Optical Transitions at the Neutral and Charged Vacancies in Diamond

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Abstract

We used the exact eigenvectors of the generalized Hubbard Hamiltonian solution to predict the transition intensities of the well known GR1 and ND1 transitions at the neutral and charged vacancies in diamond. In addition to using exact eigenvectors, the method of the calculation is more precise than already reported calculations. The quantitative results can explain recent experimental data very good.

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I. INTRODUCTION

Coulson and Kearsly introduced molecule concept for vacancy defect in diamond.\textsuperscript{1} As the results they used the common method for calculation the electronic structure of the molecules in quantum chemistry i.e. Molecular orbital and Configuration interaction.

In essence in this model, molecular orbital method gives correct symmetry and spin of the electronic levels of the vacancy and configuration interaction considers the correlation effects of e-e interaction and gives correct sequence of the electronic levels. Molecular orbital method has been the only way for accounting the appropriate symmetry and spin of the vacancy since Coulson and Kearsly model.

Already we showed that generalized Hubbard Hamiltonian can also take into account correct symmetry and spin of the system in a completely different manner.\textsuperscript{2} This approach has origin in the solid state physics and pay more attention to include symmetry and spin considerations of the system in the form of a Hubbard like Hamiltonian. This is in contrast to quantum chemistry approaches (molecular orbital theory) which pay more attention to the construction of the symmetry and spin adapted wavefunctions.

Both approaches use some parameters (8 parameters for exact evaluation of e-e interaction) which can be obtained by theory or semi empirical methods. These parameters can be obtained by choosing appropriate atomic orbital function for dangling bonds of the vacancy.

This evaluation is independent of the model of Hamiltonian calculation. These parameters are the origin of the one type of approximations which exist in the molecular model. These parameters are the only input of the generalized Hubbard Hamiltonian formalism and after evaluating and putting them into the computational scheme the exact eigenvalues and eigenstates can be obtained.

The situation in the Coulson and Kearsly model and its related models\textsuperscript{3} which
use molecular orbital techniques is different. These models not only use these approximate parameters but also apply some approximation in constructing appropriate CI wavefunctions as the bases for calculating the Hamiltonian. Basically the coefficients of the manually constructed CI wavefunctions in these models should be calculated by variational principal. Also there should be some hypothesis about the most probable configurations in the ground and excited electronic state of the system. Additionally there should be an assumption for truncation limit in accounting a high number of determinant Slaters in the expansion of the CI wavefunctions.

These assumptions and approximations, results to a approximately CI wavefunction for the electronic states of the vacancy in Coulson and Kearsly and related models.\textsuperscript{1,3} In our last communication\textsuperscript{2} we also explained some ambiguity of the applying such CI method for this problem.

In summary, it is obvious that the CI wavefunctions in molecular models are not the exact eigenvectors of the Hamiltonian of the vacancy system.

Here we should note to this point that our using bases consist of the atomic bases i.e. $a, b, c, d$ in contrast to Coulson and Kearsly and all of the previous related models which use molecular symmetric and anti symmetric bases i.e. $a, t_x, t_y, t_z$. These bases are more convenient in explaining physical properties of the system. For example ionization of the atoms of the vacancy or possibility of the existence of the hunds configurations in the ground state can be verified very easily.

Previously we reported\textsuperscript{2} the probability of finding each electronic configuration in the exact eigenstates of the generalized Hubbard Hamiltonian for the GR1 transition in the neutral vacancy. (Fig.1)

These results and similar results for the ground and excited states of ND1 transition are free from approximations of the manually constructing CI wavefunctions. In this method the origin of the error only is in the Hamiltonian parameters.
FIG. 1: Percentage of each possible electronic configurations in the ground and excited states of the neutral vacancy in diamond

For verifying the validity of these new results we have used them to calculate some experimentally observable data.

II. MODEL

Similar to Lowther and Mainwood and Stoneham we attempted to calculate the relative intensities of the well known GR1 and ND1 transitions in the neutral and charged vacancies in diamond. These intensities are calculated directly with the resultant eigenvectors of the generalized Hubbard Hamiltonina and can be a reliable check for the validity of the physics which is included in the exact eigenstates of the model.

