Probabilities of population of rotational levels by electron excitation of diatomic molecules

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The rotation group representations are used to express, in the adiabatic approximation, the excitation cross sections of individual rotational levels in terms of the *j* symbols and of quantities representing the scattering of electrons by a molecule with fixed positions of the nuclei. Formulas are also derived for the case when the Born approximation is applicable and the cross section depends only on the difference between the momenta of the incident and scattered electrons.

The distribution of diatomic molecules over the rotational levels after electron excitation is not only of intrinsic interest but also of practical importance in connection with the spectroscopic method for the determination of gas temperatures.[1] In this method the analysis of the experimental results is complicated considerably by the absence of simple relationships for the excitation probabilities of individual rotational levels, similar to the formulas for the intensity factors of radiative transitions.1) Although the problem of electron excitation of molecules has no simple general solution, the use of rotational symmetry makes it possible to obtain some information on the relative probabilities of excitation of the rotational levels, which is the subject of the present paper. A similar approach has been used by Baltayan and Nédélec[3] in the problem of the depolarization radiation emitted by molecules excited by electron impact; however, Baltayan and Nédélec have restricted their treatment by several assumptions relating to the electron part of the transition matrix.

We shall use the adiabatic approximation[4] because during a collision of an electron with a molecule the nuclei are not greatly displaced. We must stress that this approximation is not always valid and, in particular, we cannot use it if the scattering is of a resonant nature leading to the formation of an intermediate long-lived metastable state. We shall exclude such scattering from the scope of our analysis. The adiabatic approximation has been applied to the scattering of electrons by molecules in several papers[5-8] but they have only been concerned with transitions within one electron state and have ignored the exchange scattering and multipole order.

It is convenient to express the excitation cross sections in terms of the T matrix.[9] We shall separate clearly the dependence of this matrix on the spin operator of the incident electron. Since the T matrix is a scalar operator and, moreover, any function of the spin matrices of an electron can be reduced to the linear form, we can write

$$T=\hat{t}+s_\lambda\hat{\theta}^\lambda. \tag{1}$$

Here, $\hat{t}$ is a scalar operator independent of the spin of the incident electron; s_λ are the covariant components of the spin operator of the incident electron; $\hat{\theta}^\lambda$ is a contravariant operator, which is a vector relative to spin transformations (but not to spatial rotation[10]); the index λ has the values –1, 0, and 1. We shall use the spherical components of the vectors (see[10,11]) and, as usual, we shall assume that doubling of the upper or lower indices respresents summation. The relationship between the covariant and contravariant components is given by a "metric" tensor (see, for example,[10]).

Since the spin-orbit interaction can be ignored in the first approximation, the total spin of the molecule and the incident electron should be conserved. Hence, it follows readily that the operator $\hat{t}$ commutes with the spin of the molecule and, consequently, the intercombination transitions occur only because of the second term in Eq. (1). However, Eq. (1) does not represent the splitting of the T matrix into the direct and exchange components because $\hat{t}$ includes generally the contribution of the exchange scattering.

We shall determine the dependence of the T-matrix element on the rotational quantum numbers of a molecule in two stages. We shall first consider the matrix element only for the variables of a free electron. Let χ_μ and $\Phi_{\mathbf{k}}$ be, respectively, the spin and coordinate (normalized to the δ function of the momentum) wave functions of an electron whose wave vector is $\mathbf{k}$ and whose z projection of the spin is μ. It follows from2)

$$\langle\chi_\mu|s_\lambda|\chi_{\mu_0}\rangle=-\sqrt{\frac{3}{2}}\begin{pmatrix}\mu & 1 & 1/2\\ 1/2 & \lambda & \mu_0\end{pmatrix} \tag{2}$$

that

$$\langle\chi_\mu\Phi_{\mathbf{k}}|T|\chi_{\mu_0}\Phi_{\mathbf{k}_0}\rangle=\delta_{\mu\mu_0}t(\mathbf{k},\mathbf{k}_0)-\sqrt{6}\begin{pmatrix}\mu & 1 & 1/2\\ 1/2 & \lambda & \mu_0\end{pmatrix}\theta^\lambda(\mathbf{k},\mathbf{k}_0). \tag{3}$$

Here, $t(\mathbf{k},\mathbf{k}_0)$ and $\theta^\lambda(\mathbf{k},\mathbf{k}_0)$ are the operators which apply to the molecule alone. The numerical coefficient in front of the 3j symbol is selected so as to simplify the final expressions.

