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Effective Hamiltonians, two-level systems, and generalized Maxwell-Bloch equations

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A new method is proposed which involves a canonical transformation leading to the nonsecular part of time-independent perturbation calculus. The method is used to derive expressions for effective Shen-Walls Hamiltonians which, taken in the two-level approximation and on the inclusion of non-Hamiltonian terms into the dynamics of the system, lead to generalized Maxwell-Bloch equations. The rotating-wave approximation is written anew within the framework of our formalism.

I. INTRODUCTION

There are several ways in which an effective Hamiltonian can be defined. Accordingly, at the outset we must clearly state what meaning we shall be ascribing to the word. Here are some examples of its various connotations.

Consider a Hamiltonian in the form

$$H=H_0+H_{\text{int}}\ ,$$

where (1)

$$H_0=\sum_{i=1}^{n}\omega_i a_i^{\dagger}a_i\ ,$$

$$H_{\text{int}}=\lambda^* a_1^{\dagger k_1}\cdots a_m^{\dagger k_m}a_{m+1}^{k_{m+1}}\cdots a_n^{k_n}+\text{H.c}\ .$$

It is applied in quantum optics in the description of the n modes of a field interacting under conditions of generalized resonance:

$$k_1\omega_1+\cdots+k_m\omega_m=k_{m+1}\omega_{m+1}+\cdots+k_n\omega_n\ . \tag{2}$$

Note that this Hamiltonian is characterized by the fact that the interaction H_{int} leads to transition *within* the energy shells of the free Hamiltonian H_0. This is ensured by the condition (2).

Another case is that of generalization of the well-known Dicke Hamiltonian in the rotating-wave approximation to k-photon resonance:

$$H=H_0+H_{\text{int}}\ ,$$

where (3)

$$H_0=\epsilon S_3+\omega a^{\dagger}a,\quad H_{\text{int}}=\lambda^* a^{\dagger k}S^{-}+\lambda a^{k}S^{+},\quad \epsilon=k\omega\ .$$

Here also, interaction leads to transitions within the energy shells of the free (noninteractive) part only.

Hamiltonians of the forms (1) and (3) are attributed to Shen and Walls[1] and are referred to as effective Hamiltonians. Notwithstanding that the authors who took recourse to these Hamiltonians are truly innumerable, owing to the great advantages they present in practice, the procedure of their derivation has hardly been given the attention which it merits.

Formally similar "resonance" Hamiltonians have been applied by Senitzky,[2] albeit in the context of the interaction of a system of harmonic oscillators and the radiation field. The relationship between his resonance and our "effective" Hamiltonians will be the subject of a separate paper.

Consider a Hamiltonian, given as the sum of a free (noninteractive) part $\mathscr{H}_0$ and the interactive part $\mathscr{H}_{\text{int}}$:

$$\mathscr{H}=\mathscr{H}_0+\mathscr{H}_{\text{int}}\ . \tag{4}$$

In this paper we shall be using the words "effective Hamiltonian" to denote the Hamiltonian arising from $\mathscr{H}$ on application of the canonical transformation, diagonalizing $\mathscr{H}$ with respect to the energy shells of the free (noninteractive) part $\mathscr{H}_0$. We show that Hamiltonians of the types (1) and (3) are encompassed by the above definition and can be derived from standard forms of Hamiltonians describing mutually interacting matter and radiation by applying a time-independent canonical transformation. In the course of our proof we shall derive a form of perturbation calculus particularly well adapted to problems in which the spectrum of the initial operator presents a high degree of degeneracy.

The structure of our paper is as follows. In Sec. II we derive the necessary canonical transformation, equivalent to diagonalization of the initial Hamiltonian with respect to the energy shells of the free part, i.e., the canonical transformation equivalent to the nonsecular part of perturbation calculus. In Sec. III we make more concrete the physical assumptions regarding the systems under consideration. The next step in the derivation, in Sec. IV, consists in finding effective Hamiltonians for the material system interacting with the radiation field. Finally, in Sec. V, these Hamiltonians are given in the two-level approximation of the material system; in particular, for cases of multiphoton resonances. They are then applied to a discussion of the generalized Dicke super-radiant state. Section VI gives generalized Maxwell-Bloch equations as well as a discussion of the realization of the rotating-wave approximation within the framework of the formalism applied by us.

The method we propose for deriving the effective Hamiltonians has some rather loose connections with the work of other authors. The idea itself is similar to that of Heitler,[3] who uses the interaction picture, in his form of perturbation calculus. There are, as well, certain connections with the ideas put forward by Michels and Suttorp[4] in a number of papers concerning degenerate perturbation calculus, where stress is laid on the use of field-theory methods; since they deal with time-dependent perturbation calculus, they too apply the interaction picture.

By using effective Hamiltonians in the two-level approximation it is relatively easy to derive Maxwell-Bloch equations, generalized for nonlinear processes. In the case of the classical field, one can go over to Bloch equations for multiphoton processes along the path opened by the work of Grischkowsky, Loy, and Liao,[5] and expanded by Nayfeh and Nayfeh[6] and by Friedmann and Wilson-Gordon.[7]

We should also mention the approach of Rosenberg,[8] which is an extension of scattering theory and which takes recourse to the resolvent operator.

II. CANONICAL-TRANSFORMATION FORMALISM

We consider a Hermitian operator of the form

$$\mathscr{H}(\epsilon)=\mathscr{H}_0+\epsilon\mathscr{H}_1+\epsilon^2\mathscr{H}_2+\cdots=\sum_{m=0}^{\infty}\epsilon^m\mathscr{H}_m\ , \tag{5}$$

where ϵ is the perturbation parameter ($\epsilon\ll 1$). Assume $\mathscr{H}_0$ to have a discrete (and, in general, degenerate) spectrum and the solution of its eigenvalue problem

$$(\mathscr{H}_0-E_{0i})\,|\,i,\alpha\rangle=0 \tag{6}$$

to be known. The coefficient α labels states belonging to the subspace L_i, corresponding to the eigenvalue E_{0i} (if $\mathscr{H}_0$ denotes a Hamiltonian, L_i is an energy shell). The states $|\,i,\alpha\rangle$ form an orthonormal basis in the Hilbert space in which the operators $\mathscr{H}_m$ act:

$$\langle i,\alpha\,|\,j,\beta\rangle=\delta_{ij}\delta_{\alpha\beta},\ \sum_{i,\alpha}|\,i,\alpha\rangle\langle i,\alpha\,|=I\ . \tag{7}$$

In other words, we have assumed the following spectral distribution of the operator $\mathscr{H}_0$:

$$\mathscr{H}_0=\sum_i E_{0i}P_i\ , \tag{8}$$

where the projector operators P_i are expressed by the formulas

$$P_i=\sum_{\alpha}|\,i,\alpha\rangle\langle i,\alpha\,|\ . \tag{9}$$

Henceforth (unless specified otherwise) the term (non)diagonality will be used in the meaning of (non)diagonality with respect to the subspaces L_i. We shall be applying the following superscripts to denote the operation of taking the diagonal and nondiagonal parts:

$$O^{\{d\}}\equiv\sum_i P_iOP_i,\ \ O^{\{n\}}\equiv\sum_{i,j}{}'P_iOP_j\ , \tag{10}$$

respectively, where O is an arbitrary operator. The prime on the summation symbol in the preceding formula stands for summation over $i\neq j$. Also, we shall often be applying an energy-weighted operation (of taking the nondiagonal part) defined as follows:

$$O^{\{n,E\}}\equiv\sum_{i,j}{}'(E_{0i}-E_{0j})^{-1}P_iOP_j \tag{11}$$

(which is the same as the "dash operation" denoted by a long overbar in previous work). Moreover, some readily proved relations will be helpful later on. For arbitrary operators X and Y the following formulas hold:

$$
\begin{aligned}
&(XY)^{\{d\}}=(X^{\{n\}}Y^{\{n\}})^{\{d\}}+X^{\{d\}}Y^{\{d\}}\ ,\\
&(XY)^{\{n\}}=(X^{\{n\}}Y^{\{n\}})^{\{n\}}+X^{\{n\}}Y^{\{d\}}+X^{\{d\}}Y^{\{n\}}\ ,\\
&(X^{\{n,E\}})^{\{d\}}=(X^{\{d\}})^{\{n,E\}}=0\ ,\\
&(X^{\dagger})^{\{n,E\}}=-(X^{\{n,E\}})^{\dagger}\ ,\\
&(X^{\{n,E\}}Y^{\{n\}})^{\{n,E\}}+(X^{\{n\}}Y^{\{n,E\}})^{\{n,E\}}\\
&\qquad=(X^{\{n,E\}}Y^{\{n,E\}})^{\{n\}}\ ,\\
&(XY^{\{d\}})^{\{n,E\}}=X^{\{n,E\}}Y^{\{d\}}\ ,\\
&(X^{\{d\}}Y)^{\{n,E\}}=X^{\{d\}}Y^{\{n,E\}}\ ,\\
&(X^{\{n,E\}}Y)^{\{d\}}+(XY^{\{n,E\}})^{\{d\}}=0\ ,\\
&[\mathscr{H}_0,X]^{\{d\}}=0,\ \ [\mathscr{H}_0,X]^{\{n,E\}}=X^{\{n\}}\ .
\end{aligned}
\tag{12}
$$

The canonical transformation, diagonalizing the operator $\mathscr{H}(\epsilon)$, is quite easy to perform, i.e., a unitary operator $\mathscr{U}(\epsilon)$ can be defined such that the operator

$$\mathscr{K}(\epsilon)=\mathscr{U}(\epsilon)\mathscr{H}(\epsilon)\mathscr{U}^{\dagger}(\epsilon) \tag{13}$$

shall be diagonal (only with respect to linear subspaces L_i):

$$\mathscr{K}(\epsilon)=\mathscr{K}^{\{d\}}(\epsilon)\ . \tag{14}$$

The meaning of this procedure is obvious. We have here a perturbation-calculus procedure conceived in the spirit of the canonical transformation method proposed by Heitler[3] (cf. also Ref. 9). To achieve this aim, it suffices to choose $\mathscr{U}(\epsilon)$ in the following form:

$$\mathscr{U}(\epsilon)=\lim_{n\to\infty}\mathscr{U}^{(n)}(\epsilon)\ , \tag{15}$$

where

$$\mathscr{U}^{(n)}(\epsilon)=e^{\epsilon^nX_n}e^{\epsilon^{n-1}X_{n-1}}\cdots e^{\epsilon^2X_2}e^{\epsilon X_1}\ . \tag{16}$$

The operators $X_1,X_2,\ldots,X_m,\ldots$ are anti-Hermitian:

$$X_m^{\dagger}=-X_m\ . \tag{17}$$

Since we are interested in diagonalizing $\mathscr{H}(\epsilon)$ with respect to the linear subspaces L_i only, these operators can be assumed to be nondiagonal without loss of generality:

$$X_m^{\{d\}}=0\ . \tag{18}$$

Obviously, the condition of unitarity of the operator $\mathscr{U}(\epsilon)$,

$$\mathscr{U}(\epsilon)\mathscr{U}^{\dagger}(\epsilon)=\mathscr{U}^{\dagger}(\epsilon)\mathscr{U}(\epsilon)=I\ , \tag{19}$$

is satisfied automatically. Moreover, $\mathscr{K}(\epsilon)$, and likewise $\mathscr{H}(\epsilon)$, can be expressed in the form of a series:

$$\mathscr{K}(\epsilon)=\sum_{n=0}^{\infty}\epsilon^m\mathscr{K}_m\ . \tag{20}$$

We now proceed to determine the explicit form of the operators $\mathcal{K}_m$. To this aim, it is useful to transform (13) to the equivalent form:

$$\mathcal{K}(\epsilon)\mathcal{U}(\epsilon)=\mathcal{U}(\epsilon)\mathcal{H}(\epsilon) . \tag{21}$$

On equating the term at identical powers of the perturbation parameter ϵ we obtain the formula

$$\sum_{l=0}^{m}(\mathcal{K}_{m-l}\mathcal{U}_l-\mathcal{U}_l\mathcal{H}_{m-l})=0, \quad m=0,1,2,\ldots \tag{22}$$

where the operators $\mathcal{U}_l$ are defined as the expansion coefficients of $\mathcal{U}(\epsilon)$ in powers of ϵ,

$$\mathcal{U}(\epsilon)=\sum_{m=0}^{\infty}\epsilon^m\mathcal{U}_m , \tag{23}$$

and are given by the expressions

$$\mathcal{U}_0=I ,$$

$$\mathcal{U}_l=\sum_{\substack{m_1,m_2,\ldots,m_l\geq 0\\ \sum_{k=1}^{l}km_k=l}}\frac{X_l^{m_l}}{m_l!}\frac{X_{l-1}^{m_{l-1}}}{m_{l-1}!}\cdots\frac{X_2^{m_2}}{m_2!}\frac{X_1^{m_1}}{m_1!}, \quad l\geq 1 . \tag{24}$$

The summation is taken over all sets of l non-negative integers $m_1,m_2,m_3,\ldots,m_l$, fulfilling the condition

$$\sum_{k=1}^{l}km_k=m_1+2m_2+3m_3+\cdots+lm_l=l . \tag{25}$$

The successive operators $\mathcal{U}_l$ can be obtained by applying a simple recurrence relation. Let $\mathcal{D}$ denote the operation of ordering the products of operators X_m by writing them from left to right in the order of decreasing indices m:

$$\mathcal{U}_l=X_l+\sum_{m=1}^{l-1}(m/l)\mathcal{D}(X_m\mathcal{U}_{l-m}) . \tag{26}$$

A proof of this equality and of the formulas for $\mathcal{U}_l$ at $l\leq 5$ is given in the Appendix.

