Double-Photon Contributions to Multiple-Photon Processes*

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It is shown that in evaluating the transition probability of a multiple-photon process involving more than two photons, the contributions of the part of the interaction Hamiltonian that is quadratic in the vector potential of the radiation field may be neglected if the vector potential of the radiation field is assumed to be constant in the region of space that we consider.

I. INTRODUCTION

WE consider a system of particles that is described by a Hamiltonian $\mathcal{H}_m$, having a set of eigenvalues ϵ_n and corresponding eigenfunctions Φ_n, and a radiation field that is described by a Hamiltonian $\mathcal{H}_f$. We denote the stationary states of $\mathcal{H}_f$ by comparing them with a reference state (0), which we assume to have zero energy. For example, in the state $(\lambda_1,\cdots,\lambda_n;\bar{\mu}_1,\cdots,\bar{\mu}_m)$ the photons λ_i are added to the radiation field and the photons μ_j are removed from the radiation field with respect to the state (0). A photon λ is determined by a unit vector $\mathbf{e}_\lambda$, which represents the direction of polarization, and by a vector $\mathbf{k}_\lambda$ with a magnitude k_λ equal to the photon energy and with a direction which describes the direction of propagation. The stationary states of the operator $\mathcal{H}=\mathcal{H}_m+\mathcal{H}_f$ can now be represented as (n,Λ), where the first symbol refers to the stationary states of $\mathcal{H}_m$ and the second symbol refers to the radiation field.

We take it that at a time $t=0$ our system is in the state (0,0) and we seek to determine the time proportional transition probability to an arbitrary different state (n,Λ). It can be derived[1] that this transition probability is given by

$$W_{n,\Lambda}=(2\pi/\hbar)U_{n,\Lambda}(E_{0,0})U_{n,\Lambda}^*(E_{0,0})\delta(E_{0,0}-E_{n,\Lambda}), \quad (1)$$

where the quantities U_α are determined by the set of equations

$$U_\alpha(E)=H_{\alpha;0,0}+\sum_{\beta\neq(0,0)}H_{\alpha;\beta}U_\beta(E)\zeta(E-E_\beta). \quad (2)$$

Here ζ is the Dirac ζ function

$$\zeta(x)=\lim_{\gamma\to0}(x+i\gamma)^{-1}, \quad (3)$$

and the summation is to be performed over all stationary states of $\mathcal{H}_m+\mathcal{H}_f$ except the initial state (0,0). The $H_{\alpha;\beta}$ are matrix elements of the interaction operator $\mathcal{H}_{\text{int}}$, which is given as

$$\mathcal{H}_{\text{int}}=\mathcal{H}'+\mathcal{H}'',$$
$$\mathcal{H}'=\sum_j(iQ_j\hbar/M_jc)(\mathbf{A}_j\cdot\nabla_j), \quad (4)$$
$$\mathcal{H}''=\sum_j{}_j(Q_j^2/2M_jc^2)(\mathbf{A}_j)^2.$$

Here the summations are to be performed over all particles with charges Q_j and masses M_j, and $\mathbf{A}_j$ is the vector potential of the radiation field acting on the jth particle.

Let us now make the customary assumption that our system of particles is confined to a finite region of space and that the vector potential $\mathbf{A}$ of the radiation field may be taken as constant in this region. In that case the first term, $\mathcal{H}'$, of $\mathcal{H}_{\text{int}}$ has nonzero matrix elements $H_{\alpha;\beta}'$ only if the states α and β differ by one photon and if they have different eigenstates of $\mathcal{H}_m$. The second term, $\mathcal{H}''$, of $\mathcal{H}_{\text{int}}$ has nonzeromatrix elements $H_{\alpha;\beta}''$ only if the states α and β differs by two photons and if they contain the same eigenstates $\mathcal{H}_{\alpha;\alpha}''$ are also different from zero but they do not play any role in our present discussion.

