

ELECTRONIC PROPERTIES
OF SOLID

Construction of a Multielectron Basis for Mott Insulators
with Strong Electron Correlations, Spin–Orbit Interaction,
and Covalence

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Abstract—We propose that the apparatus of quantum mechanics of a free atom (in particular, the theory of nj symbols and Rakh–Wigner genealogic coefficients generalized to the case of point groups and widely used in crystal field theory) be used for constructing multielectron bases with allowance for covalence and spin–orbit interaction. This allows us to take into account the electron–electron interaction for $3d$ ions the most comprehensively. The basis constructed in this way can be used in the generalized strong coupling method for the multiband p – d model in describing the structure of the quasiparticle energy spectrum and physical properties of systems with strong electron correlations. The procedure of construction and computation is demonstrated for the ${}^5T_{2g}$ term in the d^6 configuration of the transition metal atom in an octahedral field. The mechanism for the emergence of magnetic anisotropy in S ions (Fe^{3+} and Mn^{2+}) due to covalent mixing of $d^6\bar{L}$ configurations with a nonzero orbital angular momentum (\bar{L} is a hole in ligands) is demonstrated.

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1. INTRODUCTION

Electron correlations play an important role in the formation of various magnetic and transport properties of transition metal oxides. Many attempts have been made in recent years to describe these properties and primarily the metal–insulator transition, superconductivity in cuprates, and colossal magnetoresistance in manganites. Mott [1, 2] and Hubbard [3] demonstrated that it is the strong Coulomb d – d interaction that explains the existence of many transition $3d$ metal oxide compounds with a partly filled $3d$ band in the form of magnetic insulators. A compound is a metal if the width of the $3d$ band is larger than the Coulomb d – d interaction. If, however, the Coulomb interaction exceeds the bandwidth, $3d$ electrons are localized, the compound becomes an insulator with localized magnetic moments, and the dielectric gap width is determined by the intensity of the electron–electron interaction. In other words, a strong (Coulomb or exchange) interaction of $3d$ electrons exceeding or on the order of their kinetic energy (with the electron bandwidth as a measure) renders these substances the properties of strongly correlated systems, which complicates theoretical description of their physical properties.

In [4], a scheme was proposed in which transition metal compounds can be classified in accordance with two regimes depending on the relation between ligand-to-metal charge transfer energy Δ_{tr} and Coulomb energy U . In the Mott–Hubbard regime, in which $D_{\text{tr}} > U$, band splitting occurs due to charge fluctuations of the d – d and $d^n + d^n \rightarrow d^{n+1} + d^{n-1}$ types, while the splitting is proportional to U . In the charge transfer regime, when $\Delta_{\text{tr}} < U$, the $d^n + d^n \rightarrow d^{n+1} + d^n\bar{L}$ type fluctuations (\bar{L} denotes a hole in ligands) form the p – d type splitting, and the bandwidth is proportional to Δ_{tr} .

The Mott–Hubbard single-band model (even with charge-transfer effects included) is insufficient for a comprehensive description of the properties of many compounds of transition metals [5–8]. It has become clear that these properties can be described only with allowance for orbital, spin, charge, and lattice degrees of freedom. For example, orbital and charge ordering of manganese ions in manganites plays an important role in the colossal magnetoresistance effect [9–13]. Usually, the electron–electron interaction is described using (at best) the Kanamori approximation in which only the density–density and exchange interactions are preserved from the entire set of matrix elements. The importance of including the total Hamiltonian for

the electron–electron interaction was emphasized and substantiated in [14], where the role of multiplicity effects in the formation of the energy gap was demonstrated:

$$U(d^n) = E(d^{n-1}) + E(d^{n+1}) - 2E(d^n).$$

In [15], it was shown how spin crossovers change the value of $U(d^n)$ and affect the Mott–Hubbard transition. In other words, a comprehensive multiband theory taking into account the Coulomb interaction completely is required for describing real multiorbital Mott–Hubbard systems.

A cluster approach based on multiband Hubbard models or p – d models was proposed for cuprates [16, 17] and manganates [18] for describing the structure of the quasiparticle energy spectrum for systems with strong electron correlations. In this approach, the technique of Hubbard X operators and exact diagonalization of MeO_6 clusters (Me is a transition metal) is used. However, two of the main advantages in using the X operators is the conservation of a regular structure of the Hilbert space at all computational stages and automatic fulfillment of the condition excluding energy-wise disadvantageous configurations with double filling of the same quantum state of one-electron orbitals. The construction of Hubbard operators requires knowledge of the wave eigenfunctions for each sector (d^{n-1} , d^n , d^{n+1}) of the Hilbert space under investigation. The algorithm for constructing such functions for ionic crystals is well known, but allowance for covalence requires its elaboration. Here, we propose that the apparatus of the η_j symbols and Racah–Wigner genealogical coefficients (generalized to the case of point groups and widely used in crystal field theory [19]) be employed for constructing multielectron bases taking into account covalence and the spin–orbit interaction. The basis constructed in this way will be used for calculating the electron structure of transition metal oxides with crossovers of multielectron terms with different spins. This study contains the required methodological material.

2. MODEL OF THE MeO_6 CLUSTER TAKING INTO ACCOUNT ELECTRON CORRELATIONS AND COVALENCE

Let us first recall the main concepts and notation in the theory of molecular orbitals. To describe the electron states of a cluster, the molecular orbital method, applicable to any multiatomic systems and widely used in quantum chemistry, is employed [20, 21]. It is well known that group theory makes it possible to draw a number of general conclusions on the properties of molecular orbitals. Knowing the symmetry of a cluster, we can immediately establish the classification of

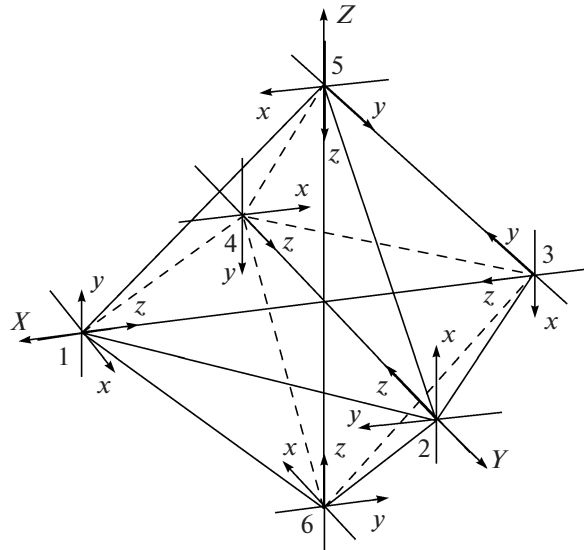


Fig. 1. Coordinate axes chosen for ligands in a MeO_6 octahedral molecule (complex).

molecular terms from irreducible representations of the corresponding point group [22]. Each electron of the cluster can be described by a one-electron spin orbital

$$\Psi_{\omega\gamma\mu m_s} = \Phi_{\omega\gamma\mu}(r)\chi_{1/2, m_s}(\sigma),$$

where $\Phi_{\omega\gamma\mu}(r)$ is a molecular orbital transformed in accordance with the μ line of irreducible representation γ of the point group under investigation; identical representations γ are distinguished by index ω . In the molecular orbital approximation in the form of a linear combination of atomic orbitals, we have

$$\Phi_{\omega\gamma\mu}(r) = A_{n'l\gamma}\phi_{\gamma\mu}^{n'l}(r) + \sum_{n'l'} B_{n'l'\gamma}\chi_{\gamma\mu}^{n'l'}(r),$$

where $\phi_{\gamma\mu}^{n'l}(r)$ is a combination of atomic functions of the $n'l$ shell of a metal ion, which is transformed in accordance with irreducible representation γ (i.e., it is a crystal harmonic) and $\chi_{\gamma\mu}^{n'l'}(r)$ is an analogous combination of atomic ligands (i.e., it is a linear combination of crystal harmonics of each ligand). Thus, the correspondence between the orbitals of the central atom and group orbitals of the ligands, which can be combined to form molecular orbitals, has been established (Table 1).

We will consider the orbitals of ligands with octahedral symmetry using the system of coordinates shown in Fig. 1. A schematic of molecular orbitals formed in this case is shown in Fig. 2.

In crystal field theory, it is assumed that the t_{2g} level becomes lower, whereas the e_g level becomes higher in an octahedral field as compared to their positions in a

Table 1. Classification of central atom orbitals and group orbitals of ligands in accordance with irreducible representations for an octahedral molecule [21]

Representation	Central atom orbitals	Group orbitals of ligands
a_{1g}	s	$\sigma_1 + \sigma_2 + \sigma_3 + \sigma_4 + \sigma_5 + \sigma_6$
e_g	$\begin{cases} d_z^2 \\ d_{x^2-y^2} \end{cases}$	$2\sigma_5 + 2\sigma_6 - \sigma_1 - \sigma_2 - \sigma_3 - \sigma_4$ $\sigma_1 - \sigma_2 + \sigma_3 - \sigma_4$
t_{2g}	$\begin{cases} d_{xz} \\ d_{yz} \\ d_{xy} \end{cases}$	$\pi_{y1} + \pi_{x5} + \pi_{x3} + \pi_{y6}$ $\pi_{x2} + \pi_{y5} + \pi_{y4} + \pi_{x6}$ $\pi_{x1} + \pi_{y2} + \pi_{y3} + \pi_{x4}$
t_{1u}	$\begin{cases} p_x \\ p_y \\ p_z \end{cases}$	$\sigma_1 - \sigma_3$ $\pi_{y2} + \pi_{x5} - \pi_{x4} - \pi_{y6}$ $\sigma_2 - \sigma_4$ $\pi_{x1} + \pi_{y5} - \pi_{y3} - \pi_{x6}$ $\sigma_5 - \sigma_6$ $\pi_{y1} + \pi_{x2} - \pi_{x3} - \pi_{y4}$
t_{1g}	—	$\begin{cases} \pi_{y1} - \pi_{x5} + \pi_{x3} - \pi_{y6} \\ \pi_{x2} - \pi_{y5} + \pi_{y4} - \pi_{x6} \\ \pi_{x1} - \pi_{y2} + \pi_{y3} - \pi_{x4} \end{cases}$
t_{2u}	—	$\begin{cases} \pi_{y2} - \pi_{x5} - \pi_{x4} + \pi_{y6} \\ \pi_{x1} - \pi_{y5} - \pi_{y3} + \pi_{x6} \\ \pi_{y1} - \pi_{x2} - \pi_{x3} + \pi_{x4} \end{cases}$

Note: Notations σ and π is used for p orbitals of ligands in accordance with the type of bonds in which they participate.

free ion. Analysis of the effect of the electrostatic potential produced by six point charges located at the vortices of the octahedron around the central ion shows that the former level becomes lower by $4Dq$, while the latter level becomes higher by $6Dq$, where $10Dq$ is the splitting between the t_{2g} and e_g levels. In the molecular orbital method, the change in the energy of atomic orbitals upon the formation of a molecule is more complicated and cannot be described so easily (see Fig. 2). As a result, total splitting Δ of the levels acquires a covalent correction.

We will describe the MeO_6 complex as a mixture of ionic ($d^N(S\Gamma)$) and covalent states with R holes in the anion subsystem,

$$d^{N+R} \underline{L}^R(S\Gamma), \quad R = 1, 2, \dots, 10 - N,$$

or

$$e_g^l(S_1\Gamma_1)t_{2g}^m(S_2\Gamma_2)S\Gamma$$

and

$$e_g^{l+n}(S_1\Gamma_1)t_{2g}^{m+k}(S_2\Gamma_2)\{\tilde{S}_1\tilde{\Gamma}_1\}\tilde{p}_\sigma^n(S_3\Gamma_3) \\ \times \tilde{p}_\pi^k(S_4\Gamma_4)\{\tilde{S}_2\tilde{\Gamma}_2\}S\Gamma,$$

where $m + l = N$ and $n + k = R$. This notation indicates that $l + n$ electrons on the e_g orbital form the $S_1\Gamma_1$ state, while $m + k$ electrons on the t_{2g} orbital form the $S_2\Gamma_2$ state; in turn, the $S_1\Gamma_1$ and $S_2\Gamma_2$ states form the $\tilde{S}_1\tilde{\Gamma}_1$ state. Analogously, the states of ligands $\tilde{p}_\sigma^n(S_3\Gamma_3)$ and $\tilde{p}_\pi^k(S_4\Gamma_4)$ are combined into the $\tilde{S}_2\tilde{\Gamma}_2$ state and, finally, $\tilde{S}_1\tilde{\Gamma}_1$ and $\tilde{S}_2\tilde{\Gamma}_2$ form the complete wavefunction of the $\tilde{S}_2\tilde{\Gamma}_2$ state. We choose the scheme with a strong crystal field because it allows us to approach in a natural way the covalence problem, since one-electron wavefunctions with the same point symmetry of the ion and its ligands are mixed in crystals (see above). Symbols \tilde{p}_σ and \tilde{p}_π denote a hole on oxygen group

