

Electronegativity: Quantum Observable

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Received 2 October 2008; accepted 6 October 2008

Published online 21 November 2008 in Wiley InterScience (www.interscience.wiley.com).

DOI 10.1002/qua.21957

ABSTRACT: The question whether electronegativity may be considered as quantum observable is responded in positive by a special ionization-affinity wave function construction within the fermionic Fock space for the valence state of a chemical system. The present approach consecrates electronegativity as the minus eigen-energy of the unperturbed occupied valence state involved in addition and release of electrons by atoms-in-molecules interactions. This way, the earlier crisis raised by Bergmann and Hinze concerning the assignment of chemical potential to electronegativity quantification is here solved in the favor of Parr density functional picture. © 2008 Wiley Periodicals, Inc. *Int J Quantum Chem* 109: 733–738, 2009

Key words: chemical potential; Fock space; creation and annihilation operators; eigen-energy

Introduction

Since the birth of modern chemistry due to the Berzelius works [1], the electroaffinity or electronegativity (χ) concept had proved its reliability in quantifying the chemical domains of the atomic and molecular structure and reactivity. Being considered as primarily of ionic type by Pauling [2], electronegativity acquired both the accepting and donating characters with introduced spectroscopic definition of Mulliken [3]. Although many qualitative features were then revealed through the pioneering works of Gordy, Sanderson, and Allred–Rochow [4–6], the quantum consecration of electronegativity comes with the Iczkowski–Mar-

grave [7], Hinze–Whitehead–Jaffé, [8] Huheey [9], and Klopman [10], pictures in which it directly relates with the total energy of the system throughout the engaged charges with environment. Still, the simpler and most meaningful form of electronegativity was achieved with the advent of density functional theory when identified by Parr et al. with the chemical potential μ of the system [11],

$$\chi = -\mu = -\left(\frac{\partial E}{\partial N}\right)_{V(\mathbf{r})} = -\left(\frac{\delta E}{\delta \rho(\mathbf{r})}\right)_{V(\mathbf{r})}, \quad (1)$$

being last identity assured since the density functional basic relationship between number of particles (N) and electronic density $\rho(\mathbf{r})$ was employed for the fixed external applied potential $V(\mathbf{r})$. However, because the chemical potential places electronegativity among global indices characterizing a

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chemical structure the question whether electrophilic and nucleophilic issues of reactivity may still be encompassed had generate a sort of a crisis as emphasized by the Bergmann and Hinze treatment in the context of basic quantum mechanics [12].

The present work like to put some light on the quantum mechanical description of electronegativity through field perturbations that may arise on a valence state for a chemical system; it eventually leads with a rigorous assessment of electronegativity as quantum observable while preserving its inner character in describing reactivity by means of electronic exchange of atoms and molecules.

Method

FERMIONS IN FOCK SPACE

Among the systems with indiscernible particles, fermions have a special character driven by the Pauli exclusion principle [13]. As a consequence, the fermionic space is built on mixing only two Hilbert (sub) spaces [14], namely that one containing the vacuum state only

$$\mathcal{H}_{(0)} \supset \{|0\rangle\}, \langle 0|0\rangle = 1 \quad (2a)$$

together with that one accounting for single particle occupancy

$$\mathcal{H}_{(1)} \supset \{|\phi_s\rangle\}_{s \in N}, \langle \phi_s | \phi_t \rangle = \delta_{s,t} = \begin{cases} 1, s = t \\ 0, s \neq t \end{cases} \quad (2b)$$

with the states being of the hydrogenic wave functions type, $|\phi_s\rangle \sim |\phi_{nml}\rangle$, for instance. In these conditions, the fermionic states are contained by the so-called Fock space:

$$\begin{aligned} \mathcal{F}_N &= \underbrace{(\mathcal{H}_{(0)} \oplus \mathcal{H}_{(1)}) \otimes (\mathcal{H}_{(0)} \oplus \mathcal{H}_{(1)}) \otimes \dots \otimes (\mathcal{H}_{(0)} \oplus \mathcal{H}_{(1)})}_{N \text{ times}} \\ & \quad (3a) \end{aligned}$$

here written for an N-electronic system, with the basic one-particle composed Hilbert space containing the set of virtual states as

$$\mathcal{H}_{(0)} \oplus \mathcal{H}_{(1)}: |0\rangle, |\phi_1\rangle, \dots, |\phi_s\rangle, \dots \quad (3b)$$