It is well known[1] that in two-photon absorption or emission processes H'' does not contribute to the transition probability. Recently[2] we found that in three-photon absorption processes H'' does not contribute either. We began to wonder whether these seemingly accidental cancellations could perhaps be special cases of a more universal property of multiple-photon processes. As a result of our subsequent investigations we prove the following interesting theorem in this paper:

In evaluating the time-proportional transition probability between two states (0,0) and (n,Λ), where Λ and 0 differ by a number of photons that is different from two, all contributions of the operator H'' may be disregarded.

This means that in deriving theoretical descriptions of multiple-photon processes it is allowed to replace $\mathcal{H}_{\text{int}}$ by $\mathcal{H}'$ at the outset of the calculation; in most cases this will simplify the problem to a great extent. We should add that our theorem is valid only if the vector potential $\mathbf{A}$ of the radiation field may be taken to be constant, but this condition is satisfied in most cases that we are concerned with.

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II. GENERAL CONSIDERATIONS AND THREE-PHOTON PROCESSES

We have seen that the transition probability $W_{n,\Lambda}$ of Eq. (1) may be derived from the solution of the set of equations (2). Since we assume that the matrix elements $H_{\alpha,\beta}$ and the quantities $U_\alpha(E_{0,0})$ are significantly smaller than the terms $\zeta(E_{0,0}-E_\alpha)$ we follow the customary iterative procedure for solving Eq. (2). If we substitute Eq. (2) into itself we obtain

$$U_\alpha(E)=H_{\alpha;0,0}+\sum_{\beta\neq 0,0} H_{\alpha;\beta}\zeta(E-E_\beta) \times[H_{\beta;0,0}+\sum_{\gamma\neq(0,0)} H_{\beta;\gamma}\zeta(E-E_\gamma)]. \quad (5)$$

We have to repeat these substitutions as many times as is necessary, that is until we have obtained a direct path from the state $\alpha=(n,\Lambda)$ to the state (0,0) by means of a succession of steps via intermediate states β, γ, δ, $\cdots$, etc.:

$$(n,\Lambda)\rightarrow\beta\rightarrow\gamma\rightarrow\delta\rightarrow\cdots\rightarrow(0,0). \quad (6)$$

Each term that occurs in the final expression for $U_\alpha(E)$ is completely determined by the intermediate states and by their order. Remember here, that each successive pair of intermediate states, for instance β and γ, can only differ by one photon if they are connected by a matrix element $H_{\beta;\gamma}'$ or by two photons if they are connected by $H_{\beta;\gamma}''$. In the latter case the states β and γ should not differ in their electronic parts. It proves to be convenient to identify the various terms of $U_{n,\Lambda}$ by pathways that are the reverse of Eq. (6). Naturally these two descriptions are entirely equivalent.

In order to introduce our notation for the subsequent general proofs let us first discuss briefly the case of a three-photon process, where we calculate the probability of a transition between the states (0,0) and $(1, \pm\mu_1, \pm\mu_2, \pm\lambda_1)$. Here U is a sum of terms that are constructed from one-photon jumps only, such as

$$(0,0)\rightarrow(m,\pm\mu_1)\rightarrow(m',\pm\mu_1,\pm\mu_2)\rightarrow (1,\pm\mu_1,\pm\mu_2,\pm\lambda_1), \quad (7)$$

and terms that contain both a two-photon and a one-photon jump. The latter terms can be combined in pairs: A typical example is

$$\begin{aligned}&(0,0)\rightarrow(0,\pm\mu_1,\pm\mu_2)\rightarrow(1,\pm\mu_1\pm\mu_2,\pm\lambda_1),\\ &(0,0)\rightarrow(1,\pm\lambda_1)\rightarrow(1,\pm\mu_1,\pm\mu_2,\pm\lambda_1).\end{aligned} \quad (8)$$