The rate of the electronic dipole transition between an initial state $\Psi_i$ and final state $\Psi_f$ is proportional to square of the dipole transition amplitude i.e.

$$I(i \rightarrow f) \propto |<\Psi_i|\vec{r}|\Psi_f>|^2$$

(1)

Accordingly for the dipole transition from the ground states of the $V^0$ and $V^-$ i.e. GR1 and ND1 we have two allowed transitions which their intensities or
transition rates are as follow:

\[ I(\text{GR}1) \propto |<^{1} E | \vec{r} |^{1} T >|^2 \] (2)

\[ I(\text{ND}1) \propto |<^{4} A | \vec{r} |^{4} T >|^2 \] (3)

The eigenvectors of the generalized Hubbard Hamiltonian have correct degeneracy due to symmetry and spin multiplicity of the states. So we obtain two and three independent eigenvectors for the ground and excited state of \text{GR}1 i.e. \(^1E\) and \(^1T\) respectively and we will have four and twelve independent eigenvectors for the ground and excited states of \text{ND}1 i.e. \(^4A\) and \(^4T\). In the end of this paper we have listed (Table a, b) the expansion coefficients of each of these degenerate levels on the \(S_z\) representation of the solving Fock space bases of the Hamiltonian. In the first quantization language this bases are determinant Slaters which are correspond to each electronic configuration. For simplicity, in this paper we will call these bases as determinant.

In Table a, b all of the exact eigenstates are orthonormal. An interesting point in the Table a is high contribution of the Hunds configurations i.e. \((1,1,1,1)\) in the ground state of \(V^0\) and also the absence of this configuration (model gives exactly zero!) in the the excited state of \text{GR}1.

At the begining of the calculation we note that this transition amplitudes has the same value for the two set of the eigenvectors which transform with a unitary matrix. So the value of the transition amplitude is the same for \(S_z\) or \(S^2\) representation of the Hamiltonian eigenvectors.

For \text{GR}1 transition only the bases or determinant Slaters which have \(S_z = 0\) enter into the calculation. (Table a)

\[ |^{1}E, i > = \sum_{j=1}^{36} \alpha_j^i \varphi_j \quad (i = 1, 2) \] (4)

\[ |^{1}T, i > = \sum_{j=1}^{36} \beta_j^i \varphi_j \quad (i = 1, 2, 3) \] (5)
In the above equations, \( i \) is the degeneracy label of the states and \( \varphi_j \)'s are the \( S_z = 0 \) bases of the Fock space or determinants. The \( \alpha^i_j \) and \( \beta^i_j \) coefficients are according to Table a, b. For GR1 transition we have 6 independent transition from the two degenerate \( ^1E \) states to three degenerate \( ^1T \) states. In this work we have calculated each transition rate (intensity) separately for each of these transitions.

Here we review in detail the steps of the calculation of the intensity of one typical transition e.g. \( |^1E, 1 \rangle \rightarrow |^1T, 1 \rangle \). The amplitude of this transition from Eq. (4), Eq. (5) is:

\[
<^1E, 1 | \vec{r} | ^1T, 1 > = \sum_{i,j=1}^{36} \alpha^i_j \beta^j_i < \varphi^i | \vec{r} | \varphi^j >
\]

In this calculation we have evaluated all of the \( < \varphi^i | \vec{r} | \varphi^i > \) terms up to \( s \) order. \( s \) is the overlap integral of the two adjacent atomic orbital of the vacancy.

\[
s = < a | b >
\]

We have calculated the value of \( s \) using appropriate Slater type function with exponent 1.595 as atomic orbital. The result was 0.16 which agrees with the previous models.\(^1,3\)

These types of amplitudes i.e. \( < \varphi^i | \vec{r} | \varphi^i > \) are direct expectation values of the dipole \( \vec{r} \) operator which arise from the same determinant Slatters terms of the \(^1E \) and \(^1T \) eigenvectors in Table a. These four electronic terms can be expanded to single particle terms as follow.

\[
< abcd | \vec{r} | abcd >= < a | \vec{r} | a > + < b | \vec{r} | b > + < c | \vec{r} | c > + < d | \vec{r} | d > + O(s^2)
\]

the terms which are proportional to \( s^2 \) are neglected equation in the above equation. In addition to these direct terms we have considered the cross terms in which the \( \varphi^i \) and \( \varphi^j \) in Eq. (6) are two different determinant Slatters. The main contribution of the cross terms comes from such \( < \varphi^i | \vec{r} | \varphi^j > \) terms in
which \( \varphi_i \) and \( \varphi_j \) only differ in one orbital. These terms are proportional to \( s \).

For example for contribution of the \( abc\vec{d} \) and \( abc\vec{c} \) determinants we have:

\[
< abc\vec{d} | \vec{r} | abc\vec{c} > = s < a | \vec{r} | a > + s < b | \vec{r} | b > + s < c | \vec{r} | c > + < d | \vec{r} | c > + O(s^2)
\]  

(9)

In these expansion the sign of the terms should be considered according to the permutation of that orbital which is different in two determinants.