Let us now return to Eq. (22). For $m=0$, it simply gives $\mathcal{K}_0=\mathcal{H}_0$. Hence, for $m\geq 1$ we have

$$\mathcal{K}_m+[\mathcal{H}_0,\mathcal{U}_m]=\sum_{l=1}^{m-1}(\mathcal{U}_l\mathcal{H}_{m-l}-\mathcal{K}_{m-l}\mathcal{U}_l)+\mathcal{H}_m . \tag{27}$$

The procedure applied by us is visualized graphically in Fig. 1. Let us assume $\mathcal{K}_1,\mathcal{K}_2,\ldots,\mathcal{K}_{m-1}$ and $\mathcal{U}_1,\mathcal{U}_2,\ldots,\mathcal{U}_{m-1}$ as known. By assumption, $\mathcal{K}_m$ is diagonal. On taking the diagonal parts of both terms of Eq. (27) and applying the relations (12), we determine the operator $\mathcal{K}_m$ from the following formula:

$$\mathcal{K}_m=\sum_{l=1}^{m-1}[(\mathcal{U}_l^{\{n\}}\mathcal{H}_{m-l})^{\{d\}}+\mathcal{U}_l^{\{d\}}\mathcal{H}_{m-l}^{\{d\}}-\mathcal{K}_{m-l}^{\{d\}}\mathcal{U}_l^{\{d\}}]+\mathcal{H}_m^{\{d\}} . \tag{28}$$

Next, taking the energy-weighted nondiagonal part of the right- and left-hand sides of Eq. (27), we calculate the nondiagonal part of $\mathcal{U}_m$:

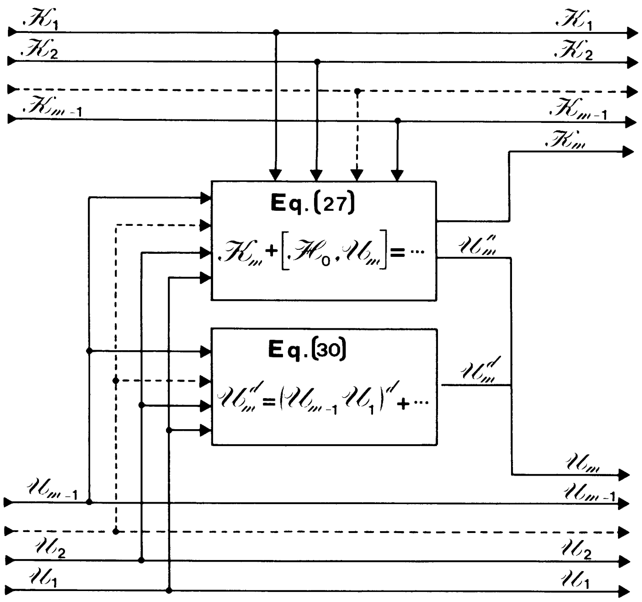


FIG. 1. Iterational scheme for the calculation of operators $\mathcal{K}_m$ and U_m.

$$\mathcal{U}_m^{\{n\}}=\sum_{l=1}^{m-1}[(\mathcal{U}_l^{\{n\}}\mathcal{H}_{m-l})^{\{n,E\}}+\mathcal{U}_l^{\{d\}}\mathcal{H}_{m-l}^{\{n,E\}}-\mathcal{K}_{m-l}\mathcal{U}_l^{\{n,E\}}]+\mathcal{H}_m^{\{n,E\}} . \tag{29}$$

We still have to determine the diagonal part of the operator $\mathcal{U}_m$. To this aim, it suffices to invert the relations (26), i.e., to write the X_m's as functions of $\mathcal{U}_1,\mathcal{U}_2,\mathcal{U}_3,\ldots,\mathcal{U}_m$ and to apply the operation of taking the diagonal part, keeping in mind the assumption $X_m^{\{d\}}=0$ [Eq. (18)]. We thus obtain expressions for the diagonal part of the operators $\mathcal{U}_m$ in the following form:

$$\mathcal{U}_1^{\{d\}}=0 ,$$

$$\mathcal{U}_2^{\{d\}}=\tfrac{1}{2}(\mathcal{U}_1^2)^{\{d\}} ,$$

$$\mathcal{U}_3^{\{d\}}=(\mathcal{U}_2\mathcal{U}_1)^{\{d\}}-\tfrac{1}{3}(\mathcal{U}_1^3)^{\{d\}} ,$$

$$\mathcal{U}_4^{\{d\}}=(\mathcal{U}_3\mathcal{U}_1)^{\{d\}}+\tfrac{1}{2}(\mathcal{U}_2^2)^{\{d\}}-\tfrac{3}{4}(\mathcal{U}_2\mathcal{U}_1^2)^{\{d\}}-\tfrac{1}{4}(\mathcal{U}_1^2\mathcal{U}_2)^{\{d\}}+\tfrac{1}{4}(\mathcal{U}_1^4)^{\{d\}} , \tag{30}$$

and so forth. For higher and higher orders of the expansions of $\mathcal{K}(\epsilon)$ and $\mathcal{U}(\epsilon)$, we have

$$\mathcal{K}_1=\mathcal{H}_1^{\{d\}} ,$$

$$\mathcal{U}_1=\mathcal{U}_1^{\{n\}}=\mathcal{H}_1^{\{n,E\}} , \tag{31a}$$

$$\begin{aligned}
&\mathscr{K}_2=(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1)^{\{d\}}+\mathscr{H}_2^{\{d\}}\ ,\\
&\mathscr{U}_2^{\{n\}}=(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1)^{\{n,E\}}-\mathscr{H}_1^{\{d\}}(\mathscr{H}_1^{\{n,E\}})^{\{n,E\}}+\mathscr{H}_2^{\{n,E\}}\ ,\\
&\mathscr{U}_2^{\{d\}}=\tfrac{1}{2}(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1^{\{n,E\}})^{\{d\}}\ ,
\end{aligned}\tag{31b}$$

$$\begin{aligned}
\mathscr{K}_3=&[(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1)^{\{n,E\}}\mathscr{H}_1]^{\{d\}}+\tfrac{1}{2}(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1^{\{n,E\}})^{\{d\}}\mathscr{H}_1^{\{d\}}+\tfrac{1}{2}\mathscr{H}_1^{\{d\}}(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1^{\{n,E\}})^{\{d\}}\\
&+(\mathscr{H}_2^{\{n,E\}}\mathscr{H}_1)^{\{d\}}+(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_2)^{\{d\}}+\mathscr{H}_3^{\{d\}}\ ,
\end{aligned}\tag{31c}$$

$$\begin{aligned}
\mathscr{K}_4=&\{[(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1)^{\{n,E\}}\mathscr{H}_1]^{\{n,E\}}\mathscr{H}_1\}^{\{d\}}\\
&+[(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1)^{\{n,E\}}\mathscr{H}_1^{\{n,E\}}]^{\{d\}}\mathscr{H}_1^{\{d\}}-\mathscr{H}_1^{\{d\}}[\mathscr{H}_1^{\{n,E\}}(\mathscr{H}_1\mathscr{H}_1^{\{n,E\}})^{\{n,E\}}]^{\{d\}}\\
&-\tfrac{1}{3}(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1^{\{n,E\}})^{\{d\}}\mathscr{H}_1^{\{d\}}+\tfrac{1}{3}\mathscr{H}_1^{\{d\}}(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1^{\{n,E\}})^{\{d\}}\\
&+\tfrac{1}{2}(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1)^{\{d\}}(\mathscr{H}^{\{n,E\}}\mathscr{H}^{\{n,E\}})^{\{d\}}-\tfrac{1}{2}(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1^{\{n,E\}})^{\{d\}}(\mathscr{H}_1\mathscr{H}_1^{\{n,E\}})^{\{d\}}\\
&+\mathscr{H}_1^{\{d\}}[\mathscr{H}_1^{\{n,E\}}(\mathscr{H}_1^{\{n,E\}})^{\{n,E\}}]^{\{d\}}\mathscr{H}_1^{\{d\}}+[(\mathscr{H}_2^{\{n,E\}}\mathscr{H}_1)^{\{n,E\}}\mathscr{H}_1]^{\{d\}}+[(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_2)^{\{n,E\}}\mathscr{H}_1]^{\{d\}}\\
&+[(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1)^{\{n,E\}}\mathscr{H}_2]^{\{d\}}+\tfrac{1}{2}\mathscr{H}_2^{\{d\}}(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1^{\{n,E\}})^{\{d\}}+\tfrac{1}{2}(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_1^{\{n,E\}})^{\{d\}}\mathscr{H}_2^{\{d\}}\\
&+(\mathscr{H}_2^{\{n,E\}}\mathscr{H}_1^{\{n,E\}})^{\{d\}}\mathscr{H}_1^{\{d\}}+\mathscr{H}_1^{\{d\}}(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_2^{\{n,E\}})^{\{d\}}+(\mathscr{H}_2^{\{n,E\}}\mathscr{H}_2)^{\{d\}}+(\mathscr{H}_3^{\{n,E\}}\mathscr{H}_1)^{\{d\}}\\
&+(\mathscr{H}_1^{\{n,E\}}\mathscr{H}_3^{\{n,E\}})^{\{d\}}+\mathscr{H}_4^{\{d\}}\ ,
\end{aligned}\tag{31d}$$

where Eqs. (31a), (31b), (31c), and (31d) refer to orders 1, 2, 3, and 4, respectively. Obviously, our chief aim is to obtain expressions for the $\mathscr{K}_m$'s. The $\mathscr{U}_m$'s are but supplementary. However, for the sake of clarity we preferred to give the expressions for the $\mathscr{U}_m$'s as well, for $m \leq 2$.

From the practical viewpoint, the most common case is when $\mathscr{H}(\epsilon)$ is a linear function of ϵ:

$$\mathscr{H}(\epsilon)=\mathscr{H}_0+\epsilon\mathscr{V}\ ,\tag{32}$$

i.e., $\mathscr{H}_1=\mathscr{V}$, $\mathscr{H}_2=\mathscr{H}_3=\cdots=0$. The following formulas now hold:

$$\mathscr{K}_1=\mathscr{V}^{\{d\}}\ ,\tag{33a}$$

$$\mathscr{K}_2=(\mathscr{V}^{\{n,E\}}\mathscr{V})^{\{d\}}\ ,\tag{33b}$$

$$\mathscr{K}_3=[(\mathscr{V}^{\{n,E\}}\mathscr{V})^{\{n,E\}}\mathscr{V}]^{\{d\}}+\tfrac{1}{2}(\mathscr{V}^{\{n,E\}}\mathscr{V}^{\{n,E\}})^{\{d\}}\mathscr{V}^{\{d\}}+\tfrac{1}{2}\mathscr{V}^{\{d\}}(\mathscr{V}^{\{n,E\}}\mathscr{V}^{\{n,E\}})^{\{d\}}\ ,\tag{33c}$$

$$\begin{aligned}
\mathscr{K}_4=&\{[(\mathscr{V}^{\{n,E\}}\mathscr{V})^{\{n,E\}}\mathscr{V}]^{\{n,E\}}\mathscr{V}\}^{\{d\}}+[(\mathscr{V}^{\{n,E\}}\mathscr{V})^{\{n,E\}}\mathscr{V}^{\{n,E\}}]^{\{d\}}\mathscr{V}^{\{d\}}-\mathscr{V}^{\{d\}}[\mathscr{V}^{\{n,E\}}(\mathscr{V}\mathscr{V}^{\{n,E\}})^{\{n,E\}}]^{\{d\}}\\
&+\tfrac{1}{3}\mathscr{V}^{\{d\}}(\mathscr{V}^{\{n,E\}}\mathscr{V}^{\{n,E\}}\mathscr{V}^{\{n,E\}})^{\{d\}}-\tfrac{1}{3}(\mathscr{V}^{\{n,E\}}\mathscr{V}^{\{n,E\}}\mathscr{V}^{\{n,E\}})^{\{d\}}\mathscr{V}^{\{d\}}+\tfrac{1}{2}(\mathscr{V}^{\{n,E\}}\mathscr{V})^{\{d\}}(\mathscr{V}^{\{n,E\}}\mathscr{V}^{\{n,E\}})^{\{d\}}\\
&-\tfrac{1}{2}(\mathscr{V}^{\{n,E\}}\mathscr{V}^{\{n,E\}})^{\{d\}}(\mathscr{V}\mathscr{V}^{\{n,E\}})^{\{d\}}+\mathscr{V}^{\{d\}}[\mathscr{V}^{\{n,E\}}(\mathscr{V}^{\{n,E\}})^{\{n,E\}}]^{\{d\}}\mathscr{V}^{\{d\}}\ ,
\end{aligned}\tag{33d}$$

etc. This case, with the additional assumption of $\mathscr{V}^{\{d\}}=0$, has been described by Heitler[3] with accuracy up to terms of order 4 and applying the interaction picture.