It follows from Eq. (5) that the explicit form of these terms is

$$\begin{aligned}&H_{0,0;0,\pm\mu_1,\pm\mu_2}\quad H_{0,\pm\mu_1,\pm\mu_2;1,\pm\mu_1,\pm\mu_2,\pm\lambda_1}\ \zeta(E_{0,0}-E_{0,\pm\mu_1,\pm\mu_2})\\ &H_{0,0;1,\pm\lambda_1}\quad H_{1,\pm\lambda_1;1,\pm\mu_1,\pm\mu_2,\pm\lambda_1}\ \zeta(E_{0,0}-E_{1,\pm\lambda_1}).\end{aligned} \quad (9)$$

We have now

$$\begin{aligned}H_{0,0;0,\pm\mu_1,\pm\mu_2}&=H_{1,\pm\lambda_1;1,\pm\mu_1,\pm\mu_2,\pm\lambda_1},\\ H_{0,0;1,\pm\lambda_1}&=H_{0,\pm\mu_1,\pm\mu_2;1,\pm\mu_1,\pm\mu_2,\pm\lambda_1},\end{aligned} \quad (10)$$

so that the sum of the two terms of Eq. (9) can be written as

$$H_1S_1, \quad (11)$$

where H_1 is immaterial and S_1 is

$$S_1=\zeta(E_{0,0}-E_{0,\pm\mu_1,\pm\mu_2})+\zeta(E_{0,0}-E_{1,\pm\lambda_1}). \quad (12)$$

We may assume that the arguments of the ζ functions are always different from zero; otherwise we would have direct transitions to one of the intermediate states and that is not what we want to consider. Hence

$$\begin{aligned}S_1&=a_1{}^1+a_2{}^1,\\ a_1{}^1&=-(\pm k_{\mu_1}\pm k_{\mu_2})^{-1},\\ a_2{}^1&=(\epsilon_0-\epsilon_1\mp k_{\lambda_1})^{-1}.\end{aligned} \quad (13)$$

We see that these terms depend only on the photon energies and consequently we introduce a more convenient notation. We write

$$a_1{}^1=-(k_{\mu_1}+k_{\mu_2})^{-1},\quad a_2{}^1=(\epsilon_0-\epsilon_1-k_{\lambda_1}), \quad (14)$$

with the understanding that for positive values of k the corresponding photon is added to the radiation field and for negative values of k the corresponding photon is removed from the radiation field with respect to the state (0). It is obvious that

$$S_1=-(\epsilon_0-\epsilon_1-k_{\mu_1}-k_{\mu_2}-k_{\lambda_1}) \times[(k_{\mu_1}+k_{\mu_2})(\epsilon_0-\epsilon_1-k_{\lambda_1})]^{-1}. \quad (15)$$

It also follows from Eq. (1) that $W_{1,\mu_1,\mu_2,\lambda_1}$ contains a factor $\delta(\epsilon_0-\epsilon_1-k_{\mu_1}-k_{\mu_2}-k_{\lambda_1})$, where the argument of the δ function is the same as the factor that is contained in S_1. Since always

$$x\delta(x)=0, \quad (16)$$

we conclude that the term H_1S_1 does not contribute to the transition probability. Consequently all contributions of the operator H'' to the transition probability of a three-photon process cancel.

In the following section we will generalize the above proof to multiple-photon process containing one double-photon jump. This result can be used to extend the proof to photon processes with two double-photon jumps. Finally we will show how the result can be generalized to all multiple-photon processes.