For simplifying the last term in Eq. (9) we have used well known Mullikens’s approximation:

\[
< d | \vec{r} | c > = \frac{s}{2}( < d | \vec{r} | d > + < c | \vec{r} | c > )
\]  

(10)

Now by these rules we can calculate the rate of the transition between each degenerate ground and excited states by squaring each transition amplitude. For simplifying the writing of the formula we have used the abbreviation:

\[
\vec{r}_{\alpha\alpha} = < a | \vec{r} | a >
\]  

(11)

and therefore the final amplitude of the dipole transition will be in the form:

\[
< ^1 E, 1 | \vec{r} | ^1 T, 1 > = \alpha_1 \vec{r}_{\alpha\alpha} + \alpha_2 \vec{r}_{\beta\beta} + \alpha_3 \vec{r}_{\gamma\gamma} + \alpha_4 \vec{r}_{\delta\delta}
\]  

(12)

The \( \alpha_i \) are obtained from coefficients which are listed in Table a and also using value of \( s = 0.16 \).

Since the \( \vec{r}_{\alpha\alpha} \), \( \vec{r}_{\beta\beta} \), ... are not orthogonal and they have equal length, after squaring the amplitude to find the transition rates we will have:

\[
| < ^1 E, 1 | \vec{r} | ^1 T, 1 > |^2 = ( \sum_{i=1}^{4} \alpha_i^2 + \cos(\angle 109.5^o) \sum_{i,j=1}^{4} \alpha_i \alpha_j ) | \vec{r}_{\alpha\alpha} |^2
\]  

(13)

III. RESULTS AND DISCUSSIONS

The intensities of each six and twelve allowed dipole transitions from the two and four fold degenerate ground state of \( V^0 \) and \( V^- \) ( GR1 and
ND1 ) are summarized in Table I, II. These transitions are calculated by the method which was explained in the last section. In the absence of $L - S$ coupling, transition between ground and excited states with different $S_z$ should be zero, therefore as the results of our calculations show, Table II should be block diagonal. The total rates of the transitions from each of the degenerate ground state of the $V^0$ and $V^-$ are summarized in the Table I, II.

| Excited states | $S_z$ | 0.185 | 0.051 |
|----------------|------|-------|-------|
| $|^1T,1>$      | 0    | 0.185 | 0.051 |
| $|^1T,2>$      | 0    | 0.025 | 0.092 |
| $|^1T,3>$      | 0    | 0.055 | 0.067 |
| Total Rate:    | -    | 0.26  | 0.21  |

TABLE I: Intensities of the transitions in the unit of $|r_{aa}|$($|r_{aa}| = 1$) from the twofold degenerate ground state $^1E$ to threefold degenerate exited state $^1T$ with $S_z = 0$ for the GR1 line.

As it is seen from the tables the total rates of the transition from each degenerate ground state are nearly equal. Physically this sounds good. We expect that in an spectroscopic experiment which does not distinguish the degeneracy of the ground and excited states, one can not create a change in the relative population of the balanced degenerate ground states by illumination of light.\textsuperscript{6}

For obtaining the effective transition rate between degenerate ground states we should average over the the total transition rates.\textsuperscript{6} This arises from this point that the probability of the existence of the system in each of the degenerate states is equal. In other hand the number of the electrons which populate the degenerate ground states are equal.

Total transition rates of the GR1 and ND1 degenerate ground states are listed in the last line of the Table I, II.
TABLE II: Intensities of the transitions in the unit of $|r_{aa}|(|r_{aa}| = 1)$ from the fourfold degenerate ground state $^4A$ to twelvefold degenerate exited state $^4T$ with available $S_z$ for the ND1 line.

From Table I we obtain:

$$|<^1E|\vec{r}|^1T>|^2 = \frac{1}{2} \sum_{i,j=1}^{2,3} |<^1E,i|\vec{r}|^1T,j>|^2 = 0.23|\vec{r}_{aa}|^2$$  \hspace{1cm} (14)

and from Table II we obtain:

$$|<^4A|\vec{r}|^4T>|^2 = \frac{1}{4} \sum_{i,j=1}^{4,12} |<^4A,i|\vec{r}|^4T,j>|^2 = 0.79|\vec{r}_{aa}|^2$$ \hspace{1cm} (15)

With these values we obtain the relative intensities of the GR1 and ND1 absorption lines:

$$\frac{I(ND1)}{I(GR1)} = 3.4$$  \hspace{1cm} (16)

