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The Electronic Structure and One-Electron Properties of BH Computed from SCF and CI Wave Functions

Stewart A. Houlden[★] and Imre G. Csizmadia[★★]

Department of Chemistry, University of Toronto, Toronto, Ontario,
Canada, M5S 1A1

The SCF and CI wave functions for BH, obtained in calculations described in detail elsewhere [2], are compared through their electron distributions and electron moments.

Key words: BH, electronic structure and one-electron properties of ~

1. Introduction

In a previous paper [1] an extensive series of calculations on the ground and excited states of BH were described. In this paper, we present a more detailed comparison of the state wave functions for the self-consistent field (SCF) and configuration interaction (CI) approximations. In the calculations an extensive $(16^s8^p)/(6^s1^p)$ primitive basis set as contracted to $[6^s3^p]/[3^s1^p]$ has been used. In the CI calculations all single and double replacements of the four valence electrons have been included.

2. Natural Orbital Analysis

The Natural Orbitals (NO) are very useful for describing the electronic structure of a complicated wave function such as a CI wave function. These one-electron orbitals form an independent electron picture of the many-electron wave function. Although the true electron probability is given only by $\Psi^*\Psi$, the partitioning of the wave function into doubly occupied one-electron functions results in considerable conceptual simplicity.

For the SCF calculations, the NO are the Molecular Orbitals (MO) themselves; thus the effect of a CI calculation on the wave function can be visualized by comparison to these orbitals. The MO obtained from the SCF calculation at $r = 2.3291$ Bohr are given in Table 1. The Gaussian orbitals referred to are described in previous papers [1, 2]. In this and subsequent tables all expansion coefficients are listed which have a value of 0.25 or over in the MO or NO. The first MO, composed primarily of the Bs_2 Gaussian orbital clearly corresponds to a B1*s* atomic orbital (AO). The high localiz-

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[★★] Author to whom correspondence should be addressed.

Table 1. SCF calculation on BH

Orbital energy r (Bohr)	ϵ (Hartree)	Principal coefficients of Gaussian orbitals		
a) First MO				
1.75	−7.6511	B s_2 (0.9791)		
2.3291	−7.6853	B s_2 (0.9799)		
2.75	−7.7143	B s_2 (0.9802)		
3.25	−7.7470	B s_2 (0.9804)		
3.75	−7.7768	B s_2 (0.9805)		
4.50	−7.8144	B s_2 (0.9806)		
6.00	−7.8611	B s_2 (0.9807)		
B^- 1s	−7.4247 [3]			
B 1s	−7.6953 [4]			
B^+ 1	−8.1859 [4]			
b) Second MO				
1.75	−0.7398	B s_3 (0.5464)	B s_4 (0.3465)	B s_6 (−0.3470)
			H s_1 (0.3341)	H s_2 (0.3125)
2.3291	−0.6493	B s_3 (0.5915)	H s_1 (0.2650)	H s_2 (0.3326)
2.75	−0.6007	B s_3 (0.6498)		H s_2 (0.3055)
3.25	−0.5669	B s_3 (0.7393)		
3.75	−0.5565	B s_3 (0.8203)		
4.50	−0.5648	B s_3 (0.8911)		
6.00	−0.5945	B s_3 (0.9524)		
B^-2s	−0.2424 [3]			
B 2s	−0.4947 [4]			
B^+ 2s	−0.8739 [4]			
c) Third MO				
1.75	−0.3398	B s_3 (0.4894)		
		B p_{1z} (0.3005)	B p_{2z} (0.2668)	
2.3291	−0.3474	B s_3 (0.5073)		
		B p_{1z} (0.2779)	B p_{2z} (0.2908)	
2.75	−0.3517	B p_{1z} (0.2634)	B p_{2z} (0.3199)	
			H s_2 (−0.3129)	
3.25	−0.3492	B s_3 (0.4285)	B p_{2z} (0.3336)	
			H s_2 (−0.3485)	
3.75	−0.3368	B s_3 (0.3396)	B p_{2z} (0.3572)	
			H s_2 (−0.3568)	
4.50	−0.3102		B p_{2z} (0.4004)	
			B s_2 (−3254)	
6.00	−0.2643		B p_{2z} (0.4639)	
		H s_1 (−0.2510)	H $s_3\alpha$ (0.3510)	
B^- 2p	−0.0263 [3]			
B 2p	−0.3099 [5]			
H^- 1s	−0.0447 [6]			
H 1s	−0.5000			