III. MULTIPLE-PHOTON PROCESSES WITH ONE DOUBLE-PHOTON JUMP

In this section we study the contributions to the transition probability of a multiple-photon process that can be constructed from one double-photon jump and a certain number of one-photon jumps. For a four-photon process these contributions can be separated into groups of three; one such group is characterized by the paths

$$(0,0) \to (0,\mu_1\mu_2) \to (1,\mu_1\mu_2\lambda_1) \to (2,\mu_1\mu_2\lambda_1\lambda_2),$$
$$(0,0) \to (1,\lambda_1) \to (1,\mu_1\mu_2\lambda_1) \to (2,\mu_1\mu_2\lambda_1\lambda_2), \quad (17)$$
$$(0,0) \to (1,\lambda_1) \to (2,\lambda_1\lambda_2) \to (2,\mu_1\mu_2\lambda_1\lambda_2).$$

The sum of these three contributions to $U_{2,\mu_1,\mu_2,\lambda_1,\lambda_2}(E_{0,0})$ can again be written as H_2S_2, where H_2 is immaterial and

$$\begin{aligned} S_2 &= a_1^2 + a_2^2 + a_3^2, \\ a_1^2 &= -(k_\mu)^{-1}(\delta_1 - k_\mu - k_1)^{-1}, \\ a_2^2 &= (\delta_1 - k_1)^{-1}(\delta_1 - k_\mu - k_1)^{-1}, \\ a_3^2 &= (\delta_1 - k_1)^{-1}(\delta_2 - k_1 - k_2)^{-1}. \end{aligned} \quad (18)$$

Here we have introduced the abbreviations

$$\begin{aligned} \delta_n &= \epsilon_0 - \epsilon_n, \\ k_\mu &= k_{\mu_1} + k_{\mu_2}, \\ k_j &= k_{\lambda j}. \end{aligned} \quad (19a)$$

If we compare Eqs. (14) and (18) we see that

$$\begin{aligned} a_1^2 &= a_1^1(\delta_1 - k_\mu - k_1)^{-1}, \\ a_2^2 &= a_2^1(\delta_1 - k_\mu - k_1)^{-1}. \end{aligned} \quad (19b)$$

Hence,

$$S_2 = (\delta_1 - k_\mu - k_1)^{-1}S_1 + (\delta_1 - k_1)^{-1}(\delta_2 - k_1 - k_2)^{-1}. \quad (20)$$

By substituting Eq. (15) we obtain

$$S_2 = -(\delta_2 - k_\mu - k_1 - k_2) \times [(k_\mu)(\delta_1 - k_1)(\delta_2 - k_1 - k_2)]^{-1}. \quad (21)$$

The first factor of Eq. (21) is again identical with the argument of the δ function that occurs in the expression for the transition probability and consequently H_2S_2 does not contribute to the transition probability.

Let us now proceed to a $(N+2)$-photon process. Here we combine all contributions to U where the final electronic state N is reached via a specific succession of intermediate electronic states, which we call 1, 2, 3, $\cdots$, $(N-1)$. The sum of these contributions can be written as H_NS_N, where H_N is immaterial and

$$S_N = \sum_{j=1}^{N+1} a_j^N. \quad (22)$$

In the case of a $(N+3)$-photon process we define in a similar fashion the sum

$$S_{N+1} = \sum_{j=1}^{N+2} a_j^{N+1}. \quad (23)$$

It is easily derived that

$$\begin{aligned} a_j^{N+1} &= (\delta_N - k_\mu - p_N)^{-1}a_j^N \\ &\qquad (j = 1, 2, 3, \cdots, N+1), \\ a_{N+2}^{N+1} &= (\delta_1 - p_1)^{-1}(\delta_2 - p_2)^{-1} \\ &\qquad \times (\delta_3 - p_3)^{-1} \cdots (\delta_{N+1} - p_{N+1})^{-1}, \end{aligned} \quad (24)$$

if we define

$$p_N = \sum_{i=1}^{N} k_i. \quad (25)$$

It follows easily now that

$$\begin{aligned} S_{N+1} &= (\delta_N - k_\mu - p_N)^{-1}S_N + (\Pi_{N+1})^{-1}, \\ \Pi_N &= (\delta_1 - p_1)(\delta_2 - p_2) \cdots (\delta_N - p_N). \end{aligned} \quad (26)$$

We derive the general form of S_N by way of induction. If we assume that

$$S_N = -(\delta_N - k_\mu - p_N)[k_\mu \Pi_N]^{-1}, \quad (27)$$

then it follows from Eq. (26) that

$$S_{N+1} = (\delta_{N+1} - k_\mu - p_{N+1})[k_\mu \Pi_{N+1}]^{-1}. \quad (28)$$

Since Eq. (27) is valid for $N=1$ and $N=2$ we may conclude from Eqs. (27) and (28) that our assumption of Eq. (27) is valid for all values of N.