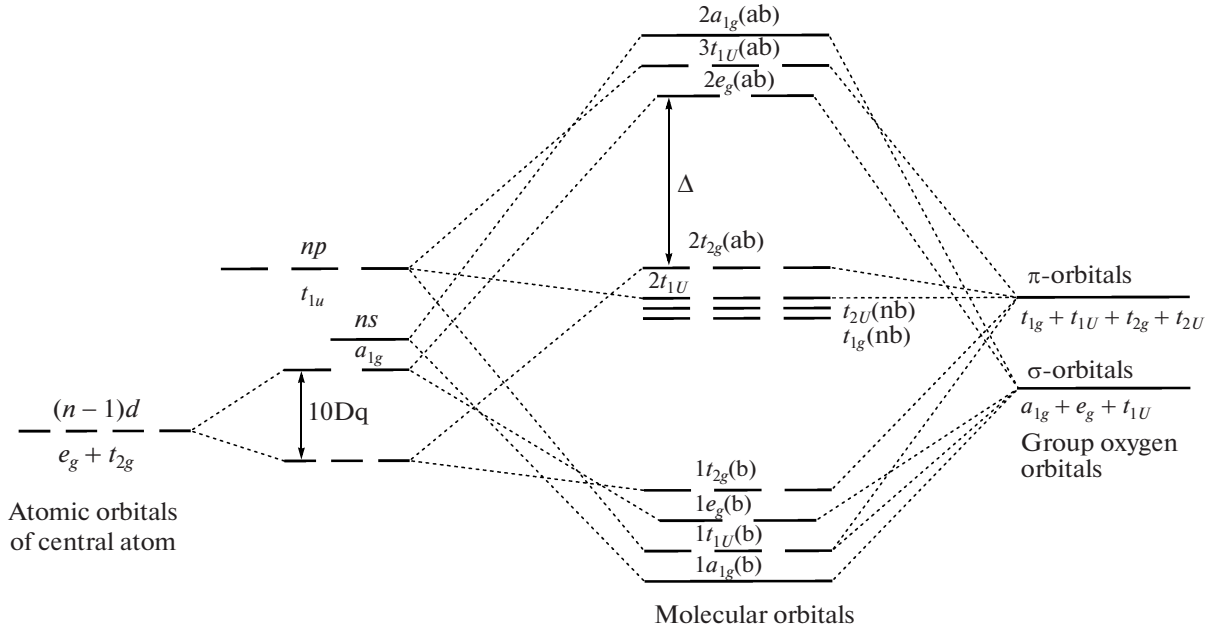


Fig. 2. Diagram of molecular orbitals and energy levels for an octahedral molecule with σ and π orbitals. Binding (b), nonbinding (nb), and antibinding (ab) molecular orbitals are indicated.

orbitals in the e_g and t_{2g} symmetries (see Table 1): $\bar{p}_\sigma^n = p_\sigma^{4-n}$ and $\bar{p}_\pi^k = p_\pi^{6-k}$. Henceforth, we will use the following notation:

$$e_g \begin{cases} d_\theta = d_{3z^2-r^2} \\ p_\theta = \frac{1}{2\sqrt{3}}(2\sigma_5 + 2\sigma_6 - \sigma_1 - \sigma_2 - \sigma_3 - \sigma_4) \\ d_\varepsilon = d_{x^2-y^2}, \quad p_\varepsilon = \frac{1}{2}(\sigma_1 - \sigma_2 + \sigma_3 - \sigma_4), \end{cases}$$

$$t_{2g} \begin{cases} d_\zeta = d_{xy}, \quad p_\zeta = \frac{1}{2}(\pi_{x1} + \pi_{y2} + \pi_{y3} + \pi_{x4}) \\ d_\xi = d_{yz}, \quad p_\xi = \frac{1}{2}(\pi_{x2} + \pi_{y5} + \pi_{y4} + \pi_{x6}) \\ d_\eta = d_{xz}, \quad p_\eta = \frac{1}{2}(\pi_{y1} + \pi_{x5} + \pi_{x3} + \pi_{y6}). \end{cases}$$

The wavefunction transformed in accordance with line M of representation Γ and characterized by total spin S and spin projection M_S will be denoted by $|\Gamma S M M_S\rangle$. This function can be written in the form

$$\begin{aligned} |\Gamma_1 S_1 \Gamma_2 S_2 \Gamma S M M_S\rangle &= \sum_{M_1 M_2} \langle \Gamma_1 \Gamma_2 M_1 M_2 | \Gamma M \rangle \\ &\times \sum_{M_{S_1} M_{S_2}} \langle S_1 S_2 M_{S_1} M_{S_2} | S M_S \rangle \\ &\times |\Gamma_1 S_1 M_1 M_{S_1}\rangle |\Gamma_2 S_2 M_2 M_{S_2}\rangle. \end{aligned}$$

Coefficients $\langle \Gamma_1 \Gamma_2 M_1 M_2 | \Gamma M \rangle$ on the right-hand side transform direct product $\Delta^{\Gamma_1} \times \Delta^{\Gamma_2}$ to a quasi-diagonal form and are analogous to the coefficients of vector summation (these coefficients are referred to as the Clebsch–Gordan coefficients of point groups), and Δ^{Γ_1} and Δ^{Γ_2} are the matrices of representations Γ_1 and Γ_2 . Wigner proved that instead of Clebsch–Gordan coefficients, we can introduce more symmetric 3Γ symbols $\left\langle \begin{matrix} \Gamma_1 & \Gamma_2 & \Gamma \\ M_1 & M_2 & M \end{matrix} \right\rangle$ for the SR group. For cubic groups, the relation between these symbols is

$$\langle \Gamma_1 \Gamma_2 M_1 M_2 | \Gamma M \rangle = [\Gamma]^{1/2} \left\langle \begin{matrix} \Gamma_1 & \Gamma_2 & \Gamma \\ M_1 & M_2 & M \end{matrix} \right\rangle,$$

where $[\Gamma]$ is the dimension of representation Γ [23]. We have also introduced the Clebsch–Gordan coefficients $\langle S_1 S_2 M_{S_1} M_{S_2} | S M_S \rangle$ for the spin component [24].

Let us write in explicit form the wavefunctions of term ${}^5T_{2g}$ for an ion with the d^6 configuration in an octahedral field taking into account covalent admixture of states with a single hole. Taking into account the conservation of total spin $S = 2$ and symmetry T_{2g} , we can form from the initial state $|t_{2g}^4({}^3T_1)e_g^2({}^3A_2){}^5T_{2g}M, M_S\rangle$ the two states,

$$|t_{2g}^4({}^3T_1)e_g^3({}^4T_1\bar{p}_\sigma){}^5T_{2g}M, M_S\rangle,$$

$$|t_{2g}^4({}^3T_1)e_g^3({}^4T_2\bar{p}_\sigma){}^5T_{2g}M, M_S\rangle,$$

while only one state, viz.,

$$|[t_{2g}^5e_g^2({}^3A_2)]^4T_1\bar{p}_\pi{}^5T_{2g}M, M_S\rangle.$$

can be formed due to the π bond. This can be represented by the following relations:

$$|[t_{2g}^4({}^3T_1)e_g^3({}^4T_1\bar{p}_\sigma)]^4T_1\bar{p}_\sigma{}^5T_{2g}M, M_S\rangle,$$

$$|[t_{2g}^4({}^3T_1)e_g^3({}^4T_2\bar{p}_\sigma)]^4T_2\bar{p}_\sigma{}^5T_{2g}M, M_S\rangle,$$

↙ due to the σ bond and

$$|t_{2g}^4({}^3T_1)e_g^2({}^3A_2){}^5T_{2g}M, M_S\rangle$$

$$\searrow$$

$$|[t_{2g}^5e_g^2({}^3A_2)]^4T_1\bar{p}_\pi{}^5T_{2g}M, M_S\rangle$$

due to the π bond.

Since

$$|t_{2g}^4({}^3T_1)[e_g^3\bar{p}_\sigma]^3A_2{}^5T_{2g}M, M_S\rangle$$

$$= \frac{1}{\sqrt{2}}|[t_{2g}^4({}^3T_1)e_g^3({}^4T_1\bar{p}_\sigma)]^4T_1\bar{p}_\sigma{}^5T_{2g}M, M_S\rangle$$

$$- \frac{1}{\sqrt{2}}|[t_{2g}^4({}^3T_1)e_g^3({}^4T_2\bar{p}_\sigma)]^4T_2\bar{p}_\sigma{}^5T_{2g}M, M_S\rangle.$$

The total state ${}^5T_{2g}$ can be written as the superposition of an ionic (d^6) and two covalent ($d^7\bar{p}_\sigma$ and $d^7\bar{p}_\pi$) components:

$$|{}^5T_{2g}M, M_S\rangle = C_1|t_{2g}^4({}^3T_1)e_g^2({}^3A_2){}^5T_{2g}M, M_S\rangle$$

$$+ C_2|t_{2g}^4({}^3T_1)[e_g^3\bar{p}_\sigma]^3A_2{}^5T_{2g}M, M_S\rangle \quad (1)$$

$$+ C_3|[t_{2g}^5e_g^2({}^3A_2)]^4T_1\bar{p}_\pi{}^5T_{2g}M, M_S\rangle,$$

where $M = \xi, \eta, \zeta$ and $M_S = -2, -1, 0, 1, 2$. Let us consider the wavefunction for $M = \xi$ and $M_S = 2$; in this case, we have

$$|t_{2g}^4({}^3T_1)e_g^2({}^3A_2){}^5T_{2g}M = \xi, M_S = 2\rangle$$

$$= \sum_{M_1M_2} \langle T_1A_2, M_1M_2 | T_2, \xi \rangle$$

$$\times \sum_{M_{S_1}M_{S_2}} \langle S_1 = 1 S_2 = 1, M_{S_1}M_{S_2} | S = 2, M_S = 2 \rangle \quad (2)$$

$$\times |t_{2g}^4({}^3T_1M_1, M_{S_1})|e_g^2({}^3A_2M_2, M_{S_2})\rangle = \xi^2\eta^+\theta^+\varepsilon^+$$

$$= d_{\xi\uparrow}^+d_{\xi\downarrow}^+d_{\eta\uparrow}^+d_{\zeta\uparrow}^+d_{\theta\uparrow}^+d_{\varepsilon\uparrow}^+|0\rangle.$$

Here, we assume that $M_1 = x$, $M_2 = a_2$, and $M_{S_1} = M_{S_2} = 1$ are the only possible values for which this expression differs from zero.

Analogously, we consider covalent component $d^7\bar{p}_\sigma$. We first form the wavefunction

$$|e_g^3\bar{p}_\sigma{}^3A_2a_2, M_S\rangle$$

$$= \sum_{M'_1M'_2} \langle EE, M'_1M'_2 | A_2, a_2 \rangle$$

$$\times \sum_{M_{S'_1}M_{S'_2}} \left\langle S'_1 = \frac{1}{2} S'_2 = \frac{1}{2}, M_{S'_1}M_{S'_2} \left| S' = 1, M_{S'} \right. \right\rangle$$

$$\times |e_g^3{}^2E_gM'_1, M_{S'_1})|\bar{p}_\sigma{}^2E_gM'_2, M_{S'_2})\rangle.$$

In this case, the ${}^5T_{2g}$ state for the $d^7\bar{p}_\sigma$ configuration with $M = \xi$ and $M_S = 2$ assumes the form

$$|t_{2g}^4({}^3T_1)[e_g^3\bar{p}_\sigma]^3A_2{}^5T_{2g}M = \xi, M_S = 2\rangle$$

$$= \sum_{M_1M_2} \langle T_1A_2, M_1M_2 | T_2, \xi \rangle$$

$$\times \sum_{M_{S_1}M_{S_2}} \langle S_1 = 1 S_2 = 1, M_{S_1}M_{S_2} | S = 2, M_S = +2 \rangle \quad (3)$$

$$\times |t_{2g}^4({}^3T_1M_1, M_{S_1})|e_g^3\bar{p}_\sigma{}^3A_2M_2, M_{S_2})\rangle$$

$$= \xi^2\eta^+\zeta^+\frac{1}{\sqrt{2}}[\theta^+\varepsilon^2\bar{p}_\varepsilon^+ - \theta^2\varepsilon^+\bar{p}_\theta^+]$$

$$= d_{\xi\uparrow}^+d_{\xi\downarrow}^+d_{\eta\uparrow}^+d_{\zeta\uparrow}^+\frac{1}{\sqrt{2}}[d_{\theta\uparrow}^+d_{\varepsilon\downarrow}^+d_{\varepsilon\downarrow}^+\bar{p}_{\varepsilon\uparrow}^+ - d_{\theta\uparrow}^+d_{\theta\downarrow}^+d_{\varepsilon\uparrow}^+\bar{p}_{\theta\uparrow}^+]|0\rangle.$$

Here, as well as in expression (2), we have $M_1 = x$, $M_2 = a_2$, and $M_{S_1} = M_{S_2} = 1$. For $d^7\bar{p}_\pi$, it is more convenient to form the wavefunction $|t_{2g}^5e_g^2({}^3A_2)^4T_1M', M_{S'}\rangle$ for the ionic d^7 configuration first:

$$|t_{2g}^5e_g^2({}^3A_2)^4T_1M', M_{S'}\rangle$$

$$\begin{aligned}
&= \sum_{M_1 M_2} \langle T_2 A_2, M_1' M_2' | T_1, M' \rangle \\
&\times \sum_{M_{S_1} M_{S_2}} \left\langle S_1' = \frac{1}{2} S_2' = \frac{1}{2}, M_{S_1} M_{S_2} \left| S' = \frac{3}{2}, M_S \right. \right\rangle \\
&\times |t_{2g}^5 T_{2g} M_1', M_{S_1}' \rangle |e_g^2 A_2 M_2', M_{S_2}' \rangle.
\end{aligned}$$

