Maximum overlap approximation calculations on polyatomic molecules. V. Principal equations of the extended-maximum-overlap-approximation method

R. BOČA

Department of Inorganic Chemistry, Slovak Technical University,
880 37 Bratislava

Received 13 December 1978

Accepted for publication 23 March 1979

The basic formulae of the EMOA method have been simplified in the present paper. This method is a general one for the construction of hybrid atomic orbitals in arbitrary polycentric molecules and is based on the maximum overlap principle. Resulting simple matrix equations for hybridization coefficients can be solved by a fast converging iterative procedure so that much computing time is saved.

Общий метод ЭМОА конструкции гибридных атомных орбиталей в любых многоатомных молекулах, основанный на принципе максимального перекрывания, был значительно упрощен. Результирующую матричную формулу для коэффициентов гибридизации можно решить быстро сходящимся итеративным процессом при большом сокращении вычислительного времени.

To the most progressive quantum chemical methods for the calculation of ground-state molecular properties proposed in the recent decade belongs the PCILO (Perturbative Configuration Interaction using Localized Orbitals) method [1—4]. In this method, the zeroth-order wave function is constructed using the basis set of strictly localized (two-centre) molecular orbitals (SLMOs). Then, the many-body perturbation theory is applied and the corrected (perturbed) wave function, the electronic energy, and electronic properties of the molecule are expressed by algebraic formulae in which only the integrals over the basis set of SLMOs occur. In order to obtain the basis set of SLMOs it is suitable to apply the well-known principle of hybridization; any SLMO can be calculated in the form of a linear combination of a pair of hybrid atomic orbitals.

However, the construction of the hybrid atomic orbitals (HAOs) from pure atomic orbitals is not a trivial problem in the general case. First of all, in a molecule which possesses little or no symmetry, the criteria for determining optimum hybrids are not unambiguously defined. The most commonly used criteria are: (i) criteria based on properties of projection operators [5—9]; (ii) the geometric criteria [10—13]; (iii) the maximum overlap criterion [14—22]. It must be pointed out that the maximum overlap criterion gives the HAOs which can be deviated from the directions of bonds.

The general method for construction of HAOs in polycentric molecules, based on the maximum overlap criterion, was proposed in paper [23]; it is called the EMOA (Extended Maximum Overlap Approximation) method. Some of its applications were described in the previous papers of this series [23—26]. In this method the hybridization coefficients are calculated solving a matrix equation by an iterative procedure. As a matter of fact, the EMOA method in its original form is rather time-consuming, as the diagonalization of a large-dimensional matrix is used in each step of the iterative procedure. In order to save the computing time, the basic matrix formulae of the EMOA method are simplified in this paper. However, only the sub-set of bonding HAOs can be obtained by the EMOA method. Then, additional criteria for construction of nonbonding HAOs (lone lobes) are to be proposed. It is the second problem on which this paper is concentrated.

Method

In the presented method, the total binding energy of a molecule, ε, is written in the form of a sum of particular binding energies, $E_{i,j}^{m,n}$, over all bonds [23, 25]

$$\varepsilon = \frac{1}{2}\sum_{m,n}^{G}\sum_{i}^{N_m}\sum_{j}^{N_n} E_{i,j}^{m,n}(1-\delta_{m,n})\,\delta_{\mathrm{f}(m,i),\,\mathrm{f}(n,j)} \qquad (1)$$

Here, G is the number of atoms and N_m is the number of bonding HAOs on the individual m-th atom. An a priori stated bonding situation in a molecule can be fully described by the discrete topological function $\mathrm{f}(m,i)$; the values of this function are equal to the ordering numbers of bonds (Fig. 1). The topological