Note that S_N contains the factor $(\delta_N - k_\mu - p_N)$, which is just the energy difference between the initial and final states of the transition. Since this factor is also the argument of the δ function, which occurs in Eq. (1) for the transition probability we conclude that the terms H_NS_N do not contribute to the transition probability.

IV. MULTIPLE-PHOTON PROCESSES WITH TWO DOUBLE-PHOTON JUMPS

The first nontrivial case where we encounter two double-photon jumps is a five-photon process. The three contributions to U that we have to consider here correspond to the paths

$$(0,0) \to (0,\mu_1\mu_2) \to (1,\mu_1\mu_2\lambda_1) \to (1,\mu_1\mu_2\rho_1\rho_2\lambda_1),$$
$$(0,0) \to (1,\lambda_1) \to (1,\mu_1\mu_2\lambda_1) \to (1,\mu_1\mu_2\rho_1\rho_2\lambda_1), \quad (29)$$
$$(0,0) \to (0,\mu_1\mu_2) \to (0,\mu_1\mu_2\rho_1\rho_2) \to (1,\mu_1\mu_2\rho_1\rho_2\lambda_1).$$

The sum of these three contributions can be written as $H_1'T_1$, where H_1' is immaterial and T_1 is

$$\begin{aligned} T_1 &= b_1^1 + b_2^1 + b_3^1, \\ b_1^1 &= -(k_\mu)^{-1}(\delta_1 - k_\mu - k_1)^{-1}, \\ b_2^1 &= (\delta_1 - k_1)^{-1}(\delta_1 - k_\mu - k_1)^{-1}, \\ b_3^1 &= (k_\mu)^{-1}(k_\mu + k_\rho)^{-1}. \end{aligned} \quad (30)$$

From a comparison of Eqs. (8) and (30) it follows easily that

$$b_1^1 = (\delta_1 - k_\mu - k_1)^{-1}a_1^1, \quad b_2^1 = (\delta_1 - k_\mu - k_1)^{-1}a_2^1, \quad (31)$$

and, consequently, that

$$T_1 = (\delta_1 - k_\mu - k_1)^{-1}S_1 + (k_\mu)^{-1}(k_\mu + k_\rho)^{-1}. \quad (32)$$

Substitution of Eq. (15) gives

$$T_1 = (\delta_1 - k_\mu - k_\rho - k_1)[k_\mu(k_\mu + k_\rho)(\delta_1 - k_1)]^{-1}. \quad (33)$$

Since the first term of T_1 is the energy difference between the initial and final states the contribution to the transition probability is zero.

It is useful to make a comparison with the next case of a six-photon process. Here we consider the six contributions

