Molecular Structure of 1-Aza-*closo*-dodecaborane(12). Experimental and Theoretical Refinement†

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Received January 29, 1992

The gas-phase electron diffraction data (GED) for 1-aza-*closo*-dodecaborane, 1-$NB_{11}H_{12}$, can be fit by four models with C_{5v} symmetry almost equally well. The MP2/6-31G* single point calculations, as well as IGLO (individual gauge for localized orbitals) computed chemical shifts were able to decide among these possibilities. The final experimental geometry was selected on the basis of the agreement between the IGLO ^{11}B chemical shifts calculated for the various GED models and the experimental values. This optimal choice was supported by the energetic criteria. The shorter N–B separation [r_g = 1.716(9) Å] and the expansion of the pentagonal belt adjacent to nitrogen [r_g = 1.825(6) Å] are the main distortions, compared with the icosahedral "parent" $B_{12}H_{12}^{2-}$ [r = 1.77 Å]. The mean length of the B–B bonds in the hemisphere opposite to nitrogen is 1.791(5) Å. The IGLO δ ^{11}B's computed for the best model agree with the experimental values. In particular, the chemical shift of the antipodal B_{12} is reproduced reasonably well (IGLO δ = 5.5 ppm; experimental δ = 2.8 ppm).

Introduction

Since the incorporation of nitrogen as a heterovertex in polyhedral borane cages is very difficult synthetically, only two *closo*-azaboranes are known, 1-NB_9H_{10}[1] and 1-$NB_{11}H_{12}$[2] (**1**; Figure 1). Both have been investigated computationally; 3-21G and 6-31G* geometries have been reported.[3,4] Although the X-ray structures of two azametalla-*closo*-dodecaboranes $MNB_{10}H_{11}$ [M = $RhH(PPh_3)_2$ and $Ru(C_6Me_6)$] have been determined,[5] the experimental geometry of the "parent" compound **1**, the first azaborane containing a six-coordinate cage nitrogen atom, is not known. Unfortunately, **1** is microcrystalline, and single crystals, needed for an X-ray structural determination, could not be grown. We now report the gas-phase geometry of **1** as deduced by a combination of electron diffraction (GED) and computational methods.

Of the ab initio optimized BB and BN distances of **1** (cf. Table I) all proved to lie within a relatively small range, ca. 0.1 Å. In contrast, 1-$SB_{11}H_{11}$, a related *closo*-heteroborane,[6] displayed more pronounced variations in the nearest-neighbor separations. Hence, **1** poses an inherently more demanding problem for the GED technique. Indeed, the refinements resulted in several competing models. These were evaluated subsequently by means of ab initio and IGLO (individual gauge for localized orbitals)[7] chemical shift calculations. The combined ab initio/IGLO/NMR method

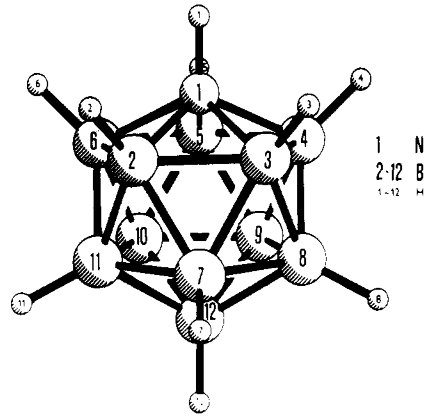

Figure 1. Molecular model of 1-$NB_{11}H_{12}$ showing the atomic numbering with a symmetry of C_{5v}.

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† A preliminary report of this work was presented at the 14th Austin Symposium on Molecular Structure, Austin, Texas, 1–3 March 1–3, 1992; Abstracts, p 66.