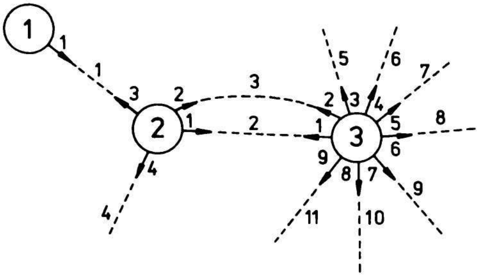

Fig. 1. Numbering system for definition of the topological function.
○ Atoms; → HAOs; ——— bonds (including lone lobes as nonconnected bonds).

function is useful for selecting a pair of HAOs ψ_i^m and ψ_j^n which "form" a given μ-th bond on the condition $\mathrm{f}(m, i)=\mu=\mathrm{f}(n, i)$. In eqn (*1*), according to the maximum overlap principle, the particular binding energies are directly proportional to the overlap integrals of optimum HAOs

$$E_{i,j}^{m,n}=K_{i,j}^{m,n} \quad \langle \psi_i^m \mid \psi_j^n \rangle \tag{2}$$

The weighing parameters $K_{i,j}^{m,n}$ are regarded as transferable from one molecule to another and they can be evaluated in a semiempirical way [23, 25]. The hybridization coefficients, $a_{i,k}^m$, which transform a basis set of atomic orbitals $\{\chi_k^m\}$ into a set of bonding HAOs $\{\psi_i^m\}$ (on each m-th atom) can be arranged into a block-diagonal (nonsquare) matrix $\boldsymbol{A}$ defined as

$$\begin{pmatrix} \psi_1^1 \\ \cdot \\ \cdot \\ \psi_{N_1}^1 \\ \cdot \\ \cdot \\ \psi_i^m \\ \cdot \end{pmatrix} = \begin{pmatrix} a_{1,1}^1 & \dots & a_{R_1,1}^1 & 0 & 0 & \cdot\cdot \\ \cdot & & \cdot & \cdot & \cdot & \\ \cdot & & \cdot & \cdot & \cdot & \\ a_{N_1,1}^1 & & a_{N_1,R_1}^1 & \dots\ 0 & 0 & \cdot\cdot \\ \cdot & & \cdot & \cdot & \cdot & \\ \cdot & & \cdot & \cdot & \cdot & \\ 0 & & 0 & a_{i,1}^m & a_{i,R_m}^m & \\ \cdot & & \cdot & \cdot & \cdot & \\ \cdot & & & & \cdot & \end{pmatrix} \begin{pmatrix} \chi_1^1 \\ \cdot \\ \cdot \\ \chi_{R_1}^1 \\ \cdot \\ \cdot \\ \chi_1^m \\ \cdot \\ \cdot \\ \chi_{R_m}^m \end{pmatrix} \tag{3}$$

or

$$\{\psi\}=\boldsymbol{A}\ \{\chi\} \tag{4}$$

In order to obtain an equation for the hybridization matrix $\boldsymbol{A}$, eqn (*1*) is to be rewritten in the matrix form as follows

$$\varepsilon=\frac{1}{2}\ \mathrm{Tr}\,(\boldsymbol{E}) \tag{5}$$

where

$$\boldsymbol{E}=\boldsymbol{A}\boldsymbol{S}\boldsymbol{A}^T\boldsymbol{Q}^T \tag{6}$$

In eqn (*6*), $\boldsymbol{S}$ is the overlap integral matrix of atomic orbitals, and the topological weighing parameters matrix $\boldsymbol{Q}$ is defined as

$$Q_{p,q}=K_{i,j}^{m,n} \quad (1-\delta_{m,n})\ \delta_{\mathrm{f}(m,i),\mathrm{f}(n,j)} \tag{7}$$

for indices

$$p = N_1 + N_2 + \ldots + N_{m-1} + i \tag{8}$$

$$q = N_1 + N_2 + \ldots + N_{n-1} + j \tag{9}$$

As it was proved in paper [23], for optimum HAOs which maximize the molecular binding energy the following equation holds

$$\boldsymbol{A} = (\boldsymbol{QAS}^T\boldsymbol{SA}^T\boldsymbol{Q}^T)^{-1/2}\boldsymbol{QAS}^T \tag{10}$$

It is the basic formula of the EMOA method which can be solved by an iterative procedure. Eqn (*6*) slightly differs from the original form [23] in the definition of the matrix $\boldsymbol{Q}$ but it is more convenient for molecules consisting of π bonds. In order to calculate the inverse square roots in eqn (*10*), the diagonalization procedure is to be used in each iteration. It is, of course, the most time-consuming step of the EMOA method. This step is tedious especially in the case of a large number of bonding HAOs in a molecule.