Having derived the formula of the canonical transformation we no longer need to write ϵ explicitly. We just assume ϵ^m as included into the $\mathscr{H}_m$, $\mathscr{K}_m$, and $\mathscr{U}_m$, respectively. This is equivalent to the current procedure of inserting a perturbation parameter equal to unity.

The advantage of the method proposed above resides in its far-reaching algebraic manipulation and simplification compared to standard perturbation calculus and, moreover, in its applicability in cases of high degeneracy of the spectrum of the initial unperturbed operator $\mathscr{H}_0$. Its range of applicability is, in fact, much wider than it might appear at first sight. Its extension to the case of a continuous spectrum of $\mathscr{H}_0$ reduces to modifying the definition of the operations of taking the diagonal part and of taking the energy-weighted nondiagonal part. In brief, diagonalization will take place with accuracy up to some interval and, instead of sums, integrals or their principal values will appear. The use of the Heisenberg picture is by no means required; just as well, recourse can be taken to, e.g., the interaction picture. Again, the form of Eq. (31) remains unchanged; only the definition of the operation of taking the energy-weighted nondiagonal part undergoes a modification (an integral over time will appear therein).

In the absence of degeneracy, Eq. (31) leads directly to the usual formulas of nondegenerate perturbation calculus for the successive corrections to the energy eigenvalues. We have

$$E_i=\sum_{n=0}^{\infty}E_i^{(n)},\quad E_i^{(n)}=\langle i\,|\,\mathscr{K}_n\,|\,i\rangle\equiv(\mathscr{K}_n)_{ii}\ .\tag{34}$$

In particular, by (31) we immediately get

$$E_i^{(0)}=E_{0i}\ ,\tag{35a}$$

$$E_i^{(1)}=(\mathscr{H}_1)_{ii}\ ,\tag{35b}$$

$$E_i^{(2)}={\sum_j}'\frac{(\mathscr{H}_1)_{ij}(\mathscr{H}_1)_{ji}}{E_{0i}-E_{0j}}+(\mathscr{H}_2)_{ii}\ ,\tag{35c}$$

$$E_i^{(3)}=\sum_j{}'\sum_k{}'\frac{(\mathscr{H}_1)_{ij}(\mathscr{H}_1)_{jk}(\mathscr{H}_1)_{kl}}{(E_{0i}-E_{0j})(E_{0i}-E_{0k})}$$
$$-(\mathscr{H}_1)_{ii}\sum_j{}'\frac{(\mathscr{H}_1)_{ij}(\mathscr{H}_1)_{ji}}{(E_{0i}-E_{0j})^2}+\sum_j{}'\frac{(\mathscr{H}_2)_{ij}(\mathscr{H}_1)_{ji}}{E_{0i}-E_{0j}}$$
$$+\sum_j{}'\frac{(\mathscr{H}_1)_{ij}(\mathscr{H}_2)_{ji}}{E_{0i}-E_{0j}}+(\mathscr{H}_3)_{ii}\ , \tag{35d}$$

etc. The primes denote omission of the index i in the sums.

Finally, as an example, we shall consider nondegenerate perturbation calculus as applied to a Hamiltonian of the form

$$\mathscr{H}=\mathscr{H}_0+\mathscr{V},\quad \mathscr{H}_0=\epsilon S_3,\quad \mathscr{V}=\lambda(S^+ + S^-)\ , \tag{36}$$

where $S_3, S^{\pm}$ are spin operators, fulfilling the usual commutation rules

$$[S^+,S^-]=2S_3,\quad [S_3,S^{\pm}]=\pm S^{\pm}\ . \tag{37}$$

By applying the unitary operator defined as

$$\mathscr{U}=\exp[\tfrac{1}{2}\varphi(S^+ - S^-)]\ , \tag{38}$$

where the parameter φ is expressed in conformity with the relations

$$\cos\varphi=\frac{\epsilon}{(\epsilon^2+4\lambda^2)^{1/2}},\quad \sin\varphi=\frac{2\lambda}{(\epsilon^2+4\lambda^2)^{1/2}}\ , \tag{39}$$

we diagonalize $\mathscr{H}$ [(Ref. 36)] with respect to the energy shells of the operator $\mathscr{H}_0$ *strictly:*

$$\mathscr{K}=\mathscr{U}\mathscr{H}\mathscr{U}^{\dagger}=(\epsilon^2+4\lambda^2)^{1/2}S_3\ . \tag{40}$$

We now proceed to apply the canonical transformation proposed by us. Taking recourse to

$$[(S^+)^m(S^-)^n]^{\{d\}}=\delta_{mn}(S^+)^m(S^-)^m\ , \tag{41}$$

$$[(S^+)^m(S^-)^n]^{\{n,E\}}=\begin{cases}\dfrac{1}{(m-n)\epsilon}(S^+)^m(S^-)^n & \text{for } m\neq n\\ 0 \text{ for } m=n\end{cases} \tag{42}$$

we obtain

$$\begin{gathered}\mathscr{K}_1=\mathscr{K}_3=\mathscr{K}_5=\cdots=0\ ,\\ \mathscr{K}_0=\epsilon S_3,\quad \mathscr{K}_2=\frac{2\lambda^2}{\epsilon}S_3,\quad \mathscr{K}_4=-\frac{2\lambda^4}{\epsilon^3}S_3\ ,\end{gathered} \tag{43}$$

etc., i.e.,

$$\mathscr{K}=\left[\epsilon+\frac{2\lambda^2}{\epsilon}-\frac{2\lambda^4}{\epsilon^3}+\cdots\right]S_3\ . \tag{44}$$

This result is in complete agreement with the strict result (40) since the expression in parentheses is a series expansion of $(\epsilon^2+4\lambda^2)^{1/2}$. In the following sections we show how the results obtained above can be used to derive the so-called effective Hamiltonians, which are widely applied in quantum optics.

III. MODEL

Basically, we shall be dealing with a system of electrons at interaction with a radiation field described in terms of a vector potential $\vec{A}$. The Hamiltonian of the system can be assumed in the form

$$\mathscr{H}=\sum_q \mathscr{H}_q+\mathscr{H}_{\text{Coul}}+\mathscr{H}_{\text{rad}}\ . \tag{45}$$

Its components have the following meaning:

$$\mathscr{H}_q=\frac{1}{2m}[\vec{p}_q-e\vec{A}(\vec{x}_q)]^2+e\varphi(\vec{x}_q) \tag{46}$$

is the nonrelativistic Hamiltonian of the qth electron moving in an external radiation field and a scalar potential φ. $\mathscr{H}_{\text{Coul}}$ refers to the electrostatic interaction of electrons and takes the form

$$\mathscr{H}_{\text{Coul}}=\tfrac{1}{2}\sum_{q_1,q_2}{}' e^2(4\pi\epsilon_0|\vec{x}_{q_1}-\vec{x}_{q_2}|)^{-1}\ , \tag{47}$$

where the prime means that summation is carried over all q_1,q_2, with $q_1\neq q_2$. From the point of view of our present aims, $\mathscr{H}_{\text{Coul}}$ is a redundant complication and we shall be omitting it throughout. In fact, in many cases of practical significance the assumption that the electrons interact by way of the radiation field and the omission of direct Coulomb interaction is fully justified. It should be stated that this simplification involves no restriction on the generality of the method but is aimed at enhancing the clarity of our presentation. Finally, $\mathscr{H}_{\text{rad}}$ is the Hamiltonian of the source-free field of radiation;

$$\mathscr{H}_{\text{rad}}=\tfrac{1}{2}\int_V d^3x(\vec{E}\cdot\vec{D}+\vec{B}\cdot\vec{H})\ , \tag{48}$$

where the fields $\vec{E}$ and $\vec{B}$ are given in terms of potential as follows:

$$\vec{B}=\vec{\nabla}\times\vec{A},\quad \vec{E}=-\frac{\partial\vec{A}}{\partial t}\ , \tag{49}$$

whence

$$\mathscr{H}_{\text{rad}}=\tfrac{1}{2}\int_V d^3x\left[\epsilon_0\left[-\frac{\partial\vec{A}}{\partial t}\right]^2+\mu_0^{-1}(\vec{\nabla}\times\vec{A})^2\right]\ . \tag{50}$$

We now write $\mathscr{H}$ in a form more adequate to our further considerations:

$$\mathscr{H}=\mathscr{H}_0+\mathscr{H}_1+\mathscr{H}_2\ , \tag{51}$$

where the "unperturbed part" $\mathscr{H}_0$ has form of the sum

$$\mathscr{H}_0=\mathscr{H}_{\text{el}}+\mathscr{H}_{\text{rad}},\quad \mathscr{H}_{\text{el}}=\sum_q\left[\frac{1}{2m}\vec{p}_q^{\,2}+e\varphi(\vec{x}_q)\right]\ . \tag{52}$$

The terms $\mathscr{H}_1$ and $\mathscr{H}_2$ are expressions, respectively, of the first and second order in the electron charge e:

$$\mathscr{H}_1=-\frac{e}{2m}\sum_q[\vec{p}_q\vec{A}(\vec{x}_q)+\vec{A}(\vec{x}_q)\vec{p}_q]\ , \tag{53}$$

$$\mathscr{H}_2=\frac{e^2}{2m}\sum_q\vec{A}^{\,2}(\vec{x}_q)\ . \tag{54}$$

The above model is of rather high generality. By an appropriate choice of the potential φ it can be fitted to the description of various real physical systems, such as the solid (when φ is the spatially periodic crystal lattice potential; the lattice vibrations can be included into the description quite easily). The Hamiltonian of the electrons of an atom, written in a coordinate system attached to the nucleus, is of the same form. Sometimes, one considers quantum structures moving—without colliding—as a whole according to the rules of classical mechanics. Then one only needs to add to $\vec{x}_q$ a given vector function of the time $\vec{\Theta}_q=\vec{\Theta}_q(t)$. The Doppler effect, which has various essential implications, i.a., for inhomogeneous line broadening, may be included by putting $\vec{\Theta}_q(t)=\vec{r}_q+\vec{v}_q t$, with $\vec{r}_q$ the positions and $\vec{v}_q$ the velocities at the initial moment of time $t=0$. The atoms are also assumed to move uniformly, rectilinearly and without colliding. Setting $\vec{v}_q=0$, one hence obtains a Hamiltonian for the case of stationary atoms well adapted to the description of admixtures in solids.

Our further considerations will be based on the second quantization formalism. The vector potential of the fields is given as follows:

$$\vec{A}(\vec{x})=\sum_{\kappa}(2\epsilon_0\omega_\kappa)^{-1/2}[\vec{u}(\vec{x})a_\kappa+\vec{u}^*(\vec{x})a_\kappa^\dagger] \,. \tag{55}$$

The $a_\kappa^\dagger,a_\kappa$ are the usual boson operators of creation and annihilation of a mode, denoted by κ, comprising the complete set of mode indices:

$$[a_\kappa,a_{\kappa'}^\dagger]=\delta_{\kappa\kappa'},\;\; [a_\kappa,a_{\kappa'}]=[a_\kappa^\dagger,a_{\kappa'}^\dagger]=0 \,. \tag{56}$$

As usual, the functions $\vec{u}_\kappa(\vec{x})$ form an orthonormal set of mode functions chosen in accordance with the volume V in which the field is to be quantized:

$$\int_V d^3x\, \vec{u}_\kappa^*(\vec{x})\vec{u}_\kappa(\vec{x})=\delta_{\kappa\kappa'} \,. \tag{57}$$

Since we apply the Coulomb gauge, it is obvious that $\operatorname{div}\vec{u}_\kappa(\vec{x})=0$. The Hamiltonian of the free radiation field now becomes

$$\mathscr{H}_{\text{rad}}=\sum_{\kappa}\omega_\kappa a_\kappa^\dagger a_\kappa \tag{58}$$

(the field energy is counted starting from the vacuum value). For the sake of convenience, we shall sometimes be using an index α equal to " + " or "−" and the notation

$$a_\kappa^\alpha=\begin{cases} a_\kappa^\dagger, & \alpha=+ \\ a_\kappa, & \alpha=- \end{cases} \tag{59a}$$

$$\vec{u}_\kappa^\alpha(\vec{x})=\begin{cases} \vec{u}_\kappa^*(\vec{x}), & \alpha=+ \\ \vec{u}_\kappa(\vec{x}), & \alpha=- \,. \end{cases} \tag{59b}$$

With this notation, e.g., the vector potential (55) assumes the form

$$\vec{A}(\vec{x})=\sum_{\alpha,\kappa}(2\epsilon_0\omega_\kappa)^{-1/2}\vec{u}_\kappa^\alpha(\vec{x})a_\kappa^\alpha \,. \tag{60}$$

Let $|\mu\rangle$ be the μth eigenstate of the one-electron Hamiltonian:

$$\left[\frac{1}{2m}\vec{p}^{\,2}+e\varphi(\vec{x})\right]|\mu\rangle=E_\mu|\mu\rangle \tag{61}$$

with μ denoting collectively the set of quantum numbers characterizing the electron. When dealing with a system of atoms μ will always be meant to include the number q, labeling the atom (each atom is understood to have but one electron). In some cases we shall separate q from μ, thus leaving μ to describe the remaining quantum numbers only; then, we shall write μq in place of the single μ.