This is in excellent agreement with the recent experimental work on the GR1 and ND1 transitions intensities.\textsuperscript{7,8,9} We have used these values from Davies\textsuperscript{10} who has calibrated them. The experimental data shows that the relative intensities of the ND1 and GR1 is equal to 3.25\textsuperscript{7,8} or 4\textsuperscript{7,9}. Our theoretical value is also consistent with the experimental error which is reported more recently.\textsuperscript{11} The
error of our calculation which arises from averaging over the total transition rates are 10 percent for ND1 and 11 percent for GR1. These errors should be due to approximations in obtaining the Hamiltonian parameters. This means that if we were able to estimate exact atomic orbitals of the vacancy and also Hamiltonian parameters then we expect that the total rates of the transition be equal. (last line of Tables I, II)

One important question in this problem has been the importance of the overlap effect of the vacancy orbitals. We have recalculated the intensities with the neglecting the overlap of the orbitals i.e. $s = 0$. The results for the GR1 intensity is 0.23 which is same as Eq. (14) and for ND1 the result is 0.87, higher than the value of the Eq. (15). By this assumption we reach to 3.8 for the relative transitions. The independent nature of the GR1 transition from the overlap parameter $s$ comes from the high contribution of the Hunds state in the ground state. As it was shown in Eq. (8) the transition amplitude of these type determinants is independent from $s$. However the value of the $s$ depends on the choosing of the atomic orbitals of the vacancy so by changing it the Hamiltonian parameters also should be changed. If we were able to include semiempirical nature of the generalized Hubbard Hamiltonian parameters to the shape of the atomic orbitals then we can be sure that the resultant overlap value is more valid. However since the Hamiltonian parameters are from Coulson and Kearsly calculation, the value of the $s$ in our calculation are choose to be 0.16. Our model is more similar to Mainwood and Stoneham model since their model does not need to detail form of atomic orbital i.e. calculation of the $|\vec{r}_{aa}|$. Also they have attributed the GR1 and ND1 transition only to extra atomic transitions. They have neglected the overlap of the vacancy orbitals ($s = 0$) maybe due to the order of the error which exist in the approximated wavefunctions of the Lannoo model.\(^3\)
IV. CONCLUSION

We applied the exact wavefunctions of the generalized Hubbard Hamiltonian to the transition intensities of the well known optical absorption lines of the diamond vacancies. This calculation can successfully explain recent experimental data. These results clearly show the advantage of using exact wavefunctions of the generalized Hubbard Hamiltonian with respect to previous CI wave functions. For comparing the different wavefunctions of the two model it is useful to compare the number of determinants which comes into the calculation of the transition rates. For example CI only predicts $1 \times 1$ determinants for ND1 ground and excited states however here we considered $8 \times 8$ determinants for the $S_z = \pm \frac{3}{2}$ and $48 \times 48$ for $S_z = \pm \frac{1}{2}$ as are reported in Table b. Previous molecular orbital approaches were not able to consider different forms of the degenerate eigenvectors of the Hamiltonian and this can be another reason for their poor quantitative results.

The results of the calculation show that the electronic configurations of the ground and excited states of GR1 (Fig. 1) can explain quantitatively experimental data. This means that the ground state of the neutral vacancy mainly comes from the Hunds rule (60 percent). High contribution of the Hunds states in the ground state of the neutral vacancy physically was expected.

New results of present model show that generalized Hubbard Hamiltonian has potential to go toward a quantitative understanding of the point defects problem in diamond.