ation of this orbital justifies the neglect of this MO in the CI calculation, since it is relatively unchanged for all geometries. The extrapolation of the orbital energy of this MO to $r = \infty$ predicts the dissociation to B^+ ion (the orbital energies of the 1*s* orbital for B^-, B and B^+ are included in Table 1).

In the bonding region, the second MO is composed primarily of the Bs_3 Gaussian orbital together with some more diffuse Boron *s* orbitals and the Hydrogen *s* orbitals. As the bond length increases, the electrons contract into the Bs_3 orbital which can be associated with the B2*s*AO. As the bond length increases, the orbital energy decreases to a minimum around 3.75 Bohr and then increases again. This suggests that initially in the dissociation this MO tends towards the B2*s*AO but, as the separation increases, it turns in the direction B^+ 2*s* AO.

The third MO is an antibonding orbital having a node through the bond. At equilibrium separation the electrons are shared between the atoms with the negative Hs_2 orbital and negative lobes of the Bp_z orbitals reinforcing each other around hydrogen. As the separation increases the electrons move into the Bp_{2z} and H s_1 and s_3 orbitals; thus, this MO is still substantially distributed over both atoms even at 6.0 Bohr but is more concentrated toward the H atom. The orbital energy has a maximum around 2.75 Bohr which suggests a tendency to approach the $E(1s)$ of the H atom. After this the energy falls below B 2*p* AO towards that of H^-s AO.

The NO for selected calculations can be compared to these MO. Since all the CI calculations were valence shell calculations [2], the lowest energy MO is unaffected by the calculations and so, in every case, occurs with occupation number exactly 1.0. This NO has been omitted from the following discussion.

Table 2 presents the NO for the $X^1\Sigma^+$ ground state and the $^1\pi$ state calculated from the CI wave functions. The sum of twice the occupation numbers of the first three NO for the $^1\Sigma^+$ state is 5.8281 which indicates these NO account for almost all the electron distribution of the wave function. The second NO (i.e. the first in the table) is

Table 2. NO for $X^1\Sigma^+$ ground state and $A^1\pi$ state for BH at equilibrium separation. (Full valence shell CI calculation using 12 NO of full double valence shell CI wave function [2]

State	Occupation Number (x2)	Principal coefficients of Gaussian orbitals	
$X^1\Sigma^+$	1.9601	B s_3 (0.3616)	
		H s_1 (0.3079)	H s_2 (0.3933)
	1.8680	B s_3 (0.7193)	B s_4 (0.3127)
$A^1\pi$	1.9339	B s_3 (0.6540)	
			H s_2 (0.2725)
	0.9879	B p_{1x} (0.4057)	B p_{2x} (0.8602)
	0.9858	B s_3 (0.4727)	
		B p_{2z} (0.3059)	B p_{3z} (0.3809)
			H s_2 (−0.3028)

similar to the second MO but with some shifting of the electrons towards the hydrogen. The third NO, however, is quite different from the third MO. The electrons are even more concentrated in the B s_3 orbital with the B s_4 orbital also strongly populated. Although the antibonding character is still there (the B p_z and H s coefficients were too small to include in Table 2), it is of minor significance in the NO. The CI calculation has therefore caused a spreading of electrons in the bonding second orbital while contracting the electrons onto B in the antibonding orbital.

The description of the $A^1\pi$ state indicates that the electron distribution can best be described as two electron pairs which doubly occupy two orbitals and one pair which is split into two other orbitals. The second NO corresponds closely to the second MO and can be identified with the boron $2s$ AO. The third NO is clearly the B $2p_x$ AO while the fourth NO is almost the same as the third MO. The electron distribution for this state corresponds very closely to a virtual orbital picture. These four NO account for a total of 5.9076 electrons.