When a hole at an oxygen atom is added, we obtain the following expression for the $d^7 \bar{p}_\pi$ component of the ${}^5T_{2g}$ state:

$$\begin{aligned}
&| [t_{2g}^5 e_g^2 ({}^3A_2)]^4 T_1 \bar{p}_\pi {}^5T_{2g} M = \xi, M_S = 2 \rangle \\
&= \sum_{M_1 M_2} \langle T_1 T_2, M_1 M_2 | T_2, \xi \rangle \\
&\times \sum_{M_{S_1} M_{S_2}} \left\langle S_1 = \frac{3}{2} S_2 = \frac{1}{2}, M_{S_1} M_{S_2} \left| S = 2, M_S = 2 \right. \right\rangle \quad (4) \\
&\times |t_{2g}^5 e_g^2 ({}^3A_2)^4 T_1 M_1, M_{S_1}' \rangle | \bar{p}_\pi^2 T_{2g} M_2, M_{S_2}' \rangle \\
&= \frac{1}{\sqrt{2}} \{ \xi^2 \eta^2 \zeta^+ \bar{p}_\pi^+ - \xi^2 \eta^+ \zeta^2 \bar{p}_\pi^+ \} \theta^+ \varepsilon^+
\end{aligned}$$

$$= \frac{1}{\sqrt{2}} d_{\xi\uparrow}^+ d_{\xi\downarrow}^+ \{ d_{\eta\uparrow}^+ d_{\eta\downarrow}^+ d_{\zeta\uparrow}^+ \bar{p}_{\eta\uparrow}^+ - d_{\eta\uparrow}^+ d_{\zeta\uparrow}^+ d_{\zeta\downarrow}^+ \bar{p}_{\zeta\uparrow}^+ \} d_{\theta\uparrow}^+ d_{\varepsilon\uparrow}^+ |0\rangle,$$

where $M_1 = y, z$; $M_2 = \zeta, \eta$; $M_{S_1} = M_{S_2} = 1$; and $|0\rangle$ is the vacuum state $d^0 \underline{L}^0$.

As a result, the wavefunction for the ${}^5T_{2g}$ term can be written in the form

$$\begin{aligned}
|{}^5T_{2g} M = \xi, M_S = 2\rangle &= C_1 d_{\xi\uparrow}^+ d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ d_{\zeta\uparrow}^+ d_{\theta\uparrow}^+ d_{\varepsilon\uparrow}^+ |0\rangle \\
&+ C_2 d_{\xi\uparrow}^+ d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ d_{\zeta\uparrow}^+ \frac{1}{\sqrt{2}} \\
&\times [d_{\theta\uparrow}^+ d_{\varepsilon\uparrow}^+ d_{\varepsilon\downarrow}^+ \bar{p}_{\varepsilon\uparrow}^+ - d_{\theta\uparrow}^+ d_{\theta\downarrow}^+ d_{\varepsilon\uparrow}^+ \bar{p}_{\theta\uparrow}^+] |0\rangle \quad (5) \\
&+ C_3 \frac{1}{\sqrt{2}} d_{\xi\uparrow}^+ d_{\xi\downarrow}^+ \{ d_{\eta\uparrow}^+ d_{\eta\downarrow}^+ d_{\zeta\uparrow}^+ \bar{p}_{\eta\uparrow}^+ - d_{\eta\uparrow}^+ d_{\zeta\uparrow}^+ d_{\zeta\downarrow}^+ \bar{p}_{\zeta\uparrow}^+ \} \\
&\times d_{\theta\uparrow}^+ d_{\varepsilon\uparrow}^+ |0\rangle.
\end{aligned}$$

Wavefunctions for the e_g^l configuration, $l = 1, \dots, 4$, and the t_{2g}^m configuration, $m = 1, \dots, 6$, are given in Table 2. The recurrence formula considered in [25] makes it possible to calculate the matrix elements of the scalar operator of the Coulomb interaction between the states originating from the $e_g^l t_{2g}^m$ configurations. Analogous formula (A.8) for matrix elements

of the Coulomb interaction $e_g^l t_{2g}^m p_\sigma^n p_\pi^{M-(l+m+n)}$ is given in the Appendix (M is the total number of electrons in the system) for the four-subshell case. In the limit $n = 0$ and total number $M = l + m$ of electrons in the system, relation (A.8) assumes the form of a recurrence relation in the two-subshell case $e_g^l t_{2g}^m$ [25].

Recurrence relation (A.8) describes the intra-atomic interactions of electrons of a transition metal (H_{dd}^{Coul}), the interaction of holes at the anion with electrons (H_{pd}^{Coul}), and the interaction between holes at oxygen (H_{pp}^{Coul}). Since covalence is reduced to mixing of ionic configurations d^N without oxygen holes and of covalent configurations with a single p hole, the role of strong correlations for p holes is much less significant than for d electrons. For this reason, instead of a complete description of Coulomb interactions H_{pd}^{Coul} and H_{pp}^{Coul} , we use a simplified scheme, retaining only the density–density interaction with parameters V_{pd} and U_p , respectively. The general scheme developed for H_{dd}^{Coul} can also be used in principle for ligand electrons; this leads to the emergence of Racah parameters for ligands and for the cation–anion interaction, which extremely complicates the problem in our opinion.

Coefficients C_1 , C_2 , and C_3 in expression (1) are obtained by diagonalization of the Hamiltonian matrix for the ${}^5T_{2g}$ term in basis $\varphi_1, \varphi_2, \varphi_3$:

$$\varphi_1 = |t_{2g}^4 ({}^3T_1) e_g^2 ({}^3A_2) {}^5T_{2g}\rangle,$$

$$\varphi_2 = |t_{2g}^4 ({}^3T_1) [e_g^3 \bar{p}_\sigma] {}^3A_2 {}^5T_{2g}\rangle,$$

$$\varphi_3 = |[t_{2g}^5 e_g^2 ({}^3A_2)]^4 T_1 \bar{p}_\pi {}^5T_{2g}\rangle,$$

$$H_{11} = E_6 - 4Dq + 15A - 35B + 7C - E_{\text{av}}(d^6),$$

$$H_{22} = E_6 + \Delta_{\text{tr}} + 2Dq + 21A - 37B + 14C - E_{\text{av}}(d^7),$$

$$H_{33} = E_6 + \Delta_{\text{tr}} - W - 8Dq$$

$$+ 21A - 40B + 14C - E_{\text{av}}(d^7),$$

$$H_{12} = -2\sqrt{2}t_{pd}^\sigma, \quad H_{13} = 2\sqrt{2}t_{pd}^\pi, \quad H_{23} = 0,$$

where t_{pd}^σ and t_{pd}^π are the jump integrals for the σ and π bonds; A , B , and C are the Racah parameters,

$$W = \varepsilon_p(e_g) - \varepsilon_p(t_{2g}), \quad \varepsilon_p(e_g) = \varepsilon_p + ((pp\sigma) - (pp\pi)),$$

$$\varepsilon_p(t_{2g}) = \varepsilon_p - ((pp\sigma) - (pp\pi)),$$

$pp\sigma$ and $pp\pi$ are the Slater–Koster parameters [26], Δ_{tr} is the charge transfer energy, and

$$E_{\text{av}}(d^n) = \frac{1}{2} n(n-1) U_{\text{av}}^{\text{eff}}, \quad U_{\text{av}}^{\text{eff}} = A - \frac{14}{9} B + \frac{7}{9} C.$$

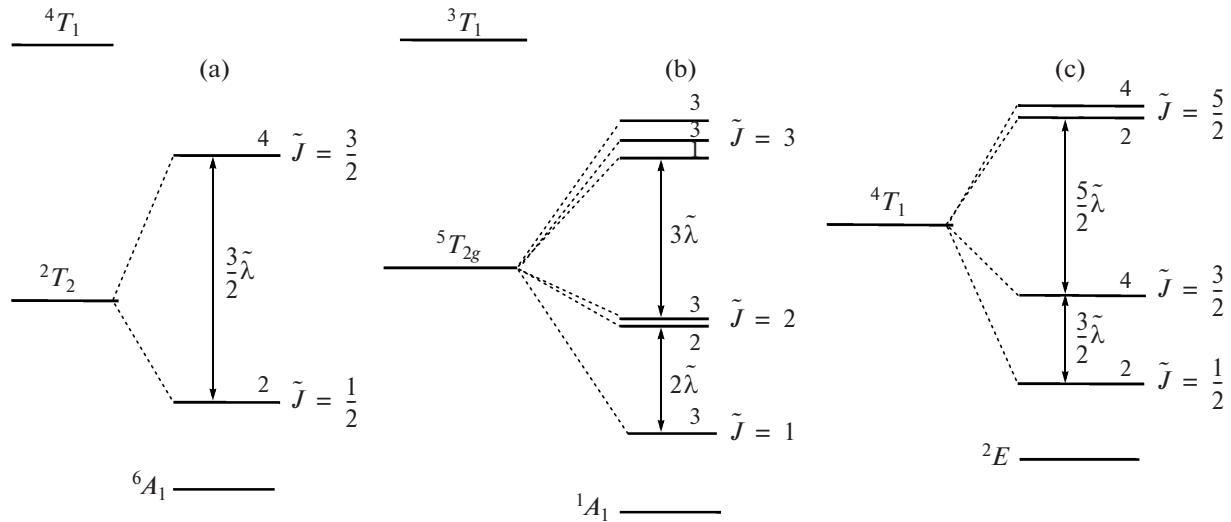


Fig. 3. Set of low-energy terms for (a) d^5 , (b) d^6 , and (c) d^7 configurations of an ion in the octahedral field taking covalence into account.

For $t_{pd}^\sigma = 0$ and $t_{pd}^\pi = 0$, we obtain, as in crystal field theory, pure state $|t_{2g}^4({}^3T_1)e_g^2({}^3A_2){}^5T_{2g}\rangle$ without a covalent admixture.

Exactly the same procedure can also be used for other terms. This gives the wavefunctions of the ground and excited states of configurations d^{n-1} , d^n , and d^{n+1} .

3. SPIN-ORBIT INTERACTION

The components of 3D representation of Γ_4 are transformed analogously to vector components. This means that the matrix elements of the orbital angular momentum components at wavefunctions of triplet states Γ_4 and Γ_5 differ from zero. Using the Wigner-Eckart theorem, we can prove that it is convenient in this case to use a pseudoangular momentum with components that have the same matrix elements (within manifold Γ_4 and Γ_5) as the matrix elements of orbital angular momentum $L = 1$ in the p state:

$$\langle \mathbf{L} \rangle = \alpha \tilde{\mathbf{I}},$$

where coefficient α is controlled by the specific structure of orbital triplets Γ_4 and Γ_5 , and angle brackets indicate projection onto the triplet manifold. In particular, $\alpha = -1$ for triplet Γ_5 originating from the D term; $\alpha = 1/2$ if energy level Γ_5 belongs to the F term, and $\alpha = -3/2$ for triplet Γ_4 belonging to the F term [27]. The existence of a finite angular momentum $\langle \mathbf{L} \rangle = \alpha \tilde{\mathbf{I}}$ indicates the existence of a nonzero spin-orbit interaction:

$$\lambda \langle \mathbf{L} \rangle \cdot \mathbf{S} = \alpha \lambda (\tilde{\mathbf{I}} \cdot \mathbf{S}) = \tilde{\lambda} (\tilde{\mathbf{I}} \cdot \mathbf{S}).$$

If we disregard the second-order effects associated with the matrix elements of operator $\tilde{\lambda}(\tilde{\mathbf{L}} \cdot \tilde{\mathbf{S}})$ corresponding to transitions to excited cubic multiplets, we can add total spin S and pseudomomentum $\tilde{l} = 1$. As a result, we obtain multiplets \tilde{J} assuming all values from $|S - 1|$ to $S + 1$ [27, 28] (Fig. 3). The figure also shows the splitting of multiplets of ions in crystal fields of cubic symmetry; the wavefunctions of these multiplets are given in Table 3.

