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Oscillator strength calculations in color centers of diamond and the role of spin

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Abstract. A generalized Hubbard model based on a molecular approach is used to calculate many electron wavefunctions of diamond vacancies. We have calculated the oscillator strength of the dipole transition rates from the ground states of the neutral and negatively charged vacancies. The ratio of the oscillator strengths is in very good quantitative agreement with the reported optical spectroscopic data. Electronic configurations in the ground and dipole allowed excited states are presented. With the proposed picture, the much larger oscillator strength of the negatively charged vacancy with respect to other experimentally investigated color centers N-V, H3, N3 and H4 is explained.

PACS. 61.72.Bb Theories and models of crystal defects – 61.72.Ji Point defects (vacancies, interstitials, color centers, etc.) and defect clusters – 71.55.-i Impurity and defect levels

1 Introduction

Color centers in diamond have been attributed to lattice vacancy and its corresponding defects [1,2]. The simplicity and rich physical content of the lattice vacancy in diamond have attracted a lot of interest for the last 50 years [1–6]. Interaction of electromagnetic field with correlated electrons of these centers can be approximated by the dipole interaction. Dipole transition is the main photon absorption mechanism in optical absorption spectroscopy, luminescence and hole burning measurements. These experiments are used to investigate the excited states of these correlated electrons [7–11].

Despite significant progress in the theoretical calculations of optical excitation energies of the diamond vacancies [12,13], there is no quantitative results on their oscillator strength which is defined as dipole transition intensity (rate) per center [14,15]. This is due to the difficulty of a computational method that introduces electron correlation into wavefunctions.

Calibrating the experimental data of dipole transition intensities or oscillator strengths for vacancy related defects in diamond, has been used widely to empirically estimate the concentration of vacancy related defects in synthesized diamond [16].

In addition to the conventional spectroscopic experiments for the quantification of absorption centers, color centers in diamond have been used as Qbits in quantum information technology (QIT) [18,17]. The interest in the excitations of these centers lies in the fact that they allow spin coherence to be effectively excited and manipulated by the use of optical laser fields while coupling to the environment, and hence the long spin coherence lifetime needed for optical memories and quantum computing is maintained [18].

For theoretical estimation of the oscillator strengths, one needs to calculate the Hamiltonian of the system to obtain accurate eigenfunctions of the vacancy electrons.

Experimental data of dipole transition rates provide an opportunity to test proposed theories which attempt to explain electronic states of the vacancies. The resulting eigenfunctions can be checked by this means.

Until now, Molecular Orbital (MO) and Configuration Interaction (CI) have been the methods used to manipulate excited states and to include tetrahedral symmetry, T_d, (Fig. 1) and spin properties of the vacancy electrons in their wavefunctions [1].

Construction of a CI wavefunction depends on the selection of the more probable configurations and the resultant wavefunctions are not exact eigenfunctions of the Hamiltonian.

We report results of the oscillator strength calculation for the dipole transitions of the neutral and negatively charged vacancies. Explicit forms of the degenerate ground and excited states's wavefunctions are used to do this calculation. No such calculation exists for these centers. Quantitative results are in very good agreement

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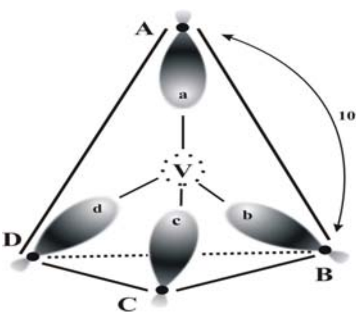

Fig. 1. Dangling orbitals of first nearest neighbor atoms of undistorted vacancy in diamond with tetrahedral symmetry.