In order to use the properties of the rotational symmetry of the molecule, we shall expand the operators $t(\mathbf{k},\mathbf{k}_0)$ and $\theta^\lambda(\mathbf{k},\mathbf{k}_0)$ as series in spherical functions of the vectors $\mathbf{k}$ and $\mathbf{k}_0$ (more exactly, in terms of spherical functions of the corresponding unit vectors):

$$t(\mathbf{k},\mathbf{k}_0)=\sum_{ll_0}Y_m{}^l(\mathbf{k})Y_{m_0}^{l_0}(\mathbf{k}_0)t_{ll_0}^{mm_0}(k,k_0), \tag{4}$$

$$\theta^\lambda(\mathbf{k},\mathbf{k}_0)=\sum_{ll_0}Y_m{}^l(\mathbf{k})Y_{m_0}^{l_0}(\mathbf{k}_0)\theta_{ll_0}^{mm_0\lambda}(k,k_0). \tag{5}$$

It readily follows from the transformation properties of the spherical functions that the operators $t_{l,l_0}^{mm_0}$ and $\theta_{ll_0}^{mm_0\lambda}$ are transformed during rotation in accordance with the product of irreducible representations of the rotation group with weights l and l_0 (we are speaking only of the transformations of the spatial coordinates which do not affect the spin variables).

As usual, it is convenient to employ quantities which transform in accordance with the irreducible representations:

$$ {}^{ll_0}t_r^{\rho}(k, k_0)=(2r+1)^{1/2}\begin{pmatrix}\rho & l & l_0\\ r & m & m_0\end{pmatrix} t_{ll_0}^{mm_0}(k, k_0), \tag{6} $$

$$ {}^{ll_0}\theta_r^{\rho\lambda}(k, k_0)=(2r+1)^{1/2}\begin{pmatrix}\rho & l & l_0\\ r & m & m_0\end{pmatrix} \theta_{ll_0}^{mm_0\lambda}(k, k_0). \tag{7} $$

Here, ${}^{ll_0}t_r^{\rho}$ is an irreducible zero-spin operator of rank r with components ρ; ${}^{ll_0}\theta_r^{\rho\lambda}$ is a double-sided (in accordance with Wigner's terminology[10]) operator of rank r with respect to the spatial rotation and of rank 1 with respect to the spin. The inverse transformations can be obtained quite easily utilizing the orthogonality of the 3j symbols.[10]

The calculation of the dependence of the matrix elements of the operators ${}^{ll_0}t_r^{\rho}$ and ${}^{ll_0}\theta_r^{\rho\lambda}$ on the rotational quantum numbers of a molecule is given in the Appendix. Using Eqs. (A.4) and (A.8), we obtain the following expression for the T matrix:

$$ \begin{gathered} \langle\chi_\mu\Phi_{\mathbf{k}}\Psi_{M\Lambda\gamma}^{JSv}|T|\chi_{\mu_0}\Phi_{\mathbf{k}_0}\Psi_{M_0\Lambda_0\gamma_0}^{J_0S_0v_0}\rangle=[(2J+1)(2J_0+1)]^{1/2} \\ \times\sum_{ll_0r}Y_m^l(\mathbf{k})Y_{m_0}^{l_0}(\mathbf{k}_0)(2r+1)^{1/2}\begin{pmatrix}r & m & m_0\\ \rho & l & l_0\end{pmatrix}\left[\delta_{\mu\mu_0}\begin{pmatrix}M & \rho & J_0\\ J & r & M_0\end{pmatrix}\right. \\ \times\langle\varphi_vF_{\sigma\Lambda}{}^{S}|{}^{ll_0}\tilde{t}_r^{\Lambda_0-\Lambda}|\varphi_{v_0}F_{\sigma\Lambda_0}^{S_0}\rangle G_r-\sqrt{6}\begin{pmatrix}\mu & 1 & 1/2\\ 1/2 & \lambda & \mu_0\end{pmatrix} \\ \left.\times\langle S\Lambda v\|{}^{ll_0}\tilde{\theta}_r\|S_0\Lambda_0v_0\rangle\sum_j(2j+1)\begin{pmatrix}M & \omega & J_0\\ J & j & M_0\end{pmatrix}\begin{pmatrix}\rho & \lambda & j\\ r & 1 & \omega\end{pmatrix}I_{jr1}\right], \end{gathered} \tag{8} $$

