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## Application of Many-Body Rayleigh-Schrödinger Perturbation Theory to Excitations Accompanying Photoionization in Molecules

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Excitations accompanying photoionizations in molecules (shake-up processes) are studied on the basis of degenerate many-body Rayleigh-Schrödinger perturbation theory (MB — RSPT).

These excitations give rise to satellite lines accompanying ionization lines. Decomposition of correlation effects in these processes into those corresponding to single-particle excitation and single-particle ionization is discussed.

### INTRODUCTION

During a photoionization process a second electron may be promoted from a filled orbital to an unfilled but bond orbital or to continuum. The former case is named the »shake-up« process and the latter is called the »shake — off« process. In the »shake — up« process a sharp line to the low- kinetic energy side of the main photoelectron peak is observed at an energy  $E = h\nu - E_h - E^*$ , where  $E_h$  is the ionization energy and  $E^*$  is the additional excitation energy. In the »shake — off« process, two electrons are emitted and we observe a continuum to the low — kinetic energy side of the main peak is observed<sup>1</sup>. In particular growing interest in photoelectron spectroscopy requires understanding of these phenomena.

The many — body techniques are recently applied in the study of photoionization in atoms and molecules. The Green function technique<sup>2-9</sup>, equation of motion method<sup>10-13</sup>, the natural transition orbitals method<sup>14,15</sup> as well as quasi-degenerate and/or degenerate MB — RSPT<sup>16,17</sup> have been used to study processes in photoelectron spectroscopy. MB-RSPT has been used to calculate molecular excitation energies<sup>18</sup> and intermolecular interaction energies<sup>19,20</sup>.

Very recently, the Green function technique has been applied to the calculation of »shake up« energies<sup>21,22</sup>. These are found to be secondary poles of the Green function.

In the present work we study excitations accompanying photoionizations by degenerate MB-RSPT. MB-RSPT is widely used in the study of the correlation energy problem, which is of recent ab — initio numerical interest<sup>23-25</sup>. The diagrammatic technique is a very powerful tool for studying the correlation energy problem to various orders and various forms of MB-RSPT (different separation of the Hamiltonian) for ground state as well as for excitation processes. This technique provides the possibility to see »structure« of the corre-

lation contribution and also to see the dominant diagrammatic contributions, which can then be summed up to all orders of perturbation theory.

The aim of this paper is to formulate the problem of excitation processes accompanying photoionization by degenerate MB-RSPT and to show how the correlation energy can be decomposed into single-particle excitation and single particle ionization.

For the sake of completeness we briefly recapitulate the theory.

### *The Formulation of Excitations Accompanying Photoionization by Degenerate MB — RSPT*

Degenerate many-body Rayleigh-Schrödinger perturbation theory has been formulated in the »microscopic« theory of N-fermion systems as a suitable tool for the introduction of the effective interaction<sup>26-30</sup>. The first complete derivation of this many-body theory was performed by Brandow<sup>29</sup> in order to give a correct theoretical definition for the nuclear shell model potential.

Let us assume that a perturbed Hamiltonian of an atomic or molecular system may be written as

$$H = l_0 + H_0 + H_1 \quad (1)$$

where  $l_0$  is the scalar quantity,  $H_0$  is an unperturbed Hamiltonian and  $H_1$  is a perturbation. In the second quantization formalism these operators have the form

$$H_0 = \sum_A \varepsilon_A X_A^+ X_A$$

$$H_1 = (1/2) \sum_{ABCD} \langle AB | v | CD \rangle X_A^+ X_B X_D X_C - \sum_{AB} \langle A | w | B \rangle X_A^+ X_B \quad (2)$$

where  $X_A^+$  and  $X_B$  are creation and annihilation operators defined on the orthonormal set of spinorbitals which are solutions of the one — particle eigenproblem  $(h + w) | A \rangle = \varepsilon_A | A \rangle$ . One of the basic concepts of degenerate MB-RSPT is a properly preselected non-degenerate »core« state vector<sup>31</sup>.