Let $b_\mu^\dagger,b_\mu$ denote creation and annihilation operators of an electron in its μth state. They fulfill the usual anticommutation rules

$$[b_\mu,b_\mu^\dagger]_+=\delta_{\mu\mu'},\;\; [b_\mu,b_{\mu'}]_+=[b_\mu^\dagger,b_{\mu'}^\dagger]_+=0 \,. \tag{62}$$

The Hamiltonian of mutually noninteracting electrons now takes the form

$$\mathscr{H}_{\text{el}}=\sum_{\mu}E_\mu b_\mu^\dagger b_\mu \,. \tag{63}$$

Hence,

$$\mathscr{H}_0=\sum_{\kappa}\omega_\kappa a_\kappa^\dagger a_\kappa+\sum_{\mu}E_\mu b_\mu^\dagger b_\mu \,. \tag{64}$$

As for the terms $\mathscr{H}_1$ and $\mathscr{H}_2$ of the Hamiltonian, they become

$$\mathscr{H}_1=\sum_{\alpha,\kappa}\sum_{\mu,\nu}\left|\begin{matrix}\alpha\kappa\\ \mu\nu\end{matrix}\right| a_\kappa^\alpha b_\mu^\dagger b_\nu \,, \tag{65}$$

$$\mathscr{H}_2=\sum_{\alpha_1,\kappa_1}\sum_{\alpha_2,\kappa_2}\sum_{\mu,\nu}\left|\begin{matrix}\alpha_1\kappa_1\alpha_2\kappa_2\\ \mu\nu\end{matrix}\right| a_{\kappa_1}^{\alpha_1}a_{\kappa_2}^{\alpha_2}b_\mu^\dagger b_\nu \,. \tag{66}$$

The constants occurring in the preceding formulas have the explicit form

$$\left|\begin{matrix}\alpha\kappa\\ \mu\nu\end{matrix}\right|=-\frac{e}{m}(2\epsilon_0\omega_\kappa)^{-1/2}\langle\mu\,|\,\vec{p}\cdot\vec{u}_\kappa^\alpha\,|\,\nu\rangle \,, \tag{67}$$

$$\left|\begin{matrix}\alpha_1\kappa_1\alpha_2\kappa_2\\ \mu\nu\end{matrix}\right|=\frac{e}{4\epsilon_0 m}(\omega_{\kappa_1}\omega_{\kappa_2})^{-1/2}\langle\mu\,|\,\vec{u}_{\kappa_1}^{\alpha_1}\cdot\vec{u}_{\kappa_2}^{\alpha_2}\,|\,\nu\rangle \,. \tag{68}$$

When writing their products we shall be omitting the matrix brackets and separating the indices by commas.

The eigenstates of the operator $\mathscr{H}_0$ are tensor products of eigenstates of the operators $\mathscr{H}_{\text{rad}}$ and $\mathscr{H}_{\text{el}}$ and are written as follows:

$$|\ldots,n_\kappa,\ldots\rangle\otimes|\ldots,m_\mu,\ldots\rangle \,, \tag{69}$$

where n_κ is the number of photons of the mode κ and m_μ the number of electrons in the μth state. The eigenvalue corresponding to this function is equal to

$$\sum_{\kappa}\omega_\kappa n_\kappa+\sum_{\mu}E_\mu m_\mu \,. \tag{70}$$

For a system of N one-electron atoms the eigenstates are of the form

$$|\ldots,n_\kappa,\ldots\rangle\otimes|\mu\rangle_{q=1}\otimes|\mu\rangle_{q=2}\otimes\cdots\otimes|\mu\rangle_{q=N}\ , \tag{71}$$

where

$$|\mu\rangle_q\equiv|\ldots,m_{\mu q}=1,\ldots\rangle\ . \tag{72}$$

The respective eigenvalue is

$$\sum_\kappa \omega_\kappa n_\kappa+\sum_{\mu,q}E_{\mu q}m_{\mu q}\ . \tag{73}$$

Hence, one notes that the spectrum of the "free" Hamiltonian $\mathscr{H}_0$ is, in general, highly degenerate. Thus, e.g., if, for two electron states μ and ν, the condition of photon resonance

$$E_\mu-E_\nu=p\omega_\kappa \tag{74}$$

is fulfilled, the states of the system as a whole with quantum numbers $n_\kappa+p,m_\mu=0$, $m_\nu=1$ and $n_\kappa,m_\mu=1$, $m_\nu=0$ correspond to the same eigenvalue (70). Similarly, for a system of atoms, if the condition

$$E_{\mu q}-E_{\nu q}=p\omega_\kappa \tag{75}$$

is fulfilled, the states $n_\kappa+p,m_{\mu q}=0$, $m_{\nu q}=1$ and $n_\kappa,m_{\mu q}=1$, $m_{\nu q}=0$ correspond to the same energy (73). Considering that the equality (75) can hold for a very great number of atoms and even (as it is in the case of homogeneous line broadening) for all the atoms, one readily imagines the order of degeneracy involved. Hence, instead of standard perturbation calculus, it is better to have recourse to the canonical-transformation method proposed in Sec. II.

As a result of the explicit form of $\mathscr{H}_1$ and $\mathscr{H}_2$ we shall deal only with operators of the form

$$\mathscr{A}=a_{\kappa_1}^{\alpha_1}a_{\kappa_2}^{\alpha_2}\cdots a_{\kappa_r}^{\alpha_r}b_{\mu_1}^\dagger b_{\nu_1}b_{\mu_2}^\dagger b_{\nu_2}\cdots b_{\mu_s}^\dagger b_{\nu_s}\ . \tag{76}$$

For each operator of the preceding form, we define a number

$$\begin{aligned}\lambda(\mathscr{A})\equiv{}&\alpha_1\omega_{\kappa_1}+\alpha_2\omega_{\kappa_2}+\cdots+\alpha_r\omega_{\kappa_r}\\&+\omega_{\mu_1\nu_1}+\omega_{\mu_2\nu_2}+\cdots+\omega_{\mu_s\nu_s}\end{aligned} \tag{77}$$

which defines the balance of frequencies, related with the operators occurring in $\mathscr{A}$. We have made use of the notation

$$\omega_{\mu\nu}\equiv E_\mu-E_\nu\ . \tag{78}$$

If the equality $\lambda(\mathscr{A})=0$ is fulfilled we shall be referring to the situation as generalized resonance of rth order. We identify the order of the resonance with the number of photon frequencies occurring in $\lambda(\mathscr{A})$. In this meaning, e.g., the Raman effect, for which a relation of the form $\omega_{\kappa_1}-\omega_{\kappa_2}+\omega_{\mu\nu}=0$ is fulfilled, is a generalized resonance of order 2.

The operator $\mathscr{A}$ given by formula (76) is diagonal if and only if $\lambda(\mathscr{A})=0$. Hence, the result of the operation of extracting the diagonal part takes the simple form

$$\mathscr{A}^{\{d\}}=\begin{cases}\mathscr{A} & \text{if } \lambda(\mathscr{A})=0\\ 0 & \text{if } \lambda(\mathscr{A})\neq0\ .\end{cases} \tag{79}$$

The other (energy-weighted) operation is also easy to perform. One notes that

$$\mathscr{A}^{\{n,\omega\}}=\begin{cases}0 & \text{if } \lambda(\mathscr{A})=0\\ \lambda^{-1}(\mathscr{A})\mathscr{A} & \text{if } \lambda(\mathscr{A})\neq0\ .\end{cases} \tag{80}$$

Thus, e.g.,

$$(a_{\kappa_1}^\dagger a_{\kappa_2}b_\mu^\dagger b_\nu)^{\{n,\omega\}}=\begin{cases}0 \quad \text{if } \omega_{\kappa_1}-\omega_{\kappa_2}+\omega_{\mu\nu}=0\\ \dfrac{a_{\kappa_1}^\dagger a_{\kappa_2}b_\mu^\dagger b_\nu}{\omega_{\kappa_1}-\omega_{\kappa_2}+\omega_{\mu\nu}}\\ \qquad \text{if } \omega_{\kappa_1}-\omega_{\kappa_2}+\omega_{\mu\nu}\neq0\ .\end{cases}$$

If a given operator is a linear combination of operators $\mathscr{A}_i$, $i\in I$, each of which is of the form (76), we have

$$\left[\sum_i\alpha_i\mathscr{A}_i\right]^{\{d\}}=\sum_i\alpha_i\mathscr{A}_i^{\{d\}}=\sum_i\alpha_i^{\{d\}}\mathscr{A}_i\ , \tag{81}$$

$$\left[\sum_i\alpha_i\mathscr{A}_i\right]^{\{n,\omega\}}=\sum_i\alpha_i\mathscr{A}_i^{\{n,\omega\}}=\sum_i\alpha_i^{\{n,\omega\}}\mathscr{A}_i\ , \tag{82}$$

where the new coefficients of the linear combinations are defined by the equalities

$$\alpha_i^{\{d\}}\equiv\begin{cases}\alpha_i & \text{if } \lambda(\mathscr{A}_i)=0\\ 0 & \text{if } \lambda(\mathscr{A}_i)\neq0\end{cases} \tag{83}$$

and

$$\alpha_i^{\{n,\omega\}}\equiv\begin{cases}0 & \text{if } \lambda(\mathscr{A}_i)=0\\ \lambda^{-1}(\mathscr{A}_i)\alpha_i & \text{if } \lambda(\mathscr{A}_i)\neq0\ .\end{cases} \tag{84}$$

This is a very simple and, at the same time, very important result. Application of the operations represented by superscripts $\{d\}$ and $\{n,\omega\}$ to operators of the form (76) reduces to transformations of the coefficients at these operators. For simplicity, we use the same symbols to denote both transformations. For example, we have

$$\left|\begin{bmatrix}\alpha_1\kappa_1\\ \mu_1\nu_1\end{bmatrix}^{\{n,\omega\}}\begin{matrix}\alpha_2\kappa_2\alpha_3\kappa_3\\ \mu_2\nu_2\end{matrix}\right|^{\{d\}}=\begin{cases}\dfrac{\begin{vmatrix}\alpha_1\kappa_1,\alpha_2\kappa_2\alpha_3\kappa_3\\ \mu_1\nu_1,\mu_2\nu_2\end{vmatrix}}{\alpha_1\omega_{\kappa_1}+\omega_{\mu_1\nu_1}} & \text{if } \alpha_1\omega_{\kappa_1}+\omega_{\mu_1\nu_1}\neq0 \text{ and } \alpha_1\omega_{\kappa_1}+\alpha_2\omega_{\kappa_2}+\omega_{\mu_1}\nu_1+\omega_{\mu_2}\nu_2=0\\ 0 & \text{otherwise}\end{cases}$$

and

$$\left[\left[\begin{matrix}\alpha\kappa\\ \mu\nu\end{matrix}\right]^{\{n,\omega\}}\right]^{\{n,\omega\}}=\begin{cases}0 \text{ if } \alpha\omega_\kappa+\omega_{\mu\nu}=0\\ \dfrac{\left|\begin{matrix}\alpha\kappa\\ \mu\nu\end{matrix}\right|}{(\alpha\omega_\kappa+\omega_{\mu\nu})^2} \text{ otherwise .}\end{cases}$$

As we shall often require Wick's theorem in the course of the later sections of this paper, we introduce the set of double dots ": :" to denote the operation of normal ordering as well as pairs of superscript dots, double dots, etc.,

$$(a^{\alpha_1}_{\kappa_1})^{\bullet}(a^{\alpha_2}_{\kappa_2})^{\bullet}=\begin{cases}\delta_{\kappa_1\kappa_2} \text{ if } \alpha_1=- \text{ and } \alpha_2=+\\ 0 \text{ otherwise}\end{cases} \tag{85}$$

$$(b^{\alpha_1}_{\mu_1})^{\bullet}(b^{\alpha_2}_{\mu_2})^{\bullet}=\begin{cases}\delta_{\mu_1\mu_2} \text{ if } \alpha_1=- \text{ and } \alpha_2=+\\ 0 \text{ otherwise ,}\end{cases} \tag{86}$$

to denote contractions. If there is but one electron in the system,

$$\sum_\mu b^\dagger_\mu b_\mu=1 , \tag{87}$$

Wick's theorem leads immediately to the equality

$$b^\dagger_{\mu_1}b_{\nu_1}b^\dagger_{\mu_2}b_{\nu_2}\cdots b^\dagger_{\mu_m}b_{\nu_m}$$

$$=\delta_{\nu_1\mu_2}\delta_{\nu_2\mu_3}\cdots\delta_{\nu_{m-1}\mu_m}b^\dagger_{\mu_1}b_{\nu_m} . \tag{88}$$

Hence, in the case of many (one-electron) atoms we have, for arbitrary q, the following relation:

$$b^\dagger_{\mu_1 q}b_{\nu_1 q}b^\dagger_{\mu_2 q}b_{\nu_2 q}\cdots b^\dagger_{\mu_m q}b_{\nu_m q}$$

$$=\delta_{\nu_1\mu_2}\delta_{\nu_2\mu_3}\cdots\delta_{\nu_{m-1}\mu_m}b^\dagger_{\mu_1 q}b_{\nu_m q} . \tag{89}$$

In this way, we have prepared the terrain for deriving the effective Hamiltonians. This will be the subject of Sec. IV.