We will be using the formalism of a fictitious angular momentum [27], in which basis one-electron functions η_x , η_y , and η_z of triplet t_2 are connected with eigenfunctions $|\tilde{m}\rangle$ of operator \tilde{l}_z via formulas

$$|\pm 1\rangle = \mp \frac{\eta_x \pm i\eta_y}{\sqrt{2}}, \quad |\tilde{0}\rangle = \eta_z. \quad (6)$$

In writing the eigenfunctions of the fictitious orbital angular momentum of the cubic term, we will use the following notation:

$$|\overline{\pm 1}\rangle = \mp \frac{\bar{X} \pm i\bar{Y}}{\sqrt{2}}, \quad |\overline{0}\rangle = \bar{Z}. \quad (7)$$

Expressions (6) and (7) differ in two respects: the former control the one-electron wavefunctions of cubic triplet t_2 , while the latter are multielectron wavefunctions of cubic triplet term T_1 or T_2 . In this case, the expression for the wavefunctions of various multiplets can be written in the form

$$S\tilde{T}\tilde{J}, \tilde{J}_z\rangle = \sum_{\bar{L}_z S_z} \langle \bar{L} = 1S, \bar{L}_z S_z | \tilde{J}, \tilde{J}_z \rangle |S\tilde{T}, \bar{L}_z S_z\rangle,$$

Table 2. Wavefunctions for configurations e_g^l ($l = 1, \dots, 4$) and t_{2g}^m ($m = 1, \dots, 6$) [19]

Configuration	Wavefunctions	Configuration	Wavefunctions
t_2^1	${}^2T_2\frac{1}{2}\xi = \xi^+ = d_{\xi, \uparrow}^+ 0\rangle$ ${}^2T_2\frac{1}{2}\eta = \eta^+ = d_{\eta, \uparrow}^+ 0\rangle$ ${}^2T_2\frac{1}{2}\zeta = \zeta^+ = d_{\zeta, \uparrow}^+ 0\rangle$	t_2^5	${}^2T_2\frac{1}{2}\xi = \xi^+ \eta^2 \zeta^2$ ${}^2T_2\frac{1}{2}\eta = \xi^2 \eta^+ \zeta^2$ ${}^2T_2\frac{1}{2}\zeta = \xi^2 \eta^2 \zeta^+$
t_2^2	${}^3T_1 1x = \zeta^+ \eta^+$ ${}^3T_1 1y = \xi^+ \zeta^+$ ${}^3T_1 1z = \eta^+ \xi^+$ ${}^1A_1 = \frac{1}{\sqrt{3}}(\xi^2 + \eta^2 + \zeta^2)$ ${}^1E\theta = \frac{1}{\sqrt{6}}(\xi^2 + \eta^2 - 2\zeta^2)$ ${}^1E\varepsilon = \frac{1}{\sqrt{2}}(\eta^2 - \xi^2)$ ${}^1T_2\xi = -\frac{1}{\sqrt{2}}(\eta^+ \zeta^- + \zeta^+ \eta^-)$ ${}^1T_2\eta = -\frac{1}{\sqrt{2}}(\xi^+ \zeta^- + \zeta^+ \xi^-)$ ${}^1T_2\zeta = -\frac{1}{\sqrt{2}}(\xi^+ \eta^- + \eta^+ \xi^-)$	t_2^4	${}^3T_1 1x = \xi^2 \eta^+ \zeta^+$ ${}^3T_1 1y = -\xi^+ \eta^2 \zeta^+$ ${}^3T_1 1z = \xi^+ \eta^+ \zeta^2$ ${}^1A_1 = \frac{1}{\sqrt{3}}(\xi^2 \eta^2 + \eta^2 \zeta^2 + \zeta^2 \xi^2)$ ${}^1E\theta = \frac{1}{\sqrt{6}}(\zeta^2 \xi^2 + \eta^2 \zeta^2 - 2\xi^2 \eta^2)$ ${}^1E\varepsilon = \frac{1}{\sqrt{2}}(\zeta^2 \xi^2 - \eta^2 \zeta^2)$ ${}^1T_2\xi = \frac{1}{\sqrt{2}}(\xi^2 \eta^+ \zeta^- - \xi^2 \eta^- \zeta^+)$ ${}^1T_2\eta = -\frac{1}{\sqrt{2}}(\xi^+ \eta^2 \zeta^- - \xi^- \eta^2 \zeta^+)$ ${}^1T_2\zeta = \frac{1}{\sqrt{2}}(\xi^+ \eta^- \zeta^2 - \xi^- \eta^+ \zeta^2)$
t_2^3	${}^4A_2\frac{3}{2}a_2 = -\xi^+ \eta^+ \zeta^+$ ${}^2E\frac{1}{2}\theta = \frac{1}{\sqrt{2}}(\xi^+ \eta^- \zeta^+ - \xi^- \eta^+ \zeta^+)$ ${}^2E\frac{1}{2}\varepsilon = \frac{1}{\sqrt{6}}(2\xi^+ \eta^+ \zeta^- - \xi^+ \eta^- \zeta^+ - \xi^- \eta^+ \zeta^+)$ ${}^2T_2\frac{1}{2}x = \frac{1}{\sqrt{2}}(\xi^+ \eta^2 - \xi^+ \zeta^2)$ ${}^2T_2\frac{1}{2}y = \frac{1}{\sqrt{2}}(\eta^+ \zeta^2 - \xi^2 \eta^+)$ ${}^2T_2\frac{1}{2}z = \frac{1}{\sqrt{2}}(\xi^2 \zeta^+ - \eta^2 \zeta^+)$ ${}^2T_2\frac{1}{2}\xi = \frac{1}{\sqrt{2}}(\xi^+ \zeta^2 + \xi^+ \eta^2)$ ${}^2T_2\frac{1}{2}\eta = \frac{1}{\sqrt{2}}(\eta^+ \zeta^2 + \xi^2 \eta^+)$ ${}^2T_2\frac{1}{2}\zeta = \frac{1}{\sqrt{2}}(\xi^2 \zeta^+ + \eta^2 \zeta^+)$	t_2^6	${}^1A_1 = \xi^2 \eta^2 \zeta^2$
e^1	${}^2E\frac{1}{2}\theta = \theta^+$ ${}^2E\frac{1}{2}\varepsilon = \varepsilon^+$	e^3	${}^2E\frac{1}{2}\theta = \theta^+ \varepsilon^2$ ${}^2E\frac{1}{2}\varepsilon = \theta^2 \varepsilon^+$
e^2	${}^3A_2 1a_2 = \theta^+ \varepsilon^+$ ${}^1A_1 = \frac{1}{\sqrt{2}}(\theta^2 + \varepsilon^2)$ ${}^1E\theta = \frac{1}{\sqrt{2}}(\varepsilon^2 - \theta^2)$ ${}^1E\varepsilon = \frac{1}{\sqrt{2}}(\theta^+ \varepsilon^- - \theta^- \varepsilon^+)$	e^4	${}^1A_1 = \theta^2 \varepsilon^2$

Note: Here, $\xi^2 = \xi^+ \xi^-$, and so on.

Table 3. Linear combinations of states $|JM_J\rangle$ for integer $J = 1, 2, 3, 4$ and half-integer values of J , which are transformed according to irreducible representations of the cubic and the double cubic group

J	States	J	States
$J = 0$	$ A_1 a_1\rangle = 00\rangle$	$J = \frac{1}{2}$	$ E'\alpha'\rangle = \left \frac{11}{22} \right\rangle$
$J = 1$	$ T_1 1\rangle = 11\rangle$ $ T_1 0\rangle = 10\rangle$ $ T_1, -1\rangle = 1, -1\rangle$		$ E'\beta'\rangle = \left \frac{1}{2}, -\frac{1}{2} \right\rangle$
$J = 2$	$ E\theta\rangle = 20\rangle$ $ E\varepsilon\rangle = \frac{1}{\sqrt{2}} 22\rangle + \frac{1}{\sqrt{2}} 2, -2\rangle$ $ T_2 1\rangle = 2, -1\rangle$ $ T_2 0\rangle = \frac{1}{\sqrt{2}} 22\rangle - \frac{1}{\sqrt{2}} 2, -2\rangle$ $ T_2, -1\rangle = - 21\rangle$	$J = \frac{5}{2}$	$ E''\alpha''\rangle = \frac{1}{\sqrt{6}}\left \frac{55}{22} \right\rangle - \frac{\sqrt{5}}{\sqrt{6}}\left \frac{5}{2}, -\frac{3}{2} \right\rangle$ $ E''\beta''\rangle = \frac{1}{\sqrt{6}}\left \frac{5}{2}, -\frac{5}{2} \right\rangle - \frac{\sqrt{5}}{\sqrt{6}}\left \frac{53}{22} \right\rangle$ $ U^{\kappa}\rangle = -\frac{1}{\sqrt{6}}\left \frac{53}{22} \right\rangle - \frac{\sqrt{5}}{\sqrt{6}}\left \frac{5}{2}, -\frac{5}{2} \right\rangle$ $ U^{\lambda}\rangle = \left \frac{51}{22} \right\rangle$ $ U^{\mu}\rangle = -\left \frac{5}{2}, -\frac{1}{2} \right\rangle$ $ U^{\nu}\rangle = \frac{1}{\sqrt{6}}\left \frac{5}{2}, -\frac{3}{2} \right\rangle + \frac{\sqrt{5}}{\sqrt{6}}\left \frac{55}{22} \right\rangle$
	$J = 3$		$ A_2 a_2\rangle = \frac{1}{\sqrt{2}} 32\rangle - \frac{1}{\sqrt{2}} 3, -2\rangle$ $ T_1 1\rangle = -\frac{\sqrt{5}}{2\sqrt{2}} 3, -3\rangle - \frac{\sqrt{3}}{2\sqrt{2}} 31\rangle$ $ T_1 0\rangle = 30\rangle$ $ T_1, -1\rangle = -\frac{\sqrt{5}}{2\sqrt{2}} 33\rangle - \frac{\sqrt{3}}{2\sqrt{2}} 3, -1\rangle$ $ T_2 1\rangle = -\frac{\sqrt{3}}{2\sqrt{2}} 33\rangle + \frac{\sqrt{5}}{2\sqrt{2}} 3, -1\rangle$ $ T_2 0\rangle = \frac{1}{\sqrt{2}} 32\rangle + \frac{1}{\sqrt{2}} 3, -2\rangle$ $ T_2, -1\rangle = -\frac{\sqrt{3}}{2\sqrt{2}} 3, -3\rangle + \frac{\sqrt{5}}{2\sqrt{2}} 31\rangle$
$J = 4$	$ A_1 a_1\rangle = \frac{\sqrt{7}}{2\sqrt{3}} 40\rangle + \frac{\sqrt{5}}{2\sqrt{6}} 44\rangle + \frac{\sqrt{5}}{2\sqrt{6}} 4, -4\rangle$ $ E\theta\rangle = -\frac{\sqrt{5}}{2\sqrt{3}} 40\rangle + \frac{\sqrt{7}}{2\sqrt{6}} 44\rangle + \frac{\sqrt{7}}{2\sqrt{6}} 4, -4\rangle$ $ E\varepsilon\rangle = \frac{1}{\sqrt{2}} 42\rangle + \frac{1}{\sqrt{2}} 4, -2\rangle$ $ T_1 1\rangle = -\frac{1}{2\sqrt{2}} 4, -3\rangle - \frac{\sqrt{7}}{2\sqrt{2}} 41\rangle$ $ T_1 0\rangle = \frac{1}{\sqrt{2}} 44\rangle - \frac{1}{\sqrt{2}} 4, -4\rangle$ $ T_1, -1\rangle = \frac{1}{2\sqrt{2}} 43\rangle + \frac{\sqrt{7}}{2\sqrt{2}} 4, -1\rangle$ $ T_2 1\rangle = \frac{\sqrt{7}}{2\sqrt{2}} 43\rangle - \frac{1}{2\sqrt{2}} 4, -1\rangle$ $ T_2 0\rangle = \frac{1}{\sqrt{2}} 42\rangle - \frac{1}{\sqrt{2}} 4, -2\rangle$ $ T_2, -1\rangle = \frac{1}{2\sqrt{2}} 41\rangle - \frac{\sqrt{7}}{2\sqrt{2}} 4, -3\rangle$	$J = \frac{7}{2}$	$ E'\alpha'\rangle = \frac{\sqrt{5}}{\sqrt{12}}\left \frac{7}{2}, -\frac{7}{2} \right\rangle + \frac{\sqrt{7}}{\sqrt{12}}\left \frac{71}{22} \right\rangle$ $ E'\beta'\rangle = -\frac{\sqrt{5}}{\sqrt{12}}\left \frac{77}{22} \right\rangle - \frac{\sqrt{7}}{\sqrt{12}}\left \frac{7}{2}, -\frac{1}{2} \right\rangle$ $ E''\alpha''\rangle = \frac{\sqrt{3}}{2}\left \frac{75}{22} \right\rangle - \frac{1}{2}\left \frac{7}{2}, -\frac{3}{2} \right\rangle$ $ E''\beta''\rangle = -\frac{\sqrt{3}}{2}\left \frac{7}{2}, -\frac{5}{2} \right\rangle + \frac{1}{2}\left \frac{73}{22} \right\rangle$ $ U^{\kappa}\rangle = \frac{\sqrt{3}}{2}\left \frac{73}{22} \right\rangle + \frac{1}{2}\left \frac{7}{2}, -\frac{5}{2} \right\rangle$ $ U^{\lambda}\rangle = \frac{\sqrt{7}}{\sqrt{12}}\left \frac{7}{2}, -\frac{7}{2} \right\rangle - \frac{\sqrt{5}}{\sqrt{12}}\left \frac{71}{22} \right\rangle$ $ U^{\mu}\rangle = \frac{\sqrt{7}}{\sqrt{12}}\left \frac{77}{22} \right\rangle - \frac{\sqrt{5}}{\sqrt{12}}\left \frac{7}{2}, -\frac{1}{2} \right\rangle$ $ U^{\nu}\rangle = \frac{\sqrt{3}}{2}\left \frac{7}{2}, -\frac{3}{2} \right\rangle + \frac{1}{2}\left \frac{75}{22} \right\rangle$

Note: Since the inclusion of covalence preserves the total orbital and spin angular momenta and changes only wavefunctions, linear combinations of states $|JM_J\rangle$ have the same form as in the crystal field theory [19, 27].

where wavefunctions $|\Sigma\Gamma, \bar{L}_Z S_Z\rangle$ are given by a superposition of configurations $d^N(\Sigma\Gamma)$ and $d^{N+R}\underline{L}^R(\Sigma\Gamma)$ due to covalence.