$$\begin{aligned}
&(0,0)\rightarrow(0,\mu_1\mu_2)\rightarrow(1,\mu_1\mu_2\lambda_1)\rightarrow(1,\mu_1\mu_2\rho_1\rho_2\lambda_1)\rightarrow(2,\mu_1\mu_2\rho_1\rho_2\lambda_1\lambda_2),\\
&(0,0)\rightarrow(1,\lambda_1)\rightarrow(1,\mu_1\mu_2\lambda_1)\rightarrow(1,\mu_1\mu_2\rho_1\rho_2\lambda_1)\rightarrow(2,\mu_1\mu_2\rho_1\rho_2\lambda_1\lambda_2),\\
&(0,0)\rightarrow(0,\mu_1\mu_2)\rightarrow(0,\mu_1\mu_2\rho_1\rho_2)\rightarrow(1,\mu_1\mu_2\rho_1\rho_2\lambda_1)\rightarrow(2,\mu_1\mu_2\rho_1\rho_2\lambda_1\lambda_2),\\
&(0,0)\rightarrow(0,\mu_1\mu_2)\rightarrow(1,\mu_1\mu_2\lambda_1)\rightarrow(2,\mu_1\mu_2\lambda_1\lambda_2)\rightarrow(2,\mu_1\mu_2\rho_1\rho_2\lambda_1\lambda_2),\\
&(0,0)\rightarrow(1,\lambda_1)\rightarrow(1,\mu_1\mu_2\lambda_1)\rightarrow(2,\mu_1\mu_2\lambda_1\lambda_2)\rightarrow(2,\mu_1\mu_2\rho_1\rho_2\lambda_1\lambda_2),\\
&(0,0)\rightarrow(1,\lambda_1)\rightarrow(2,\lambda_1\lambda_2)\rightarrow(2,\mu_1\mu_2\lambda_1\lambda_2)\rightarrow(2,\mu_1\mu_2\rho_1\rho_2\lambda_1\lambda_2).
\end{aligned}\tag{34}$$

If we compare the first three terms with Eq. (29) we find that their sum can be written as

$$b_1^2+b_2^2+b_3^2=(\delta_1-k_\mu-k_\rho-k_1)^{-1}T_1. \tag{35}$$

The last three terms of Eq. (29) should be compared with Eq. (17) and their sum is

$$b_4^2+b_5^2+b_6^2=(\delta_2-k_\mu-k_1-k_2)^{-1}S_2. \tag{36}$$

The sum T_2 of all terms b_j^2 is therefore

$$T_2=(\delta_1-k_\mu-k_\rho-p_1)^{-1}T_1+(\delta_2-k_\mu-p_2)^{-1}S_2. \tag{37}$$

It is easily shown that in the general case we can follow the same procedure, and that the result is then

$$T_{N+1}=(\delta_N-k_\mu-k_\rho-p_N)^{-1}T_N+(\delta_{N+1}-k_\mu-p_{N+1})^{-1}S_{N+1}. \tag{38}$$

It is again possible to derive the general expression for T_N by means of induction. If we assume that

$$T_N=(\delta_N-k_\mu-k_\rho-p_N)[k_\mu(k_\mu+k_\rho)\Pi_N]^{-1}, \tag{39}$$

then it may be derived from Eqs. (27) and (39) that

$$T_{N+1}=(\delta_{N+1}-k_\mu-k_\rho-p_{N+1})[k_\mu(k_\mu+k_\rho)\Pi_{N+1}]^{-1}. \tag{40}$$

Since Eq. (39) is valid also for $N=1$ we may conclude that our assumption of Eq. (39) is correct for all values of N.

The first term of Eq. (39) represents again the energy difference between the initial and final states of the transition; it follows therefore that in multiple-photon processes the terms that correspond to two double-photon jumps do not contribute to the transition probability.

V. FURTHER GENERALIZATIONS

We are now in a position to generalize the previous results to a $(2M+N)$-photon process, where we consider M double-photon jumps and N one-photon jumps. We take it that the double-photon jumps involve the photons (μ_1,μ_1'), (μ_2,μ_2'), $\cdots$, etc., and we introduce the abbreviations

$$k_i'=k_{\mu i}+k_{\mu i}', \qquad q_M=\sum_{j=1}^{M}k_j'. \tag{41}$$

In Sec. III we considered the case where $M=1$ and it follows from Eq. (27) that the corresponding sum, which we denote now by $S_{1,N}$, may be written as

$$S_{1,N}=(-1)^1(\delta_N-k_1'-p_N)[k_1'\Pi_N]^{-1}. \tag{42}$$

The case $M=2$ was discussed in Sec. IV and the corresponding sum, which we denote now by $S_{2,N}$ instead of T_2, is according to Eq. (39)

