III. The small

Table I. Observed Rotational Transition Frequencies (MHz) of CB_5H_7[a]

Transitions	All ^{11}B	2-^{10}B	4-^{10}B	6-^{10}B	2,6-^{10}B	4,6-^{10}B
			Normal CB_5H_7			
$2_{20} \leftarrow 1_{11}$	23 042.37 (−0.30)	23 257.36 (+0.22)	23 259.02 (+0.02)	23 442.3 (+0.3)	*b*	*b*
$2_{02} \leftarrow 1_{11}$	21 603.88 (+0.02)	21 872.23 (+0.07)	21 844.29 (−0.18)	21 753.8 (0.0)	*b*	22 012.60 (+0.18)
$2_{12} \leftarrow 1_{01}$	21 686.87 (+0.16)	21 996.12 (+0.05)	21 960.07 (−0.03)	21 886.2 (+0.2)	*b*	*b*
$2_{21} \leftarrow 1_{10}$	22 467.27 (+0.07)	22 767.50 (+0.01)	22 743.40 (+0.33)	22 818.5 (+0.88)	23 113.48 (+0.88)	*b*
$3_{21} \leftarrow 2_{12}$	34 922.95 (−0.13)					
$3_{30} \leftarrow 2_{21}$	34 368.65 (+0.04)	34 715.40 (0.0)	34 709.05 (0.0)	34 947.18 (+0.02)	35 288.42 (+0.05)	35 276.69 (+0.0)
$3_{03} \leftarrow 2_{12}$	32 291.94 (−0.01)	32 734.70 (0.0)	32 682.11 (0.0)	32 525.65 (0.0)	32 995.85 (−0.01)	32 952.01 (+0.0)
$3_{12} \leftarrow 2_{21}$	32 846.10 (−0.04)	33 162.53 (0.0)	33 145.36 (0.0)	33 092.84 (0.0)	33 428.11 (−0.04)	33 393.80 (+0.0)
$3_{31} \leftarrow 2_{20}$	33 927.42 (+0.20)	34 385.04 (−0.04)	34 351.13 (0.0)	34 507.84 (+0.29)	34 949.53 (+0.40)	34 931.53 (−0.40)
$3_{12} \leftarrow 2_{02}$	32 303.99 (−0.05)	32 760.50 (+0.03)	32 704.65 (−0.06)	32 550.53 (−0.08)	33 036.50 (−0.21)	32 992.30 (+0.11)
$3_{22} \leftarrow 2_{11}$	32 115.70 (+0.08)	33 572.68 (−0.08)	33 527.68 (+0.14)	33 529.12 (+0.17)	33 992.88 (+0.23)	33 962.12 (−0.35)
$3_{31} \leftarrow 2_{02}$	35 365.73 (+0.06)					
			^{13}C Enriched CB_5H_7			
$2_{20} \leftarrow 1_{11}$	22 783.65 (−0.05)	23 001.96 (−0.09)	23 008.96 (+0.30)	23 146.83 (+0.59)		
$2_{02} \leftarrow 1_{11}$	21 493.22 (+0.17)	21 740.36 (−0.08)	21 716.23 (−0.37)	21 657.38 (+0.85)		
$2_{12} \leftarrow 1_{01}$	21 540.29 (+0.24)	21 834.60 (−0.16)	21 801.86 (+0.82)	21 766.61 (+0.15)		
$2_{21} \leftarrow 1_{10}$	22 224.79 (−0.59)	22 534.48 (+0.01)	22 510.70 (−0.16)	22 584.09 (−0.26)		
$3_{30} \leftarrow 2_{21}$	33 997.77 (0.0)	34 352.38 (0.0)	34 342.45 (0.0)	34 526.31 (0.0)		
$3_{03} \leftarrow 2_{12}$	32 115.62 (0.0)	32 526.66 (0.0)	32 481.21 (0.0)	32 389.41 (0.0)		
$3_{12} \leftarrow 2_{21}$	32 673.84 (0.0)	32 967.44 (0.0)	32 951.16 (0.0)	32 913.69 (0.0)		
$3_{31} \leftarrow 2_{20}$	33 526.70 (+0.22)	34 009.11 (+0.87)	*b*	34 118.65 (−0.63)		
$3_{13} \leftarrow 2_{02}$	32 120.02 (+0.15)	32 543.44 (+0.01)	32 495.35 (+0.10)	32 408.15 (−0.28)		
$3_{22} \leftarrow 2_{11}$	32 823.26 (+0.29)	33 276.46 (−0.25)	33 233.99 (+0.93)	33 263.43 (−0.48)		

[a] Deviations from rigid rotor calculations ($\nu_{calcd} - \nu_{obsd}$) are given in parentheses and all frequencies were measured to better than ±0.10 MHz. [b] Lines were too weak and/or obscured by neighboring lines.