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Table I. Ab Initio Optimized Geometries for 1-$NB_{11}H_{12}$[a]

(a) Bond lengths (Å)

	N–B	B_2–B_3	B_3–B_7	B_7–B_8	B_7–B_{12}	N–H	B_2–H_2	B_7–H_7	B_{12}–H_{12}
3-21G	1.741	1.852	1.777	1.817	1.808	1.008	1.174	1.179	1.179
6-31G*	1.710	1.821	1.767	1.808	1.798	1.003	1.177	1.183	1.183
GED[b]	*1.714*	*1.822*		*1.788*		*1.055*		*1.228*	

(b) Bond angles (deg)

	N–B–H	B_{12}–B_7–H
3–21G	114.2	124.3
6-31G*	113.9	124.2
GED[b]	*111.5*	*121.6*

[a] Reference 4. [b] Present work; r_a and $\angle_a$ values based on model D.

has been applied successfully for the structural elucidation of numerous boron compounds.[8] This method is employed here for the first time in conjunction with an experimental structure determination as an additional refinement condition.

Experimental and Computational Section

The sample of 1-aza-*closo*-dodecaborane(12) was prepared according to the literature.[2] The purity was established by the ^1H and ^{11}B NMR spectra.[2]

Diffraction patterns were recorded with the Balzers Eldigraph KDG-2 instrument at the University of Oslo,[9,10] on Kodak electron image plates with nozzle-tip temperature of about 120 °C. The accelerating voltage of the electron beam was 42 kV. The voltage/distance calibration was done with benzene as reference. The typical data treatment of the Norwegian group was employed.[11] Two plates from the long (498.57 mm) and three from the short (248.70 mm) nozzle-to-plate distances were selected for analysis. Data from the long and short camera distances used in the analysis were in the ranges $2.00 \leq s \leq 14.875$ and $3.75 \leq s \leq 27.25$ Å^{-1}, respectively [$s = (4\pi/\lambda) \sin(\theta/2)$, where λ, the electron wavelength, was 0.058 73 Å for both camera distances and θ is the scattering angle]. The least-squares method was applied to molecular intensities modified by s^4. Calculations were carried out using a modified PC version of the program by Seip et al.[11] Atomic scattering factors were taken from ref 12.

The ^{11}B chemical shifts were calculated in Erlangen with the CONVEX version of the IGLO program[7] employing a Huzinaga DZ basis set.[13] The Gaussian lobe contractions were as follows:[7b] B, N 7s3p (4111, 21); H 3s (21). Energy calculations were performed using standard procedures and basis sets.[14]

Structure Analysis

As mentioned above, many of the bonds are similar in length and could not be resolved by GED method. Hence, C_{5v} symmetry was assumed and the following independent parameters were selected to describe the geometry of **1**: B_2–B_3, $(B–B)_{mean}$ (i.e. a single value for all the other B–B linkages), $(B–H)_{mean}$ (i.e. a single value assumed for all the BH bonds), the $r(B_2–B_3) - r(N–B)$ difference (Dif_1), and the $r(B–H)_{mean} - r(N–H)$ difference (Dif_2), as well as the N–B–H and B_{12}–B_7–H bond angles.

Table II. Geometric Parameters[a] and *R* Factors for 1-Aza-*closo*-dodecaborane(12) As Obtained from the Competing Models

parameter	model A[b]	model B	model C	model D[b]
Dif_1[c]	0.109(2)	0.109(2)	0.104(1)	0.108(2)
Dif_2[d]	0.024(28)	0.024[f]	0.173[f]	0.173[f]
r(N–B)[e]	1.704(3)	1.715(5)	1.750(4)	1.714(5)
$r(B_2–B_3)$	1.813(3)	1.824(4)	1.854(4)	1.822(3)
$r(B–B)_{mean}$	1.792(2)	1.786(2)	1.770(2)	1.788(2)
r(N–H)[e]	1.199(24)	1.206(3)	1.067(3)	1.055(3)
$r(B–H)_{mean}$	1.223(3)	1.230(3)	1.241(3)	1.228(3)
∠N–B–H	113.9[f]	108.8(12)	104.6(12)	111.5(10)[g]
$\angle B_{12}$–B_7–H	124.2[f]	123.1(29)	123.4(24)	121.6(34)[g]
R factor	0.0456	0.0432	0.0459	0.0498

[a] Bond lengths (r_a) are given in Å, angles in deg. [b] Least-squares standard deviations in the last digit are given in parentheses. [c] $Dif_1 = r(B_2–B_3) - r(N–B)$. [d] $Dif_2 = r(B–H)_{mean} - r(N–H)$. [e] Dependent parameter. [f] Fixed parameter, with the exception of 0.024 for Dif_1 at the 6-31G* values. [g] Fixed at the 6-31G* value and then refined.