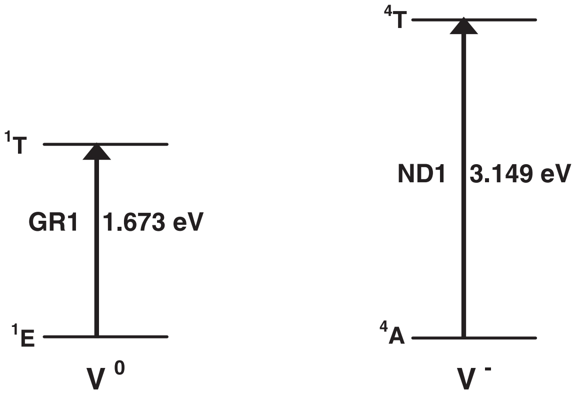

Fig. 2. Dipole transitions in the diamond vacancies are called GR1 and ND1 for the neutral and charged vacancies respectively, spatial degeneracy and spin multiplicity of the ground and excited states are labeled.

with reported optical spectroscopy data for dipole transition intensities. Electronic configurations in the ground and excited states will be reported and we will discuss the role of the ground state spin in the oscillator strength of the vacancies and other related color centers of diamond.

Two type of the vacancies i.e. neutral (V^0) and negatively charged (V^-), have been observed in the irradiated natural, synthesized and CVD diamond. The V^0 and V^- have four and five electrons which remain in the neighboring atomic sites after creation of the vacancy. In the V^0 there are four electron of four dangling bonds and in the V^-, there is one extra electron trapped in the vicinity due to presence of a donor Nitrogen impurity. The neutral and charged vacancies are characterized by the GR1 (1.673 eV) [3] and ND1 (3.149 eV) [4] absorption lines respectively. These lines are related to the first optically allowed electronic transition from the ground states of the vacancies (Fig. 2).

2 Calculations

2.1 Many body wavefunctions calculation

We use a Fock space representation in order to construct a Hilbert space of many electron basis (Eq. (1))

$$|\psi_i\rangle = |a_{i0}, a_{i1}, a_{i2}, ..., a_{i8}\rangle \tag{1}$$

a_{i1} to a_{i8} are -1 or $+1$ (for spin down and up on the site i respectively) to show the occupation condition of each vacancy atomic orbital. This basis is made from the atomic orbitals belonging to the four nearest neighbor (NN) atoms of the vacant site which are oriented inward into the center of the vacancy. The dangling orbitals of the NN atoms are labeled a, b, c and d for their connection to atoms A, B, C and D respectively in Figure 1.

Recent EPR experiments indicate that almost all of the unpaired electrons of the lattice vacancy in diamond are localized on the nearest neighbor atoms of the vacancy [20]. This implies that considering just these four dangling orbitals to calculate the electronic structure properties is reasonable [19].

To solve the problem of the many electron wavefunctions of these systems, we have introduced a generalized form of Hubbard Hamiltonian. The generalized Hubbard Hamiltonian for a four (five) electron system considers e-e correlation effects beyond the usual Hubbard Hamiltonian formalism. Correct space symmetry (T_d) and spin properties of the system can be incorporated. Solving this Hamiltonian for electronic states of the diamond vacancies allows for a unified and accurate treatment of many electron energy levels and wavefunctions for both vacancies of diamond. This is in contrast to a CI approach where its wavefunctions are not eigenstates of the Hamiltonian.

As the consequence of the spatial symmetry, T_d invariance, we were able to appropriately factorize the Hamiltonian.

$$H = t\sum_{ij,\sigma} c^{\dagger}_{i\sigma} c_{j\sigma} + U\sum_{i} n_{i\uparrow} n_{i\downarrow} + \frac{V}{2}\sum_{i\neq j,\sigma\sigma'} n_{i\sigma} n_{j\sigma'} + \frac{1}{2}\sum_{ijlm,\sigma\sigma'} X_{ijlm} c^{\dagger}_{i\sigma} c^{\dagger}_{j\sigma'} c_{m\sigma'} c_{l\sigma} \tag{2}$$

where i, j, l and m are atomic orbital indices of the vacancy ranging from 1 to 4 for a, b, c and d orbitals of Figure 1. σ, σ' are the spin indices that vary from 1 to 2. $c_{i\sigma}$ and $c^{\dagger}_{i\sigma}$ are annihilation and creation operators on the ith site with spin σ, respectively. The $n_{i\uparrow}$, $n_{i\downarrow}$ and $n_{i\sigma}$ are spin occupation number at the ith site. t is single particle hopping overlap integral. U and V are the Columbic overlap integrals. X_{ijlm} are the quantum exchange integrals that with tetrahedral symmetry (T_d), reduce to $X_1, ..., X_5$. The shape of the dangling orbitals a, b, c and d should be given before calculating Hamiltonian parameters (t, U, V, X_1,.., X_5). These parameters are defined as:

$$t = \langle i|T + V(r)|j\rangle, \quad U = \langle ii|\frac{1}{r}|ii\rangle, \quad V = \langle ij|\frac{1}{r}|ij\rangle$$

$$X_1 = \langle ij|\frac{1}{r}|ji\rangle, \qquad X_2 = \langle ii|\frac{1}{r}|ij\rangle, \quad X_3 = \langle ij|\frac{1}{r}|ik\rangle$$

$$X_4 = \langle ii|\frac{1}{r}|jk\rangle, \qquad X_5 = \langle ij|\frac{1}{r}|kl\rangle \tag{3}$$

Table 1. List of the eight parameters that are used to evaluate energy eigenfunctions of the vacancy. They are calculated based on Slater type orbital with exponent 1.562 and semi empirical value of U.

		t	U	V	$X1$	$X2$	$X3$	$X4$	$X5$
Present model	(eV)	-8.447	12.856	8.17	0.447	2.23	0.396	1.598	0.322

$T+V(r)$ is kinetic energy plus the ionic interaction energy of electron. The Hamiltonian calculation has been carried out in S_z basis. Then the Hamiltonian matrix was transformed to a new one in S^2 basis. Finally by exact diagonalization, we obtained complete eigenvectors of the Hamiltonian. Resultant eigenstates of the Hamiltonian have definite spin and symmetry degeneracy.

To calculate parameters of equation (3), we used a minimal basis set of Slater type orbitals (STO) [21]. We used an exponent of 1.562 for the vacancy orbitals of Figure 1. To obtain simultaneously GR1 and ND1 transition energies, the semi empirical value of 12.855 eV for Hubbard's U parameter (one center Coulombic integral) was used. This value is the same as that used by Coulson and Kearsley, with a Slater exponent equal to 1.595 to obtain proper GR1 transition energy [22]. Results are summarized in Table 1.

2.2 Oscillator strength calculations

To obtain dipole transition rates, one must calculate $\langle \Psi_i | \overrightarrow{r} | \Psi_f \rangle$ amplitude, where Ψ_i and Ψ_f are the initial and final many electron eigenfunctions of the Hamiltonian for degenerate ground and excited states. $\overrightarrow{r}$ is the dipole transition operator. After performing the calculation, the dipole transition intensity from each degenerate ground states of V^0 such as $|\Psi_i\rangle = |^1E,1\rangle$ to any degenerate excited state such as $|\Psi_f\rangle = |^1T,1\rangle$, was obtained from the following relation:

$$\left|\langle {}^1E,1|\overrightarrow{r}|{}^1T,1\rangle\right|^2 = \left(\sum_{i=1}^{4}\alpha_i^2 + \cos\theta\sum_{i,j=1}^{4}\alpha_i\alpha_j\right)\;|\overrightarrow{r}_m|^2 \quad (4)$$

where $\overrightarrow{r}_m$ is defined as $\langle m|\overrightarrow{r}|m\rangle$ and m represents a, b, c, and d orbitals. α's are obtained from the expansion coefficients of the Hamiltonian eigenfunctions on Fock basis. Details of the matrix element calculations that lead to equation (4) are given in Appendix A. The above relation is general and gives the optical absorption intensities as a function of the vacancy bond angle (θ) and expansion coefficients of the many electron eigenfunctions. For an undistorted vacancy in a tetrahedrally symmetric lattice such as diamond, the angle is $\theta = 109.5°$.

Equation (4) was used to calculate transition intensity of the GR1

$$I(GR1) \propto \frac{1}{2}\sum_{k=1}^{2}\sum_{l=1}^{3}\left|\langle {}^1T,l|\overrightarrow{r}|{}^1E,k\rangle\right|^2. \quad (5)$$

Table 2. Intensities of the transitions in the unit of $|r_m|$, from each twofold degenerate ground state ^1E to threefold degenerate exited state ^1T with zero S_z for the GR1 line.