Table II. Rotational Constants[a] and Moments of Inertia[b] of CB_5H_7

Isotopic species	*A*	*B*	*C*	*Ia*	*Ib*	*Ic*
Parent	5714.38	5642.20	5324.22	88.4446	89.5707	94.9202
2-^{10}B	5788.30	5687.01	5402.62	87.3099	88.8650	93.5428
4-^{10}B	5783.89	5687.82	5392.05	87.3765	88.8523	93.7261
6-^{10}B	5821.45	5710.90	5354.98	86.8127	88.4932	94.3750
2,6-^{10}B	5891.66	5756.81	5439.37	85.7782	87.7875	92.9108
4.6-^{10}B	5889.46	5753.66	5431.14	85.8102	87.8356	93.0516
^{13}C	5641.51	5598.66	5299.67	89.5817	90.2673	95.3600
^{13}C, 2-^{10}B	5721.12	5641.92	5371.11	88.3352	89.5752	94.0915
^{13}C, 4-^{10}B	5716.12	5642.13	5362.19	88.4124	89.5719	94.2481
^{13}C, 6-^{10}B	5748.09	5658.09	5339.56	87.9192	89.3192	94.6475

[a] Errors in rotational constants were estimated to be ±0.1 MHz. [b] ^{12}C scale was used.

Table III. Stark Effect and Dipole Moment Measurements

Transition	*m*	$\Delta\nu/E^2$, MHz V^{-2} cm^2
$2_{02}-1_{11}$	0	-8.17×10^{-6}
$2_{12}-1_{01}$	0	6.17×10^{-6}
$2_{12}-1_{01}$	1	3.60×10^{-4}
$3_{31}-2_{20}$	0	1.197×10^{-6}
$3_{22}-2_{11}$	0	2.209×10^{-6}
	$\mu_b = 1.43 \pm 0.04$ D	
	$\mu_c = 0.06 \pm 0.04$ D	
	$\mu_{total} = 1.43 \pm 0.04$ D	

dipole component μ_c is not determined accurately and even μ_b exhibits more than the usual error limits.

Molecular Structure. The magnitude of the coordinates of all atoms except the hydrogens were determined by the Kraitchman method.[6] The signs of the atomic positions were deduced by symmetry and other reasoning. Since some doubly substituted species were assigned, various singly substituted species as well as the normal isomer could be used as the reference molecule.

In the reference frames of the normal, ^{13}C, and 6-^{10}B isomers, the *a* components of both B(6) and C were calculated to be imaginary. This frequently occurs when atoms are close to a symmetry element. Since there is good evidence that the molecule contains a mirror plane, these values were set to zero. In the 2-^{10}B and 4-^{10}B frames, B(4) and B(2) positions, respectively, were undetermined due to the failure in assigning transitions of the 2,4-disubstituted species.

The signs of all coordinates must be chosen such that the center of mass conditions, $\Sigma m_i q_i = 0$, and the products of inertia, $\Sigma m_i p_i q_i = 0$, are roughly satisfied. Furthermore, the calculated moments of inertia must agree reasonably well with the observed values. While the signs of some of the coordinates of some of the atoms could be deduced from the symmetry and shape of the molecule, there are others that proved to be nontrivial. Arbitrarily, B(2) was positioned in the first quadrant of the principal axis system. B(3) is equivalent with respect to the reflection plane *bc* and was assigned a negative *a* coordinate. To satisfy the center of mass condition, it was necessary to place B(4) and B(5) on the opposite side of the *ac* plane. According to conventional numbering, B(4) was given a negative and B(5) a positive *a* value. Location of all four borons

Table IV. Coordinates in Principal Axis System of Different CB_5H_7 Isotopic Species