Table III. IGLO Results for $NB_{11}H_{12}$

	$\delta(^{11}B)$[a]			
level of theory	B_{2-6}	B_{7-11}	B_{12}	rel energy[b]
DZ//3-21G	−8.3	−16.6	8.9	
DZ//6-31G*	−10.4	−16.8	7.7	
DZ//GED(A)	−10.6	−17.4	4.9	21.0
DZ//GED(B)	−10.9	−18.9	2.6	27.2
DZ//GED(C)	−9.0	−21.3	0.0	24.9
DZ//GED(D)	−11.2[c]	−17.4	5.5	11.0
expt.[d]	*−9.8*	*−11.9*	*2.8*	

[a] Relative to $BF_3 \cdot OEt_2$ (ppm). [b] With respect to the 6-31G* geometry (kcal/mol). [c] −10.2 ppm when both angles were fixed at the 6-31G* values. [d] Reference 2.

Different initial starting values of the independent parameters always refined to the same geometry. Attempts to resolve the lengths of the individual B–B bonds in the hemisphere opposite to nitrogen [B_2–B_7, B_7–B_8, and B_7–B_{12}; i.e. those defining $(B–B)_{mean}$] led to divergence in the refinement. This was also true if the B–B distances was refined and the 6-31G* differences in bond lengths for the other BB bonds were assumed.

Due to the lack of spectroscopic data, some of the vibrational parameters of 1-$SB_{11}H_{11}$[6] were taken as starting values. The vibrational amplitudes, *l*, of adjacent distances were coupled in blocks and varied under the constraint of constant differences within each block. Since the use of different $l(B_2–B_3)$ and $l(B–B)_{mean}$ amplitudes did not affect the results, a single $l(B–B)_{all}$ value was employed to characterize the B–B bond vibrations. Thus, all the nearest-neighbor B–B separations are indicated to be closely spaced. In contrast, the $\Delta_1 l = l(B–B)_{all} - l(N–B)$ difference influenced the results substantially. The use of different $\Delta_2 l = l(N \cdots B_9) - l(B \cdots B)_{nnn}$ [the vibrational amplitudes of the next-nearest-neighbor boron atoms $l(B \cdots B)_{nnn}$ were assumed to be equal] and $\Delta_3 l = l(N \cdots B_{12}) - l(B_2 \ldots B_9)$ values only led to negligible changes of the resulting geometry.

In the first analysis, the simultaneous refinement of all the independent parameters, as well as a value of −0.016 Å for $\Delta_1 l$, gave the best *R* factor (0.0421). Except for the relatively small N–B–H angle (104.6°), the resulting geometric parameters were similar to those for the 3-21G values. However, the 3-21G separations for most cluster boranes,[8c] including 1-$SB_{11}H_{11}$,[6] tend to be too long. Furthermore, the IGLO ^{11}B chemical shifts calculated for this first model showed a rather large deviation from the experimental value for B_{7-11} (IGLO $\delta = -23.6$ ppm; experimental $\delta = -11.9$ ppm). Also, single point MP2/6-31G* calculations showed this first model to be 25.5 kcal/mol less stable than the 6-31G* optimized structure. Hence, this initial model was rejected.