In order to exclude the diagonalization of large-dimensional matrices, eqn (*10*) is to be modified. It was pointed out that the matrix $\boldsymbol{A}$ must take a block-diagonal form. This condition was secured in the original form of the EMOA method by additional neglecting overlap integrals between formally nonbonded HAOs at each step of the iterative procedure. However, this condition is equivalent to the other one, *e.g.* the matrix $\boldsymbol{QAS}^T$ is to be a block-diagonal one; the form of the matrix $\boldsymbol{QAS}^T$ must be the same as the form of the matrix $\boldsymbol{A}$. Owing to the block-diagonal form of matrices $\boldsymbol{A}$ and $\boldsymbol{QAS}^T$, eqn (*10*) is fully factorized on a set of the following equations

$$\boldsymbol{A}_m = (\boldsymbol{B}_m\boldsymbol{B}_m^T)^{-1/2}\boldsymbol{B}_m \tag{11}$$

Here, $\boldsymbol{A}_m$ is the matrix of hybridization coefficients, $a^m_{i,k}$; it transforms a basis set of pure atomic orbitals into a set of bonding HAOs on the given m-th atom. In eqn (*11*), the matrix $\boldsymbol{B}_m$ is defined as

$$(B_m)_{i,k} = \sum_n^G \sum_j^{N_n} \sum_l^{R_n} \langle \chi^m_k | \chi^n_l \rangle \, a^n_{j,l} \, K^{m,n}_{i,j} \, (1-\delta_{m,n}) \, \delta_{f(m,i),f(n,j)} \tag{12}$$

(R_n is the number of AOs on the n-th atom.) Then, the formula for the molecular binding energy takes the form

$$\varepsilon = \frac{1}{2} \sum_m^G \mathrm{Tr}\,[(\boldsymbol{B}_m\boldsymbol{B}_m^T)^{1/2}] \tag{13}$$

Eqns (*11*) and (*12*) can be solved by the following iterative procedure. Firstly, an initial approximation of hybridization coefficients is to be chosen. For example, we can start from canonical (*sp*, sp^2, sp^3, *etc.*) HAOs: they determine the initial approximation of matrices ${}^{(0)}\boldsymbol{A}_m$. Then, the first approximation of ${}^{(1)}\boldsymbol{B}_m$ is calculated by means of eqn (*12*), and the matrix product $(\boldsymbol{B}_m\boldsymbol{B}_m^T)$ is diagonalized with an unitary matrix $\boldsymbol{U}_m$

$$(\boldsymbol{B}_m\boldsymbol{B}_m^T)^{-1/2} = \boldsymbol{U}_m^T\boldsymbol{D}_m^{-1/2}\boldsymbol{U}_m \tag{14}$$

Here, in conformity with the postulate of the maximum trace of the matrix $\boldsymbol{E}$, we accept the positive sign of the square roots of the reciprocal elements of the diagonal matrix $\boldsymbol{D}_m$. Finally, the first approximation of the matrix ${}^{(1)}\boldsymbol{A}_m$ is calculated using the equation

$${}^{(1)}\boldsymbol{A}_m = ({}^{(1)}\boldsymbol{B}_m\,{}^{(1)}\boldsymbol{B}_m^T)^{-1/2}\,{}^{(1)}\boldsymbol{B}_m \tag{15}$$

This process is repeated up to the self-consistency and its convergence is rapid (after 4—6 iterations). By the simplified approach proposed here only the number G of matrices $\boldsymbol{B}_m\boldsymbol{B}_m^T$ is to be diagonalized; the maximum dimensions of these matrices are 4 (or less than 4) in the valence s—p basis set and 9 (or less than 9) in the valence s—p—d basis set.