Now, out of (3a) there can be abstracted the fermionic Fock $\mathcal{F}_{\{|0\rangle, |1\rangle\}}$ space only by identifying the vacuum sector:

$$|0\rangle_N = \underbrace{|0\rangle \otimes |0\rangle \otimes \dots \otimes |0\rangle}_{N \text{ times}} \quad (4a)$$

among the uni-particle sector:

$$\begin{aligned} |1(\phi_k)\rangle_N &= \frac{1}{\sqrt{N!}} \left(\underbrace{|\phi_k\rangle \otimes |0\rangle \otimes \dots \otimes |0\rangle}_{N \text{ states}} \right. \\ & \quad \left. + \dots + \underbrace{|0\rangle \otimes |0\rangle \otimes \dots \otimes |\phi_k\rangle}_{N \text{ states}} \right) \quad (4b) \end{aligned}$$

to be considered for further defining the fermionic operators.

Accordingly, we may define the annihilation and creation operators on the fermionic Fock space, $\mathbf{a}(\phi), \mathbf{a}^+(\phi): F_{\{|0\rangle, |1\rangle\}} \rightarrow F_{\{|0\rangle, |1\rangle\}}$, respectively as:

$$\mathbf{a} = |0\rangle\langle 1|, \quad (5a)$$

$$\mathbf{a}^+ = |1\rangle\langle 0|, \quad (5b)$$

since only two projections are possible within the bi-dimensional unitary operator:

$$1 = |0\rangle\langle 0| + |1\rangle\langle 1| = \mathbf{a}\mathbf{a}^+ + \mathbf{a}^+\mathbf{a} = \{\mathbf{a}, \mathbf{a}^+\} \quad (6)$$

on the vacuum and uni-particle sectors, while noting the rules:

$$\langle 0|1\rangle = \langle 1|0\rangle = 0, \quad (7a)$$

$$\langle 0|0\rangle = \langle 1|1\rangle = 1. \quad (7b)$$

The check out of these operatorial actions,

$$\mathbf{a}^+|0\rangle = |1\rangle\langle 0|0\rangle = |1\rangle, \quad (8a)$$

$$\mathbf{a}|1\rangle = |0\rangle\langle 1|1\rangle = |0\rangle \quad (8b)$$

proves the consistency of the present construction and offers the key in next description of the density functional electronegativity as given by Eq. (1).

QUANTUM PERTURBATION OF THE VALENCE STATE

Here we will consider the "valence state" reality as a sort of stationary state providing special constrains for excited states are considered so that an eigen-equation for its unperturbed state $|\psi_0\rangle$ with associated eigen-energy E_0 may be conventionally written:

$$H|\psi_0\rangle = E_0|\psi_0\rangle. \quad (9)$$

This way, the normalization condition for the valence wave-function allows the unitary operator decomposition on the vacuum and uni-particle occupancies as:

$$\begin{aligned} 1 &= \langle\psi_0|\psi_0\rangle = \langle\psi_0|1|\psi_0\rangle = \langle\psi_0|(\mathbf{a}\mathbf{a}^+ + \mathbf{a}^+\mathbf{a})|\psi_0\rangle \\ &= \langle\psi_0|\mathbf{a}\mathbf{a}^+|\psi_0\rangle + \langle\psi_0|\mathbf{a}^+\mathbf{a}|\psi_0\rangle = |\langle 0|\psi_0\rangle|^2 + |\langle 1|\psi_0\rangle|^2 \\ &= (1 - \rho_0) + \rho_0, \rho_0 \in [0,1], \quad (10) \end{aligned}$$

from where the associate wave function projections yield:

$$\langle 0|\psi_0\rangle = \langle\psi_0|0\rangle = \sqrt{1 - \rho_0}, \quad (11a)$$

$$\langle 1|\psi_0\rangle = \langle\psi_0|1\rangle = \sqrt{\rho_0}. \quad (11b)$$