IV. EFFECTIVE HAMILTONIANS

Here, we shall derive effective Hamiltonians for a variety of situations—including one and many electrons—omitting as well as taking into account the term in $\vec{A}^2$ in the interaction Hamiltonian. The results are applicable, e.g., to the description of the one-electron atom, a system of stationary atoms, and electrons in a solid. In accordance with a statement in Sec. III, the extension to atoms in motion is straightforward. We start from the simplest case.

One electron; $\vec{A}^2$-type interaction omitted. Since there is but one electron, the equality (88) holds. We perform the canonical transformation. Omitting the $\vec{A}^2$-type interaction, we are justified in applying formulas (33). We obtain

$$\mathscr{K}=\sum_{m=0}^{\infty}\mathscr{K}_m , \tag{90}$$

where

$$\mathscr{K}_0=\sum_\kappa \omega_\kappa a^\dagger_\kappa a_\kappa+\sum_\mu E_\mu b^\dagger_\mu b_\mu , \tag{91}$$

$$\mathscr{K}_m=\sum_{\substack{\alpha_1,\alpha_2,\ldots,\alpha_m\\ \kappa_1,\kappa_2,\ldots,\kappa_m}}\sum_{\mu,\nu}K^{\alpha_1\alpha_2\cdots\alpha_m}_{\kappa_1\kappa_2\cdots\kappa_m,\mu\nu}$$

$$\times a^{\alpha_1}_{\kappa_1}a^{\alpha_2}_{\kappa_2}\cdots a^{\alpha_m}_{\kappa_m}b^\dagger_\mu b_\nu, \quad m\geq 1 . \tag{92}$$

With regard to formulas (33) we have

$$K^{\alpha_1}_{\kappa_1,\mu\nu}=\left|\begin{matrix}\alpha_1\kappa_1\\ \mu\nu\end{matrix}\right|^{\{d\}} , \tag{93a}$$

$$K^{\alpha_1\alpha_2}_{\kappa_1\kappa_2,\mu\nu}=\sum_\lambda\left|\left[\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right]^{\{n,\omega\}}\begin{matrix}\alpha_2\kappa_2\\ \lambda\nu\end{matrix}\right|^{\{d\}} , \tag{93b}$$

$$\begin{aligned}K^{\alpha_1\alpha_2\alpha_3}_{\kappa_1\kappa_2\kappa_3,\mu\nu}=\sum_{\lambda,\rho}\Bigg\{&\left|\left[\left[\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right]^{\{n,\omega\}}\begin{matrix}\alpha_2\kappa_2\\ \lambda\rho\end{matrix}\right]^{\{n,\omega\}}\begin{matrix}\alpha_3\kappa_3\\ \rho\nu\end{matrix}\right|^{\{d\}}\\ &+\frac{1}{2}\left|\left[\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right]^{\{n,\omega\}}\left[\begin{matrix}\alpha_2\kappa_2\\ \lambda\rho\end{matrix}\right]^{\{n,\omega\}}\right|^{\{d\}}\left|\begin{matrix}\alpha_3\kappa_3\\ \rho\nu\end{matrix}\right|^{\{d\}}+\frac{1}{2}\left|\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right|^{\{d\}}\left|\left[\begin{matrix}\alpha_2\kappa_2\\ \lambda\rho\end{matrix}\right]^{\{n,\omega\}}\left[\begin{matrix}\alpha_3\kappa_3\\ \rho\nu\end{matrix}\right]^{\{n,\omega\}}\right|^{\{d\}}\Bigg\} ,\end{aligned} \tag{93c}$$

$$
\begin{aligned}
&K^{\alpha_1\alpha_2\alpha_3\alpha_4}_{\kappa_1\kappa_2\kappa_3\kappa_4,\mu\nu}\\
&\quad=\sum_{\lambda,\rho,\pi}\Bigg[\left|\left\{\left[\left[\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right]^{\{n,\omega\}}\begin{matrix}\alpha_2\kappa_2\\ \lambda\rho\end{matrix}\right]^{\{n,\omega\}}\begin{matrix}\alpha_3\kappa_3\\ \rho\pi\end{matrix}\right\}^{\{n,\omega\}}\begin{matrix}\alpha_4\kappa_4\\ \pi\nu\end{matrix}\right|^{\{d\}}+\left|\left[\left[\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right]^{\{n,\omega\}}\begin{matrix}\alpha_2\kappa_2\\ \lambda\rho\end{matrix}\right]^{\{n,\omega\}}\left[\begin{matrix}\alpha_3\kappa_3\\ \rho\pi\end{matrix}\right]^{\{n,\omega\}}\right|^{\{d\}}\left|\begin{matrix}\alpha_4\kappa_4\\ \pi\nu\end{matrix}\right|^{\{d\}}\\
&\qquad-\left|\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right|^{\{d\}}\left|\left[\begin{matrix}\alpha_2\kappa_2\\ \lambda\rho\end{matrix}\right]^{\{n,\omega\}}\left[\begin{matrix}\alpha_3\kappa_3\\ \rho\pi\end{matrix}\left[\begin{matrix}\alpha_4\kappa_4\\ \pi\nu\end{matrix}\right]^{\{n,\omega\}}\right]^{\{n,\omega\}}\right|^{\{d\}}+\frac{1}{3}\left|\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right|^{\{d\}}\left|\left[\begin{matrix}\alpha_2\kappa_2\\ \lambda\rho\end{matrix}\right]^{\{n,\omega\}}\left[\begin{matrix}\alpha_3\kappa_3\\ \rho\pi\end{matrix}\right]^{\{n,\omega\}}\left[\begin{matrix}\alpha_4\kappa_4\\ \pi\nu\end{matrix}\right]^{\{n,\omega\}}\right|^{\{d\}}\\
&\qquad-\frac{1}{3}\left|\left[\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right]^{\{n,\omega\}}\left[\begin{matrix}\alpha_2\kappa_2\\ \lambda\rho\end{matrix}\right]^{\{n,\omega\}}\left[\begin{matrix}\alpha_3\kappa_3\\ \rho\pi\end{matrix}\right]^{\{n,\omega\}}\right|^{\{d\}}\left|\begin{matrix}\alpha_4\kappa_4\\ \pi\nu\end{matrix}\right|^{\{d\}}+\frac{1}{2}\left|\left[\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right]^{\{n,\omega\}}\begin{matrix}\alpha_2\kappa_2\\ \lambda\rho\end{matrix}\right|^{\{d\}}\left|\left[\begin{matrix}\alpha_3\kappa_3\\ \rho\pi\end{matrix}\right]^{\{n,\omega\}}\left[\begin{matrix}\alpha_4\kappa_4\\ \pi\nu\end{matrix}\right]^{\{n,\omega\}}\right|^{\{d\}}\\
&\qquad-\frac{1}{2}\left|\left[\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right]^{\{n,\omega\}}\left[\begin{matrix}\alpha_2\kappa_2\\ \lambda\rho\end{matrix}\right]^{\{n,\omega\}}\right|^{\{d\}}\left|\begin{matrix}\alpha_3\kappa_3\\ \rho\pi\end{matrix}\left[\begin{matrix}\alpha_4\kappa_4\\ \pi\nu\end{matrix}\right]^{\{n,\omega\}}\right|^{\{d\}}\\
&\qquad+\left|\begin{matrix}\alpha_1\kappa_1\\ \mu\lambda\end{matrix}\right|^{\{d\}}\left|\left[\begin{matrix}\alpha_2\kappa_2\\ \lambda\rho\end{matrix}\right]^{\{n,\omega\}}\left[\left[\begin{matrix}\alpha_3\kappa_3\\ \rho\pi\end{matrix}\right]^{\{n,\omega\}}\right]^{\{n,\omega\}}\right|^{\{d\}}\left|\begin{matrix}\alpha_4\kappa_4\\ \pi\nu\end{matrix}\right|^{\{d\}}\Bigg], \qquad (93\text{d})
\end{aligned}
$$

and so forth. These formulas make apparent the advantages to be drawn from the method proposed in this work. The expressions for the numerical coefficients $K^{\alpha_1\cdots\alpha_m}_{\kappa_1\cdots\kappa_m,\mu\nu}$ result, as it were, automatically from the general formulas (33). Moreover, $K^{\alpha_1\cdots\alpha_m}_{\kappa_1\cdots\kappa_m,\mu\nu}\neq 0$ only if the generalized resonance condition

$$\alpha_1\omega_{\kappa_3}+\cdots+\alpha_m\omega_{\kappa_m}+\omega_{\mu\nu}=0 \qquad (94)$$

is fulfilled. Hence, on transformation, the Hamiltonian contains only such terms as to fulfill the energy conservation principle (meaning that they admit only of transitions between states belonging to the same energy shell). In a way, the result resembles that obtained when applying the rotating-wave approximation, albeit with the significant advantage of being strict. Approximations appear only when some of the terms of the series (90) are omitted.

We wish to draw attention to the following fact. Each $\mathscr{H}_m$ contains one term, describing transitions through virtual electronic levels,

$$|(\cdots\{[(\mathscr{V})^{\{n,\omega\}}\mathscr{V}]^{\{n,\omega\}}\mathscr{V}\}^{\{n,\omega\}}\ldots\mathscr{V})^{\{n,\omega\}}|^{\{d\}}\,,$$

in which m $\mathscr{V}$ operators appear in order of decreasing level of nesting within braces, brackets, etc., and terms describing compositions of generalized resonances, i.e., transitions through at least one real level. They are terms having the form of a product of two or more diagonal operators. The coefficients $K^{\alpha_1\cdots\alpha_m}_{\kappa_1\cdots\kappa_m,\mu\nu}$ contain, summed together, the contributions from the two types of transitions.

The form (90) of the transformed Hamiltonian is still not satisfactory. Identical effects are described by different terms; besides, the physical interpretation of the latter is insufficiently clear. It is desirable to have a selection of the terms of $\mathscr{H}$ in conformity with the effects for which they account. The difficulties vanish on performing normal ordering of the field operators (the electron operators are already normally ordered). Through Wick's theorem we have

$$
\begin{aligned}
a^{\alpha_1}_{\kappa_1}\cdots a^{\alpha_m}_{\kappa_m}&=:a^{\alpha_1}_{\kappa_1}\cdots a^{\alpha_m}_{\kappa_m}:+\sum_{\substack{i,j\\ i<j}}:a^{\alpha_1}_{\kappa_1}\cdots(a^{\alpha_i}_{\kappa_i})^{\bullet}\cdots(a^{\alpha_j}_{\kappa_j})^{\bullet}\cdots a^{\alpha_m}_{\kappa_m}:\\
&\quad+\sum_{\substack{i,j,k,l\\ i<j<k<l}}[a^{\alpha_1}_{\kappa_1}\cdots(a^{\alpha_i}_{\kappa_i})^{\bullet}\cdots(a^{\alpha_j}_{\kappa_j})^{\bullet}\cdots(a^{\alpha_k}_{\kappa_k})^{\bullet\bullet}\cdots(a^{\alpha_l}_{\kappa_l})^{\bullet\bullet}\cdots a^{\alpha_m}_{\kappa_m}:\\
&\qquad+:a^{\alpha_1}_{\kappa_1}\cdots(a^{\alpha_i}_{\kappa_i})^{\bullet}\cdots(a^{\alpha_j}_{\kappa_j})^{\bullet\bullet}\cdots(a^{\alpha_k}_{\kappa_k})^{\bullet\bullet}\cdots(a^{\alpha_l}_{\kappa_l})^{\bullet}\cdots a^{\alpha_m}_{\kappa_m}:\\
&\qquad+:a^{\alpha_1}_{\kappa_1}\cdots(a^{\alpha_i}_{\kappa_i})^{\bullet}\cdots(a^{\alpha_j}_{\kappa_j})^{\bullet\bullet}\cdots(a^{\alpha_k}_{\kappa_k})^{\bullet}\cdots(a^{\alpha_l}_{\kappa_l})^{\bullet\bullet}\cdots a^{\alpha_m}_{\kappa_m}:]+\cdots\,. \qquad (95)
\end{aligned}
$$

Moreover, the following formulas hold:

$$\sum_{\substack{\alpha_1,\ldots,\alpha_m\\ \kappa_1,\ldots,\kappa_m}}K^{\alpha_1\cdots\alpha_m}_{\kappa_1\cdots\kappa_m,\mu\nu}:a^{\alpha_1}_{\kappa_1}\cdots a^{\alpha_m}_{\kappa_m}:=\sum_{\kappa_1,\ldots,\kappa_m}\sum_{n=0}^{m}\frac{1}{n!(m-n)!}\sum_{P_2(\kappa_1,\ldots,\kappa_m)}K^{\alpha_1\cdots\alpha_m}_{\kappa_1\cdots\kappa_m,\mu\nu}a^{\dagger}_{\kappa_1}\cdots a^{\dagger}_{\kappa_n}a_{\kappa_{n+1}}\cdots a_{\kappa_m}\,. \qquad (96)$$

The symbol $P_n(\kappa_1,\ldots,\kappa_m)$ stands for summation over all permutations of pairs of indices $\tilde{\alpha}_r,\tilde{\kappa}_r$ of the form

$$\begin{bmatrix} \tilde{\alpha}_1\tilde{\kappa}_1,\ldots,\tilde{\alpha}_n\tilde{\kappa}_n, & \tilde{\alpha}_{n+1}\tilde{\kappa}_{n+1},\ldots,\tilde{\alpha}_m\tilde{\kappa}_m \\ +\kappa_1,\ldots,+\kappa_n, & -\kappa_{n+1},\ldots,-\kappa_m \end{bmatrix} .$$

Similar expressions are obtained for field-operator products involving 1, 2, 3, and more contractions. Products of the m field operators appear in the expressions of order $m+2k$ on performing k-fold contraction. We redefine the coupling constants so that they shall comprise the higher-order contributions.