The calculation off the lone lobe functions (nonbonding HAOs) is the second problem dealt with in this paper. By the EMOA method only the number N_m of bonding HAOs has been calculated on each m-th atom. Full basis set of orthonormal HAOs (that includes both the sub-set of bonding HAOs and the sub-set of lone lobes) contains R_m functions on the m-th atom. It is important to point out that, in the general case, an infinite number of different sub-sets of lone lobes can be constructed for which the orthogonality conditions are well fulfilled, *e.g.* the lone lobes are orthogonal with bonding HAOs and also with themselves. For example, we can apply the Schmidt's orthogonalization process starting from the sub-set of bonding HAOs to which the number $R_m - N_m$ of linear independent functions is added. However, the finding of these linear independent functions is not a trivial problem, in the general case.

In order to obtain the lone lobe functions, the use of the projection operators' technique is proposed here. The matrix

$$\boldsymbol{P}_m = \boldsymbol{A}_m^T\boldsymbol{A}_m \tag{16}$$

is a projector defined in the sub-space of bonding HAOs as the following equations are fulfilled

$$\boldsymbol{P}_m^2 = \boldsymbol{P}_m \tag{17}$$

$$\mathrm{Tr}(\boldsymbol{P}_m) = N_m \tag{18}$$

Then, the complementary projector

$$\boldsymbol{L}_m = \boldsymbol{I}_m - \boldsymbol{P}_m \tag{19}$$

($\boldsymbol{I}_m$ is the unit matrix) is defined in the sub-space of the lone lobes and it holds

$$\mathbf{L}_m^2 = \mathbf{L}_m \tag{20}$$

$$\mathrm{Tr}(\mathbf{L}_m) = R_m - N_m \tag{21}$$

From the total number R_m, only the number $R_m - N_m$ of rows in the matrix $\mathbf{L}_m$ is linearly independent. In practice, we construct a trial square matrix

$$\mathbf{A}'_m = \begin{pmatrix} \mathbf{A}_m \\ \mathbf{L}'_m \end{pmatrix} \tag{22}$$

where the nonsquare matrix $\mathbf{L}'_m$ is represented by the selected number $R_m - N_m$ of rows of the square matrix

$$\mathbf{L}_m = \mathbf{I}_m - \mathbf{A}_m^T \mathbf{A}_m \tag{23}$$

If the trial matrix $\mathbf{A}'_m$ has the nonzero determinant, the Schmidt's orthogonalization process can be applied on rows of the matrix $\mathbf{A}'_m$ and the resulting matrix (after normalization of rows) is obtained.

We often wish to construct a set of lone lobes which are equivalent in their s, p or d characters. In the valence s—p basis set the s character, n_s, and the p character, n_p, of a lone lobe are defined as

$$n_s = \left\{ \left[1 - \sum_{i=1}^{N_m} (a_{i,s}^m)^2 \right] \Big/ (R_m - N_m) \right\}^{1/2} \tag{24}$$

$$n_p = (1 - n_s^2)^{1/2} \tag{25}$$

The construction of the equivalent sub-set of lone lobes is usually based on the geometric ideas [27]. Here, the alternative algebraic way is proposed. The resulting (equivalent) lone lobe $\mathbf{A}_r^m \equiv \{a_{r,s}^m;\ a_{r,x}^m;\ a_{r,y}^m;\ a_{r,z}^m\}$ is calculated in the form of a linear combination of two normalized initial lone lobes as

$$\mathbf{A}_r^m = \alpha\, \mathbf{A}_t^m + \beta\, \mathbf{A}_u^m \tag{26}$$

for $r, t \neq u \in \langle N_m + 1, R_m \rangle$. From the condition of the given n_s or n_p character of the lone lobe $\mathbf{A}_r^m$, the following equations are obtained for the coefficients α and β