$$|\bar{\Phi}_0\rangle = \prod_{A \in \text{FS}} X_A^+ | 0 \rangle \quad (3)$$

$$H_0 |\bar{\Phi}_0\rangle = E_0^{(0)} |\bar{\Phi}_0\rangle; E_0^{(0)} = \sum_{A \in \text{FS}} \varepsilon_A \quad (4)$$

where the index A runs over all occupied (taken from the Fermi sea) one-particle functions, and  $| 0 \rangle$  is the normalized vacuum state vector. Using this core state vector we generate a certain finite d-dimensional model space

$$\Omega_0 \equiv \{ |\bar{\Phi}_u\rangle = \prod_u^+ |\bar{\Phi}_0\rangle \} \quad (5)$$

where  $\prod_u^+$  are ordered products with fixed numbers of creation and annihilation operators. We assume that the model space  $\Omega_0$  is an eigenspace of the unperturbed Hamiltonian with an eigenenergy  $E_\alpha^{(0)}$ . Let  $P_0$  be a projector onto  $\Omega_0$ , then  $\text{Tr}(P_0) = d$  and  $\bar{H} = P_0 H_0 = H_0 P_0 = E_\alpha^{(0)} P_0$ . If we assume that the perturbation  $H_1$  is an »analytical perturbation«<sup>32</sup>, then there exist d perturbed eigenvalues

$$\{ E_{\lambda_1}, E_{\lambda_2}, \dots, E_{\lambda_d} \} = \{ E_\lambda; \lambda \in \mathbf{M} \} \quad (6)$$

tending to the unperturbed energy  $E_\alpha^{(0)}$  when the perturbation  $H_1$  is »switched — off«, i. e.  $H_1 \rightarrow 0$ . The energy difference  $\Delta E_\lambda = E_\lambda - E_0$ , where  $\lambda \in M$  and  $E_0$  is the perturbed energy of the core subsystem, are then determined as eigenvalues of the following non — hermitian  $d$  — dimensional model eigenproblem

$$H_{RS} |\Psi_\lambda\rangle = \Delta E_\lambda |\Psi_\lambda\rangle \tag{7}$$

$$H_{RS} = (E_\lambda^{(0)} - E_0^{(0)}) P_0(\alpha) + G_{RS} \tag{8}$$

$H_{RS}$  is the model Hamiltonian defined in the model space  $\Omega_0$ , and  $G_{RS}$  is the model interaction defined in the framework of the diagrammatic technique as follows

$$\begin{aligned} G_{RS} &= \{P_0 H_1 P_0\}_{LC} + \sum_{n=1}^{\infty} \sum_{k_1 k_2 \dots k_n} \{P_0 H_1 G^{k_1} H_1 G^{k_2} H_1 \dots G^{k_n} H_1 P_0\}_{LC} = \\ &= \{P_0 H_1 P_0\}_{LC} + \{P_0 H_1 \frac{1 - P_0}{E_\alpha^{(0)} - H_0} H_1 P_0\}_{LC} + \\ &+ \{P_0 H_1 \frac{1 - P_0}{E_\alpha^{(0)} - H_0} H_1 \frac{1 - P_0}{E_\alpha^{(0)} - H_0} H_1 P_0\}_{LC} - \\ &- \{P_0 H_1 \frac{1 - P_0}{(E_\alpha^{(0)} - H_0)^2} H_1 P_0 H_1 P_0\}_{LC} + \dots \end{aligned} \tag{9}$$

where the subscript LC means that only linked connected diagrams<sup>33</sup> are contributing, and nonnegative summation indices are determined by the two conditions

$$\begin{aligned} \sum_{i=1}^n K_i &= n \\ \sum_{i=1}^m K_i &\geq m \text{ for } m = 1, 2, \dots, n-1 \end{aligned} \tag{10}$$

The powers of the unperturbed propagator  $G^k$  from (9) are defined by

$$G^K = \begin{cases} -P_0 & \text{for } K = 0 \\ \frac{1 - P_0}{(E_\alpha^{(0)} - H_0)^K} & \text{for } K \geq 1 \end{cases} \tag{11}$$

The model space  $\Omega_0$  for a »shake-up« process may be spanned by the following unperturbed state vector:

$$|\Phi'\rangle = X_K^+ X_I X_H |\Phi_0\rangle \tag{12}$$

$$H_0 |\Phi'\rangle = (E_0^{(0)} + \varepsilon_K - \varepsilon_I - \varepsilon_H) |\Phi'\rangle \tag{13}$$

where  $E_0^{(0)}$  is the unperturbed eigenenergy defined by (4) and  $I, H \in FS, K \notin FS, I \neq H$ . Generally speaking, this unperturbed state vector  $|\Phi'\rangle$  may be interpreted in two alternative ways: (i) as ionization of an H electron and simultaneous excitation of an I electron to a K state, or (ii) as ionization of an I electron and excitation of an H electron to a K state. It should be noted

that these two possible mechanisms of the given »shake-up« process cannot be distinguished in the frame work of the time-independent formalism.