where the components of the operators $\tilde{t}$ and $\tilde{\theta}$ apply to a moving system of coordinates (see Appendix) and the corresponding electron matrix elements describe the scattering of electrons of a given energy by a molecule with fixed positions of the nuclei. The dependence of the matrix element on the vibrational quantum numbers can be dealt with, as in the emission of light, with the help of the Franck-Condon principle.[1]

The total cross section can be expressed quite simply in terms of an element of the T matrix.[9] After the prescribed integration over the angles and summation over the projections of the moments, we obtain (m is the electron mass)

$$ \begin{gathered} \sigma_{J_0\Lambda_0\to J\Lambda}=4\pi^3m^2\hbar^2\frac{k}{k_0}(2J+1)\left[\sum_r G_r^2\sum_{ll_0}|\langle F_{\sigma\Lambda}{}^{S}\varphi_v|{}^{ll_0}\tilde{t}_r^{\Lambda_0-\Lambda}|F_{\sigma\Lambda_0}^{S}\varphi_{v_0}\rangle|^2\right. \\ \left.+\sum_{jr}(2j+1)I_{jr1}^2\sum_{ll_0}|\langle S\Lambda v\|{}^{ll_0}\tilde{\theta}_r\|S_0\Lambda_0v_0\rangle|^2\right]. \end{gathered} \tag{9} $$

In practical applications of Eq. (9) we must bear in mind that in the case of terms exhibiting the Λ doubling we must associate a Λ doublet with two states having opposite values of the projection of the electron momentum on the axis of the molecule. Strictly speaking, this approach is incorrect (see Appendix) but if the components of the Λ cannot be resolved spectroscopically and have the same populations in the initial state, the approach is permissible. Then, if $r \geq |\Lambda| + |\Lambda_0|$ and if Λ as well as Λ_0 do not vanish, the electron matrix elements $\langle S\Lambda v \| {}^{ll_0}\tilde{\theta}_r \| S_0\Lambda_0 v_0\rangle$ and $\langle S\Lambda v \| {}^{ll_0}\tilde{\theta}_r \| S_0, -\Lambda_0, v_0\rangle$ are not linked to one another by a simple symmetry relationship. This also applies to the electron matrix elements of the operators ${}^{ll_0}t_r^{\rho}$. However, if at least one of the electron states is of the Σ type, the situation simplifies because of the inversion relationship (see Appendix).

The electron matrix elements in Eq. (9) can be determined theoretically or experimentally. The theoretical methods of calculation of the electron functions are not yet sufficiently accurate but it is possible to determine the unknown quantities by an experimental investigation of the intensities of the rotational lines resulting from the excitation of molecules by electron impact. In this case, the unknown quantities are the terms on the right-hand side of Eq. (9), which are the sums (over l and l_0) of the squares of the electron matrix element corresponding to fixed values of r; the corresponding angular factors I_{jr1} and G_r are given by Eqs. (A.5)–(A.7) and (A.9). However, we must bear in mind the following point. We can easily check (by applying the rules for the summation of the j symbols[11]) that

$$ \sum_{J_0}\frac{2J_0+1}{2J+1}\sigma_{J_0\to J} $$

is independent of J. Therefore, if the groups of spectroscopic terms which make the principal contribution to the population of the upper levels are themselves populated in proportion to their statistical weights (for example, in the case of equilibrium over the rotational levels and a small—compared with the rotational temperature—difference between energies within a group), the excitation of the upper levels is independent of the rotational structure of the electron terms (in this case, the rotational temperature of the upper state is different, see[1]). However, this dependence may appear when the rotational momentum and rotational quanta become greater.