$$a_{r,s}^m = n_s = \alpha\, a_{t,s}^m + \beta\, a_{u,s}^m \tag{27}$$

$$\alpha^2 + \beta^2 = 1 \tag{28}$$

Eqns (*27*) and (*28*) have the solutions

$$\alpha = (n_s a_{t,s}^m \pm a_{u,s}^m \sqrt{D}) / [(a_{u,s}^m)^2 + (a_{t,s}^m)^2] \tag{29}$$

$$\beta = (n_s - \alpha\, a_{t,s}^m) / a_{u,s}^m \tag{30}$$

if the following conditions are fulfilled

$$D=(a^{m}_{u,s})^2+(a^{m}_{t,s})^2-n_s^2\geq 0 \tag{31}$$

$$-1\leq\alpha,\beta\leq+1 \tag{32}$$

In the case of the valence *s—p—d* basis set the process proposed here can be generalized: in the combination *via* eqn (*26*) more initial normalized vectors are to be considered.

Results and discussion

In order to demonstrate applications of the EMOA method on calculations of polyatomic molecules, $Cu(NH_3)_2(NCX)_2$ complexes were chosen ($X=O$, S). Here, 15 atoms, 47 AOs, and 36 bonding HAOs are to be considered. Calculations in plane geometries were performed using the *Slater* [28] basis set of AOs with exponents for the Cu-AOs according to *Gouterman* [29]. The bonding situation with 14σ and 4π bonds was considered and the corresponding bonding model is illustrated in Fig. 2. The topological function describing this bonding situation is given in Table 1. All weighing parameters $K^{m;n}_{i,j}$ were equal to 1.0 in the calculations, *e.g.* eqns (*1*), (*5*) or (*13*) correspond to the sum of the overlap integrals of bonding HAOs over bonds. For the individual iterations, the trace of the matrix $\boldsymbol{E}$ is listed in Table 2. Starting from the basis set of canonical HAOs the convergence of the iterative procedure is good for the complex when $X=S$. Using the calculated EMOA-HAOs of the complex for $X=S$ as a starting set in calculations of the complex for $X=O$, the convergence of the EMOA method is accelerated (Table 2). The resulting HAOs in the $d^{\beta}sp^{\alpha}$ form are listed in Table 3.

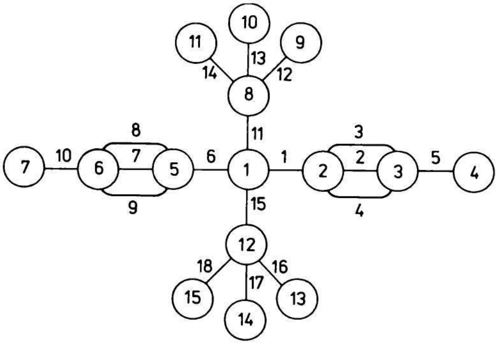

Fig. 2. Bonding model of the $Cu(NH_3)_2(NCX)_2$ complexes ($X=O$, S).

In the presented calculations a diagonalization of large-dimensional matrices is unnecessary; only 15 small matrices are to be diagonalized in each step of the

Table 1

Topological function f(*m*, *i*) describing the bonding situation in $Cu(NH_3)_2(NCX)_2$ complexes

Atom	*m*	*i*-th bonding HAO on the *m*-th atom				
		1	*2*	*3*	*4*	
Cu	1	6	1	11	15	f(*m*, *i*)
N	2	1	2	3	4	
C	3	2	5	3	4	
X	4	5				
N	5	7	6	8	9	
C	6	10	7	8	9	
X	7	10				
N	8	11	12	13	14	
H	9	12				
H	10	13				
H	11	14				
N	12	15	16	17	18	
H	13	16				
H	14	17				
H	15	18				

Table 2

Convergence of the EMOA method for $Cu(NH_3)_2(NCX)_2$ complexes

Iteration	Trace of the matrix $\boldsymbol{E}$	
	$X = S^{a}$	$X = O^{b}$
1	16.646000	19.831090
2	19.447232	19.859515
3	19.642283	19.859547
4	19.661161	19.859547
5	19.662689	
6	19.662856	
7	19.662879	
8	19.662882	
9	19.662883	

a) Start from canonical HAOs.
b) Start from resulting HAOs of X = S.