	Ground States	$\lvert{}^1E,1\rangle$	$\lvert{}^1E,2\rangle$
Excited states	S_z	0	0
$\lvert{}^1T,1\rangle$	0	0.071	0.074
$\lvert{}^1T,2\rangle$	0	0.069	0.016
$\lvert{}^1T,3\rangle$	0	0.15	0.16
Total Rate:	-	0.29	0.25

In equation (5), l and k are degeneracy labels. Here we have summed over all degenerate three fold final states and averaged over two fold degenerate ground states. At thermal equilibrium, the occupation probability of all n degenerate ground states are equal and is $\frac{1}{n}$. Therefore, the transition rate should be averaged over all these states [23].

3 Results and discussions

All calculated values of the $|\langle {}^1T,l|\overrightarrow{r}|{}^1E,k\rangle|^2$ for different degenerate ground and excited states are listed in Table 2. The numbers in Table 2 are for $|\overrightarrow{r}_m| = 1$. By putting these numbers into equation (5), we obtain:

$$I(GR1) \propto 0.27\,|\overrightarrow{r}_m|^2. \quad (6)$$

With a similar calculation for the $ND1$ transition we obtained the values of $|\langle {}^4A,l|\overrightarrow{r}|{}^4T,k\rangle|^2$ as summarized in Table 3. These transitions are from the four-fold degenerate ground state $|^4A,k\rangle$ to the twelvefold excited state $|^4T,l\rangle$, all with $S=\frac{3}{2}$. In the absence of L-S coupling the transition between states with the same S_z are allowed and their transition rates are nonzero. Our results in Table 3 are consistent with this. From Tables 2, 3, we found that for the transitions from any degenerate ground state to all degenerate excited states, total transition rates for each vacancy are almost equal. This means that, in a typical spectroscopic experiment, the balanced population of the degenerate ground states does not change with illumination of light [23]. The slight difference can be attributed to the approximation in using STO for dangling bonds of Figure 1.

Table 3. Intensities of the transitions in the unit of $|r_m|$, from each fourfold degenerate ground state ${}^4A$ to twelvefold degenerate exited state ${}^4T$ with available S_z for the ND1 line.

Excited states	Ground states S_z	$\vert{}^4A,1\rangle$ $-3/2$	$\vert{}^4A,2\rangle$ $-1/2$	$\vert{}^4A,3\rangle$ $1/2$	$\vert{}^4A,4\rangle$ $3/2$
$\vert{}^4T,1\rangle$	$-3/2$	0.33	0	0	0
$\vert{}^4T,2\rangle$	$-3/2$	0.33	0	0	0
$\vert{}^4T,3\rangle$	$-3/2$	0.33	0	0	0
$\vert{}^4T,4\rangle$	$-1/2$	0	0.05	0	0
$\vert{}^4T,5\rangle$	$-1/2$	0	0.47	0	0
$\vert{}^4T,6\rangle$	$-1/2$	0	0.31	0	0
$\vert{}^4T,7\rangle$	$1/2$	0	0	0.38	0
$\vert{}^4T,8\rangle$	$1/2$	0	0	0.00	0
$\vert{}^4T,9\rangle$	$1/2$	0	0	0.46	0
$\vert{}^4T,10\rangle$	$3/2$	0	0	0	0.49
$\vert{}^4T,11\rangle$	$3/2$	0	0	0	0.12
$\vert{}^4T,12\rangle$	$3/2$	0	0	0	0.22
Total Rate:	-	0.99	0.83	0.84	0.83