We shall conclude by considering the scattering in the case when the Born approximation is applicable and the exchange can be ignored. In this case, $\theta^{\lambda} = 0$ and t is solely a function of $\mathbf{q} = \mathbf{k}_0 - \mathbf{k}$ which is the difference between the electron momenta before and after scattering. Instead of Eq. (4) we now have

$$ t(\mathbf{q})=\sum_l Y_m^l(\mathbf{q})t_l^m(q). \tag{10} $$

The expression for the differential cross section becomes

$$ \frac{d\sigma_{J_0\to J}}{dO}=4\pi^3m^2\hbar^2\frac{k}{k_0}(2J+1)\sum_l G_l^2|\langle\varphi_vF_{\sigma\Lambda}^{S}|t_l^{\Lambda_0-\Lambda}|\varphi_{v_0}F_{\sigma\Lambda_0}\rangle|^2. $$

The total cross section can be found by integrating the transferred momentum.[12]

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APPENDIX

Before we consider the total matrix elements of a molecule, we shall give the factorization relationship for the electron (more exactly, electron-vibrational) matrix elements. It is convenient to use a moving system of coordinates linked to the nuclei. This system can be defined in various ways; we shall use the definition given in[13]. Let $A_{rc}^{\rho\lambda}$ be the components of a two-sided[10] electron tensor operator of rank r with respect to the spatial rotation and of rank c with respect to the spin ($\rho = -r, -r + 1, \ldots, r$; $\lambda = -c, -c + 1, \ldots, c$). A tilde above an operator means that its spatial components are taken in a moving system of coordinates. However, the spin components will be referred to a laboratory system of coordinates at rest.

It follows from the Wigner-Eckart theorem applied to the spin variables and from the axial symmetry of a diatomic molecule that (see[13] 3)

$$\langle F^{S}_{\sigma\Lambda}\varphi_v|\tilde{A}^{\rho\lambda}_{rc}|F^{S_0}_{\sigma_0\Lambda_0}\varphi_{v_0}\rangle=\delta_{\rho,\Lambda_0-\Lambda}\begin{pmatrix}\sigma & \lambda & S_0\\ S & c & \sigma_0\end{pmatrix}\langle S\Lambda v\|\tilde{A}_{rc}\|S_0\Lambda_0 v_0\rangle; \quad (A.1)$$

Here, $F^{S}_{\sigma\Lambda}$ and φ_v are the electron and vibrational wave functions; S is the electron spin; σ is the z projection of the spin; Λ is the projection of the orbital momentum of electrons on the axis of the molecule; v is the vibrational quantum number; the subscript "0" represents the corresponding quantity in the other state of the molecule; $\langle S\Lambda v \| \tilde{A}_{rc} \| S_0\Lambda_0 v_0\rangle$ is the "reduced" matrix element; $\delta_{\rho,\Lambda_0-\Lambda}$ is the Kronecker delta. If $c = \lambda = 0$, i.e., in the case of a zero-spin operator, Eq. (A.1) reduces to

$$\langle F^{S}_{\sigma\Lambda}\varphi_v\|\tilde{A}^{\rho}_{r}\|F^{S_0}_{\sigma_0\Lambda_0}\varphi_{v_0}\rangle=\frac{\delta_{SS_0}\delta_{\sigma\sigma_0}}{(2S+1)^{1/2}}\langle S\Lambda v\|\tilde{A}_r\|S_0\Lambda_0 v_0\rangle. \quad (A.2)$$

If $\Lambda \neq 0$, the electron functions $F^{S}_{\sigma\Lambda}$ and $F^{S}_{\sigma,-\Lambda}$ are different. It is convenient to select their phases so that on reflection in the $\xi 0 \eta$ plane of a coupled system of coordinates (see[13]) they transform into one another ("reflection" means only a spatial transformation which does not affect the spin variables). If $\Lambda = 0$, we must distinguish between the Σ^+ and Σ^- states. In the case of the electron matrix elements we have an easily verifiable equality which we shall call the inversion relationship (see[13]):

$$\langle S\Lambda v\|\tilde{A}_{rc}\|S_0\Lambda_0 v_0\rangle=\pm(-1)^{r+p}\langle S, -\Lambda, v\|\tilde{A}_{rc}\|S_0, -\Lambda_0, v_0\rangle. \quad (A.3)$$

Here, p = 0 if $\tilde{A}^{\rho\lambda}_{rc}$ is an even operator and p = 1 if this operator is odd; the minus sign of the right-hand side is used if one of the two electron states is Σ^- and the plus is taken in all other cases (both or neither of the states are of the Σ^- type). If $\Lambda = \Lambda_0 = 0$, Eq. (A.3) transforms into the corresponding selection rule.