Table 3

Resulting hybridization in $d^{\beta}sp^{\alpha}$ on individual atoms of $Cu(NH_3)_2(NCX)_2$ complexes

Bond		X = O			X = S		
A—B	No.	A(α)	B(α)	B(β)	A(α)	B(α)	B(β)
N—Cu	*11*	2.406	2.227	1.227	2.408	2.239	1.239
N—Cu	*1*	0.864	1.896	0.896	0.802	1.886	0.886
N—C	*2*	1.157	0.821		1.247	1.111	
C—X	*5*	1.218	0.602		0.900	1.192	

iterative procedure. There are 7 matrices of the dimension 4 × 4 (1 for Cu, 4 for N, and 2 for C atoms) and 8 matrices of the dimension 1 × 1 (6 for H and 2 for X = O, S atoms). Results obtained by means of eqns (*11*) and (*12*) are exactly the same as those produced by the original form of the EMOA method, but the computing time is much shorter. It is the principal result of this communication.

The second application presented here concerns the dependence of the PCILO method on the use of various sets of lone lobes. In the PCILO method, the second-order ground-state molecular energy can be written in the form

$$E_T^{(2)} = E_T^{(0)} + E_{\mathrm{del}}^{(2)} + E_{\mathrm{cor}}^{(2)} \qquad (33)$$

where $E_T^{(0)}$ is the zeroth-order ground-state molecular energy (that corresponds to the single Slater determinant of occupied SLMOs), $E_{\mathrm{del}}^{(2)}$ is the second-order delocalization energy, and $E_{\mathrm{cor}}^{(2)}$ is the second-order correlation energy. The method for calculating the individual energy terms (the modified PCILO method) is described elsewhere [3, 4] on the CNDO/2 or INDO levels of Hamiltonian approximations. In the HF molecule (with the bonding model H—$\overline{\underline{\mathrm{F}}}|$) the bonding HAO on the fluorine atom was maintained in $sp^{0.595}$ as it follows from the EMOA method. Three different sub-sets of lone lobes were considered: (i) a set with two pure AOs of the p type; (ii) a set of nonequivalent lone lobes produced by the Schmidt's orthogonalization procedure; and (iii) a set of equivalent lone lobes. The resulting individual energy terms obtained by the modified PCILO method are listed in Table 4. As the single Slater determinant is invariant with respect to unitary transformation applied on the set of occupied spin-orbitals, so the zeroth-order ground-state energy $E_T^{(0)}$ has to be maintained by all sub-sets of lone lobes. It is well fulfilled in our calculations (Table 4). However, the corrected second-order ground-state energy depends on the use of lone lobes. As for

Table 4

Molecular energy terms (eV) of HF molecule in the second order of the perturbation theory obtained by the modified PCILO method using various basis sets of HAOs

Hybridization in sp^{α} form		Basis set of HAOs		
		a	*b*	*c*
Bonding HAO	*1*:	0.595	0.595	0.595
Lone lobe	*2*:	1.680	7.041	7.041
Lone lobe	*3*:	Pure p_x	15.082	7.041
Lone lobe	*4*:	Pure p_y	4.361	7.041
A) CNDO/2 Hamiltonian				
$-E_T^{(0)}$		770.162	770.162	770.162
$-E_{\mathrm{del}}^{(2)}$		1.793	2.124	2.157
$-E_{\mathrm{cor}}^{(2)}$		0.175	0.175	0.175
$-E_T^{(2)}$		772.131	772.462	772.495
B) INDO Hamiltonian				
$-E_T^{(0)}$		734.600	734.600	734.600
$-E_{\mathrm{del}}^{(2)}$		1.820	2.142	2.174
$-E_{\mathrm{cor}}^{(2)}$		0.273	0.270	0.270
$-E_T^{(2)}$		736.693	737.013	737.044

a) Nonequivalent lone lobes; two pure AOs (p_x and p_y).
b) Nonequivalent lone lobes.
c) Equivalent lone lobes.