Let us consider in greater detail the form of the wavefunction of cubic term ${}^5T_{2g}$ (orbital triplet originating from term 5D) for the Co^{3+} ion in the octahedral surrounding. Under the action of spin-orbit coupling, the orbital triplet with fivefold degeneracy in spin ($S=2$) splits into a triplet, a quintet, and a septet (see Fig. 3). The triplet is the lowermost energy level. These results can easily be obtained considering that triplet ${}^5T_{2g}$ can be described by effective angular momentum $\tilde{l}=1$ with $\alpha=-1$. Then the "term" with $\alpha=-1$ and $S=2$ splits due to the spin-orbit interaction into states with effective total angular momenta $\tilde{J}=1, 2, 3$. Since spin-orbit interaction parameter λ is negative for configuration d^6 , we can expect that the multiplet will be reversed. In fact, we obtain a normal multiplet with the lower triplet state since $\alpha=-1$.

For the ground level, we have $\tilde{J}=1$, and the wavefunctions of states with $\tilde{J}_Z=\pm 1$ are defined by vector summation formulas:

$$\begin{aligned} & |{}^5T_2\tilde{J}=1, \tilde{J}_Z=0\rangle \\ &= -\sqrt{\frac{2}{5}}|{}^5T_2\bar{L}=1, \bar{L}_Z=0, S=2, S_Z=0\rangle \\ &+ \sqrt{\frac{3}{10}}|{}^5T_2\bar{L}=1, \bar{L}_Z=+1, S=2, S_Z=-1\rangle \\ &+ \sqrt{\frac{3}{10}}|{}^5T_2\bar{L}=1, \bar{L}_Z=-1, S=2, S_Z=+1\rangle, \\ & |{}^5T_2\tilde{J}=1, \tilde{J}_Z=\pm 1\rangle \\ &= -\sqrt{\frac{1}{10}}|{}^5T_2\bar{L}=1, \bar{L}_Z=\pm 1, S=2, S_Z=0\rangle \\ &- \sqrt{\frac{3}{10}}|{}^5T_2\bar{L}=1, \bar{L}_Z=0, S=2, S_Z=\pm 1\rangle \\ &+ \sqrt{\frac{3}{5}}|{}^5T_2\bar{L}=1, \bar{L}_Z=\mp 1, S=2, S_Z=\pm 2\rangle, \end{aligned}$$

where the components on the right-hand side are given by

$$\begin{aligned} & |{}^5T_2\bar{L}=1, \bar{L}_Z, S=2, S_Z\rangle \\ &= C_1|t_{2g}^4({}^3T_1)e_g^2({}^3A_2){}^5T_{2g}\bar{L}_Z, S_Z\rangle \\ &+ C_2|t_{2g}^4({}^3T_1)[e_g^3\bar{p}_\sigma]^3A_2{}^5T_{2g}\bar{L}_Z, S_Z\rangle \\ &+ C_3|[t_{2g}^5e_g^2({}^3A_2)]^4T_1\bar{p}_\pi{}^5T_{2g}\bar{L}_Z, S_Z\rangle. \end{aligned} \quad (8)$$

By way of example, we set $\bar{L}_Z=0$ and $S_Z=0$. Using expression (7), we obtain

$$\begin{aligned} & |t_{2g}^4({}^3T_1)e_g^2({}^3A_2){}^5T_{2g}\bar{L}_Z=0, S_Z=0\rangle \\ &= \frac{1}{\sqrt{6}}\zeta^2(\xi^+\eta^+\theta^-\varepsilon^- + \xi^+\eta^-\theta^+\varepsilon^- + \xi^-\eta^+\theta^+\varepsilon^- \\ &+ \xi^+\eta^-\theta^-\varepsilon^+ + \xi^-\eta^+\theta^-\varepsilon^+ + \xi^-\eta^-\theta^+\varepsilon^+). \end{aligned}$$

Analogously, for states with a single hole, we have

$$\begin{aligned} & |t_{2g}^4({}^3T_1)[e_g^3\bar{p}_\sigma]^3A_2{}^5T_{2g}\bar{L}_Z=0, S_Z=0\rangle \\ &= \frac{1}{\sqrt{6}}\zeta^2\left(\xi^+\eta^+\frac{1}{\sqrt{2}}[\theta^-\varepsilon^2\bar{p}_\varepsilon^- - \theta^2\varepsilon^-\bar{p}_\theta^-] + \xi^+\eta^-\frac{1}{\sqrt{2}} \right. \\ &\times [\theta^+\varepsilon^2\bar{p}_\varepsilon^- - \theta^2\varepsilon^-\bar{p}_\theta^+] + \xi^+\eta^-\frac{1}{\sqrt{2}}[\theta^-\varepsilon^2\bar{p}_\varepsilon^+ - \theta^2\varepsilon^+\bar{p}_\theta^-] \\ &+ \xi^-\eta^+\frac{1}{\sqrt{2}}[\theta^+\varepsilon^2\bar{p}_\varepsilon^- - \theta^2\varepsilon^-\bar{p}_\theta^+] \\ &+ \xi^-\eta^+\frac{1}{\sqrt{2}}[\theta^-\varepsilon^2\bar{p}_\varepsilon^+ - \theta^2\varepsilon^+\bar{p}_\theta^-] \\ &\left. + \xi^-\eta^-\frac{1}{\sqrt{2}}[\theta^+\varepsilon^2\bar{p}_\varepsilon^+ - \theta^2\varepsilon^+\bar{p}_\theta^+]\right) \end{aligned}$$

in the case of a σ bond and

$$\begin{aligned} & |[t_{2g}^5e_g^2({}^3A_2)]^4T_1\bar{p}_\pi{}^5T_{2g}\bar{L}_Z=0, S_Z=0\rangle \\ &= \frac{1}{\sqrt{6}}\zeta^2\left\{\frac{1}{\sqrt{2}}(\xi^2\eta^-\bar{p}_\xi^- - \xi^-\eta^2\bar{p}_\eta^-)\theta^+\varepsilon^+ \right. \\ &+ \frac{1}{\sqrt{2}}(\xi^2\eta^+\bar{p}_\xi^+ - \xi^+\eta^2\bar{p}_\eta^+)\theta^-\varepsilon^- \\ &+ \frac{1}{\sqrt{2}}(\xi^2\eta^+\bar{p}_\xi^- - \xi^+\eta^2\bar{p}_\eta^-)\theta^+\varepsilon^- \\ &+ \frac{1}{\sqrt{2}}(\xi^2\eta^-\bar{p}_\xi^+ - \xi^-\eta^2\bar{p}_\eta^+)\theta^+\varepsilon^- \\ &+ \frac{1}{\sqrt{2}}(\xi^2\eta^+\bar{p}_\xi^- - \xi^+\eta^2\bar{p}_\eta^-)\theta^-\varepsilon^+ \\ &\left. + \frac{1}{\sqrt{2}}(\xi^2\eta^-\bar{p}_\xi^+ - \xi^-\eta^2\bar{p}_\eta^+)\theta^-\varepsilon^+\right\} \end{aligned}$$

in the case of a π bond. Ultimately, we can write function (8) in the form

$$\begin{aligned}
& |^5T_2\bar{L} = 1, \bar{L}_Z = 0, S = 2, S_Z = 0\rangle \\
& = C_1 \frac{1}{\sqrt{6}} d_{\zeta\uparrow}^+ d_{\zeta\downarrow}^+ (d_{\xi\uparrow}^+ d_{\eta\uparrow}^+ d_{\theta\downarrow}^+ d_{\varepsilon\downarrow}^+ + d_{\xi\uparrow}^+ d_{\eta\downarrow}^+ d_{\theta\uparrow}^+ d_{\varepsilon\downarrow}^+ \\
& + d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ d_{\theta\uparrow}^+ d_{\varepsilon\downarrow}^+ + d_{\xi\uparrow}^+ d_{\eta\downarrow}^+ d_{\theta\downarrow}^+ d_{\varepsilon\uparrow}^+ + d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ d_{\theta\downarrow}^+ d_{\varepsilon\uparrow}^+ \\
& + d_{\xi\downarrow}^+ d_{\eta\downarrow}^+ d_{\theta\uparrow}^+ d_{\varepsilon\uparrow}^+) |0\rangle + C_2 \frac{1}{\sqrt{6}} d_{\zeta\uparrow}^+ d_{\zeta\downarrow}^+ \left(d_{\xi\uparrow}^+ d_{\eta\uparrow}^+ \frac{1}{\sqrt{2}} \right. \\
& \quad \times [d_{\theta\downarrow}^+ d_{\varepsilon\uparrow}^+ d_{\varepsilon\downarrow}^+ \bar{p}_{\varepsilon\downarrow}^+ - d_{\theta\uparrow}^+ d_{\theta\downarrow}^+ d_{\varepsilon\downarrow}^+ \bar{p}_{\theta\downarrow}^+] \\
& \quad + d_{\xi\uparrow}^+ d_{\eta\downarrow}^+ \frac{1}{\sqrt{2}} [d_{\theta\uparrow}^+ d_{\varepsilon\uparrow}^+ d_{\varepsilon\downarrow}^+ \bar{p}_{\varepsilon\downarrow}^+ - d_{\theta\uparrow}^+ d_{\theta\downarrow}^+ d_{\varepsilon\downarrow}^+ \bar{p}_{\theta\downarrow}^+] \\
& \quad + d_{\xi\uparrow}^+ d_{\eta\downarrow}^+ \frac{1}{\sqrt{2}} [d_{\theta\downarrow}^+ d_{\varepsilon\uparrow}^+ d_{\varepsilon\downarrow}^+ \bar{p}_{\varepsilon\uparrow}^+ - d_{\theta\uparrow}^+ d_{\theta\downarrow}^+ d_{\varepsilon\uparrow}^+ \bar{p}_{\theta\downarrow}^+] \\
& \quad + d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ \frac{1}{\sqrt{2}} [d_{\theta\uparrow}^+ d_{\varepsilon\uparrow}^+ d_{\varepsilon\downarrow}^+ \bar{p}_{\varepsilon\downarrow}^+ - d_{\theta\uparrow}^+ d_{\theta\downarrow}^+ d_{\varepsilon\downarrow}^+ \bar{p}_{\theta\uparrow}^+] \\
& \quad + d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ \frac{1}{\sqrt{2}} [d_{\theta\downarrow}^+ d_{\varepsilon\uparrow}^+ d_{\varepsilon\downarrow}^+ \bar{p}_{\varepsilon\uparrow}^+ - d_{\theta\uparrow}^+ d_{\theta\downarrow}^+ d_{\varepsilon\uparrow}^+ \bar{p}_{\theta\downarrow}^+] \\
& \quad \left. + d_{\xi\downarrow}^+ d_{\eta\downarrow}^+ \frac{1}{\sqrt{2}} [d_{\theta\uparrow}^+ d_{\varepsilon\uparrow}^+ d_{\varepsilon\downarrow}^+ \bar{p}_{\varepsilon\uparrow}^+ - d_{\theta\uparrow}^+ d_{\theta\downarrow}^+ d_{\varepsilon\uparrow}^+ \bar{p}_{\theta\downarrow}^+] \right) |0\rangle \\
& + C_3 \frac{1}{\sqrt{6}} d_{\zeta\uparrow}^+ d_{\zeta\downarrow}^+ \left\{ \frac{1}{\sqrt{2}} (d_{\xi\uparrow}^+ d_{\xi\downarrow}^+ d_{\eta\downarrow}^+ \bar{p}_{\xi\downarrow}^+ - d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ d_{\eta\downarrow}^+ \bar{p}_{\eta\downarrow}^+) \right. \\
& \times d_{\theta\uparrow}^+ d_{\varepsilon\uparrow}^+ + \frac{1}{\sqrt{2}} (d_{\xi\uparrow}^+ d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ \bar{p}_{\xi\uparrow}^+ - d_{\xi\uparrow}^+ d_{\eta\uparrow}^+ d_{\eta\downarrow}^+ \bar{p}_{\eta\uparrow}^+) d_{\theta\downarrow}^+ d_{\varepsilon\downarrow}^+ \\
& \quad + \frac{1}{\sqrt{2}} (d_{\xi\uparrow}^+ d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ \bar{p}_{\xi\downarrow}^+ - d_{\xi\uparrow}^+ d_{\eta\uparrow}^+ d_{\eta\downarrow}^+ \bar{p}_{\eta\downarrow}^+) d_{\theta\uparrow}^+ d_{\varepsilon\downarrow}^+ \\
& \quad + \frac{1}{\sqrt{2}} (d_{\xi\uparrow}^+ d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ \bar{p}_{\xi\uparrow}^+ - d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ d_{\eta\downarrow}^+ \bar{p}_{\eta\uparrow}^+) d_{\theta\uparrow}^+ d_{\varepsilon\downarrow}^+ \\
& \quad + \frac{1}{\sqrt{2}} (d_{\xi\uparrow}^+ d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ \bar{p}_{\xi\downarrow}^+ - d_{\xi\uparrow}^+ d_{\eta\uparrow}^+ d_{\eta\downarrow}^+ \bar{p}_{\eta\downarrow}^+) d_{\theta\downarrow}^+ d_{\varepsilon\uparrow}^+ \\
& \quad \left. + \frac{1}{\sqrt{2}} (d_{\xi\uparrow}^+ d_{\xi\downarrow}^+ d_{\eta\downarrow}^+ \bar{p}_{\xi\uparrow}^+ - d_{\xi\downarrow}^+ d_{\eta\uparrow}^+ d_{\eta\downarrow}^+ \bar{p}_{\eta\uparrow}^+) d_{\theta\downarrow}^+ d_{\varepsilon\uparrow}^+ \right\} |0\rangle.
\end{aligned}$$

4. MAGNETIC ANISOTROPY OF S IONS

Magnetic anisotropy of ferrimagnets with a high symmetry of the crystal structure is assumed to be one-ion type. Ferrites with the spinel or garnet structure belong to this group of crystals.