Four additional models (**A–D**) were then considered. These differ in the constraints and in the conditions of the refinement

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Table IV. Final Molecular Parameters[a] for 1-Aza-*closo*-dodecaborane(12) As Obtained from Refinement D

(a) Distances r_a[b] and r_g[c] and Mean Amplitudes of Vibration (Å)[b]

atomic pair	multiplicity	r_g	r_a	l	coupling of the amplitudes refined as groups
N–B	5	1.716(9)	1.714(5)[d]	0.062(1)	i
B_2–B_3	5	1.825(6)	1.822(3)	0.074	i
$(B–B)_{mean}$	20	1.791(5)	1.788(2)	0.074	i
$(B–H)_{mean}$	11	1.235(6)	1.228(3)	0.089(4)	ii
N–H_1	1	1.060(76)	1.055(3)[d]	0.074	ii
N···B_7	5		2.713(3)[d]	0.089(2)	iii
N···B_{12}	1		3.186(4)[d]	0.056(6)	iv
B_2···B_9	5		3.423(1)[d]	0.076	iv
B_2···B_8	10		2.909(1)[d]	0.084	iii
B_2···B_4	5		2.947(6)[d]		
B_7···B_9	5		2.892(3)[d]		
B_2···B_{12}	5		2.903(2)[d]		
N···H_2	5		2.446(14)[d]	0.112	iii
N···H_7	5		3.831(27)[d]	0.113(6)	v
N···H_{12}	1		4.413(4)[d]	0.116(23)	vi
(B···H)[e]	$4 \times 10 + 2 \times 5$		(2.596–2.797)[d]	0.112–0.132	iii
(B···H)[f]	$4 \times 10 + 2 \times 5$		(3.899–4.103)[d]	0.100–0.130	v
(B···H)[g]	2×5		(4.611–4.651)[d]	0.116	vi
(B···H_1)[e]	5		2.365(6)[d]	0.117	iii
(B···H_1)[f]	5		3.634(4)[d]	0.118	iv
(B···H_1)[g]	1		4.241(4)[d]	0.116	vi

(b) Bond Angles (deg) and Differences between Bond Distances (Å)

parameter	value	
$\angle_a$N–B–H	111.5(39)	111.5(10)[k]
$\angle_a B_{12}$–B_7–H	121.6(50)	121.6(34)[k]
Dif_1[h]		0.108(2)
Dif_2[j]		0.173[l]

[a] The H···H distances were included in the refinement but they are not listed here. Their amplitudes were within the range 0.142–0.20 Å. [b] Least-squares standard deviations in the last digit are given in parentheses. [c] Total errors for r_g bonds and for $\angle_a$ bond angles correspond to the following expressions: $\sigma_t = [(0.001r)^2 + 2\sigma^2 + (\Delta/2)^2]^{1/2}$ and $\sigma_t = [2\sigma^2 + (\Delta/2)^2]^{1/2}$, respectively, where 0.001 means experimental scale error and σ is the standard deviation from the least-squares refinement (see also Table II) multiplied by $2^{1/2}$ to take the consequences of data correlation into account. The correlation coefficients exceeding 0.6 in their absolute values are $r(B_2–B_3)/r(B–B)_{mean} = -0.94$, $l^i/r(B–B)_{mean} = -0.74$, $l^i/r(B_2–B_3) = 0.71$, $l^i/Dif_1 = -0.62$, and $\angle N–B–H/\angle B_{12}–B_7–H = 0.66$. Δ is the maximum difference between the parameters referring to the three models A, B, and D in Table II. [d] Dependent parameter. [e] Two bonds removed. [f] Three bonds removed. [g] Diametrically opposite. [h] $Dif_1 = r(B_2–B_3) - r(N–B)$. [j] $Dif_2 = r(B–H)_{mean} - r(N–H)$. [k] Fixed at the 6-31G* value and then refined. [l] Fixed parameter at the 6-31G* value.