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| $\varphi_1$ | $^1E, 1 >$ | $^1E, 2 >$ | $^1T, 1 >$ | $^1T, 2 >$ | $^1T, 3 >$ |
|------------|-------------|-------------|-------------|-------------|-------------|
| aabd       | 0.169       | 0.038       | -0.323      | -0.061      | 0.024       |
| aacc       | 0.102       | 0.023       | 0.106       | 0.226       | -0.066      |
| aabc       | 0.118       | -0.127      | 0.323       | 0.059       | -0.022      |
| abcd       | 0.429       | -0.133      | 0.000       | 0.000       | 0.000       |
| aacd       | 0.051       | 0.166       | 0.323       | 0.058       | -0.025      |
| abcc       | 0.051       | 0.166       | -0.168      | 0.274       | -0.072      |
| aacd       | -0.051      | -0.166      | -0.323      | -0.058      | 0.025       |
| abcd       | -0.100      | 0.438       | 0.000       | 0.000       | 0.000       |
| abcd       | -0.330      | -0.305      | 0.000       | 0.000       | 0.000       |
| abdd       | 0.051       | 0.166       | -0.073      | -0.093      | 0.307       |
| aadd       | -0.071      | 0.076       | 0.170       | -0.024      | 0.193       |
| accd       | 0.118       | -0.127      | -0.168      | 0.273       | -0.074      |
| aabb       | -0.031      | -0.100      | 0.163       | -0.122      | -0.159      |
| accd       | -0.169      | -0.038      | -0.073      | -0.095      | 0.306       |
| abc c       | 0.169       | 0.038       | 0.084       | 0.240       | 0.209       |
| abbd       | -0.118      | 0.127       | 0.084       | 0.238       | 0.211       |
| abcc       | -0.051      | -0.166      | 0.168       | -0.274      | 0.072       |
| abbc       | -0.169      | -0.038      | -0.084      | -0.240      | -0.209      |
| abcd       | -0.330      | -0.305      | 0.000       | 0.000       | 0.000       |
| abcd       | -0.100      | 0.438       | 0.000       | 0.000       | 0.000       |
| abdd       | -0.051      | -0.166      | 0.073       | 0.093       | -0.307      |
| abcd       | 0.429       | -0.133      | 0.000       | 0.000       | 0.000       |
| accd       | -0.118      | 0.127       | 0.168       | -0.273      | 0.074       |
| accd       | 0.169       | 0.038       | 0.073       | 0.095       | -0.306      |
| bcc c       | -0.071      | 0.076       | -0.170      | 0.024       | -0.193      |
| aabd       | -0.169      | -0.038      | 0.323       | 0.061       | -0.024      |
| bccd       | 0.051       | 0.166       | -0.082      | -0.239      | -0.211      |
| bcd d       | -0.051      | -0.166      | 0.082       | 0.239       | 0.211       |
| bbd d       | 0.102       | 0.023       | -0.106      | -0.226      | 0.066       |
| abbd       | 0.118       | -0.127      | -0.084      | -0.238      | -0.211      |
| bc c c       | -0.169      | -0.038      | -0.166      | 0.275       | -0.073      |
| abc c       | -0.118      | 0.127      | -0.323      | -0.059      | 0.022       |
| bc d d       | 0.118       | -0.127      | -0.071      | -0.094      | 0.307       |
| bc d d       | 0.169       | 0.038       | 0.166       | -0.275      | 0.073       |
| bc d d       | -0.118      | 0.127      | 0.071      | 0.094      | -0.307       |
| cc d d       | -0.031      | -0.100      | -0.163      | 0.122       | 0.159       |

Table a. Expansion coefficients of the $^1E$ and $^1T$ states of the GR1 on the $S_z$ representation of the Fock space bases (determinants)
| $S_a$ | $\psi_i$ | $|A, 1\rangle$ | $|A, 2\rangle$ | $|A, 3\rangle$ | $|A, 4\rangle$ | $|T, 1\rangle$ | $|T, 2\rangle$ | $|T, 3\rangle$ | $|T, 4\rangle$ |
|------|----------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| -3/2 | $abcd$   | -0.500         | 0.000          | 0.000          | 0.000          | 0.866          | 0.000          | 0.000          | 0.000          |
| -3/2 | $abcd$   | 0.500          | 0.000          | 0.000          | 0.000          | 0.249          | 0.289          | 0.777          | 0.000          |
| -3/2 | $abcd$   | -0.500         | 0.000          | 0.000          | 0.000          | 0.549          | -0.289         | 0.605          | 0.000          |
| -3/2 | $abcd$   | 0.500          | 0.000          | 0.000          | 0.000          | 0.798          | 0.289          | -0.173         | 0.000          |
| -1/2 | $abcd$   | 0.000          | 0.289          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          | -0.173         |
| -1/2 | $abcd$   | 0.000          | 0.289          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          | -0.173         |
| -1/2 | $abcd$   | 0.000          | 0.289          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          |
| -1/2 | $abcd$   | 0.000          | 0.289          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          |
| -1/2 | $abcd$   | 0.000          | 0.289          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          |
| -1/2 | $abcd$   | 0.000          | 0.289          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          |
| -1/2 | $abcd$   | 0.000          | 0.289          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          |
| -1/2 | $abcd$   | 0.000          | 0.289          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          | 0.000          |

Table b. Expansion coefficients of the $^1A$ and $^1T$ states of the ND1 on
the S, representation of the Fock space bases (determinants)
| $S_a$ | $\psi_1$ | $^4T, 5 >$ | $^4T, 6 >$ | $^4T, 7 >$ | $^4T, 8 >$ | $^4T, 9 >$ | $^4T, 10 >$ | $^4T, 11 >$ | $^4T, 12 >$ |
|------|----------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |
| 1/2  | abcd     | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       | 0.000       |

Table b. (continued) Expansion coefficients of the $^4A$ and $^4T$ states of the NDI on the $S_a$ representation of the Fock space bases (determinants).