Again with the same procedure as we used for the $GR1$, we will end up with:

$$I(ND1) \propto \frac{1}{4}\sum_{k=1}^{4}\sum_{l=1}^{12}\left|\langle {}^4T,l|\overrightarrow{r}|{}^4A,k\rangle\right|^2 \qquad (7)$$

$$I(ND1) \propto 0.87\,|\overrightarrow{r}_m|^2 . \qquad (8)$$

So the relative intensities of the ND1 to GR1 line is:

$$\frac{I(ND1)}{I(GR1)} = 3.2. \qquad (9)$$

The strength A of the absorption spectra in a zero phonon line is defined as the integral over the zero phonon line:

$$A = \int \mu(\nu)d(h\nu) \qquad (10)$$

where $\mu(\upsilon)$ is the absorption coefficient in cm^{-1}, measured at 77 K and integrated over the zero phonon absorption line, and $h\nu$ is the photon energy in meV. In these units the concentration in cm^{-3} of the neutral and negatively charged vacancies are:

$$\left[N^0\right] = A_{GR1}/f_{GR1} \qquad (11)$$

$$\left[N^-\right] = A_{ND1}/f_{ND1} \qquad (12)$$

where f_{GR1} is the oscillator strength of the GR1 that is $(1.2 \pm 0.3) \times 10^{-16}$ [16] and f_{ND1} is oscillator strength of ND1 transition which is equal to $(4.8 \pm 0.2) \times 10^{-16}$ [16] or $(3.9 \pm 1) \times 10^{-16}$ [11,16,24].

Our calculated value from equation (9) is in very good agreement with the reported experimental data 3.25 [10,24] and 4 [10,11] for the relative intensity of the ND1 to GR1.

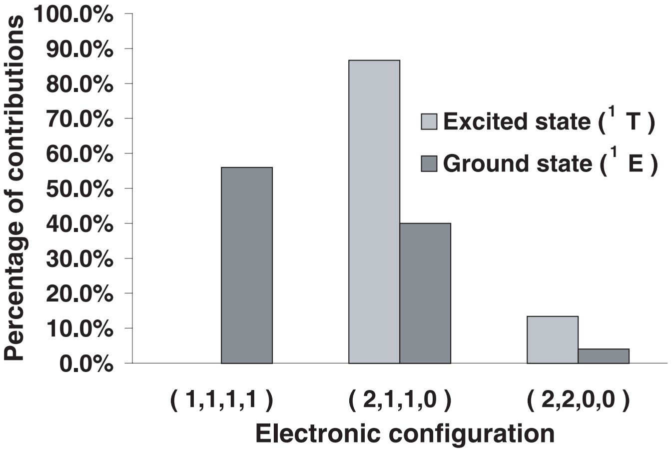

Fig. 3. Percentage of the contribution of each possible electronic configurations in the ground and excited states of the neutral vacancy in diamond.

The contribution of the allowed electronic configuration in the ground and dipole allowed excited states of the V^0 are summarized in Figure 3. Numbers in the parenthesis in this figure are the occupation numbers of atomic orbitals of the vacancy. For the V^0, our calculation shows that the ground state is the spin singlet state ${}^1E$ in agreement with experiment (Fig. 2). Therefore, all possible orbital occupations are allowed in the ground state. The allowed electronic configurations of four electrons in four NN site are (1,1,1,1), (2,1,1,0) and (2,2,0,0). They are Hund , singly paired and doubly paired configurations respectively.

As shown in Figure 3, all possible electronic configurations are present in the ground state ${}^1E$.

From this figure, the ground state is mainly (1,1,1,1) configuration (56%), where all of the vacancy atomic orbitals are half filled. This means that the probability of finding the Hund configuration in the ground state of V^0 is 56%. In $(1,1,1,1)$ or Hund configuration, the

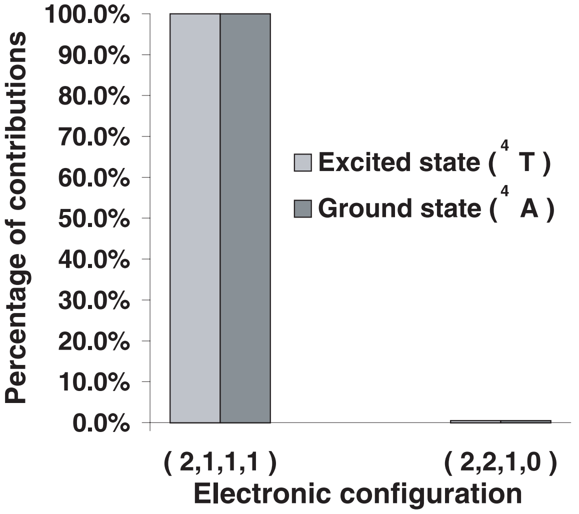

Fig. 4. Percentage of the contribution of each possible electronic configurations in the ground and excited states of the negative vacancy in diamond.