Atom	Normal	^{13}C	2-^{10}B	4-^{10}B	6-^{10}B
B(2) *a*	0.937 ± 0.004	0.933 ± 0.006	0.444 ± 0.003		0.937 ± 0.004
b	0.733 ± 0.003	0.679 ± 0.006	1.067 ± 0.003		0.776 ± 0.002
c	0.418 ± 0.005	0.506 ± 0.004	0.547 ± 0.006		0.327 ± 0.006
B(4) *a*	−0.859 ± 0.005	−0.852 ± 0.006		−0.444 ± 0.003	−0.861 ± 0.004
b	−0.708 ± 0.003	−0.672 ± 0.005		−0.973 ± 0.003	−0.776 ± 0.006
c	−0.482 ± 0.004	−0.545 ± 0.004		−0.607 ± 0.005	−0.366 ± 0.003
B(6) *a*	i	i	0.371 ± 0.010	0.317 ± 0.012	i
b	−0.808 ± 0.004	−0.915 ± 0.004	−0.784 ± 0.004	−0.833 ± 0.004	−0.674 ± 0.003
c	0.980 ± 0.003	0.900 ± 0.003	0.924 ± 0.003	0.904 ± 0.003	1.095 ± 0.002
C *a*	i	i	−0.278 ± 0.006	−0.259 ± 0.006	i
b	0.623 ± 0.003	0.705 ± 0.005	0.640 ± 0.004	0.629 ± 0.004	0.490 ± 0.004
c	−0.875 ± 0.003	−0.793 ± 0.003	−0.817 ± 0.003	−0.829 ± 0.003	−0.937 ± 0.003

Table V. Atomic Coordinates in the Principal Axis Frame of Normal CB_5H_7

Atom	Normal	^{13}C	2-^{10}B	4-^{10}B	6-^{10}B	Average
B(2) *a*	0.937 ± 0.004	0.933 ± 0.006	0.938 ± 0.003		0.937 ± 0.004	0.936 ± 0.004
b	0.733 ± 0.003	0.737 ± 0.006	0.732 ± 0.003		0.735 ± 0.002	0.734 ± 0.004
c	0.418 ± 0.005	0.419 ± 0.004	0.418 ± 0.006		0.417 ± 0.006	0.418 ± 0.005
B(4) *a*	−0.859 ± 0.005	−0.852 ± 0.006		−0.859 ± 0.003	−0.861 ± 0.004	−0.858 ± 0.005
b	−0.708 ± 0.003	−0.718 ± 0.005		−0.708 ± 0.003	−0.707 ± 0.006	−0.710 ± 0.005
c	−0.482 ± 0.004	−0.482 ± 0.004		−0.489 ± 0.005	−0.482 ± 0.003	−0.484 ± 0.004
B(6) *a*	0	0	−0.005 ± 0.010	−0.000 ± 0.012	0	−0.001 ± 0.011
b	−0.808 ± 0.004	−0.806 ± 0.004	−0.806 ± 0.004	−0.806 ± 0.004	−0.807 ± 0.003	−0.807 ± 0.004
c	0.980 ± 0.003	0.981 ± 0.003	0.980 ± 0.003	0.972 ± 0.003	0.979 ± 0.002	0.978 ± 0.003
C *a*	0	0	−0.003 ± 0.006	−0.008 ± 0.006	0	−0.002 ± 0.005
b	0.623 ± 0.003	0.625 ± 0.005	0.623 ± 0.004	0.623 ± 0.004	0.625 ± 0.004	0.624 ± 0.004
c	−0.875 ± 0.003	−0.875 ± 0.003	−0.874 ± 0.003	−0.873 ± 0.003	−0.874 ± 0.003	−0.874 ± 0.003

on the same side of the *ab* plane would have resulted in unrealistic bond lengths between the apical atoms and equatorial atoms regardless of where the B(6) and carbon atoms were placed. Thus, B(4) and B(5) were assigned negative *c* coordinates. Then B(6) was first put above the *ab* plane and C below it to satisfy the center of mass conditions. For reasonable bond lengths, the *b* coordinate of B(6) had to be negative and that of carbon positive, thereby giving structure A in Figure 1. However, when the position of B(6) and carbon were reversed with respect to the *ab* plane, the procedure above yielded a different, yet equally sound, structure B.

To solve this dilemma, coordinates of all atoms obtained in the isotopic reference frames were transformed into that of the normal species for both possible structures. This approach was used to determine an unambiguous structure in $C_4B_2H_6$.[7] By comparing the coordinates for the same atom originating from different reference frames, structure A proved to be far more consistent than structure B. Table IV shows the coordinates in principal axis systems of different isotopic species and Table V gives transformed coordinates in the frame of the normal species using signs consistent with structure A. Bond distances were calculated from the average values of the coordinates and are shown in Figure 1A. Errors were estimated from an error of ±0.1 MHz in rotational constants.