However, when electronegativity field of the system-environment (bath) complex is acting on this valence state it has to be calculated throughout the perturbation factor λ as:

$$\chi_\lambda = -\frac{\partial\langle E_\lambda\rangle}{\partial\rho_\lambda} = -\frac{\partial\langle E_\lambda\rangle}{\partial\lambda} \frac{\partial\lambda}{\partial\rho_\lambda}, \quad (12)$$

while the modified wave-state may be seen either along the uni-particle creation state through adding an electron to the system,

$$\begin{aligned} |\psi_\lambda^A\rangle &= (1 + \lambda\mathbf{a}^+\mathbf{a})|\psi_0\rangle = |\psi_0\rangle + \lambda|1\rangle\langle 0|0\rangle\langle 1|\psi_0\rangle = |\psi_0\rangle \\ &\quad + \lambda\sqrt{\rho_0}|1\rangle, \quad (13a) \end{aligned}$$

or along the vacuum sector through giving up an electron from the system:

$$\begin{aligned} |\psi_\lambda^I\rangle &= (1 + \lambda\mathbf{a}\mathbf{a}^+)|\psi_0\rangle = |\psi_0\rangle + \lambda|0\rangle\langle 1|1\rangle\langle 0|\psi_0\rangle = |\psi_0\rangle \\ &\quad + \lambda\sqrt{1 - \rho_0}|0\rangle, \quad (13b) \end{aligned}$$

properly called as affinity and ionization wave-functions, respectively.

Note that both these states perturb the same initial state $|\psi_0\rangle$ characterized by the eigen-equation (9). Therefore, it makes sense to consider them at once when computing the electronegativity as the power of ionization-affinity perturbation of the concerned valence state. In other words, the perturbed quantum occupancy and energy are to be written as a sort of transition amplitudes of the Hamiltonian and of particle operators, between the virtually possible ionization and affinity wave states, no matter in which (temporal) order, normalized at their scalar product for limiting occupancy $0 \ll \rho_0 \leq 1$ (since they are not necessary orthogonal while related), written respectively as:

$$\langle E_{\lambda \in \mathfrak{R}}^{I \leftrightarrow A} \rangle = \frac{\langle \psi_\lambda^I | H | \psi_\lambda^A \rangle}{\langle \psi_\lambda^I | \psi_\lambda^A \rangle_{0 \ll \rho_0 \leq 1}}, \quad (14)$$

$$\rho_{\lambda \in \mathfrak{R}}^{I \leftrightarrow A} = \frac{\langle \psi_\lambda^I | \mathbf{a}^+ \mathbf{a} | \psi_\lambda^A \rangle}{\langle \psi_\lambda^I | \psi_\lambda^A \rangle_{0 \ll \rho_0 \leq 1}}. \quad (15)$$

Starting with computing the perturbed occupancy, we get successively:

$$\begin{aligned} \rho_\lambda &= \frac{\langle \psi_0 | (1 + \lambda\mathbf{a}\mathbf{a}^+) \mathbf{a}^+ \mathbf{a} (1 + \lambda\mathbf{a}^+\mathbf{a}) | \psi_0 \rangle}{\langle \psi_0 | (1 + \lambda\mathbf{a}\mathbf{a}^+) (1 + \lambda\mathbf{a}^+\mathbf{a}) | \psi_0 \rangle_{0 \ll \rho_0 \leq 1}} \\ &= \frac{\langle \psi_0 | (\mathbf{a}^+ \mathbf{a} + \lambda\mathbf{a}^+ \mathbf{a}\mathbf{a}^+ \mathbf{a} + \lambda\mathbf{a}\mathbf{a}^+ \mathbf{a}^+ \mathbf{a} + \lambda^2 \mathbf{a}\mathbf{a}^+ \mathbf{a}^+ \mathbf{a}\mathbf{a}^+ \mathbf{a}) | \psi_0 \rangle}{\langle \psi_0 | (1 + \lambda\mathbf{a}^+ \mathbf{a} + \lambda\mathbf{a}\mathbf{a}^+ + \lambda^2 \mathbf{a}\mathbf{a}^+ \mathbf{a}^+ \mathbf{a}) | \psi_0 \rangle_{0 \ll \rho_0 \leq 1}} \\ &= \frac{1 + \lambda}{\rho_0(1 + \lambda\rho_0)} \quad (16) \end{aligned}$$

based on the above rules and on their extensions:

$$\langle \psi_0 | \mathbf{a}\mathbf{a}^+ \mathbf{a}^+ \mathbf{a} | \psi_0 \rangle = \langle \psi_0 | 0 \rangle \langle 1 | 1 \rangle \langle 0 | 0 \rangle \langle 0 | 0 \rangle \langle 1 | \psi_0 \rangle = 0, \quad (17a)$$

$$\langle \psi_0 | \mathbf{a}^+ \mathbf{a}\mathbf{a}^+ \mathbf{a} | \psi_0 \rangle = \langle \psi_0 | 1 \rangle \langle 0 | 0 \rangle \langle 1 | 1 \rangle \langle 0 | 0 \rangle \langle 1 | \psi_0 \rangle = \rho_0, \quad (17b)$$

$$\begin{aligned} \langle \psi_0 | \mathbf{a}\mathbf{a}^+ \mathbf{a}^+ \mathbf{a}\mathbf{a}^+ \mathbf{a} | \psi_0 \rangle &= \langle \psi_0 | 0 \rangle \langle 1 | 1 \rangle \langle 0 | 1 \rangle \langle 0 | 0 \rangle \langle 1 | 1 \rangle \langle 0 | 0 \rangle \\ &\quad \times \langle 1 | \psi_0 \rangle = 0. \quad (17c) \end{aligned}$$

With this expression the perturbation factor is firstly obtained

$$\lambda = \frac{\rho_\lambda - \rho_0}{\rho_0(1 - \rho_\lambda)} \quad (18)$$

to provide the expression:

$$\frac{\partial \lambda}{\partial \rho_\lambda} = \frac{(1 + \lambda \rho_0)^2}{\rho_0(1 - \rho_0)}. \quad (19)$$

$$\langle E_\lambda \rangle = \frac{\langle \psi_0 | (1 + \lambda \mathbf{a} \mathbf{a}^+) H (1 + \lambda \mathbf{a}^+ \mathbf{a}) | \psi_0 \rangle}{\langle \psi_0 | (1 + \lambda \mathbf{a} \mathbf{a}^+) (1 + \lambda \mathbf{a}^+ \mathbf{a}) | \psi_0 \rangle_{\lambda_0 \ll \rho_0 \leq 1}} = \frac{\langle \psi_0 | H | \psi_0 \rangle + \lambda \langle \psi_0 | H \mathbf{a}^+ \mathbf{a} | \psi_0 \rangle + \lambda \langle \psi_0 | \mathbf{a} \mathbf{a}^+ H | \psi_0 \rangle + \lambda^2 \langle \psi_0 | \mathbf{a} \mathbf{a}^+ H \mathbf{a}^+ \mathbf{a} | \psi_0 \rangle}{1 + \lambda \rho_0} = E_0 \frac{1 + \lambda}{1 + \lambda \rho_0} \quad (20)$$

since the eigen-equation of the nonperturbed valence state by means of its energy is accommodated with the creation-annihilation quantum rules:

$$\begin{aligned} \langle \psi_0 | H \mathbf{a}^+ \mathbf{a} | \psi_0 \rangle &= \langle \psi_0 | H | 1 \rangle \langle 0 | 0 \rangle \langle 1 | \psi_0 \rangle \\ &= E_0 \langle \psi_0 | 1 \rangle \langle 1 | \psi_0 \rangle = E_0 \rho_0, \end{aligned} \quad (21a)$$

$$\begin{aligned} \langle \psi_0 | \mathbf{a} \mathbf{a}^+ H | \psi_0 \rangle &= \langle \psi_0 | 0 \rangle \langle 1 | 1 \rangle \langle 0 | H | \psi_0 \rangle \\ &= \langle \psi_0 | 0 \rangle \langle 0 | \psi_0 \rangle E_0 = E_0 (1 - \rho_0), \end{aligned} \quad (21b)$$

while the term

$$\langle \psi_0 | \mathbf{a} \mathbf{a}^+ H \mathbf{a}^+ \mathbf{a} | \psi_0 \rangle = \langle \psi_0 | 0 \rangle \langle 1 | 1 \rangle \langle 0 | H | 1 \rangle \langle 0 | 0 \rangle \langle 1 | \psi_0 \rangle \quad (22)$$

is set to zero based on the usual second quantization form of the Hamiltonian [15],

$$H = \sum_{pq} h_{pq} \mathbf{a}_p^+ \mathbf{a}_q + \frac{1}{2} \sum_{pqts} g_{pqts} \mathbf{a}_p^+ \mathbf{a}_t^+ \mathbf{a}_q \mathbf{a}_s. \quad (23)$$

Note that Eq. (23) through its one and two particle terms (with the corresponding integrals h_{pq} and g_{pqts} over the $p, q, t,$ and s orbitals) will produce the zero giving quantum operation

$$\langle 0 | H | 1 \rangle \sim \langle 0 | \mathbf{a}_p^+ \dots | 1 \rangle = \langle 0 | 1 \rangle \langle 0 | \dots | 1 \rangle = 0. \quad (24)$$

Finally, from Eq. (20) we get:

$$\frac{\partial \langle E_\lambda \rangle}{\partial \lambda} = E_0 \frac{1 - \rho_0}{(1 + \lambda \rho_0)^2}, \quad (25)$$

while when combined with the Eq. (19) the electronegativity (12) is simple shaped as:

$$\begin{aligned} \chi_\lambda &= -\frac{E_0}{\rho_0} = -\mu_0 \\ &= \begin{cases} \infty & , \rho_0 \rightarrow 0 (E_0 < 0) \\ -E_0 = -\langle \psi_0 | H | \psi_0 \rangle, & \rho_0 \rightarrow 1 \end{cases} \end{aligned} \quad (26)$$

Then, going to the perturbed energy calculation the specific relationships are:

this way revealing itself as a quantum (chemical potential) observable; i.e. electronegativity is spanned between the pure electronic attraction and either electronic attraction or release characters for the states described by the upper and lower branches of (26), respectively.

Discussion

Since the result of Eq. (26) is obtained no matter which transition is considered, from ionization to affinity or vice-versa, we may widely interpret the electronegativity as the quantum state (energy) either (or equally in the phenomenological sense) to be added or depleted with an electron in an inter-electronic field; with this the conventional Mulliken spectroscopic definition as the semi-sum of the ionization and affinity energy naturally follows [3]. Worth observing that seeing quantum electronegativity as an eigen-energy is not in contradiction with its genuine chemical potential nature; enough recalling that the chemical potential stands in solid state for the Fermi level, i.e. the last possible filled level with bounded electrons (at 0 K) [16]. This way, we may still interpret electronegativity as the quantum state able to either attract or to cede electrons in fermionic Fock space. Therefore, the electronegativity was proved as a viable quantum concept, with an observable character since it may be associated with the eigen-energy of the unperturbed valence state ready to be involved in charge exchange without photochemical stimulation (i.e., in atoms-in-molecules interactions).