electrons have maximum separation distance, which result to lowering the Coulombic repulsion energy of the system. The contribution of the paired configuration, (2,1,1,0) is around 40%. As we expect, the contribution of the high energy (doubly paired) (2,2,0,0) configuration in the ground state is negligible at 4%. This is due to the high Coulombic repulsion of this fully paired configuration.

For the ND1 (V^-), available electronic configurations for distributing the five electrons of the V^- in four nearest neighbor sites are (2,1,1,1) and (2,2,1,0). Our calculation gives $\frac{3}{2}$ for the spin of the ground state ^4A in agreement with experiment (Fig. 2). Hence the dipole allowed excited state should have the same spin (^4T state in Fig. 2). Due to this fact, the only allowed configuration for these states is (2,1,1,1) which is a Hund state with maximum value of spin. Hence there is no room for configuration change via ND1 optical excitation (Fig. 4). In other words, the highest value of the ground state spin of the V^- restricts the allowed spatial distribution of the ground and the dipole allowed excited state to the (2,1,1,1) configuration, which has a minimum number of paired electrons. This significantly increases the overlap of the ground and excited state wavefunctions and the corresponding dipole transition amplitude $\langle \Psi_i | \overrightarrow{r} | \Psi_f \rangle$ for ND1 transition. This is in contrast to the GR1 transition in V^0 in which the ground and excited states have lowest value of the spin, among the allowed values 0, 1 and 2. There will be a different contribution of these configurations in the ground and excited state (Fig. 3). This reduces the overlap integral of the wavefunctions and consequently the related dipole transition amplitude in the low spin transition GR1.

Let us discuss the relative oscillator strength in other color centers of diamond. By following the loss of GR1 and ND1 absorption during annealing and with a further assumption that all vacancies are trapped at nitrogen in type Ia diamond, Davies et al. were able to measure the relative oscillator strengths of the V^-, Nitrogen vacancy (N-V) and aggregations of the one paired (H3) and two paired impurity nitrogen atoms (H4) with a single vacancy [10].

Table 4. Calibrated oscillator strength of the different color centers in diamond [10,16] and the spin values of their ground state.

Color center	V^-	NV	N3	V^0	H3	H4
Ground state spin	$\frac{3}{2}$	1	$\frac{1}{2}$	0	0	0
Calibrated strength	4.8, 3.9	1.4	0.9	1.2	1	1

Their findings are summarized in Table 4. The complementary work for obtaining absolute value of the ND1 transition strength based on EPR experiment [11,24] and the absorption intensity of the N3 center (3 nitrogen atoms around one vacancy) are also summarized in this table.

The value of the oscillator strength of N-V, N3, V^0, H3 and H4 centers are almost the same but for ND1 case, the value is four times larger (Tab. 4). Our calculation (Eq. (9)) agrees quantitatively with the resultant strength ratio of f_{ND1}/f_{GR1} [10,11,24] which one can obtain from Table 4. Calculation results also can explain the much greater oscillator strength of the ND1 among other optical absorption lines. The spin of the ground state for N-V and N3 are 1 [26,27] and $\frac{1}{2}$ [28] respectively (Tab. 4). The V^0, H3 and H4 are non paramagnetic ($S = 0$) [25]. Hence it is the V^- transition whose ground state has the highest available spin value (Hund law). The low values of spin in the ground state of the other color centers reduce the overlap integral of their ground and excited state wavefunctions for the dipole amplitude. This is much smaller than the dipole amplitude of Hund states of the V^-.