In the one-ion approach to anisotropy analysis, ions are classified into two categories: (i) ions with an orbital angular momentum in the ground state and (ii) ions with zero orbital angular momentum (i.e., ions in the S state, or S ions). Accordingly, two different theoretical approaches have been developed for describing magnetic anisotropy: the method of spin Hamiltonian for S ions [29, 30] and the method of Hamiltonian in the coordinate–momentum representation [31, 32] for ions with a nonzero orbital angular momentum.

In the spin Hamiltonian method, the reasons for anisotropy are not considered. It is only assumed that the coefficients in the expression for the Hamiltonian depend on the properties of cations and their surroundings. These coefficients are estimated by fitting the Hamiltonian spectrum to experimental spectra (e.g., EPR spectra) without explaining their relation to the physical properties of ions and crystals.

The knowledge of the mechanisms or processes of the orbital angular momentum formation for S ions would enable us to use a unified theoretical approach for describing magnetic anisotropy of the entire set of d ions [33].

Let us consider by way of example a d^5 ion (Fe^{3+} , Mn^{2+}) in cubic surroundings, in which the orbital angular momentum in the ground state 6A_1 is zero and the spin–orbit interaction makes zero contribution. As a consequence, the problem of magnetic anisotropy arises. The emergence of anisotropy in the theory of magnetism is usually considered in perturbation theory in covalence. We will show that covalence effects make a nonzero contribution to the spin–orbit interaction even in the ground state.

Covalent admixture for the 6A_1 multielectron term follows the following scheme:

$$\begin{aligned}
& [t_2^3({}^4A_2)e^3]^5 E\bar{p}_\sigma {}^6A_1\rangle \text{ due to the } \sigma \text{ bond and} \\
& \nearrow \\
& |t_2^3({}^4A_2)e^2({}^3A_2){}^6A_1\rangle \\
& \searrow \\
& [t_2^4({}^3T_1)e^2({}^3A_2)]^5 T_2\bar{p}_\pi {}^6A_1\rangle \text{ due to the } \pi \text{ bond.}
\end{aligned}$$

The total state can be written in the form

$$\begin{aligned}
|{}^6A_1 M, M_S\rangle & = C_1 |t_2^3({}^4A_2)e^2({}^3A_2){}^6A_1 M, M_S\rangle \\
& + C_2 [t_2^3({}^4A_2)e^3]^5 E\bar{p}_\sigma {}^6A_1 M, M_S\rangle \\
& + C_3 [t_2^4({}^3T_1)e^2({}^3A_2)]^5 T_2\bar{p}_\pi {}^6A_1 M, M_S\rangle,
\end{aligned}$$

where $M = a_1$, $M_S = -5/2, -3/2, -1/2, 1/2, 3/2$, and $5/2$; coefficients C_1 , C_2 , and C_3 are determined by exact diagonalization. For example, the contribution of the state $[t_2^4({}^3T_1)e^2({}^3A_2)]^5 T_2\bar{p}_\pi {}^6A_1\rangle$ we are interested

in to the total state ${}^6A_1M, M_S$) is 10% for parameters typical of iron oxides.

A state with the $t_{2g}^4e_g^2$ electron configuration is formed as a result of transfer to the t_{2g} shell at the d ion, which exhibits an energy behavior in the crystal field analogous to the behavior of the $\Gamma_5({}^5T_{2g}) t_{2g}^4e_g^2$ term; as a result, the Fe^{3+} ion (which is a classical representative of S ions) passes to the effective P state, acquiring an orbital angular momentum and changing its spin. The effective P state is described by orbital angular momentum $\tilde{l} = 1$ and orbital factor $\alpha = -1$ (see Section 3).

5. CONCLUSIONS

We have proposed a method for constructing the wavefunctions of multielectron terms of a transition metal ion taking into account covalence and the spin-orbit interaction. For this purpose, we generalized the apparatus of quantum mechanics for a free atom to the case of point groups. This approach enabled us to take into consideration the electron-electron interaction for $3d$ ions most comprehensively. A recurrence formula has been derived for calculating matrix elements of the Coulomb interaction between multielectron configurations with allowance for covalence. The procedure of construction and calculation was carried out for the ${}^5T_{2g}$ term in the d^6 configuration of the transition metal ion in the octahedral field. In each sector (d^{n-1} , d^n , and d^{n+1}) of the Hilbert space considered here, a certain set of energy terms exists (see, for example, Fig. 3), for which intersections and crossovers may take place. Applying the above technique for each term, we can determine the eigenstates of a cell or cluster with various numbers of electrons. Eigenstates $|p\rangle$ obtained in this way can be used for constructing Hubbard's X operators. The explicit form of eigenstates in the secondary quantization representation makes it possible to calculate matrix elements

$\gamma_{\lambda\sigma}(m) = \langle p|a_{\lambda\sigma}|q\rangle$ (and $\gamma_{\lambda\sigma}^\dagger(m) = \langle q|a_{\lambda\sigma}^\dagger|p\rangle$) of the jump amplitudes of the corresponding root vectors $\vec{\alpha}_m(pq)$. Such matrix elements could be calculated taking advantage of the fact that annihilation operator $a_{\mu\sigma}^{\gamma\Sigma}$ (or production operator $a_{\mu\sigma}^{+\gamma\Sigma}$, where index σ denoted spin projection Σ and μ denotes the line of irreducible representation γ) is a double cubic tensor. Using the Wigner-Eckart theorem, we then obtain

$$\langle \Gamma S M M_S | a_{\mu\sigma}^{\gamma\Sigma} | \Gamma' S' M' M'_S \rangle = (-1)^{S'+\Sigma+M_S} \times \left\langle \begin{array}{ccc} \Gamma' & \gamma & \Gamma \\ M' & \mu & M \end{array} \right\rangle \left(\begin{array}{ccc} S' & \sigma & S \\ M'_S & -\sigma & M_S \end{array} \right) \langle \Gamma S || a^{\gamma\Sigma} || \Gamma' S' \rangle,$$

where the explicit form of wavefunctions $|\Gamma S M M_S\rangle$ and $|\Gamma' S' M' M'_S\rangle$ is obviously unneeded. However, such an approach requires knowledge of the reduced matrix elements $\langle \Gamma S || a^{\gamma\Sigma} || \Gamma' S' \rangle$, the calculation of which is a cumbersome and complicated process. The multielectron basis constructed by us makes it possible to consider real multiorbital systems with various interactions using the generalized multiband Hubbard model and to apply a large number of methods developed for the Hubbard model for studying such systems (in particular, the generalized strong coupling method for calculating the band structure of a quasiparticle with allowance for the above-mentioned crossovers emerging upon a change in external conditions).

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APPENDIX

Recurrence Formula for the Matrix Elements of the Coulomb Interaction

Since the wavefunction is completely antisymmetric, we can write

$$\begin{aligned} & \langle e_g^l(S_1\Gamma_1)t_{2g}^m(S_2\Gamma_2)\{\tilde{S}_1\tilde{\Gamma}_1\}p_\sigma^n(S_3\Gamma_3)p_\pi^{M-(l+m+n)}(S_4\Gamma_4)\{\tilde{S}_2\tilde{\Gamma}_2\}S\Gamma|G_M\rangle \\ & \times e_g^l(S_5\Gamma_5)t_{2g}^{m'}(S_6\Gamma_6)\{\tilde{S}_3\tilde{\Gamma}_3\}p_\sigma^{n'}(S_7\Gamma_7)p_\pi^{M-(l'+m'+n')}(S_8\Gamma_8)\{\tilde{S}_4\tilde{\Gamma}_4\}S\Gamma\rangle \\ & = \frac{M}{M-2} \langle e_g^l(S_1\Gamma_1)t_{2g}^m(S_2\Gamma_2)\{\tilde{S}_1\tilde{\Gamma}_1\}p_\sigma^n(S_3\Gamma_3)p_\pi^{M-(l+m+n)}(S_4\Gamma_4)\{\tilde{S}_2\tilde{\Gamma}_2\}S\Gamma|G_{M-1}| \\ & \times e_g^l(S_5\Gamma_5)t_{2g}^{m'}(S_6\Gamma_6)\{\tilde{S}_3\tilde{\Gamma}_3\}p_\sigma^{n'}(S_7\tilde{\Gamma}_7)p_\pi^{M-(l'+m'+n')}(S_8\Gamma_8)\{\tilde{S}_4\tilde{\Gamma}_4\}S\Gamma\rangle, \end{aligned} \quad (\text{A.1})$$

where

$$G_M = \sum_{1 \leq i < j \leq M} G(ij) = \sum_{1 \leq i < j \leq M} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|}$$

is the Coulomb interaction operator.

The total M -electron functions with configuration $e_g^l t_{2g}^m p_\sigma^n p_\pi^{M-(l+m+n)}$ can be represented as the sum of the products of the wavefunctions for the d and p states,

$$\begin{aligned} |e_g^l(S_1\Gamma_1)t_{2g}^m(S_2\Gamma_2)\{\tilde{S}_1\tilde{\Gamma}_1\}p_\sigma^n(S_3\Gamma_3)p_\pi^{M-(l+m+n)}(S_4\Gamma_4)\{\tilde{S}_2\tilde{\Gamma}_2\}S\Gamma\rangle &= \{M!(l+m)!(M-(l+m))!\}^{1/2} \\ &\times \sum_{\substack{\nu \tilde{m}_1 \tilde{m}_2 \\ \tilde{\gamma}_1 \tilde{\gamma}_2}} (-1)^\nu P_\nu(T_{11} - T_{12} - T_{13} - \dots - T_{1M}) \langle \tilde{S}_1\tilde{S}_2, \tilde{m}_1\tilde{m}_2S, m \rangle \langle \tilde{\Gamma}_1\tilde{\Gamma}_2, \tilde{\gamma}_1\tilde{\gamma}_2\Gamma, \gamma \rangle \\ &\times |e_g^l(S_1\Gamma_1)t_{2g}^m(S_2\Gamma_2)\tilde{S}_1\tilde{\Gamma}_1\tilde{m}_1\tilde{\gamma}_1\rangle |p_\sigma^n(S_3\Gamma_3)p_\pi^{M-(l+m+n)}(S_4\Gamma_4)\tilde{S}_2\tilde{\Gamma}_2\tilde{m}_2\tilde{\gamma}_2\rangle, \end{aligned} \quad (\text{A.2})$$

where ν runs through all transpositions of $M-1$ electrons except the first one, T_{1i} denoting the transposition of the i th and first electrons. Wavefunctions $|e_g^l(S_1\Gamma_1)t_{2g}^m(S_2\Gamma_2)\tilde{S}_1\tilde{\Gamma}_1\tilde{m}_1\tilde{\gamma}_1\rangle$ and $|p_\sigma^n(S_3\Gamma_3)p_\pi^{M-(l+m+n)}(S_4\Gamma_4)\tilde{S}_2\tilde{\Gamma}_2\tilde{m}_2\tilde{\gamma}_2\rangle$ on the right-hand side contain the coordinates of the first $(l+m)$ and last $(M-(l+m))$ electrons, both functions being antisymmetric. Consequently, expression (A.2) can be decomposed into two parts, $|X_{et_2}\rangle$ and $|X_{p_\sigma p_\pi}\rangle$; for the first term, we obtain

$$\begin{aligned} |X_{et_2}\rangle &= \{M!(l+m)!(M-(l+m))!\}^{1/2} \sum_{\substack{\nu \tilde{m}_1 \tilde{m}_2 \\ \tilde{\gamma}_1 \tilde{\gamma}_2}} (-1)^\nu P_\nu(T_{11} - T_{12} - \dots - T_{1l+m}) \\ &\times \langle \tilde{S}_1\tilde{S}_2, \tilde{m}_1\tilde{m}_2 | S, m \rangle \langle \tilde{\Gamma}_1\tilde{\Gamma}_2, \tilde{\gamma}_1\tilde{\gamma}_2 | \Gamma, \gamma \rangle \\ &\times |e_g^l(S_1\Gamma_1)t_{2g}^m(S_2\Gamma_2)\tilde{S}_1\tilde{\Gamma}_1\tilde{m}_1\tilde{\gamma}_1\rangle |p_\sigma^n(S_3\Gamma_3)p_\pi^{M-(l+m+n)}(S_4\Gamma_4)\tilde{S}_2\tilde{\Gamma}_2\tilde{m}_2\tilde{\gamma}_2\rangle \\ &= \frac{l+m}{\{M!(l+m)!(M-(l+m))!\}^{1/2}} \sum_{\substack{\nu \tilde{m}_1 \tilde{m}_2 \\ \tilde{\gamma}_1 \tilde{\gamma}_2}} (-1)^\nu P_\nu \langle \tilde{S}_1\tilde{S}_2, \tilde{m}_1\tilde{m}_2 | S, m \rangle \langle \tilde{\Gamma}_1\tilde{\Gamma}_2, \tilde{\gamma}_1\tilde{\gamma}_2 | \Gamma, \gamma \rangle \\ &\times |e_g^l(S_1\Gamma_1)t_{2g}^m(S_2\Gamma_2)\tilde{S}_1\tilde{\Gamma}_1\tilde{m}_1\tilde{\gamma}_1\rangle |p_\sigma^n(S_3\Gamma_3)p_\pi^{M-(l+m+n)}(S_4\Gamma_4)\tilde{S}_2\tilde{\Gamma}_2\tilde{m}_2\tilde{\gamma}_2\rangle. \end{aligned} \quad (\text{A.3})$$