We shall now consider the total matrix elements of a molecule.[13,3] We shall first consider the transitions between the states having fixed (including sign) values of the projection of the electron momentum on the axis of the molecule. These states, with the exception of the Σ ones, cannot be attributed a definite sign because of the inversion transformation. The changes associated with allowance for the Λ doubling and for the inversion symmetry are considered below.

A matrix element of the operator $A^{\rho\lambda}_{rc}$ can be represented in the form

$$\langle JM\Lambda Sv\gamma|A^{\rho\lambda}_{rc}|J_0M_0\Lambda_0S_0v_0\gamma_0\rangle=[(2J+1)(2J_0+1)]^{1/2}\langle S\Lambda v\|\tilde{A}_{rc}\|S_0\Lambda_0 v_0\rangle$$
$$\times\sum_j(2j+1)\begin{pmatrix}M & \omega & J_0\\ J & j & M_0\end{pmatrix}\begin{pmatrix}\rho & \lambda & j\\ r & c & \omega\end{pmatrix}I_{jrc}. \quad (A.4)$$

Here, J and M are, respectively, the angular momentum and its projection along the 0Z axis; γ is the set of quantum numbers which are not written out explicitly and which depend, in particular, on the coupling scheme; j and ω are the summation indices corresponding to the irreducible representations into which $A^{\rho\lambda}_{rc}$ splits in the case of simultaneous spin and spatial rotation; the limits of summation over j follow automatically from the condition that the 3j symbols in Eq. (A.4) should not vanish and that the angular factors occurring in I_{jrc} should not vanish either. Here, I_{jrc} is a coefficient which depends on the coupling scheme and we shall now give expressions for this coefficient.

If both states have the a-type coupling (according to the Hund nomenclature), we find that

$$I_{jrc}=(-1)^{J-\Omega+S_0-\Sigma_0}\begin{pmatrix}J & j & J_0\\ -\Omega & \Omega-\Omega_0 & \Omega_0\end{pmatrix}$$
$$\times\begin{pmatrix}r & c & j\\ \Lambda_0-\Lambda & \Sigma_0-\Sigma & \Omega-\Omega_0\end{pmatrix}\begin{pmatrix}S & c & S_0\\ \Sigma & \Sigma_0-\Sigma & -\Sigma_0\end{pmatrix}. \quad (A.5)$$

Here, Σ is the projection of the electron spin onto the axis of the molecule and $\Omega = \Lambda + \Sigma$. If both states have b-type coupling, then

$$I_{jrc}=(-1)^{K_0-\Lambda_0}[(2K+1)(2K_0+1)]^{1/2}\begin{pmatrix}K & r & K_0\\ \Lambda & \Lambda_0-\Lambda & -\Lambda_0\end{pmatrix}\begin{Bmatrix}K & r & K_0\\ S & c & S_0\\ J & j & J_0\end{Bmatrix}. \quad (A.6)$$

Here, K is the quantum number of the orbital angular momentum.[1] The 9j symbol is enclosed in braces.[11] Finally, if the first state is the a-type according to Hund and the second (identified by the subscript 0) is the b-type, we have

$$I_{jrc}=(-1)^{J+\Omega+K_0-\Lambda_0}(2K_0+1)^{1/2}\sum_{\varkappa}(2\varkappa+1)\begin{pmatrix}J & \varkappa & S\\ -\Omega & \Lambda & \Sigma\end{pmatrix}$$
$$\times\begin{pmatrix}\varkappa & r & K_0\\ \Lambda & \Lambda_0-\Lambda & -\Lambda_0\end{pmatrix}\cdot\begin{Bmatrix}J & j & J_0\\ \varkappa & r & K_0\\ S & c & S_0\end{Bmatrix}. \quad (A.7)$$