To specify clearly the states K, I, and H, we assume that in both cases ionization is described by H. Since the model space  $\Omega_0$  spanned by  $|\Phi'\rangle$  is a one-dimensional space ( $d = 1$ ) it is not necessary to solve the model eigenproblem (7), its eigenvector  $|\Psi_\lambda\rangle$  can be simply identified with  $|\Phi'\rangle$ ,

$$\Delta E' = \langle \Phi' | H_{RS} | \Phi' \rangle = \varepsilon_K - \varepsilon_I - \varepsilon_H + \langle \Phi' | G_{RS} | \Phi' \rangle, \quad (14)$$

where  $E' = \Delta E' + E_0$  is the energy of the »shake-up« state described in the zero-order approximation by  $|\Phi'\rangle$ , i. e. the difference  $\Delta E'$  is direct measure of the energy of the »shake-up« process. Following the diagrammatic rules the matrix elements  $\langle \Phi' | G_{RS} | \Phi' \rangle$  from (14) can be divided into three contributions (see Figure 1.)

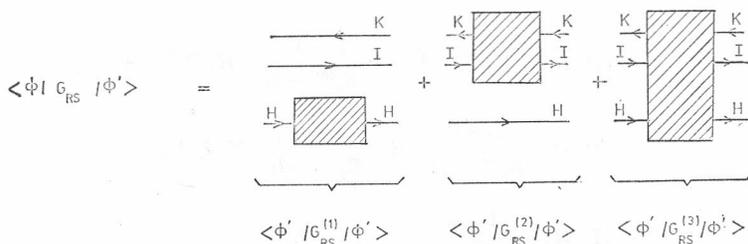


Figure 1. The diagrammatic decomposition of the matrix elements  $\langle \Phi' | G_{RS} | \Phi' \rangle$

$$\begin{aligned} \langle \Phi' | G_{RS} | \Phi' \rangle &= \langle \Phi' | G_{RS}^{(1)} | \Phi' \rangle + \\ &+ \langle \Phi' | G_{RS}^{RS(2)} | \Phi' \rangle + \langle \Phi' | G_{RS}^{(3)} | \Phi' \rangle \end{aligned} \quad (15)$$

Here the matrix element  $\langle \Phi' | G_{RS}^{(1)} | \Phi' \rangle$  represents the sum of all possible diagrammatic contributions with free particle K and hole I lines. These diagrams contribute<sup>16</sup> to the ionization potential  $I_H = E_H - E_0$ , where  $E_H$  is the energy of single ionized systems described in the zero-order approximation by  $X_H | \Phi_0 \rangle$ ,

$$I_H = -\varepsilon_H + \langle \Phi' | G_{RS}^{(1)} | \Phi' \rangle \quad (16)$$

The matrix element  $\langle \Phi' | G_{RS}^{(1)} | \Phi' \rangle$  represents the correlation contribution to the single particle ionization process. The matrix element  $\langle \Phi' | G_{RS}^{(2)} | \Phi' \rangle$  represents the sum of all possible diagrammatic contributions with a free hole H line. These diagrams contribute<sup>18</sup> to the excitation energy  $\Delta E_{I \rightarrow K} = E_{I \rightarrow K} - E_0$ , where  $E_{I \rightarrow K}$  is the energy of an excited electroneutral system described in the zero-order approximation by  $X_{K^+} X_I | \Phi_0 \rangle$ ,

$$\Delta E_{I \rightarrow K} = \varepsilon_K - \varepsilon_I + \langle \Phi' | G_{RS}^{(2)} | \Phi' \rangle \quad (17)$$

The matrix element  $\langle \Phi' | G_{RS}^{(2)} | \Phi' \rangle$  represents the correlation contribution to the single particle excitation process.