We introduce the following notation:

$$\delta E_\mu \equiv \sum_\lambda K_{\bar{\lambda}\bar{\lambda},\mu\mu}^{-+} + \sum_{\lambda,\lambda'} \left(K_{\bar{\lambda}\bar{\lambda}\bar{\lambda}'\bar{\lambda}',\mu\mu}^{-+-+} + K_{\bar{\lambda}\bar{\lambda}'\bar{\lambda}'\bar{\lambda}}^{--++} + K_{\bar{\lambda}\bar{\lambda}'\bar{\lambda}\bar{\lambda}'\mu\mu}^{--++}\right) + \cdots , \tag{97}$$

where the ellipsis stands for terms of order 6 and higher, and

$$\chi^n_{\kappa_1\cdots\kappa_m,\mu\nu} \equiv \frac{1}{n!(m-n)!} \times \sum_{P_n(\kappa_1,\ldots,\kappa_m)} \left[K^{\tilde{\alpha}_1\cdots\tilde{\alpha}_m}_{\tilde{\kappa}_1\cdots\tilde{\kappa}_m,\mu\nu} + \sum_\lambda \sum_{\substack{i,j \\ 1\le i\le j\le m+2}} K^{\tilde{\alpha}_1\cdots\tilde{\alpha}_{i-1},-,\tilde{\alpha}_i\cdots\tilde{\alpha}_{j-2},+,\tilde{\alpha}_{j-1}\cdots\tilde{\alpha}_m}_{\tilde{\kappa}_1\cdots\tilde{\kappa}_{i-1},\lambda,\tilde{\kappa}_i\cdots\tilde{\kappa}_{j-2},\lambda,\tilde{\kappa}_{j-1}\cdots\tilde{\kappa}_m,\mu\nu} + \cdots \right] , \tag{98}$$

where the last set of ellipsis stands for terms of order $m+4$ and higher, permitting the expression of the effective Hamiltonians in the general form:

$$\mathscr{K} = \sum_{m=0}^{\infty} \mathscr{K}_{\overset{\frown}{m}} , \tag{99}$$

where

$$\mathscr{K}_{\overset{\frown}{0}} = \sum_\kappa \omega_\kappa a_\kappa^\dagger a_\kappa + \sum_\mu (E_\mu + \delta E_\mu) b_\mu^\dagger b_\mu , \tag{100}$$

$$\mathscr{K}_{\overset{\frown}{m}} = \sum_{n=0}^{m} \sum_{\kappa_1,\ldots,\kappa_m} \sum_{\mu,\nu} \chi^n_{\kappa_1\cdots\kappa_m,\mu\nu} \times a_{\kappa_1}^\dagger \cdots a_{\kappa_n}^\dagger a_{\kappa_{n+1}} \cdots a_{\kappa_m} b_\mu^\dagger b_\nu , \quad m \ge 1 . \tag{101}$$

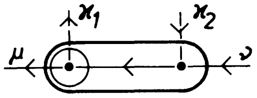
Obviously, the coefficients $\chi^n_{\kappa_1\cdots\kappa_m,\mu\nu}$ are symmetric in the first n and remaining m-n photon indices. The property

$$(\chi^n_{\kappa_1\cdots\kappa_m,\mu\nu})^* = \chi^{m-n}_{\kappa_m\cdots\kappa_1,\nu\mu} \tag{102}$$

ensures Hermiticity of the $\mathscr{K}_{\overset{\frown}{m}}$'s. In contradistinction to the $\mathscr{K}_m$'s, the $\mathscr{K}_{\overset{\frown}{m}}$'s are not terms of the mth order in the perturbation but contain corrections of higher orders. The transition from the $\mathscr{K}_m$'s to the $\mathscr{K}_{\overset{\frown}{m}}$'s corresponds to a partial resummation of the series composing the Hamiltonian $\mathscr{K}$. Clearly, the coupling constant $\chi^n_{\kappa_1\cdots\kappa_m,\mu\nu}$ is nonzero only if

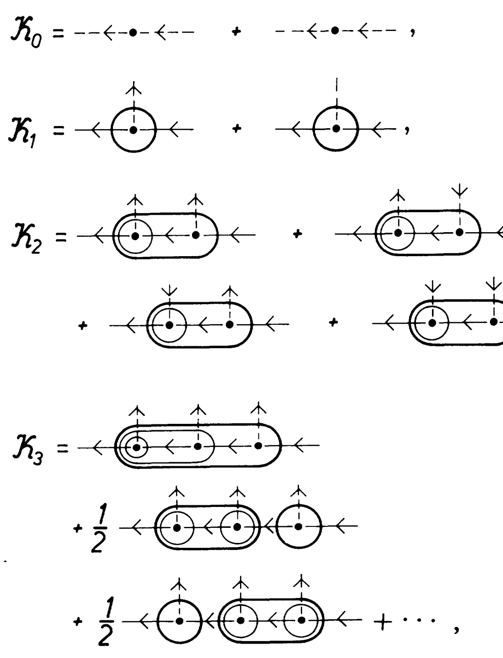
$$\omega_{\kappa_1} + \cdots + \omega_{\kappa_n} - \omega_{\kappa_{n+1}} - \cdots - \omega_{\kappa_m} + \omega_{\mu\nu} = 0 . \tag{103}$$

Diagram representation. The results derived by us above are well adapted to graphical representation using graphical symbols of Table I. Moreover, a diagram lacking one of the indices will be assumed to denote the sum of diagrams over the lacking index. For example,



$$\equiv \sum_\lambda \left| \begin{pmatrix} +\kappa_1 \\ \mu\lambda \end{pmatrix}^{\{n,\omega\}} \begin{matrix} -\kappa_2 \\ \lambda\nu \end{matrix} \right|^{\{d\}} a_{\kappa_1}^\dagger a_{\kappa_2} b_\mu^\dagger b_\nu .$$

The terms $\mathscr{K}_m$ of the Hamiltonian $\mathscr{K}$ now take the following form:



(104a)

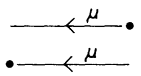
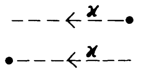
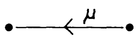
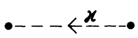
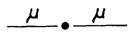
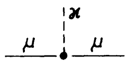
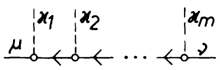
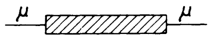
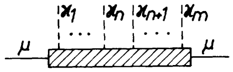
(104b)

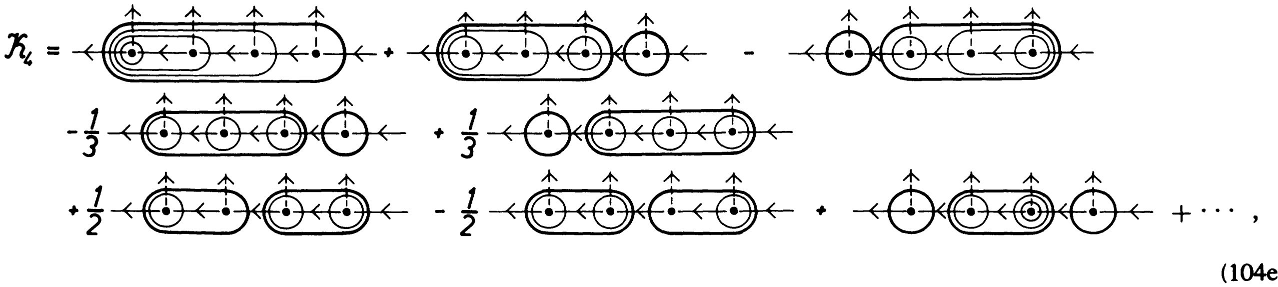
(104c)

(104d)

where the ellipsis stands for the remaining $(2^3-1)3=21$ diagrams,

TABLE I. Graphical symbols.

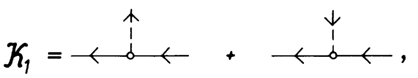
Mathematical quantity	Graphical symbol	Verbal description
electron $\left\{\begin{matrix}\text{creation}\\ \text{annihilation}\end{matrix}\right\}$ operators $\left\{\begin{matrix}b_\mu^\dagger\\ b_\mu\end{matrix}\right.$		continuous line directed $\left\{\begin{matrix}\text{away from}\\ \text{towards}\end{matrix}\right\}$ dot
photon $\left\{\begin{matrix}\text{creation}\\ \text{annihilation}\end{matrix}\right\}$ operators $\left\{\begin{matrix}a_\kappa^\dagger\\ a_\kappa\end{matrix}\right.$		dashed line directed $\left\{\begin{matrix}\text{away from}\\ \text{towards}\end{matrix}\right\}$ dot
electron contraction $(b_\mu)^\bullet(b_\mu^\dagger)^\bullet$		continuous line connecting two dots
photon contraction $(a_\kappa)^\bullet(a_\kappa^\dagger)^\bullet$		dashed line connecting two dots
E_μ		dot in which two continuous lines merge
ω_κ		dot in which two dashed lines merge
$\begin{vmatrix}\alpha\kappa\\ \mu\nu\end{vmatrix}$		dot in which two continuous lines and one dashed line merge
$(\cdots)^{\{d\}}$		thickly drawn cartouche
$(\cdots)^{\{n,\omega\}}$		thinly drawn cartouche
coupling constant $K^{\alpha_1\alpha_2\cdots\alpha_m}_{\kappa_1\kappa_2\cdots\kappa_m,\mu\nu}$		m little circles, in each of which two continuous lines and one dashed line merge, connected chainwise by dashed lines
$E_\mu+\delta E_\mu$		shaded rectangle, into which two (continuous) electron lines merge
coupling constant $\chi^n_{\kappa_1\cdots\kappa_m,\mu\nu}$		shaded rectangle, into which two continuous lines and m dashed lines merge



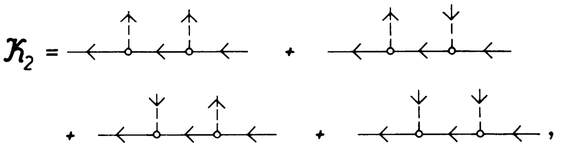
(104e)

where the ellipsis stands for the remaining $(2^4-1)8=120$ diagrams, and so forth. In the formulas for $\mathscr{K}_3$ and $\mathscr{K}_4$ we refrained, for the sake of conciseness, from plotting diagrams differing from those shown by the direction of at least one photon line (21 and 120 diagrams, respectively).