If it is necessary to consider transitions between the components of the Λ doublets, the wave functions of the molecule should be taken in the form of linear combinations of functions with opposite values of the projection of the angular momentum of electrons onto the axis of the molecule (see[13]). The corresponding matrix elements are linear combinations or quantities of the type in Eq. (A.4). They can be partly simplified by the inversion relationship (A.3) and by the rules governing the changes in the signs of the projections of the momenta.[10,11]

Since these transformations are simple, we shall not give them here because of lack of space (see[13]).

It follows from Eqs. (A.4)–(A.7) and (A.2) that we can again obtain easily the formulas for the electron zero-spin operators (see[13]). Let us assume that B^{ρ}_{r} is such an operator. Let

$$\langle JM\Lambda Sv\gamma|B_r{}^{\rho}|J_0M_0\Lambda_0S_0v_0\gamma_0\rangle=\delta_{SS_0}[(2J+1)(2J_0+1)]^{1/2}$$
$$\times\begin{pmatrix}M & \rho & J_0\\ J & r & M_0\end{pmatrix}\langle F^{S}_{\sigma\Lambda}\varphi_v|B^{\Lambda_0-\Lambda}_{r}|F^{S_0}_{\sigma\Lambda_0}\varphi_{v_0}\rangle G_r, \quad (A.8)$$

where the angular brackets $\langle \dots \rangle$ on the right-hand side denote an electron-vibrational matrix element; G_r is a factor which depends on the coupling scheme:

$$G_r=(-1)^{J_0+\Omega_0}\delta_{\Sigma\Sigma_0}\begin{pmatrix}J & r & J_0\\ \Omega & \Omega_0-\Omega & -\Omega_0\end{pmatrix},$$
$$G_r=(-1)^{J+r+S-\Lambda_0}[(2K+1)(2K_0+1)]^{1/2}\begin{pmatrix}K & r & K_0\\ \Lambda & \Lambda_0-\Lambda & -\Lambda_0\end{pmatrix}\begin{Bmatrix}r & J & J_0\\ S & K_0 & K\end{Bmatrix}$$
$$G_r=(2K_0+1)^{1/2}\begin{pmatrix}J & r & J_0\\ \Lambda+\Sigma & \Lambda_0-\Lambda & -\Lambda_0-\Sigma\end{pmatrix}\begin{pmatrix}J_0 & K_0 & S\\ -\Lambda_0-\Sigma & \Lambda_0 & \Sigma\end{pmatrix} \quad (A.9)$$

which apply to the transitions between states with the a and a, b and b, and a and b Hund couplings. The inversion relationship for the zero-spin operators is fully analogous to Eq. (A.3) [see Eq. (A.2)] and can be used to simplify the matrix elements of the transitions between the components of the Λ doublets.

We shall now consider the operators t(k, k₀) and $\theta^{\lambda}(k, k_0)$. Using the standard separation of the nuclear and electron coordinates employed in the theory of molecules,[1,14] we can easily show that in the adiabatic approximation $t(k, k_0)$ and $\theta^{\lambda}(k, k_0)$, if expressed in terms of the coordinates of the momentum vectors $\underset{\sim}{k}$

and $\widetilde{\mathbf{k}}_0$ in a moving system of coordinates, cease to depend on the angular coordinates of the nuclear axis although they include the internuclear distance as a parameter. Hence, it follows that if we allow for the transformation properties of ${}^{ll_0}t_{\mathbf{r}}^{\rho}$ and ${}^{ll_0}\theta_{\mathbf{r}}^{\rho\lambda}$, we find that Eqs. (A.8) and (A.4) are applicable to these quantities.

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1)The assumption, which is occasionally made, that the probabilities of population of rotational levels are proportional to the probabilities of radiative transitions [2] is not always justified.

2)This equality is the special case of the Wigner-Eckart theorem; here and later we shall use covariant as well as contravariant components in the 3j symbols. [10]

3)The notation used in [13] differs slightly from that adopted here (see also Footnote 2).

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