The NO formed for the widest separation will give an indication of the atomic species to which the states are dissociating. Table 3 gives the principal coefficients of the first five states calculated at $r = 6.0$ Bohr. As before, the first NO is omitted from the table since it was unchanged from the first MO.

For the $X^1\Sigma^+$ ground state four NO are required to account for 5.8738 electrons. The first NO closely resembles the B $2s$ although it is slightly more diffuse than the corresponding MO for this geometry. The third NO is similar to the third MO which has antibonding character but with the B $2p_z$ having the largest coefficient. For the CI calculation, this NO is described with only 1.3827 electrons with the fourth NO containing 0.6084. This latter NO is also made up of B $2p_z$ and H $1s$ type Gaussian orbitals but with the electrons equally distributed between the atoms. This function will have two nodes: the $2p_z$ node through B and one closer to H since the negative lobe of the B $2p_z$ is between the atoms but the H $1s$ is a positive orbital. The nature of these NO indicates that there is still much interaction between the two atoms even at 6.0 Bohr separation. It is apparent that the outer electron pair is splitting into two electrons in different orbitals as is expected from the dissociation products for this state.

On the other hand, the $A^1\pi$ state appears completely dissociated. The second NO is identical to the second NO for the ground state while the third and fourth NO, which are each occupied by almost exactly one electron, are situated one on each atom. One electron is in a H $1s$ orbital and one in the B $2p_x$. This accounts for the shallowness of the minimum for the $A^1\pi$ profile. In separating the two electrons only the distribution of one electron has changed drastically. The electron in the fourth NO at equilibrium geometry was shared between the two atoms while at this separation it is centred exclusively on hydrogen. These four NO for the $A^1\pi$ state account for 5.8747 electrons.

The next two states, the $C^1\Delta$ and $B^1\Sigma^+$ have almost identical NO, which is to be expected since they dissociate to the same species. Three of the NO can be easily identified with B $2s^2$, $2p_x^1$ and $2p_y^1$ (or $2p_z^1$). The remaining NO, which would be expected to become H $1s^1$, is still very much centred on boron. These five NO account for 5.9579 and 5.9832 electrons respectively for the two states.

Table 3. NO of first five singlet electronic states of BH at a separation of 6.0 Bohr. (Full valence shell CI calculated using 12 ANO of full double valence shell CI wave function)

State	Occupation Number (x2)	Principal coefficients of Gaussian orbitals	
$X^1\Sigma^+$	1.8827	B s_3 (0.8742)	
	1.3827	B p_{1z} (0.2663)	B p_{2z} (0.4440)
		H s_1 (−0.2794)	H s_2 (−0.3824)
	0.6084	B p_{1z} (0.3231)	B p_{2z} (0.5124)
		H s_1 (0.3174)	H s_2 (0.5260)
$A^1\pi$	1.8848	B s_3 (0.8687)	
	1.0001	H s_1 (0.4081)	H s_2 (0.6349)
	0.9899	B p_{1x} (0.4207)	B p_{2x} (0.6939)
$C^1\Delta$	1.2015	B s_3 (0.6844)	B s_4 (0.2946)
		H s_1 (0.2716)	H s_2 (0.4291)
	0.98137	B p_{1x} (0.4192)	B p_{2x} (0.1878)
	0.98131	B p_{1y} (0.4144)	B p_{2y} (0.8163)
	0.7937	B s_3 (0.6669)	
		B s_1 (−0.3014)	B s_2 (−0.4730)
$B^1\pi^+$	1.2278	B s_3 (0.5991)	
		B p_{3z} (−0.2954)	
		H s_1 (0.2551)	H s_2 (0.3902)
	1.1014	B s_3 (0.3544)	
		B p_{1z} (0.3727)	B p_{2z} (0.7225)
	0.7830	B s_3 (0.6533)	
		H s_1 (−0.3061)	H s_2 (0.4782)
	0.7710	B p_{1x} (0.4206)	B p_{2x} (0.8095)
$C^1\Sigma^+$	1.9015	B s_4 (−0.8056)	B s_5 (0.6841)
		B s_6 (0.3131)	
		H s_1 (0.2662)	H s_3 (0.5617)
	1.8307	B s_3 (0.9741)	B s_4 (0.3871)