Finally, the matrix element  $\langle \Phi' | G_{RS}^{(3)} | \Phi' \rangle$  represents the sum of all possible remaining diagrams that are not included in the previous two matrix elements. These diagrams may, of course contain also free particle and/or hole

lines, but of such types which have not been included in the above — mentioned cases (16) and (17). Then the »shake-up« energy determined by (14) can be rewritten in the following form

$$\Delta E' = I_H + \Delta E_{I \rightarrow K} + \langle \Phi' | G_{RS}^{(3)} | \Phi' \rangle \quad (18)$$

This means that when the »shake-up« process is treated in the framework of degenerate MB-RSPT it is possible to extract the ionization potential  $I_H$  and the excitation energy  $\Delta E_{I \rightarrow K}$ . This fact might be of value for (i) simple interpretation of the given »shake-up« process, and (ii) also for a less elaborate calculation of the »shake-up« energy  $\Delta E'$ , since the quantities  $I_H$  and  $\Delta E_{I \rightarrow K}$  can be properly estimated from photoelectron and UV spectroscopy, respectively. In order to simplify the calculation it is advantageous to formulate the theory in such a way as to calculate that the energy difference between the »shake-up« energy and the ionization potential  $\Delta E' - I_H$  instead of performing the direct calculation of »shake-up« energies.

$$\Delta E' - I_H = \Delta E_{I \rightarrow K} + \langle \Phi' | G_{RS}^{(3)} | \Phi' \rangle \quad (19)$$

The theory formulated in this way is true of spinorbitals. Its application to a concrete molecular system is more complicated, because the model space  $\Omega_0$ , as will be outlined below, is not a one-dimensional space. This model space can, of course, be factorized on the subspaces with lower dimension using the constants of motion  $S^2$  and  $S_Z$ .

In the following part we give a short outline of the present theory of closed-shell molecular systems for which the solution of restricted Hartree-Fock equations<sup>34</sup> is known. Let us study N-electron ( $N = 2n$ ) molecular systems which are in the zero-order approximation described by the closedshell »core« state vector

$$|\Phi_0\rangle = \prod_{i=1}^n (X_{i\alpha}^+ X_{i\beta}) 0 \quad (20)$$

where  $X_{i\alpha}^+$  ( $X_{i\beta}$ ) is the creation operator of the spin-orbital

$$|A\rangle = |i\rangle |\alpha\rangle \quad (|A'\rangle = |i\rangle |\beta\rangle).$$

Assuming that the orbitals  $\{|i\rangle\}$  are solution of the restricted HF equations in the canonical form, we can express the Hamiltonian in the so called normal form

$$H = \langle \Phi_0 | H | \Phi_0 \rangle + H_0 + H_1 \quad (21)$$

$$H_0 = \sum_A \varepsilon_A N [X_A^+ X_A] \quad (22)$$

$$H_1 = (1/2) \sum_{ABCD} \langle AB | v | CD \rangle N [X_A^+ X_B^+ X_D X_C] \quad (23)$$

where  $\langle \Phi_0 | H | \Phi_0 \rangle$  is the restricted HF energy,  $\varepsilon_A$  is the HF orbital energy which does not depend on the spin part, i. e.  $\varepsilon_A = \varepsilon_{A'} = \varepsilon_i$ . The expression  $N[\dots]$  is the normal product defined with respect to  $|\Phi_0\rangle$ <sup>35</sup>. The model space  $\Omega_0$  for the calculation of the »shake-up« process may now be spanned by three unperturbed state vectors with  $E_\alpha^{(0)} = E_0^{(0)} + \varepsilon_k - \varepsilon_i - \varepsilon_h$  and  $S_Z = 1/2$ .

$$|A_1\rangle = X_{k\beta}^+ X_{i\beta} X_{h\beta} |\Phi_0\rangle \quad (24)$$

$$|A_2\rangle = X_{k\alpha}^+ X_{i\alpha} X_{h\beta} |\Phi_0\rangle \quad (25)$$

$$|A_3\rangle = X_{k\alpha}^+ X_{i\beta} X_{h\alpha} |\Phi_0\rangle \quad (26)$$

The spin-symmetry adapted functions are the following:

$$|\Phi_Q'\rangle = \frac{1}{\sqrt{3}} (|A_1\rangle + |A_3\rangle - |A_2\rangle) \quad (\text{quartet}) \quad (27)$$

$$|\Phi_{D_1}'\rangle = \frac{1}{\sqrt{6}} (2|A_1\rangle + |A_2\rangle - |A_3\rangle) \quad (\text{doublet}) \quad (28)$$

$$|\Phi_{D_2}'\rangle = \frac{1}{\sqrt{2}} (|A_1\rangle + |A_2\rangle) \quad (\text{doublet}) \quad (29)$$