$$S_{2,N}=(-1)^2(\delta_N-q_2-p_N)[q_1q_2\Pi_N]^{-1}. \tag{43}$$

In deriving this result we made use of Eq. (32), which we write in our new notation as

$$S_{2,1}=(\delta_1-q_1-p_1)^{-1}S_{1,1}+(-1)^2(\chi_2)^{-1}, \tag{44}$$

with

$$\chi_M=q_1q_2\cdots q_M. \tag{45}$$

It can be derived from Eq. (29) that

$$S_{3,1}=(\delta_1-q_2-p_1)^{-1}S_{2,1}+(-1)^3(\chi_3)^{-1}, \tag{46}$$

and, in general, we have

$$S_{M+1,1}=(\delta_1-q_M-p_1)^{-1}S_{M,1}-(-1)^M(\chi_{M+1})^{-1}. \tag{47}$$

By induction it can be shown that $S_{M,1}$ is given by

$$S_{M,1}=(-1)^M(\delta_1-q_M-p_1)[\chi_M(\delta_1-p_1)]^{-1}. \tag{48}$$

Let us now rewrite Eq. (38) in our new notation:

$$S_{2,N+1}=(\delta_N-q_2-p_N)^{-1}S_{2,N}+(\delta_{N+1}-q_1-p_{N+1})S_{1,N+1}. \tag{49}$$

It may be verified that this relationship can be generalized to

$$S_{M+1,N+1}=(\delta_N-q_{M+1}-p_N)^{-1}S_{M+1,N}+(\delta_{N+1}-q_M-p_{N+1})S_{M,N+1}. \tag{50}$$

By means of induction we can now derive the general expression

$$S_{M,N}=(-1)^M(\delta_N-q_M-p_N)[\chi_M\Pi_N]^{-1}. \tag{51}$$

We see that $S_{M,N}$ always contains a factor which is equal to the energy difference between the initial and final states and therefore, that it does not contribute to the transition probability. It follows that the term $\mathcal{H}''$ of Eq. (4) may always be disregarded in evaluating transition probabilities of the type $(0,0)\rightarrow(n,\Lambda)$ if $n\neq0$, and if the vector potential of the radiation field may be assumed to be constant in the region that we consider.

VI. THE CASE $n=0$

In the previous derivations it was more or less understood that we intended the material states 0 and n of the transition $(0,0)\rightarrow(n,\Lambda)$ to be different. Even though we did not use this assumption explicity, we considered only pathways between 0 and n that contained at least one intermediate electronic state. If $n=0$ this procedure does not cover all possibilities; in this case we can construct a path

$$(0,0)\rightarrow(0,\mu_1\mu_1')\rightarrow (0,\mu_1\mu_1'\mu_2\mu_2')\rightarrow\cdots(0,\mu_1\mu_1'\mu_2\mu_2'\cdots,\mu_N\mu_N') \quad (52)$$

if we consider a $(2N)$-photon process. We may conclude therefore that our previous considerations are valid also for $n=0$ if the path between the initial and final states contains at least one intermediate material state that is different from the state 0, but that in the case $n=0$ we should in addition investigate also pathways of the type of Eq. (52).

Let us therefore set out to study the contributions to $U_{0,\mu_1\mu_1'\mu_2\mu_2'\cdots\mu_N,\mu_N'}$ that result from the terms of the type of Eq. (52). If N is equal to unity the only term of this type corresponds to

$$(0,0)\rightarrow(0,\mu_1\mu_1'), \quad (53)$$

and its contribution is not necessarily zero. Remarkably enough this is the only case where the term $\mathcal{H}''$ can contribute to the transition probability.