calculations it can be concluded that the best molecular energy after perturbative configuration interaction is produced with the sub-set of equivalent lone lobes. It is the second principal result of this communication.

References

1. Diner, S., Malrieu, J. P., Jordan, F., and Gilbert, M., *Theor. Chim. Acta 15,* 100 (1969).
2. Jordan, F., Gilbert, M., Malrieu, J. P., and Pincelli, U., *Theor. Chim. Acta 15,* 211 (1969).
3. Boča, R. and Pelikán, P., *Theor. Chim. Acta 50,* 11 (1978).
4. Boča, R., *Collect. Czech. Chem. Commun. 44,* 3141 (1979).
5. Polák, R., *Int. J. Quantum Chem. 4,* 271 (1970).
6. Polák, R., *Int. J. Quantum Chem. 6,* 1077 (1972).
7. Roby, K. R., *Mol. Phys. 28,* 1441 (1974).

8. Boyle, L. L., *Chem. Phys. Lett. 5,* 493 (1970).
9. Deplus, A., Leroy, G., and Peeters, D., *Theor. Chim. Acta 36,* 109 (1974).
10. Pauling, L., *J. Amer. Chem. Soc. 53,* 1367, 3225 (1931).
11. Slater, J. C., *Phys. Rev. 38,* 1109 (1931).
12. Del Re, G., *Theor. Chim. Acta 1,* 188 (1963).
13. Del Re, G., *Int. J. Quantum Chem. 1,* 293 (1967).
14. Murrell, J. N., *J. Chem. Phys. 32,* 767 (1960).
15. Golebiewski, A., *Acta Phys. Pol. 23,* 243 (1963).
16. Gilbert, T. L. and Lykos, P. G., *J. Chem. Phys. 34,* 2199 (1961).
17. Lykos, P. G. and Schmeising, H. N., *J. Chem. Phys. 33,* 288 (1961).
18. Mårtensson, O. and Öhrn, Y., *Theor. Chim. Acta 9,* 133 (1967).
19. Lindner, P. and Mårtensson, O., *Acta Chem. Scand. 23,* 429 (1969).
20. Randič, M. and Maksič, Z. B., *Theor. Chim. Acta 3,* 59 (1965).
21. Randič, M. and Maksič, Z. B., *Chem. Rev. 72,* 43 (1972).
22. Hubač, I., Laurinc, V., and Kvasnička, V., *Chem. Phys. Lett. 13,* 357 (1972).
23. Boča, R., Pelikán, P., Valko, L., and Miertuš, S., *Chem. Phys. 11,* 229 (1975).
24. Miertuš, S., Boča, R., Pelikán, P., and Valko, L., *Chem. Phys. 11,* 237 (1975).
25. Boča, R., Pelikán, P., and Valko, L., *Chem. Zvesti 33,* 289 (1979).
26. Boča, R., Pelikán, P., and Valko, L., *J. Mol. Struct. 50,* 161 (1978).
27. Claverie, P., Daudey, J. P., Diner, S., Giessner-Prettre, Cl., Gilbert, M., Langlet, J., Malrieu, J. P., Pincelli, U., and Pullman, B., *QCPE, Program No. 220.* Indiana University, Indiana.
28. Slater, J. C., *Phys. Rev. 36,* 57 (1930).
29. Zerner, M. and Gouterman, M., *Theor. Chim. Acta 4,* 44 (1966).

Translated by R. Boča