Nevertheless, since quantum electronegativity as a global measure for the valence was previously criticized by Bergmann and Hinze [12] grounded on a similar construction worth pointing here in detail their flaws. Actually, they thought to consider only the ionization type of perturbation working with a perturbed wave function as

$$\begin{aligned}
 |\psi_\lambda^I\rangle &= (1 + \lambda \mathbf{a})|\psi_0\rangle = |\psi_0\rangle + \lambda|0\rangle\langle 1|\psi_0\rangle \\
 &= |\psi_0\rangle + \lambda\sqrt{\rho_0}|0\rangle \quad (27a)
 \end{aligned}$$

while using implicitly (though conjugation) the counterpart affinity-perturbed state as well:

$$\begin{aligned}
 |\psi_\lambda^A\rangle &= (1 + \lambda \mathbf{a}^+)|\psi_0\rangle = |\psi_0\rangle + \lambda|1\rangle\langle 0|\psi_0\rangle \\
 &= |\psi_0\rangle + \lambda\sqrt{1 - \rho_0}|1\rangle. \quad (27b)
 \end{aligned}$$

However, such construction suffers from a qualitative consistency since association of the vacuum sector $|0\rangle$ with uni-particle occupancy ρ_0 in (27a) and the uni-particle sector $|1\rangle$ with vacuum occupancy $(1 - \rho_0)$ in (27b) for the ionization and affinity wave vectors; such construction is peculiar being avoided by the present one, see relations (13a) and (13b), respectively. Moreover, also quantitatively, there seems that Bergmann and Hinze have neglected the mix products $\langle\psi_0|0\rangle\langle 1|\psi_0\rangle$ and $\langle\psi_0|1\rangle\langle 0|\psi_0\rangle$ like the vacuum projected probability $|\langle 0|\psi_0\rangle|^2$ in the above employed limiting occupancy, $0 \ll \rho_0 \leq 1$, from where the result in that electronegativity cannot be considered like the global (observable) quantity of chemical potential—an error later noticed by the Parr and Yang [17]. The present approach widely clarifies these aspects.

Still, we may notice that electronegativity is a versatile quantity in quantum chemistry, but always with a strong relationship with total or valence eigen-energy or with the global characters of a concerned system, e.g. effective potential of the nuclei, total number of particles involved, chemical action, chemical hardness [18, 19]. However, it was also shown that density functional electronegativity may systematically be expressed according with the total energy expansion in charge and applied potential, with an absolute unfold (the so-called Putz electronegativity) closely related with chemical potential (the so-called Parr or differential electronegativity) and with Mulliken (or chemical) one [20, 21]. Further studies on deriving total energy functionals [22] based on electronegativity are thus possible and are currently in progress.

Conclusion

According to the accepted epistemological rules, science has to have both a study object and specific tools for endeavoring it. In this respect, while chemistry is mainly devoted to study the electronic struc-

ture and reactivity it still develops specific tools among which electronegativity stands as a crucial concept in assessing stability and transformations of atoms and molecules. With this motivation, electronegativity acquired many qualitative and quantitative forms until the recent density functional chemical potential assignment of Parr [11]. However, it was argued that although most useful as chemical potential such identification lacks for quantum mechanical support as inferred by Bergmann and Hinze [12]. Whishing to clarify the nature of quantum electronegativity, the present approach exposes how the ionization-affinity perturbation on the valence state of a chemical system may be conducted to recover the eigen-energy of that state as an observable quantity for the electronegativity within the density functional chemical potential framework; the Parr picture is thus confirmed with fundamental quantum mechanical arguments while the flaws of Bergmann-Hinze treatment are here identified. Therefore, electronegativity may provide a viable quantum rationale for future chemical analysis of bond and bonding throughout energy density functional and its observable character [23, 24].

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