(see Table II). A value of 0.012 Å for $\Delta_1 l$ was found to be optimal under these refinement conditions. However, the *R* factors of **A**–**D** all differed insignificantly. Hence, we performed IGLO chemical shift calculations for **A**–**D** (Table III) in order to decide among these possibilities. The IGLO values for **A** and **D** fit the experimental chemical shifts slightly better (maximum deviations $\Delta\delta_{max}$ = 5.5 ppm) than those of **B** and **C** ($\Delta\delta_{max}$ = 7.0 and 9.4 ppm, respectively). Note also that the N–B and B_2–B_3 separations of **C** are very close to the 3-21G dimensions. Model **D** was computed to be ca. 10 kcal/mol more stable than model **A** (DZ basis set, Table III). Finally, the more refined MP2/6-31G* single point calculations showed that the model **D** geometry gave an energy only 6.3 kcal/mol higher than that of the 6-31G* optimized structure. Therefore, both the NMR and in particular the energetic criteria indicate that the model **D** parameters afford the best representation of the experimental structure. The final geometry we deduce is summarized in Table IV (same as **D** in Table II), together with the vibrational amplitudes. Figures 2 and 3 show the molecular scattering intensity curves and the radical distribution curve, respectively.

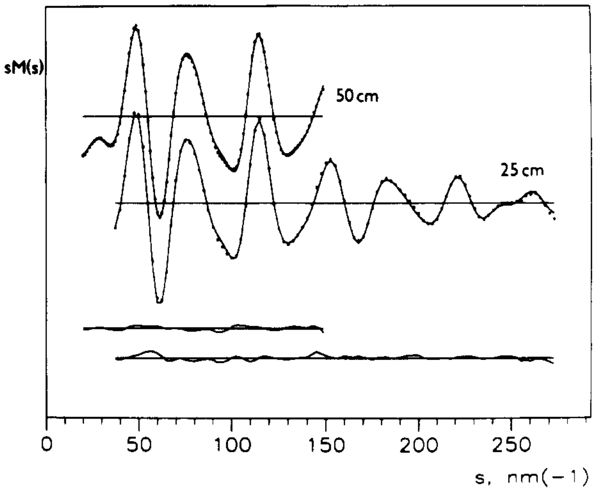

Figure 2. Experimental (●) and theoretical (—) molecular intensity curves for the two camera distances for the model obtained from refinement D with difference curves given below.

Discussion

The somewhat larger total errors, estimated using the results of models **A** and **B** as well (**C** is not considered for the above-mentioned geometric reasons), indicate that the structure of **1** could not be determined unambiguously by GED. However, several interesting features are revealed:

The elongation of the B_2–B_3 distance with respect to the parent $B_{12}H_{12}^{2-}$ (r = 1.77 Å)[15] is apparent in **1** [$r_g(B_2–B_3)$ = 1.825(6) Å] but to a lesser extent than in compounds with second-row heterovertices [e.g. 1.867(7) Å (mean value) in $PCH_3B_{11}H_{11}$[16] and 1.905(4) Å in 1-$SB_{11}H_{11}$[6]].

That the terminal B–H bonds in **1**, ca. 1.23 Å, are somewhat longer than those in 1-$SB_{11}H_{11}$, ca. 1.19 Å, can be ascribed to

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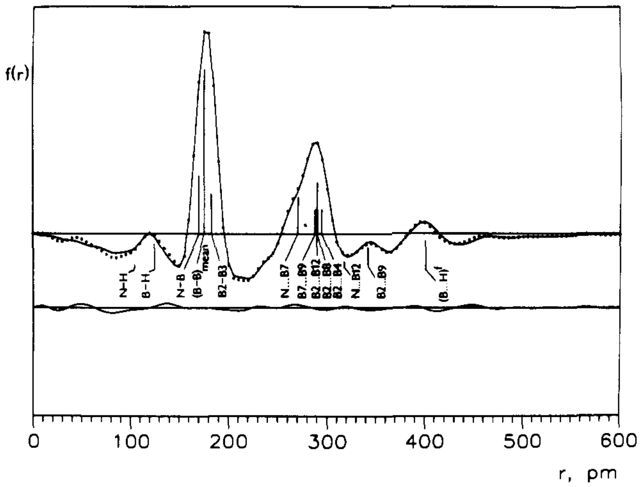