4 Conclusion

The paper reports a computational method to calculate dipole transition rates in color centers of diamond. Correlation effects are also included in this calculation. We calculated the many electron wavefunctions of the generalized Hubbard Hamiltonian using an atomic orbital basis for the diamond vacancies. Our approach, unlike configuration interaction, can give different forms of degenerate wavefunction. The oscillator strength of the dipole transition intensities for the well known GR1 and ND1 optical absorption bands were calculated based on these many electron wavefunctions. Result is in good quantitative agreement with recent experimental data for relative oscillator strengths of the V^0 and V^-. The higher absorption intensity of the negatively charged vacancy is related to the implementation of Hund rule in this center. The highest value of spin for the ground and excited states restricts the orbital occupation condition to only one available configuration with a minimum number of paired electrons. This increases the overlap integral of the ground and excited state of V^- wavefunctions. This is in contrast to the situation in the centers with a low spin value such as N-V, N3, H3, H4.

We would like to thank Prof. J.E. Lowther and Prof. M. Djafari Rouhani for their comments.

Appendix A

For V^0 and V^- both ground and excited states have definite degeneracy which we must consider. Therefore for $|\Psi_i\rangle$ and $|\Psi_f\rangle$ we define:

$$\left|\Psi_i^k\right\rangle = \sum_j \alpha_j^{ik} \left|\varphi_j\right\rangle \tag{13}$$

$$\left|\Psi_f^l\right\rangle = \sum_j \alpha_j^{fl} \left|\varphi_j\right\rangle \tag{14}$$

where $|\varphi_j\rangle$ are Hilbert space bases. We used a Fock representation of equation (1) which is equivalent to determinant Slaters [21]. α_j^{ik}'s and α_j^{fk}'s are calculated expansion coefficients of the Hamiltonian eigenfunctions for each degenerate state. k and l are degeneracy labels for initial and final states with their upper limits of 2, 3 (V^0) and 4, 12 (V^-) respectively. With the above definition we have:

$$\left\langle \Psi_f^l \left| \overrightarrow{r} \right| \Psi_i^k \right\rangle \equiv \sum_{jj'} \alpha_j^{ik} \alpha_{j'}^{fl} \left\langle \varphi_j \left| \overrightarrow{r} \right| \varphi_{j'} \right\rangle . \tag{15}$$

For simplifying calculations, we considered that $\langle \varphi_j | \overrightarrow{r} | \varphi_{j'} \rangle$ terms were dominant. In such conditions φ_j and $\varphi_{j'}$ have the same orbital occupation condition. For example one class of such terms are $\langle abcd | \overrightarrow{r} | abcd \rangle$ terms. These many electron integrals can be calculated by expanding them to single electron integrals as follows:

$$\begin{aligned}\langle abcd \,|\overrightarrow{r}|\, abcd\rangle &= \langle a \,|\overrightarrow{r}|\, a\rangle + \langle b \,|\overrightarrow{r}|\, b\rangle \\ &\quad + \langle c \,|\overrightarrow{r}|\, c\rangle + \langle d \,|\overrightarrow{r}|\, d\rangle + O(s^2).\end{aligned} \tag{16}$$

In the above equation we neglect terms which are proportional to s^2 where s is the overlap integral of dangling orbitals $\langle a|b\rangle$. Finally we have:

$$\left\langle {}^1T, l \left| \overrightarrow{r} \right| {}^1E, k \right\rangle = \sum_m \beta_m^{lk} \overrightarrow{r}_m \tag{17}$$

where $\overrightarrow{r}_m \equiv \langle m | \overrightarrow{r} | m \rangle$ and m represents a, b, c, and d orbitals. Since the $\overrightarrow{r}_m$'s are not orthogonal and they have equal length, after squaring the amplitude of equation (17) to find the transition rates, we will have:

$$\left| \left\langle {}^1E, 1 \left| \overrightarrow{r} \right| {}^1T, 1 \right\rangle \right|^2 = \left(\sum_{i=1}^{4} \alpha_{i,lk}^2 + \cos\theta \sum_{i,j=1}^{4} \alpha_i^{lk} \alpha_j^{l'k'} \right) \left| \overrightarrow{r}_{aa} \right|^2 \tag{18}$$

where α^{lk} are coefficients which were obtained after squaring equation (17).

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