The last state is the $C^1\Sigma^+$ state which has the maximum in the energy profile described previously [1, 7, 8]. In the analysis of the NO, an explanation for this behaviour is forthcoming. The expected dissociation products for this state are $B\ ^2S$ ($1s^2\ 2s^2\ 3s^1$) and H ^2S ($1s^1$); thus four NO are expected: two doubly occupied (B $1s^2$ $2s^2$) and two singly occupied (B $3s^1$, H $1s^1$). In fact, 5.7322 electrons are accounted for by three NO with the fourth NO (not shown on Table 3) having only 0.1091 electrons. The first NO, as explained above, is assigned B $1s^2$ while the third is clearly B $2s^2$. The second NO, however, is a bonding orbital delocalized over the two atoms. The coefficient of H s_2, which is the closest Gaussian orbital to H $1s$, is very small while the H s_3 is large. On the other hand, the more diffuse B s orbitals have large coefficients indicating some tendency toward occupying the B $3s$. The negative B s_4 effectively cancels all electron distribution near the boron nucleus. Thus the molecule is able to adopt a lower energy configuration with the atomic open shell electrons paired in a bonding MO. As the separation increases farther this MO will become unstable and the correct dissociation products will be assumed.

3. One-Electron Properties

The desirability of several criteria for judging a new computational procedure is quite clear. Unfortunately no experimental quantities are available for BH with the exception of dipole moment [9] and so the properties calculated must be compared to intuitive expectations. In this regard it was decided to calculate selected properties which reflect the electron distribution, namely the electronic moments.

Table 4 gives the unique moments of BH up to octopole. As some previous results on BH [10, 11] are of comparable quality to the present wave function, values obtained earlier are also included in Table 4. All moments except the dipole moment are dependent on choice of origin relative to which they are calculated; hence, by convention the centre of mass was chosen (0.1984 Bohr from boron on the BH bond). As correlation was introduced, the dipole moment decreased in magnitude.

The second moment is a measure of the absolute size of the charge distribution described by the wave function, whereas the quadrupole moment is a measure of both size and shape [16]. From the values obtained it is seen that the charge distribution increases as correlation effects are accounted for. This is in agreement with other calculations on other systems [17, 18]. Previous calculations [19, 20] have shown that the calculated quadrupole moment is strongly influence by choice of basis set. The smallest basis set [19] referred to gave a poorer value for the energy and dipole moment yet a better value for the quadrupole moment. The values obtained in the present calculation show that the SCF values tend to the Hartree-Fock (HF) value while the introduction of CI caused a decrease in absolute value. In other calculations the Valence Bond [14] calculation gave a result higher than HF, and SOGI [15] a value below HF.

Table 5 shows the values of the same properties calculated from SCF wave functions (Basis 3) for different geometries. The most interesting feature of this table is the dipole moment which goes from negative at shorter distances (B^-H^+) to positive at longer (B^+H^-). This agrees with the behaviour of the orbital energy of the second MO which first tends toward B 2*s* (i.e. dissociation to B and H, dipole moment zero) and later to B^+ 2*s*. The values calculated for CI wave functions for different geometries are included in Table 6. With the exception of the dipole moment, all properties show the same trends as the HF results; however, their behaviour is much more restrained. The dipole moment is quite different in that instead of increasing to infinity it now appears to be converging on zero. This is the value expected from dissociation into two neutral atoms. It is interesting to note that the molecule at large internuclear separation does have $B^+ H^-$ charge distribution. A plot of dipole moment versus separation for both SCF and CI wave functions is given in Fig. 1.

The calculated bond moments can be used to give further indications of the electronic distribution. The components of the dipole moment define the centroid of charge for the distribution. In Table 7 the centroids of charge for the NO of the two wave functions are given. For the SCF approximation, the charge of the first MO is centred on the B nucleus, the second MO is centred between the two atoms, while the third is centred on the opposite side of the B atom. This agrees with a description of the electronic configuration having a B inner shell, a bonding orbital and a B lone pair.