Figure 3. Radial distribution curves [(●) experimental; (—) theoretical] calculated with an artificial damping factor $\exp(-0.002s^2)$. Theoretical values are used in the region $0.00 \le s \le 1.75$ Å^{-1}. The positions of the more important distances are marked with the vertical bars of height proportional to the weight of the distances. Only the mean value for $(B\cdots H)^f$ is marked. The meaning of $(B\cdots H)^f$ as well as the positions of the other distances can be deduced from Table IV. The difference curve is given below.

the larger vibrational amplitude in **1** (ca. 0.09 Å), as compared to the thiaborane (ca. 0.06 Å).[6] The 6-31G* optimized mean B–H length in **1** (r_e value) is 1.18 Å, 0.05 Å shorter than the experimental r_g value. This difference between $r_e(B–H)_{mean}$ and $r_g(B–H)_{mean}$ may be responsible for most of the ca. 6 kcal/mol "excess energy" of the GED structure with respect to the ab initio optimized geometry, as assessed by the MP2/6-31G* single points. Evidence is provided by optimization at 6-31G* of the hydrogen positions of the GED structure, keeping the NB_{11} cluster fixed at its experimental geometry (model **D**). The MP2/6-31G* single point energy of this "partially optimized experimental" structure is only 0.9 kcal/mol above that of the 6-31G* fully optimized geometry. Single point energies calculated using experimental borane and carborane geometries can be much higher (30–60 kcal/mol) than the theoretical, fully optimized energies.[8b,e] In such cases, the experimental determinations may be suspect. Inclusion of the $\Delta BH = r(B_{12}–H_{12}) - r(B_2–H_2)$ constraint (0.006 Å, 6-31G* basis set; see Table I) in order to resolve the individual B–H bonds did not improve the agreement at all.

The N–H bond length is uncertain due to the very large error. The arrangement at nitrogen is flattened. As a consequence, the $N\cdots H_{2-6}$ distances, ca. 2.45 Å, are shorter than the sum of the corresponding van der Waals radii (2.66 Å). Hence, the nitrogen is "shielded" by the five adjacent H atoms. Experimental observations are consistent: **1** is known to be quite acidic (a stronger acid than $HNEt_3^+$),[2] and the corresponding $NB_{11}H_{11}^-$ anion can only be alkylated with less bulky, strong electrophiles (e.g. triflates $(RO)O_2SCF_3$, where R = Me, Et).[18]

Note that $l(N\cdots B_{12}) < l(N–B)$ and $l(B_2\cdots B_9) \approx l(B–B)_{all}$. This reflects the specific internal motion within rigid clusters and is in line with the known relationship between vibrational amplitudes of bonding and nonbonding distances.[6,19]

The IGLO ^{11}B chemical shifts, computed for the best theoretical (6-31G*) and the best experimental geometries (GED, model **D**), compare reasonably well with the experimental $\delta(^{11}B)$ values. The somewhat larger discrepancy found for B_{7-11} was decisive for the selection of the best model. The downfield shift of B_{12} with respect to $B_{12}H_{12}^{2-}$ ($\delta = -15.3$ ppm),[20] the antipodal effect,[21] is well reproduced by the calculations (DZ//GED $\delta = 5.5$ ppm, experiment 2.8 ppm).

Acknowledgment. Financial support from the Czechoslovak Academy of Science (Grant No 43202) is highly appreciated. M.B. acknowledges a grant from the Studieststiftung des Deutschen Volkes. We thank Professor A. Haaland for his interest in this work. We also thank Professor W. Kutzelnigg, Dr. M. Schindler, and Mr. U. Fleischer for the IGLO program. The calculations in Erlangen were performed on a Convex C220s computer and were supported by Convex Computer Corp., the Deutsche Forschunsgemeinschaft, and the Fonds der Chemischen Industrie. Computer time was granted on a Cray Y-MP of the Höchstleistungsrechenzentrum Jülich. We thank V. Balaji and Professor J. Michl for sending the 6-31G* geometry prior to publication, and a reviewer for helpful comments.

Supplementary Material Available: Listings of total electron diffraction intensities multiplied by s^4 for both camera distances and atomic coordinates (3 pages). Ordering information is given on any current masthead page.

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