If $N=2$ we have to take the sum of the two contributions

$$\begin{aligned}&(0,0)\rightarrow(0,\mu_1\mu_1')\rightarrow(0,\mu_1\mu_1'\mu_2\mu_2'),\\&(0,0)\rightarrow(0,\mu_2\mu_2')\rightarrow(0,\mu_1\mu_1'\mu_2\mu_2').\end{aligned} \quad (54)$$

This sum can again be written as H_2K_2, where H_2 is immaterial and

$$K_2(1,2)=-[(k_1')^{-1}+(k_2')^{-1}] =-(k_1'+k_2')(k_1'k_2')^{-1}. \quad (55)$$

Here k_i' is defined in Eq. (41).

In the case $N=3$ we should take the sum of the six contributions

$$\begin{aligned}&(0,0)\rightarrow(0,\mu_1\mu_1')\rightarrow(0,\mu_1\mu_1'\mu_2\mu_2')\rightarrow(0,\mu_1\mu_1'\mu_2\mu_2'\mu_3\mu_3'),\\&(0,0)\rightarrow(0,\mu_2\mu_2')\rightarrow(0,\mu_1\mu_1'\mu_2\mu_2')\rightarrow(0,\mu_1\mu_1'\mu_2\mu_2'\mu_3\mu_3'),\\&(0,0)\rightarrow(0,\mu_3\mu_3')\rightarrow(0,\mu_2\mu_2'\mu_3\mu_3')\rightarrow(0,\mu_1\mu_1'\mu_2\mu_2'\mu_3\mu_3'),\\&(0,0)\rightarrow(0,\mu_2\mu_2')\rightarrow(0,\mu_2\mu_2'\mu_3\mu_3')\rightarrow(0,\mu_1\mu_1'\mu_2\mu_2'\mu_3\mu_3'),\\&(0,0)\rightarrow(0,\mu_1\mu_1')\rightarrow(0,\mu_1\mu_1'\mu_3\mu_3')\rightarrow(0,\mu_1\mu_1'\mu_2\mu_2'\mu_3\mu_3'),\\&(0,0)\rightarrow(0,\mu_3\mu_3')\rightarrow(0,\mu_1\mu_1'\mu_3\mu_3')\rightarrow(0,\mu_1\mu_1'\mu_2\mu_2'\mu_3\mu_3').\end{aligned} \quad (56)$$

It is easily seen that the sum $K_3(1,2,3)$ that corresponds to this situation can be written as

$$K_3(1,2,3)=-[(k_1'+k_2')^{-1}K_2(1,2) +(k_3'+k_2')^{-1}K_2(3,2)+(k_1'+k_3')^{-1}K_2(1,3)]. \quad (57)$$

The result is

$$K_3(1,2,3)=(k_1'+k_2'+k_3')(k_1'k_2'k_3')^{-1}. \quad (58)$$

We may write Eq. (56) also as

$$K_3(1,2,3)=-[(k_1'+k_2')^{-1}K_2(1,2)] +[P(1,3)+P(2,3)][(k_1'+k_2')^{-1}K_2(1,2)], \quad (59)$$

where $P(i,j)$ is an operator which means that we have to replace k_i' by k_j' in the expression upon which it works. It can be verified that this result may be generalized to

$$k_{N+1}(1,2,\cdots,N+1)=-[1+\sum_{j=1}^{N}P(j,N)] \times[(k_1'+k_2'+\cdots+k_N')^{-1}K_N(1,2,\cdots,N)]. \quad (60)$$

From this recurrence relation and from Eq. (58) it can now be derived by means of induction that

$$K_N(1,2,\cdots,N)=(-1)^{N-1}(k_1'+k_2'+\cdots+k_N') \times(k_1'k_2'\cdots k_N')^{-1}. \quad (61)$$

Since the factor $(k_1'+k_2'+\cdots+k_N')$ is the energy difference between the initial and final states of the transition we may conclude that K_N does not contribute to the transition probability.

VII. CONCLUSION

The above derivations show that for any multiple-photon process that involves more than two photons the operator $\mathcal{H}''$ of Eq. (4) does not contribute in any way to the transition probability if the vector potential of the radiation field is assumed to be constant in the region of space that we are concerned with.