Table 4. Electron moments for BH wave functions calculated at centre of mass

Wave Function		Dipole Moment $\langle z\rangle$	Second moments $\langle x^2\rangle$	$\langle z^2\rangle$	Third moments $\langle z^3\rangle$	$\langle y^2z\rangle$	$\langle r^2z\rangle$	Quadrupole Moment $\frac{1}{2}\langle 3z-r^2\rangle$	Octopole Moment $\frac{1}{2}\langle 5z^2-3r^2z\rangle$	$\langle r^2\rangle$
This work	SCF	−0.6980	−4.9620	−7.8133	−3.7802	−1.3223	−7.4248	−2.8513	0.1867	−17.7572
	CI	−0.5575	−4.9653	−7.4621	−2.3442	−1.0025	−4.3472	−2.4968	0.6603	−17.3926
CI [11]		−0.490						−2.359[a]		
Large CI [10]		−0.5785								
HF [13]		−0.682								
VB [14]		−0.702						−2.901		
SOGI [15]		−0.702						−2.72		
Exp. [9]		−0.492								

[a] Calculated with respect to the B nucleus.

Table 5. Electron moments for BH calculated at centre of mass for different internuclear separations for SCF (near HF) wave functions

r (Bohr)	Dipole Moment $\langle z\rangle$	Second Moments $\langle x^2\rangle$	$\langle z^2\rangle$	Third Moments $\langle z^3\rangle$	$\langle y^2z\rangle$	$\langle r^2z\rangle$	Quadrupole moment $\frac{1}{2}\langle 3z^2-r^2\rangle$	Octopole moment $\frac{1}{2}\langle 5z^2-3r^2z\rangle$	$\langle r^2\rangle$
1.75	−0.9839	−4.8363	− 7.3842	−7.1176	−2.4966	−12.1108	− 2.5479	0.3721	−17.0569
2.3291	−0.6980	−4.9620	− 7.8133	−3.7802	−1.3223	− 6.4248	− 2.8513	0.1867	−17.7372
2.75	−0.4007	−5.0881	− 8.2548	−0.0542	−0.1625	− 0.3791	− 3.1677	0.4332	−18.4310
3.25	0.0163	−5.2764	− 8.9900	5.9555	1.4536	8.8627	− 3.7137	1.5948	−19.5428
3.75	0.4603	−5.4889	− 9.9996	13.7612	3.2120	20.1853	− 4.5107	4.1251	−20.9774
4.50	1.1087	−5.8063	−12.0422	29.1660	4.8974	40.9608	− 6.2359	11.4738	−23.6548
6.00	2.2065	−6.2051	−17.6678	75.5000	10.5953	96.6906	−11.4627	43.7140	−30.0779

Table 6. Electron moments for BH calculated at centre of mass for different internuclear separations for CI wave functions

r (Bohr)	Dipole moment $\langle z \rangle$	Second Moments $\langle x^2 \rangle$	$\langle z^2 \rangle$	Third Moments $\langle z^3 \rangle$	$\langle y^2 z \rangle$	$\langle y^2 z \rangle$	Quadrupole moment $\frac{1}{2}\langle 3z^2 - r^2 \rangle$	Octopole moment $\frac{1}{2}\langle 5z^2 - 3r^2 z \rangle$	$\langle r^2 \rangle$
1.75	−0.8519	−4.8355	−7.0672	−5.7377	−2.1310	−9.9998	−2.2317	0.6554	−16.7383
2.3291	−0.6127	−4.9700	−7.4946	−2.8016	−1.0817	−4.9650	−2.5246	0.4437	−17.4346
2.75	−0.3631	−5.0865	−7.8995	0.3370	−0.0635	0.2100	−2.8130	0.5275	−18.0724
3.25	0.0071	−5.2338	−8.4566	5.3851	1.3488	8.0826	−3.2228	1.3387	−18.9243
3.75.	0.3340	−5.3621	−8.9991	10.6621	2.6899	16.0418	−3.6370	2.5924	−19.7231
4.50	0.6071	−5.4974	−9.6716	17.1181	4.2020	25.5320	−4.1742	4.4973	−20.6664
6.00	0.5258	−4.5340	−9.6720	20.4779	5.2462	30.9703	−4.1380	4.7393	−20.7401