Analogously, wavefunction $|e_g^l(S_1\Gamma_1)t_{2g}^m(S_2\Gamma_2)\tilde{S}_1\tilde{\Gamma}_1\tilde{m}_1\tilde{\gamma}_1\rangle$ for d electrons itself can be decomposed into two parts, $|Y_e\rangle$ and $|Y_{t_2}\rangle$; for the first part, we have

$$\begin{aligned} |Y_e\rangle &= \frac{l}{\{(l+m)!l!m!\}^{1/2}} \sum_{\substack{\nu' m_1 m_2 \\ \gamma_1 \gamma_2}} (-1)^{\nu'} P_{\nu'} \\ &\times \langle S_1 S_2, m_1 m_2 | \tilde{S}_1, \tilde{m}_1 \rangle \langle \Gamma_1 \Gamma_2, \gamma_1 \gamma_2 | \tilde{\Gamma}_1, \tilde{\gamma}_1 \rangle |e_g^l S_1 \Gamma_1 m_1 \gamma_1\rangle |t_{2g}^m S_2 \Gamma_2 m_2 \gamma_2\rangle, \end{aligned} \quad (\text{A.4})$$

where ν' runs through all transpositions of $l + m - 1$ electrons except the first one. Substituting expression (A.4) into (A.3), we decompose $|X_{e_t}\rangle$ into $|X_e\rangle$ and $|X_{t_e}\rangle$. For $|X_e\rangle$, we obtain

$$\begin{aligned}
|X_e\rangle &= \frac{l+m}{\{M!(l+m)!(M-(l+m))!\}^{1/2}} \frac{1}{\{(l+m)!l!m!\}^{1/2}} \sum_{\substack{\tilde{m}_1\tilde{m}_2 \\ \tilde{\gamma}_1\tilde{\gamma}_2}} (-1)^\nu P_\nu \sum_{\substack{\nu'm_1m_2 \\ \gamma_1\gamma_2}} (-1)^{\nu'} P_{\nu'} \\
&\times \langle \tilde{S}_1\tilde{S}_2, \tilde{m}_1\tilde{m}_2 | S, m \rangle \langle \tilde{\Gamma}_1\tilde{\Gamma}_2, \tilde{\gamma}_1\tilde{\gamma}_2 | \Gamma, \gamma \rangle \langle S_1S_2, m_1m_2 | \tilde{S}_1, \tilde{m}_1 \rangle \langle \Gamma_1\Gamma_2, \gamma_1\gamma_2 | \tilde{\Gamma}_1, \tilde{\gamma}_1 \rangle \\
&\times |e'_g S_1\Gamma_1 m_1\gamma_1\rangle |t_{2g}^m S_2\Gamma_2 m_2\gamma_2\rangle |p_\sigma^n(S_3\Gamma_3) p_\pi^{M-(l+m+n)}(S_4\Gamma_4) \tilde{S}_2\tilde{\Gamma}_2 \tilde{m}_2\tilde{\gamma}_2\rangle \\
&= \frac{l+m}{\{M!(l+m)!(M-(l+m))!\}^{1/2}} \frac{l}{\{(l+m)!l!m!\}^{1/2}} \sum_{\substack{\tilde{m}_1\tilde{m}_2 \\ \tilde{\gamma}_1\tilde{\gamma}_2}} (-1)^\nu P_\nu \sum_{\substack{\nu'm_1m_2 \\ \gamma_1\gamma_2}} (-1)^{\nu'} P_{\nu'} \\
&\times \langle \tilde{S}_1\tilde{S}_2, \tilde{m}_1\tilde{m}_2 | S, m \rangle \langle \tilde{\Gamma}_1\tilde{\Gamma}_2, \tilde{\gamma}_1\tilde{\gamma}_2 | \Gamma, \gamma \rangle \langle S_1S_2, m_1m_2 | \tilde{S}_1, \tilde{m}_1 \rangle \langle \Gamma_1\Gamma_2, \gamma_1\gamma_2 | \tilde{\Gamma}_1, \tilde{\gamma}_1 \rangle \\
&\times \sum_{\substack{S'_1\Gamma'_1 m'_1\gamma'_1 \\ m_{1/2}\gamma_e}} (ee^{l-1}(S'_1\Gamma'_1)S_1\Gamma_1 |) e' S_1\Gamma_1 \langle \frac{1}{2} S'_1, m_{1/2} m'_1 | S_1, m_1 \rangle \langle E\Gamma'_1, \gamma_e \gamma'_1 | \Gamma_1, \gamma_1 \rangle \\
&\times |e \frac{1}{2} E m_{1/2} \gamma_e \rangle |e'_g{}^{l-1} S'_1\Gamma'_1 m'_1\gamma'_1\rangle |t_{2g}^m S_2\Gamma_2 m_2\gamma_2\rangle |p_\sigma^n(S_3\Gamma_3) p_\pi^{M-(l+m+n)}(S_4\Gamma_4) \tilde{S}_2\tilde{\Gamma}_2 \tilde{m}_2\tilde{\gamma}_2\rangle \\
&= \frac{l+m}{\{M!(l+m)!(M-(l+m))!\}^{1/2}} \left(\frac{l}{l+m}\right)^{1/2} \sum_{\substack{\tilde{m}_1\tilde{m}_2 \\ \tilde{\gamma}_1\tilde{\gamma}_2}} (-1)^\nu P_\nu \sum_{\substack{m_1m_2 S'_1\Gamma'_1 m'_1\gamma'_1 \\ \gamma_1\gamma_2 m_{1/2}\gamma_e \\ \tilde{S}_1\tilde{\Gamma}_1 \tilde{m}_1\tilde{\gamma}_1}} \langle \tilde{S}_1\tilde{S}_2, \tilde{m}_1\tilde{m}_2 | S, m \rangle \\
&\times \langle \tilde{\Gamma}_1\tilde{\Gamma}_2, \tilde{\gamma}_1\tilde{\gamma}_2 | \Gamma, \gamma \rangle \langle S_1S_2, m_1m_2 | \tilde{S}_1, \tilde{m}_1 \rangle \langle \Gamma_1\Gamma_2, \gamma_1\gamma_2 | \tilde{\Gamma}_1, \tilde{\gamma}_1 \rangle \langle \frac{1}{2} S'_1, m_{1/2} m'_1 | S_1, m_1 \rangle \\
&\times \langle S\Gamma'_1, \gamma_e \gamma'_1 | \Gamma_1, \gamma_1 \rangle \langle S'_1S_2, m'_1m_2 | \tilde{S}_1, \tilde{m}_1 \rangle \langle \Gamma'_1\Gamma_2, \gamma'_1\gamma_2 | \tilde{\Gamma}_1, \tilde{\gamma}_1 \rangle (ee^{l-1}(S'_1\Gamma'_1)S_1\Gamma_1 |) e' S_1\Gamma_1 \\
&\times |e \frac{1}{2} E m_{1/2} \gamma_e \rangle |e'_g{}^{l-1} (S'_1\Gamma'_1) t_{2g}^m (S_2\Gamma_2) \tilde{S}_1\tilde{\Gamma}_1 \tilde{\gamma}_1 \tilde{m}_1 \rangle |p_\sigma^n(S_3\Gamma_3) p_\pi^{M-(l+m+n)}(S_4\Gamma_4) \tilde{S}_2\tilde{\Gamma}_2 \tilde{m}_2\tilde{\gamma}_2\rangle \\
&= \left(\frac{l+m}{M}\right)^{1/2} \left(\frac{l}{l+m}\right)^{1/2} \sum_{\substack{\tilde{m}_1\tilde{m}_2 m_1 m_2 S'_1\Gamma'_1 m'_1\gamma'_1 \tilde{S}_1\tilde{\Gamma}_1 \tilde{S}\tilde{\Gamma}' \\ \tilde{\gamma}_1\tilde{\gamma}_2 \gamma_1\gamma_2 m_{1/2}\gamma_e \tilde{m}_1\tilde{\gamma}_1 \tilde{m}\tilde{\gamma}'}} \sum \sum \sum \sum \langle \tilde{S}_1\tilde{S}_2, \tilde{m}_1\tilde{m}_2 | S, m \rangle \langle \tilde{\Gamma}_1\tilde{\Gamma}_2, \tilde{\gamma}_1\tilde{\gamma}_2 | \Gamma, \gamma \rangle \\
&\times \langle S_1S_2, m_1m_2 | \tilde{S}_1, \tilde{m}_1 \rangle \langle \Gamma_1\Gamma_2, \gamma_1\gamma_2 | \tilde{\Gamma}_1, \tilde{\gamma}_1 \rangle \langle \frac{1}{2} S'_1, m_{1/2} m'_1 | S_1, m_1 \rangle \langle E\Gamma'_1, \gamma_e \gamma'_1 | \Gamma_1, \gamma_1 \rangle \\
&\times \langle S'_1S_2, m'_1m_2 | \tilde{S}_1, \tilde{m}_1 \rangle \langle \Gamma'_1\Gamma_2, \gamma'_1\gamma_2 | \tilde{\Gamma}_1, \tilde{\gamma}_1 \rangle \langle \tilde{S}_1\tilde{S}_2, \tilde{m}_1\tilde{m}_2 | \tilde{S}, \tilde{m} \rangle \langle \tilde{\Gamma}_1\tilde{\Gamma}_2, \tilde{\gamma}_1\tilde{\gamma}_2 | \tilde{\Gamma}, \tilde{\gamma} \rangle \\
&\times \langle \frac{1}{2} \tilde{S}, m_{1/2} \tilde{m} | S', m' \rangle \langle E\tilde{\Gamma}, \gamma_e \tilde{\gamma} | \Gamma', \gamma' \rangle (ee^{l-1}(S'_1\Gamma'_1)S_1\Gamma_1 |) e' S_1\Gamma_1 \\
&\times |ee'_g{}^{l-1} (S'_1\Gamma'_1) t_{2g}^m (S_2\Gamma_2) \{ \tilde{S}_1\tilde{\Gamma}_1 \} p_\sigma^n(S_3\Gamma_3) p_\pi^{M-(l+m+n)}(S_4\Gamma_4) \{ \tilde{S}_2\tilde{\Gamma}_2 \} \tilde{S}\tilde{\Gamma}' S'\Gamma' m'\gamma' \rangle.
\end{aligned} \tag{A.5}$$

It should be noted in this connection that for all values of $S'\Gamma'm'\gamma'$, a nonzero contribution to (A.1) comes only from $S' = S$, $\Gamma' = \Gamma$, $m' = m$, and $\gamma' = \gamma$.

Expressing summation over indices m and γ in terms of the Racah coefficients, we can write $|X_e\rangle$ in the form

$$\begin{aligned}
 |X_e\rangle = & \left(\frac{l}{M}\right)^{1/2} \sum_{\substack{S_1 \bar{S}_1 \bar{S} \\ \Gamma_1 \bar{\Gamma}_1 \bar{\Gamma}}} (e^l S_1 \Gamma_1 \{ | e e^{l-1} (S_1' \Gamma_1') S_1 \Gamma_1 \} \left(\left[\frac{1}{2} S_1' (S_1), S_2 \right] \{ \bar{S}_1 \} \bar{S}_2 S \middle| \frac{1}{2}, [S_1' S_2 (\bar{S}_1) \bar{S}_2] \{ \bar{S} \} S \right) \\
 & \times ([E \Gamma_1' (\Gamma_1), \Gamma_2] \{ \tilde{\Gamma}_1 \} \tilde{\Gamma}_2 \Gamma | E, [\Gamma_1' \Gamma_2 (\bar{\Gamma}_1) \bar{\Gamma}_2] \{ \tilde{\Gamma} \} \Gamma) \\
 & \times | e e_g^{l-1} (S_1' \Gamma_1') t_{2g}^m (S_2 \Gamma_2) \{ \bar{S}_1 \bar{\Gamma}_1 \} p_\sigma^n (S_3 \Gamma_3) p_\pi^{M-(l+m+n)} (S_4 \Gamma_4) \{ \tilde{S}_2 \tilde{\Gamma}_2 \} \bar{S} \bar{\Gamma} \}.
 \end{aligned} \tag{A.6}$$

Functions $|X_{t_2}\rangle$, $|X_{p_\sigma}\rangle$, and $|X_{p_\pi}\rangle$ can be found analogously.