An expression analogous to (19) can now be obtained. Of course, the model space  $\Omega_0$  can now be factorized onto quartet (one-dimensional) and doublet (two-dimensional) subspaces. Substituting (26) into (18), we obtain the energy difference between the quartet »shake-up« peak and the ionization peak as a one-dimensional problem. The quartet  $\Delta E_Q' - I_H$  energy is then given by

$$\Delta E_Q' - I_H = \Delta E_{I \rightarrow K} + \langle \Phi_Q' | G_{(3)}^{RS} | \Phi_Q' \rangle \quad (30)$$

In order to obtain the energy difference between the doublet states and the ionization peak, the two by two model characteristic problem (7) must be solved, since the doublet subspace is two-dimensional.

#### DISCUSSION

In recent work of Cederbaum<sup>21</sup> and Purvis and Öhrn<sup>22</sup> the Green functions were used to study the »shake-up« processes in photoelectron spectroscopy. »Shake-up« energies were found as secondary poles of the Green function. The present approach is based on degenerate BM-RSPT.

This approach provides the possibility of obtaining explicit formulae for »shake-up« energies or even more, for the energy difference between the »shake-up« and ionization peaks. It also shows that the decomposition of the correlation energy can be diagrammatically treated exactly.

For the application of the present theory to molecular systems which will be our next aim, let us say a few words about the structure of the basic formula (19). The energy difference between the »shake-up« peak and ionization peak is given by two terms. The first represents the simple excitation energy, while the second represents correlation and reorganization effects which are not included in the given ionization potential  $I_H$  and in the excitation energy  $\Delta E_{I \rightarrow K}$ . This fact can be very useful for application of this theory, since the term  $\Delta E_{I \rightarrow K}$  may be estimated from separate theoretical calculation which simplifies the calculations. This fact reduces the number of terms occurring in the model interaction  $G_{RS}$ .

In some cases, especially in the valence region (correlation satellites<sup>21</sup>), satellites are not due to ionization out of a specific shell nor to simultaneous excitation. The ionic state corresponding to the shake-up line »borrows«<sup>21</sup> intensity from several ionic states of the unperturbed form  $X_I |\Phi_0\rangle$ . This would mean that the space  $\Omega_0$  must be enlarged, e. g.  $\{X_{K_1}^+ X_{I_1}, X_{H_1}, X_{K_2}^+ X_{I_2}, X_{H_2}, \dots\}$

$|\Phi_0\rangle$  <sup>36</sup> We think that for these cases the formulation of the problem through quasi-degenerate MB-RSPT<sup>16,30</sup> might be of value because this fact can be taken into account through construction of unperturbed model space  $\Omega_0$  by  $P_0$  defined as

$$P_0 = \sum_1 |\Phi'_e\rangle \langle \Phi'_e| \quad (31)$$

where  $|\Phi'_e\rangle$  is identical with  $|\Phi'\rangle$  i. e.

$$|\Phi'_e\rangle \equiv |\Phi'\rangle = X_K^+ X_I X_H |\Phi_0\rangle \quad (32)$$

Note that the formulation of the problem in this paper corresponds to the case

$$P_0 |\Phi'_e\rangle \langle \Phi'_e| \quad (33)$$

Due to the fact that in many cases satellite lines are located in a small energy range, their identification can hardly be made on the ground of an approximate energy calculation alone. In these cases it is as necessary to know the intensities as is to know the energies<sup>36</sup>.

To conclude this article we note that it was not the aim of this work to present here the numerical results, but to formulate the problem of excitations accompanying photoionizations by degenerate MB-RSPT and to show how on the basis of this theory the correlation and reorganization effects corresponding to single particle ionization and to single particle excitation can be treated exactly using the diagrammatic technique. We believe that this should be useful for application of this theory and also for understanding of these phenomena.

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### SAŽETAK

**Primjena mnogo-čestične Rayleigh-Schrödingerove perturbacijske teorije na studij pobuđenja koja slijede fotoionizacijske procese u molekulama**

*I. Hubač i V. Kvasnička*

Pobuđenja koja slijede fotoionizacijske procese u molekulama uzrokuju pojavu satelitskih linija u spektrima. One su studirane primjenom mnogo-čestične Rayleigh-Schrödingerove perturbacijske teorije. Diskutirano je rastavljanje korelacijskih efekata na procese koji odgovaraju jednočestičnim pobuđenjima.

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