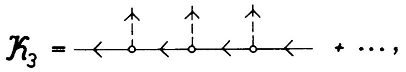
Overlooking the internal structure of the diagrams, i.e., going over to a description in terms of the coupling constants $K^{\alpha_1\cdots\alpha_m}_{\kappa_1\cdots\kappa_m,\mu\nu}$, we obtain



(105a)



(105b)

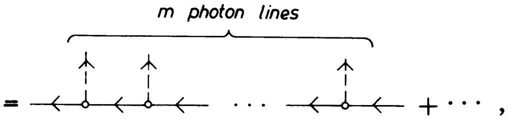


(105c)

where the ellipsis stands for the remaining 7 diagrams,

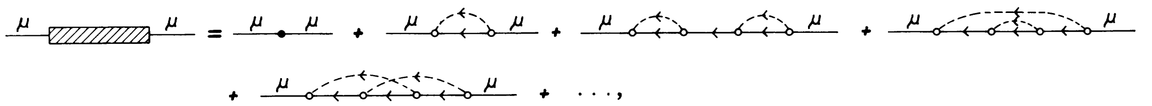
(105d)

where the ellipsis stands for the remaining 15 diagrams, etc. In general, for arbitrary $m\geq 1$, we have

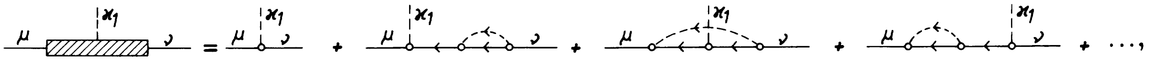


(106)

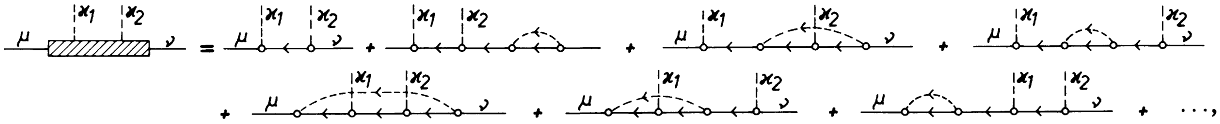
where the ellipsis stands for the remaining (2^m-1) diagrams. The final step of the preceding part of this section consisted in the application of normal ordering of the field operators. Taking recourse to expressions of the form



(107a)

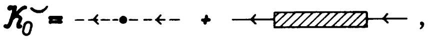


(107b)

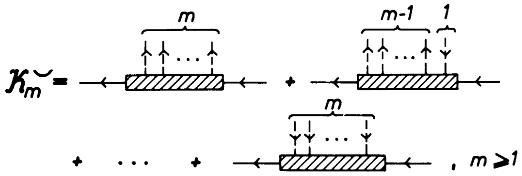


(107c)

etc., which graphically represent the relationships between the constants $E_\mu+\delta E_\mu$ and $X^n_{\kappa_1\cdots\kappa_m,\mu\nu}$ and the coefficients $K^{\alpha_1\cdots\alpha_m}_{\kappa_1\cdots\kappa_m,\mu\nu}$ we finally arrive at the following diagrammatic representation of the terms $\mathscr{K}^{\smile}_m$ [Eqs. (100) and (101)]:

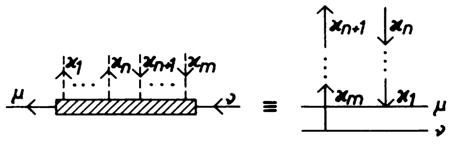


(108)



(109)

Yet another, maybe even more convincing visualization is worth mentioning. It consists in the assignment to each $\mathscr{K}^{\smile}_m$ of $\mathscr{K}$ of the corresponding scheme of transitions of the electron spectrum. This is a *one-to-one* assignment; it can be expressed as follows:



$$\equiv X^n_{\kappa_1\cdots\kappa_m,\mu\nu}a^\dagger_{\kappa_1}\cdots a^\dagger_{\kappa_n}a_{\kappa_{n+1}}\cdots a_{\kappa_m}b^\dagger_\mu b_\nu\ . \qquad (110)$$

The horizontal lines, denoting electronic levels, are connected by arrows which define the electron transition. On the other hand, the number and lengths of the arrows correspond to the number and frequencies of the photons taking part in the process. An arrow pointing upwards denotes absorption whereas one pointing downwards denotes emission of the corresponding photon. Intermediate levels have been omitted intentionally. They can be

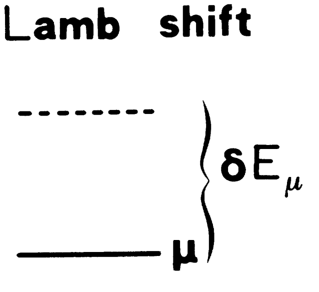


FIG. 2. Effect described by δE_{μ}.

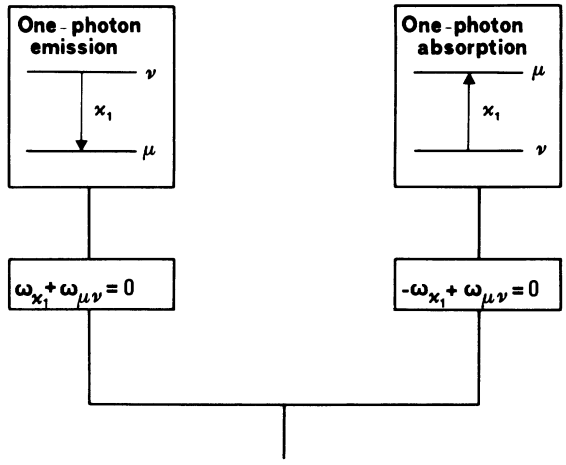


FIG. 3. Effects described by $\chi^{n}_{\kappa_1,\mu\nu}$.

real or virtual. The existence of transitions by way of real intermediate states is equivalent to the existence of compositions of generalized resonances. The structure of these compositions, defined by the formulas (33), is complicated and it is by no means easy to point to some simple procedure leading to it. Within the framework of the present theory, the question as to whether a given transition involves a real or a virtual level affects the dynamics of the system but does so indirectly—by affecting the values of the coefficients δE_{μ} and $\chi^{n}_{\kappa_1\cdots\kappa_m,\mu\nu}$. Diagrams of the above form readily permit the classification of the processes of quantum optics. Figures 2—5 represent such a classification of the phenomena under consideration, described by δE_{μ} and $\chi^{n}_{\kappa_1\cdots\kappa_m,\mu\nu}$, for $m \leq 3$.

One electron; $\vec{\mathrm{A}}^{2}$*-type interaction taken into account.* Taking into consideration the interaction term in $\vec{\mathrm{A}}^{2}$ causes but one complication: instead of the simpler formulas (33), the more highly complicated ones (31) have to be used. In all other respects the procedure is the same; even the final result is of the same form [Eqs. (100) and (101)], albeit the numerical values of the δE_{μ} and $\chi^{n}_{\kappa_1\cdots\kappa_m,\mu\nu}$ will contain corrections for the $\vec{\mathrm{A}}^{2}$-type interactions. Only the numerical values of the coefficients at the $K^{\alpha_1\cdots\alpha_m}_{\kappa_1\cdots\kappa_m,\mu\nu}$'s undergo a change, whereas the subsequent procedure of going over from the $\mathscr{K}_m$'s to $\mathscr{K}_{\breve{m}}$'s remains the same. Matters are similar regarding the diagrammatic representation. Apart from their internal structure, the diagrams will be the same in either case [formulas (105) *et seq.*]. This may suffice, as there is no need to reproduce essentially identical considerations.

Many electrons; cooperative effects. This case is more highly complicated. Although the free Hamiltonian $\mathscr{K}_0$ has the same unchanged form (64), the relation (88) is, in general, no longer valid. The present considerations refer to, e.g., a system of the electrons of a solid at interaction with a radiation field, a system of electrons of a many-electron atom, or a system of many atoms interacting with a field. On performing the canonical transformation as for the case of one electron we obtain, instead of (91) and (92), the following expressions:

$$\mathscr{K}_{\breve{0}} = \sum_{\kappa} \omega_{\kappa} a^{\dagger}_{\kappa} a_{\kappa} + \sum_{\mu} E_{\mu} b^{\dagger}_{\mu} b_{\mu} \,, \tag{111}$$

$$\mathscr{K}_{\breve{m}} = \sum_{\alpha_1,\alpha_2,\ldots,\alpha_m} \sum_{r=0}^{[m/2]} \sum_{\substack{\mu_1,\mu_2,\ldots,\mu_{m-r} \\ \nu_1\nu_2,\ldots,\nu_{m-r}}} K^{\alpha_1\alpha_2\cdots\alpha_m}_{\kappa_1\kappa_2\cdots\kappa_m,\mu_1\nu_1\mu_2\nu_2\cdots\mu_{m-r}\nu_{m-r}} a^{\alpha_1}_{\kappa_1} a^{\alpha_2}_{\kappa_2} \cdots a^{\alpha_m}_{\kappa_m} b^{\dagger}_{\mu_1} b_{\nu_1} b^{\dagger}_{\mu_2} b_{\nu_2} \cdots b^{\dagger}_{\mu_{m-r}} b_{\nu_{m-r}} \,. \tag{112}$$

The occurrence of more than two electron operators in a single term is due to the fact that, here, Eq. (89) no longer holds.

On omission of the $\vec{\mathrm{A}}^{2}$ interaction term, each of the $\mathscr{K}_m$'s will contain only terms with $r=0$. One notes that the principle previously applied to the one-electron case is still successfully operative here. The above form of the Hamiltonian is unsatisfactory now for the same reasons as it was then: it lacks a sufficiently clear physical interpretation. The difficulty is removed by performing a normal ordering of the operators. We obtain

$$\mathscr{K}_{\breve{0}} = \sum_{\kappa} \omega_{\kappa} a^{\dagger}_{\kappa} a_{\kappa} + \sum_{\mu} (E_{\mu} + \delta E_{\mu}) b^{\dagger}_{\mu} b_{\mu} \,, \tag{113}$$

$$\mathscr{K}_{\breve{m}} = \sum_{n=0}^{m} \sum_{\kappa_1,\ldots,\kappa_m} \sum_{r=1}^{m} \sum_{\substack{\mu_1,\ldots,\mu_r \\ \nu_1,\ldots,\nu_r}} \chi^{n}_{\kappa_1\cdots\kappa_m,\mu_1\cdots\mu_r\nu_1\cdots\nu_r} a^{\dagger}_{\kappa_1} \cdots a^{\dagger}_{\kappa_n} a_{\kappa_{n+1}} \cdots a_{\kappa_m} b^{\dagger}_{\mu_1} \cdots b^{\dagger}_{\mu_r} b_{\nu_1} \cdots b_{\nu_r} \,, \quad m \geq 1 \tag{114}$$

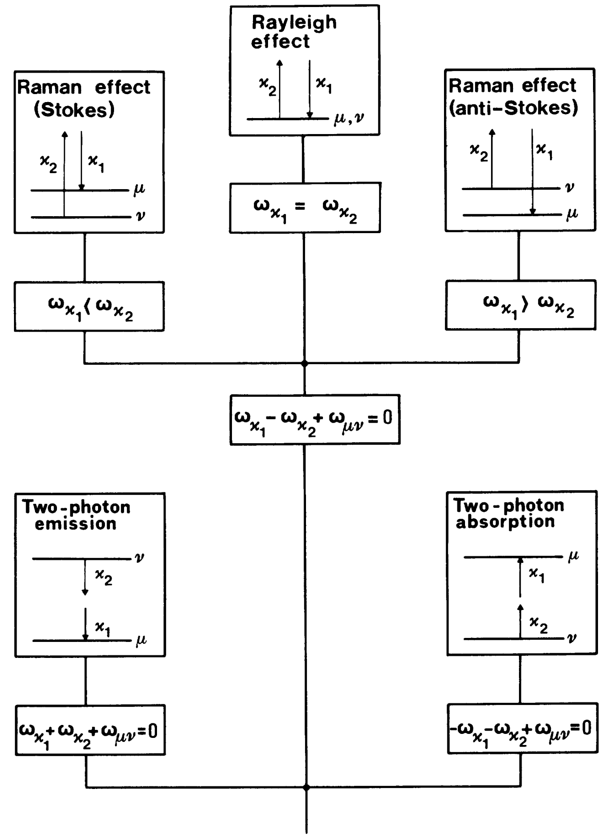


FIG. 4. Effects described by $\chi^{n}_{\kappa_1\kappa_2,\mu\nu}$.

where the $\chi^{n}_{\kappa_1\cdots\kappa_m,\mu_1\cdots\mu_r\nu_1\cdots\nu_r}$ are nonzero constants only if

$$\omega_{\kappa_1}+\cdots+\omega_{\kappa_n}-\omega_{\kappa_{n+1}}-\cdots-\omega_{\kappa_m}$$
$$+E_{\mu_1}+\cdots+E_{\mu_r}-E_{\nu_1}-\cdots-E_{\nu_r}=0 \quad (115)$$

and their explicit form results from the application of Wick's theorem. Compared to the previous case, a novel nontrivial element emerges here: terms occur, each containing $r\geq 2$ electron creation and annihilation operators. With regard to the Pauli principle they differ from zero only provided that no two creation operators or annihilation operators of electrons concern the same state. Thus, these terms describe cooperative pairwise ($r=2$), triple ($r=3$), etc., interaction of the electrons with the field. That is to say that the electrons do not interact with the field independently (individually) but in complexes of two, three, or more electrons. As an example, we shall consider the effect of cooperative two-photon absorption, given by terms of the form

$$\chi^{0}_{\kappa_1\kappa_2,\mu_1\mu_2\nu_1\nu_2}a_{\kappa_1}a_{\kappa_2}b^{\dagger}_{\mu_1}b^{\dagger}_{\mu_2}b_{\nu_1}b_{\nu_2} .$$

The electrons go over from the states ν_1 and ν_2 to the states μ_1 and μ_2, and simultaneously two photons with the frequencies ω_{κ_1} and ω_{κ_2} are absorbed. At the same time, the condition

$$-\omega_{\kappa_1}-\omega_{\kappa_2}+E_{\mu_1}+E_{\mu_2}-E_{\nu_1}-E_{\nu_2}=0$$