Since orbitals d and p are mutually orthogonal, matrix element (A.1) can be split into components, each of which is a matrix element of the Coulomb interaction among $M-1$ electrons:

$$\langle X | G_M | X' \rangle = \langle X_e | G_{M-1} | X'_e \rangle + \langle X_{t_2} | G_{M-1} | X'_{t_2} \rangle + \langle X_{p_\sigma} | G_{M-1} | X'_{p_\sigma} \rangle + \langle X_{p_\pi} | G_{M-1} | X'_{p_\pi} \rangle. \tag{A.7}$$

The final expression for the matrix elements has the form

$$\begin{aligned}
 & \langle e_g^l (S_1 \Gamma_1) t_{2g}^m (S_2 \Gamma_2) \{ \tilde{S}_1 \tilde{\Gamma}_1 \} p_\sigma^n (S_3 \Gamma_3) p_\pi^{M-(l+m+n)} (S_4 \Gamma_4) \{ \tilde{S}_2 \tilde{\Gamma}_2 \} S \Gamma | G_M | \\
 & \times e_g^{l'} (S_5 \Gamma_5) t_{2g}^{m'} (S_6 \Gamma_6) \{ \tilde{S}_3 \tilde{\Gamma}_3 \} p_\sigma^{n'} (S_7 \Gamma_7) p_\pi^{M-(l'+m'+n')} (S_8 \Gamma_8) \{ \tilde{S}_4 \tilde{\Gamma}_4 \} S \Gamma \rangle = \frac{\sqrt{l l'}}{M-2} \\
 & \times \sum_{\substack{S_1 \Gamma_1 \bar{S}_1 \bar{\Gamma}_1 \\ \bar{S} \bar{\Gamma} S_3 \Gamma_3 \bar{S}_3 \bar{\Gamma}_3}} (e^l S_1 \Gamma_1 \{ | e e^{l-1} (S_1' \Gamma_1') S_1 \Gamma_1 \} \left(\left[\frac{1}{2} S_1' (S_1), S_2 \right] \{ \bar{S}_1 \} \bar{S}_2 S \middle| \frac{1}{2}, [S_1' S_2 (\bar{S}_1) \bar{S}_2] \{ \bar{S} \} S \right) \\
 & \times ([E \Gamma_1' (\Gamma_1), \Gamma_2] \{ \tilde{\Gamma}_1 \} \tilde{\Gamma}_2 \Gamma | E, [\Gamma_1' \Gamma_2 (\bar{\Gamma}_1) \bar{\Gamma}_2] \{ \tilde{\Gamma} \} \Gamma) \\
 & \times \langle e_g^{l-1} (S_1' \Gamma_1') t_{2g}^m (S_2 \Gamma_2) \{ \bar{S}_1 \bar{\Gamma}_1 \} p_\sigma^n (S_3 \Gamma_3) p_\pi^{M-(l+m+n)} (S_4 \Gamma_4) \{ \tilde{S}_2 \tilde{\Gamma}_2 \} \bar{S} \bar{\Gamma} | G_{M-1} | \\
 & \times e_g^{l'-1} (S_5' \Gamma_5') t_{2g}^{m'} (S_6 \Gamma_6) \{ \bar{S}_3 \bar{\Gamma}_3 \} p_\sigma^{n'} (S_7 \Gamma_7) p_\pi^{M-(l'+m'+n')} (S_8 \Gamma_8) \{ \tilde{S}_4 \tilde{\Gamma}_4 \} \bar{S} \bar{\Gamma} \rangle \\
 & \times \left(\left[\frac{1}{2} S_5' (S_5), S_6 \right] \{ \bar{S}_3 \} \bar{S}_4 S \middle| \frac{1}{2}, [S_5' S_6 (\bar{S}_3) \bar{S}_4] \{ \bar{S} \} S \right) \\
 & \times ([E \Gamma_5' (\Gamma_5), \Gamma_6] \{ \tilde{\Gamma}_3 \} \tilde{\Gamma}_4 \Gamma | E, [\Gamma_5' \Gamma_6 (\bar{\Gamma}_3) \bar{\Gamma}_4] \{ \tilde{\Gamma} \} \Gamma) (e^l S_5 \Gamma_5 \{ | e e^{l-1} (S_5' \Gamma_5') S_5 \Gamma_5 \} \\
 & + \frac{\sqrt{m m'}}{M-2} \sum_{\substack{S_2 \Gamma_2 \bar{S}_1 \bar{\Gamma}_1 \\ \bar{S} \bar{\Gamma} S_6 \Gamma_6 \bar{S}_3 \bar{\Gamma}_3}} (t_2^m S_2 \Gamma_2 \{ | t_2 t_2^{m-1} (S_2' \Gamma_2') S_2 \Gamma_2 \} \left(\left[\frac{1}{2} S_2' (S_2), S_1 \right] \{ \bar{S}_1 \} \bar{S}_2 S \middle| \frac{1}{2}, [S_2' S_1 (\bar{S}_1) \bar{S}_2] \{ \bar{S} \} S \right) \\
 & \times ([T_2 \Gamma_2' (\Gamma_2), \Gamma_1] \{ \tilde{\Gamma}_1 \} \tilde{\Gamma}_2 \Gamma | T_2, [\Gamma_2' \Gamma_1 (\bar{\Gamma}_1) \bar{\Gamma}_2] \{ \tilde{\Gamma} \} \Gamma) \\
 & \times \langle e_g^l (S_1 \Gamma_1) t_{2g}^{m-1} (S_2' \Gamma_2') \{ \bar{S}_1 \bar{\Gamma}_1 \} p_\sigma^n (S_3 \Gamma_3) p_\pi^{M-(l+m+n)} (S_4 \Gamma_4) \{ \tilde{S}_2 \tilde{\Gamma}_2 \} \bar{S} \bar{\Gamma} | G_{M-1} | \\
 & \times e_g^{l'} (S_5 \Gamma_5) t_{2g}^{m'-1} (S_6' \Gamma_6') \{ \bar{S}_3 \bar{\Gamma}_3 \} p_\sigma^{n'} (S_7 \Gamma_7) p_\pi^{M-(l'+m'+n')} (S_8 \Gamma_8) \{ \tilde{S}_4 \tilde{\Gamma}_4 \} \bar{S} \bar{\Gamma} \rangle \\
 & \times \left(\left[\frac{1}{2} S_6' (S_6), S_5 \right] \{ \bar{S}_3 \} \bar{S}_4 S \middle| \frac{1}{2}, [S_6' S_5 (\bar{S}_3) \bar{S}_4] \{ \bar{S} \} S \right) ([T_2 \Gamma_6' (\Gamma_6), \Gamma_5] \{ \tilde{\Gamma}_3 \} \tilde{\Gamma}_4 \Gamma | T_2, [\Gamma_6' \Gamma_5 (\bar{\Gamma}_3) \bar{\Gamma}_4] \{ \tilde{\Gamma} \} \Gamma) \tag{A.8}
 \end{aligned}$$

$$\begin{aligned}
& \times (t_2^m S_6 \Gamma_6 \{ |t_2^{m-1} (S_6' \Gamma_6') S_6 \Gamma_6 \} + \frac{\sqrt{nn'}}{M-2} \sum_{\substack{S_3 \Gamma_3 \bar{S}_2 \bar{\Gamma}_2 \\ \bar{S} \bar{\Gamma} S_7 \Gamma_7 \bar{S}_4 \bar{\Gamma}_4}} (p_\sigma^n S_3 \Gamma_3 \{ |p_\sigma p_\sigma^{n-1} (S_3' \Gamma_3') S_3 \Gamma_3 \}) \\
& \times \left(\tilde{S}_1 \left[S_4, S_3' \frac{1}{2} (S_3) \right] \{ \tilde{S}_2 \} S \left[\tilde{S}_1, S_4 S_3' (\bar{S}_2) \right] \{ \bar{S} \} \frac{1}{2} S \right) (\tilde{\Gamma}_1 [\Gamma_4, \Gamma_3' E(\Gamma_3)] \{ \tilde{\Gamma}_2 \} \Gamma \left[\tilde{\Gamma}_1, \Gamma_4 \Gamma_3' (\bar{\Gamma}_2) \right] \{ \bar{\Gamma} \} E \Gamma) \\
& \quad \times \langle e_g^l (S_1 \Gamma_1) t_{2g}^m (S_2 \Gamma_2) \{ \tilde{S}_1 \tilde{\Gamma}_1 \} p_\sigma^{n-1} (S_3' \Gamma_3') p_\pi^{M-(l+m+n)} (S_4 \Gamma_4) \{ \bar{S}_2 \bar{\Gamma}_2 \} \bar{S} \bar{\Gamma} | G_{M-1} | \\
& \quad \times e_g^l (S_5 \Gamma_5) t_{2g}^m (S_6 \Gamma_6) \{ \tilde{S}_3 \tilde{\Gamma}_3 \} p_\sigma^{n'-1} (S_7' \Gamma_7') p_\pi^{M-(l'+m'+n')} (S_8 \Gamma_8) \{ \bar{S}_4 \bar{\Gamma}_4 \} \bar{S} \bar{\Gamma} \rangle \\
& \times \left(\tilde{S}_3 \left[S_8, S_7' \frac{1}{2} (S_7) \right] \{ \tilde{S}_4 \} S \left[\tilde{S}_3, S_8 S_7' (\bar{S}_4) \right] \{ \bar{S} \} \frac{1}{2} S \right) (\tilde{\Gamma}_3 [\Gamma_8, \Gamma_7' E(\Gamma_7)] \{ \tilde{\Gamma}_4 \} \Gamma \left[\tilde{\Gamma}_3, \Gamma_8 \Gamma_7' (\bar{\Gamma}_4) \right] \{ \bar{\Gamma} \} E \Gamma) \\
& \quad \times (p_\sigma^n S_7 \Gamma_7 \{ |p_\sigma p_\sigma^{n-1} (S_7' \Gamma_7') S_7 \Gamma_7 \} + \frac{\sqrt{\{M-(l+m+n)\} \{M-(l'+m'+n')\}}}{M-2} \\
& \quad \times \sum_{\substack{S_4 \Gamma_4 \bar{S}_2 \bar{\Gamma}_2 \\ \bar{S} \bar{\Gamma} S_8 \Gamma_8 \bar{S}_4 \bar{\Gamma}_4}} (p_\pi^{M-(l+m+n)} S_4 \Gamma_4 \{ |p_\pi p_\pi^{M-(l+m+n)-1} (S_4' \Gamma_4') S_4 \Gamma_4 \}) \\
& \times \left(\tilde{S}_1 \left[S_3, S_4' \frac{1}{2} (S_4) \right] \{ \tilde{S}_2 \} S \left[\tilde{S}_1, S_3 S_4' (\bar{S}_2) \right] \{ \bar{S} \} \frac{1}{2} S \right) (\tilde{\Gamma}_1 [\Gamma_3, \Gamma_4' T_2(\Gamma_4)] \{ \tilde{\Gamma}_2 \} \Gamma \left[\tilde{\Gamma}_1, \Gamma_3 \Gamma_4' (\bar{\Gamma}_2) \right] \{ \bar{\Gamma} \} T_2 \Gamma) \\
& \quad \times \langle e_g^l (S_1 \Gamma_1) t_{2g}^m (S_2 \Gamma_2) \{ \tilde{S}_1 \tilde{\Gamma}_1 \} p_\sigma^n (S_3 \Gamma_3) p_\pi^{M-(l+m+n)-1} (S_4' \Gamma_4') \{ \bar{S}_2 \bar{\Gamma}_2 \} \bar{S} \bar{\Gamma} | G_{M-1} | \\
& \quad \times e_g^l (S_5 \Gamma_5) t_{2g}^m (S_6 \Gamma_6) \{ \tilde{S}_3 \tilde{\Gamma}_3 \} p_\sigma^{n'} (S_7 \Gamma_7) p_\pi^{M-(l'+m'+n')-1} (S_8' \Gamma_8') \{ \bar{S}_4 \bar{\Gamma}_4 \} \bar{S} \bar{\Gamma} \rangle \\
& \times \left(\tilde{S}_3 \left[S_7, S_8' \frac{1}{2} (S_8) \right] \{ \tilde{S}_4 \} S \left[\tilde{S}_3, S_7 S_8' (\bar{S}_4) \right] \{ \bar{S} \} \frac{1}{2} S \right) (\tilde{\Gamma}_3 [\Gamma_7, \Gamma_8' T_2(\Gamma_8)] \{ \tilde{\Gamma}_4 \} \Gamma \left[\tilde{\Gamma}_3, \Gamma_7 \Gamma_8' (\bar{\Gamma}_4) \right] \{ \bar{\Gamma} \} T_2 \Gamma) \\
& \quad \times (p_\pi^{M-(l'+m'+n')} S_8 \Gamma_8 \{ |p_\pi p_\pi^{M-(l'+m'+n')-1} (S_8' \Gamma_8') S_8 \Gamma_8 \}).
\end{aligned}$$

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