Discussion

Though essentially of octahedral form, the molecule is quite distorted. The B(2)–B(3), B(2)–B(6), and B(3)–B(6) distances are all considerably larger than any other B–B bonds in the compound and are comparable to hydrogen bridged B–B distances in CB_5H_9. Thus a four-center bonded hydrogen could conceivably be located above the faces B(2)–B(3)–B(6).

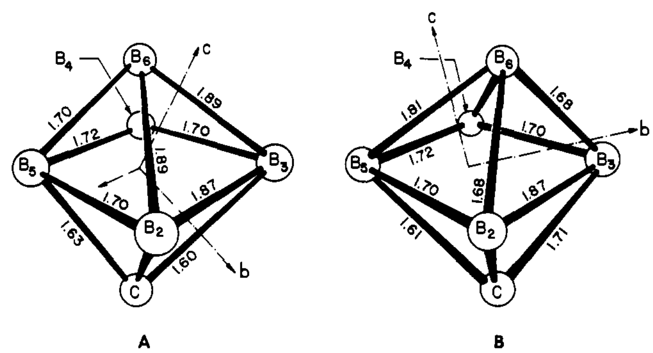

Figure 1. (A) Structure with signs of coordinates consistent with measurements in principal axis system of all isotopic species observed. (B) Structure with signs of the *a*, *b* components of B(6) and C interchanged and proven to be incorrect.

To further substantiate our interpretation, CNDO[8] calculations were performed on the molecule. The experimentally determined bond lengths of all heavy atoms were used as the starting point. The terminal hydrogens were assumed to have a B–H distance of 1.20 Å and the bridged hydrogen to be at an equatorial position bonded to B(2) and B(3) with a B–H distance of 1.36 Å. The CNDO program then proceeded to vary parameters until a minimum in energy was obtained, while the symmetry of the molecule was retained at all times.

Two minima were obtained by the program: one when the dihedral angle between the B(6)–B(2)–B(3) plane and the B(2)–B(3)–H_{br} plane was 70° (Figure 2a) and another when

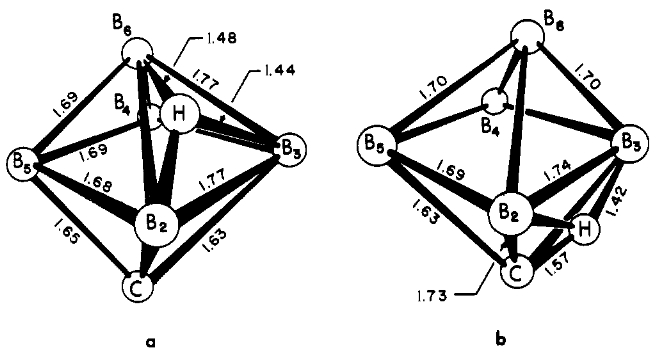

Figure 2. Structures predicted by CNDO: a was calculated to have an energy 27 kcal/mol lower than b.

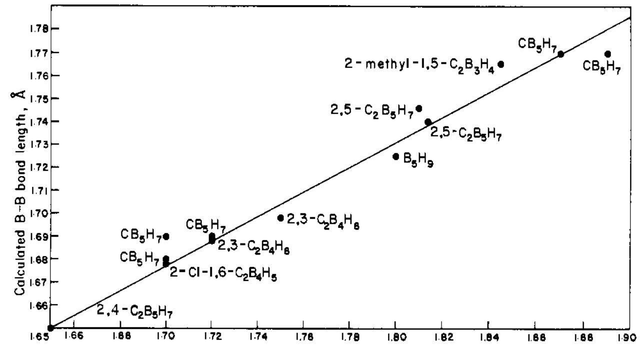

Figure 3. Experimental B-B bond length, Å.

Table VI. Comparison of Observed and CNDO Predicted Structural Parameters of CB_5H_7

	Experimental values[a]	CNDO predicted values
B(2)-B(3)	1.87	1.77
B(2)-B(6)	1.89	1.77
B(4)-B(5)	1.72	1.69
B(5)-B(6)	1.70	1.69
B(2)-B(5)	1.70	1.68
B(2)-C	1.60	1.63
B(4)-C	1.63	1.65
B(2)-H(2)	*b*	1.20
B(4)-H(4)	*b*	1.20
B(6)-H(6)	*b*	1.21
C-H_c	*b*	1.11
B(2)-H_b	*b*	1.44
B(6)-H_b	*b*	1.48
∠B(6)B(2)H_b	*b*	61°

[a] The experimental errors were estimated to be less than 0.01 Å. [b] Values undetermined experimentally.