Table 7. The centroid of charge and "size"[a] of the NO of various wave functions at equilibrium separation[b]

Wave Function	NO	Centroid of charge x_a	y_a	z_a	$\langle r_i^2 \rangle_a$
SCF	1	0.0	0.0	1.1638	−0.1435
	2	0.0	0.0	−0.2196	−3.2546
	3	0.0	0.0	1.7339	−5.7383
CI[c]	2	0.0	0.0	−0.5933	−2.7336
	3	0.0	0.0	2.0677	−4.5233

[a] Size defined as $|(\chi | r_i^2 \chi)_a|^{1/2}$ – see text.

[b] Coordinates: B 0.0, 0.0, 1.1645, H 0.0, 0.0, −1.1645, centre of mass 0.0, 0.0, 0.9661.

[c] First NO for CI wave functions same as first SCF MO for same basis.

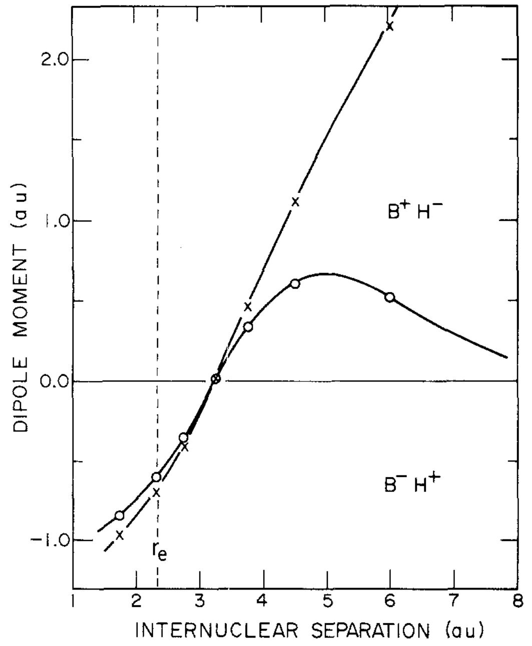

Fig. 1. The variation of the dipole moment of BH with internuclear separation. (x = SCF, o = CI)

For the CI wave function, the same configuration is observed with a slight shifting of the centres.

Table 8 gives the centroids of charge at a separation of 6 Bohr for these wave functions. The SCF function shows two distributions centred on B and one nearer the H atom, which corresponds to a B^+H^- picture. The CI function shows the outer SCF orbital dividing into two charge distributions centred one on each atom in agreement with the observed dissociation products, B and H neutral atoms.

Table 8. The centroid of charge and "size"[a] of the NO of various wave functions at separation of 6.0 a.u.[b]

Wave Function	NO	Centroid of charge			
		x_a	y_a	z_a	$\langle r_i^2\rangle_a$
SCF	1	0.0	0.0	2.9997	− 0.1429
	2	0.0	0.0	3.0763	− 4.3804
	3	0.0	0.0	−1.1742	−12.2082
CI D	1	0.0	0.0	2.9997	− 0.1429
	2	0.0	0.0	3.0222	− 4.4342
	3	0.0	0.0	−0.4488	−13.1120
	4	0.0	0.0	2.2698	−18.0350

[a] Size defined as $|(\chi|r_i^2\chi)_a|^{1/2}$ – see text.

[b] Coordinates: B 0.0, 0.0, 3.0, H 0.9, 0.0, −3.0, centre of mass 0.0, 0.0, 2.4888.

In addition the "size" of the electron distributions in the orbitals is given by the root mean square distance of an electron associated with that distribution from the centroid of charge [17]. This is given mathematically by the integral $|(\chi|r_i^2\chi)_a|^{1/2}$. Tables 7 and 8 include the values of the integral $(\chi|r_i^2\chi)_a$. As expected, the first MO is very localized while the outer orbitals are somewhat larger. On going to a CI wave function, the outer orbitals contract slightly, especially the third one. Since the centroids of these orbitals are further apart for the CI wave function, this does not conflict with the earlier observation that the *total* electron distribution increases with inclusion of CI.

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