Table VII. Results of Positioning Bridge Hydrogen Using Observed Moments of Inertia

	Atomic coordinates		
	a	*b*	*c*
B(2)[a]	0.937	0.733	0.410
B(4)[a]	−0.859	−0.708	−0.482
B(6)[a]	0	−0.808	0.980
C[a]	0	0.623	−0.875
H(2)[b]	1.81	1.49	0.73
H(4)[b]	−1.70	−1.34	−1.05
H(6)[b]	0	−1.47	1.99
H_c[b]	0	1.31	−1.75
H_b[c]	0.13	0.46	1.57

Moments		Bridge hydrogen parameters	
$\Sigma m_i a_i$	= 0.1310	B(2)-H_b	1.51
$\Sigma m_i b_i$	= −0.2785	B(3)-H_b	1.59
$\Sigma m_i c_i$	= 0.0692	B(6)-H_b	1.40
$\Sigma m_i a_i b_i$	= 0.0603		
$\Sigma m_i b_i c_i$	= −0.4767		
$\Sigma m_i a_i c_i$	= 0.0946		

[a] Values determined experimentally. [b] Values from CNDO prediction. [c] Values determined by fitting observed moments of inertia.

this angle was 162 °C (Figure 2b). The former is preferred, since it was predicted to be about 27 kcal/mol lower in energy than the latter. Furthermore, structure b calls for a large B(2)-B(3)-C face on which the bridged hydrogen resides at a distance of 1.57 Å from carbon, thus implying a six-coordinate carbon atom, whereas structure a resembles the experimentally determined structure A closely. The calculated B-B distances are all shorter than the observed values, consistent with the trends observed by Beaudet et al.[9] A comparison of CNDO vs. experimental bond lengths is shown in Figure 3. The divergent nature of the calculation with increasing bond distance is evident. In view of this relationship, the principal moments of inertia were calculated using predicted bond lengths and angles of the terminal hydrogens and the experimental heavy-atom parameters (see Table VI). The position of the bridge hydrogen was fitted to the experimental moments of inertia (cf. Table VII). The deviation from both the center of mass condition, $\Sigma m_i q_i = 0$, and the products of inertia, $\Sigma m_i p_i q_i = 0$, were of the same order of magnitude as those calculated from a completely determined r_s structure of 2-chloropropane.[10] The bridge hydrogen was adjusted to a position 1.51 Å from B(2), 1.59 Å from B(3), and 1.40 Å from B(6). This result is fortuitously consistent and again supports the location of the bridge hydrogen.

Furthermore, CNDO predicted a dipole moment of 2.52 D at an angle of 29° to the four boron plane as compared to the experimentally determined value of 1.43 D at an angle of 32°. It should be noted that CNDO usually predicts dipole moments twice as large as the experimental values.

Acknowledgment. We thank Dr. T. Onak for providing the samples of CB_5H_7 and $^{13}CB_5H_7$, the Los Alamos Scientific Laboratories for a sample of $^{13}CH_3Cl$, and the U.S.C. Computer Center for computer time. We are also grateful for the support from the AFOSR (ARSC) USAR, under Grant AFOSR 72-2157, from the National Science Foundation under Grant MPS74-01364, and the United States Energy Research and Development Administration.

References and Notes

(1) (a) University of Southern California, University Park; (b) Los Alamos Scientific Laboratory.
(2) T. Onak, R. Drake, and G. Dunks, *J. Am. Chem. Soc.*, **87**, 2505 (1965).
(3) S. R. Prince and R. Schaeffer, *Chem. Commun.*, 451 (1968).
(4) E. Groszek, J. B. Leach, G. T. F. Wong, C. Ungermaner, and T. Onak, *Inorg. Chem.*, **10**, 2770 (1971).
(5) G. L. McKown, B. P. Don, R. A. Beaudet, P. J. Vergamini, and L. H. Jones, *J. Chem. Soc., Chem. Commun.*, 765 (1974).
(6) J. Kraitchman, *Am. J. Phys.*, **21**, 17 (1953).
(7) J. P. Pasinski and R. A. Beaudet, *J. Chem. Phys.*, **61**, 683 (1974).
(8) J. A. Pople and G. A. Segal, *J. Chem. Phys.*, **43**, S136 (1965).
(9) C. S. Cheung, G. A. Segal, and R. A. Beaudet, *J. Am. Chem. Soc.*, **92**, 4158 (1970).
(10) D. R. Lide, Jr., and M. A. Paul, Ed., "Critical Evaluation of Chemical and Physical Structural Information", National Academy of Sciences, Washington, D.C., 1974, p 32.