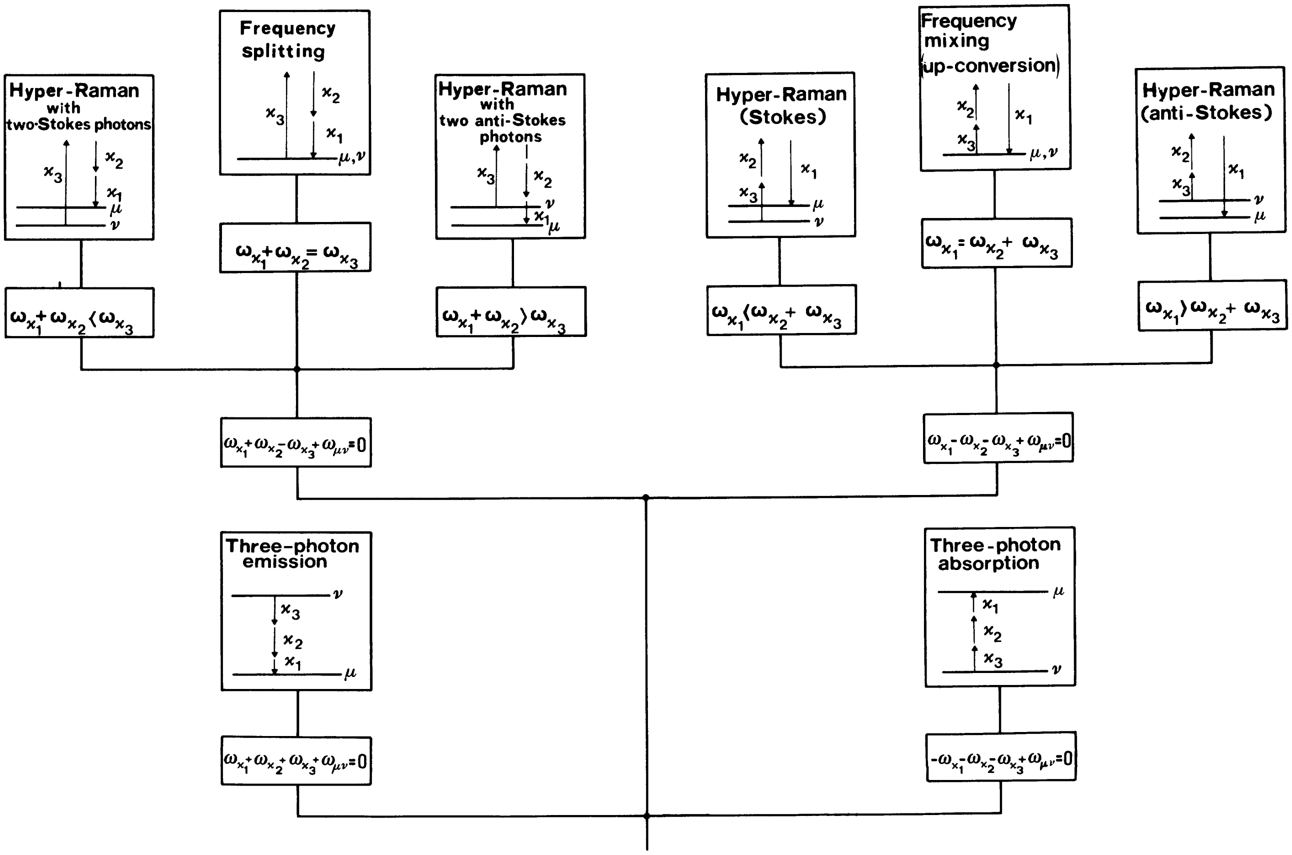


FIG. 5. Effects described by $\chi^{n}_{\kappa_1\kappa_2\kappa_3,\mu\nu}$.

is fulfilled. The essence of the phenomenon resides in the circumstance that neither of the two electrons could separately absorb a radiation field quantum of one of the above given frequencies since the conditions

$$\omega_{\mu_1\nu_1},\omega_{\mu_1\nu_2},\omega_{\mu_2\nu_1},\omega_{\mu_2\nu_2}\neq\omega_{\kappa_1},\omega_{\kappa_2}$$

are fulfilled; however, they can do so jointly.

Cooperative processes represent a highly interesting nonlinear aspect of interaction of radiation and matter. Although they doubtlessly deserve wider consideration, we refrain from their further discussion here so as not to exceed the scope of the present paper excessively.

Many electrons; cooperative effects omitted. We select this case for closer consideration as that on which we primarily concentrate our attention. As assumed by us, the atoms are one-electron ones. Hence, in the particular case of a single atom, we shall be dealing with the previously discussed one-electron system. Before going any further in our analysis we exclude the index q, labeling the atoms, from the set of electron indices μ. Neglecting cooperative interactions of electrons and the radiation field, we readily obtain from formulas (100) and (101)

$$\breve{\mathcal{K}}_0=\sum_{\kappa}\omega_\kappa a_\kappa^\dagger a_\kappa+\sum_{q\mu}(E_{\mu q}+\delta E_{\mu q})b_{\mu q}^\dagger b_{\mu q}\ , \tag{116}$$

$$\breve{\mathcal{K}}_m=\sum_{n=0}^{m}\sum_{\kappa_1,\dots,\kappa_m}\sum_{q,\mu,\nu}\chi^n_{\kappa_1\cdots\kappa_m,\mu\nu q}\times a^\dagger_{\kappa_1}\cdots a^\dagger_{\kappa_n}a_{\kappa_{n+1}}\cdots a_{\kappa_m}\times b^\dagger_{\mu q}b_{\nu q},\quad m\geq 1\ . \tag{117}$$

For a single atom in the system of reference attached to its nucleus, the form of the coupling constants in the above formulas is identical with that previously derived for the single electron [Eqs. (97) and (98)]. In general, the constants $\chi^n_{\kappa_1\cdots\kappa_m,\mu\nu q}$ can be represented in the form of a product of the constant $\chi^n_{\kappa_1\cdots\kappa_m,\mu\nu}$ and a term dependent on q. For example, on the assumption that the atoms are disposed within a box of volume V and accordingly taking mode functions in the form

$$\vec{u}_\kappa(\vec{x})=V^{-1/2}\vec{\epsilon}_\kappa e^{i\vec{k}_\kappa\cdot\vec{x}},\quad |\vec{\epsilon}_\kappa|=1 \tag{118}$$

the coupling constants take the form

$$\chi^n_{\kappa_1\cdots\kappa_m,\mu\nu q}=\exp[-i\vec{\Theta}_q\cdot(\vec{k}_{\kappa_1}+\cdots+\vec{k}_{\kappa_n}-\vec{k}_{\kappa_{n+1}}-\cdots-\vec{k}_{\kappa_m})]\chi^n_{\kappa_1\cdots\kappa_m,\mu\nu}\ . \tag{119}$$

The time-dependent vectors $\vec{\Theta}_q=\vec{\Theta}_q(t)$ correspond to positions of the atoms. If one assumes that the atoms move uniformly and rectilinearly, the vectors $\vec{\Theta}_q$ are determined by the positions $\vec{r}_q$ and velocities $\vec{v}_q$ at the initial moment of the time $t=0$:

$$\vec{\Theta}_q=\vec{r}_q+\vec{v}_q t\ . \tag{120}$$

V. APPROXIMATION OF TWO-LEVEL ATOMS

The approximation of two-level atoms consists in dealing with the atoms as if they possessed but two levels. The problem of whether a given physical system can be dealt with in this way is by no means simple and we shall not discuss it here, but refer to the book by Allen and Eberly[10] and the paper by Takatsuji.[11]

Here, we shall carry out the approximation by projecting the Hilbert spaces of the electrons of the various atoms onto spaces spanned on two selected electronic eigenstates. We label the latter 1 and 2, so as to have $E_2(q)\geq E_1(q)$. At the same time, we subject all operators, including the Hamiltonian, to the transformation

$$X\to\tilde{X}\equiv QXQ\ , \tag{121}$$

where, for the case of N atoms,

$$Q=I\otimes(|1\rangle\langle 1|+|2\rangle\langle 2|)_{q=1}\otimes(|1\rangle\langle 1|+|2\rangle\langle 2|)_{q=2}\otimes\cdots\otimes(|1\rangle\langle 1|+|2\rangle\langle 2|_{\dot{q}=N})\ . \tag{122}$$

It should be stressed that the sequence [apply the canonical transformation (13) and then perform the projection (121)] cannot be inverted. Even a superficial analysis shows that this—the sequence applied in our paper—leads to physically meaningful results: no effects "get lost."

In this section we consider atoms for solely traditional reasons. Obviously, nothing stands in the way of applying the two-level approximation to other systems as well.

Let us consider the transformed Hamiltonian of the form of Eqs. (116) and (117). The two-level approximation then leads directly to the formulas:

$$\breve{\mathcal{K}}\to\tilde{\mathcal{K}}=\sum_{m=0}^{\infty}\tilde{\mathcal{K}}_m\ , \tag{123}$$

where

$$\tilde{\mathcal{K}}_0=\sum_{\kappa}\omega_\kappa a^\dagger_\kappa a_\kappa+\sum_{q}[(E_{1q}+\delta E_{1q})b^\dagger_{1q}b_{1q}+(E_{2q}+\delta E_{2q})b^\dagger_{2q}b_{2q}]\ , \tag{124}$$

$$\tilde{\mathcal{K}}_m=\begin{cases}\displaystyle\sum_{\kappa}\sum_{q}(\chi^0_{\kappa,21q}a_\kappa b^\dagger_{2q}b_{1q}+\chi^1_{\kappa,12q}a^\dagger_\kappa b^\dagger_{1q}b_{2q}),\quad m=1\\ \displaystyle\sum_{n=0}^{m=1}\sum_{\kappa_1,\dots,\kappa_m}\sum_{q}(\chi^n_{\kappa_1\cdots\kappa_m,21q}a^\dagger_{\kappa_1}\cdots a^\dagger_{\kappa_n}a_{\kappa_{n+1}}\cdots a_{\kappa_m}b^\dagger_{2q}b_{1q}+\text{H.c.})\\ \displaystyle+\sum_{n=1}^{m-1}\sum_{\kappa_1,\dots,\kappa_m}\sum_{q}(\chi^n_{\kappa_1\cdots\kappa_m,22q}b^\dagger_{2q}b_{2q}+\chi^n_{\kappa_1\cdots\kappa_m,11q}b^\dagger_{1q}b_{1q})a^\dagger_{\kappa_1}\cdots a^\dagger_{\kappa_n}a_{\kappa_{n+1}}\cdots a_{\kappa_m},\quad m\geq 2\ .\end{cases} \tag{125}$$

Consider operators defined as follows:

$$\sigma_{3q} \equiv \tfrac{1}{2}(b_{2q}^{\dagger}b_{2q} - b_{1q}^{\dagger}b_{1q}) \,, \qquad \sigma_q^{+} \equiv b_{2q}^{\dagger}b_{1q}, \quad \sigma_q^{-} \equiv b_{1q}^{\dagger}b_{2q} \,. \tag{126}$$

They fulfill the usual spin commutation rules

$$[\sigma_q^{+},\sigma_{q'}^{-}] = 2\delta_{qq'}\sigma_{3q} \,, \qquad [\sigma_{3q},\sigma_{q'}^{\pm}] = \pm\delta_{qq'}\sigma_q^{\pm} \,. \tag{127}$$

The value of the qth spin is

$$\sigma_q \equiv \tfrac{1}{2}(b_{2q}^{\dagger}b_{2q} + b_{1q}^{\dagger}b_{1q}) \,. \tag{128}$$

This operator commutes with the Hamiltonian (129) and its eigenvalues are $\frac{1}{2}$ and 0. Strictly, the time evolution of the operator in question is described by the total Hamiltonian [Eqs. (116) and (117)]; for a given q, the spin can appear ($\frac{1}{2}$) and vanish (0) with time. The approximation applied by us has eliminated the possibility of time variation of σ_q. In this way we ignore the possibility of the atoms performing transitions between 1 or 2 and the other levels. Maybe the two-level approximation could be enhanced by postulating a time dependence of the value of σ_q. We would then be dealing with a system of stochastically appearing and disappearing spins $\frac{1}{2}$, i.e., with a sort of "spin glass," interacting nonlinearly with the radiation field. Here, however, we shall restrict ourselves to the case of frozen spins. The Hamiltonian, described by the formulas (124) and (125), can be expressed in terms of the spin operators as follows:

$$\tilde{\mathscr{K}}_0 = \sum_{\kappa} a_{\kappa}^{\dagger}a_{\kappa} + \sum_{q} [\Omega_q\sigma_q + (\epsilon_q + \delta\epsilon_q)\sigma_{3q}] \,, \tag{129}$$

$$\tilde{\mathscr{K}}_m = \begin{cases} \displaystyle\sum_{\kappa}\sum_{q} (\chi^{0}_{\kappa,21q}a_{\kappa}\sigma_q^{+} + \chi^{1}_{\kappa,12q}a_{\kappa}^{\dagger}\sigma_q^{-}), \quad m=1 \\ \displaystyle\sum_{n=0}^{m-1}\sum_{\kappa_1,\ldots,\kappa_m}\sum_{q} (\chi^{n}_{\kappa_1\cdots\kappa_m,21q}a_{\kappa_1}^{\dagger}\cdots a_{\kappa_n}^{\dagger}a_{\kappa_{n+1}}\cdots a_{\kappa_m}\sigma_q^{+} + \text{H.c.}) \\ \displaystyle+\sum_{n=1}^{m-1}\sum_{\kappa_1,\ldots,\kappa_m}\sum_{q} [\chi^{n}_{\kappa_1\cdots\kappa_m,22q}(\sigma_q + \sigma_{3q}) + \chi^{n}_{\kappa_1\cdots\kappa_m,11q}(\sigma_q - \sigma_{3q})]a_{\kappa_1}^{\dagger}\cdots a_{\kappa_n}^{\dagger}a_{\kappa_{n+1}}\cdots a_{\kappa_m}, \quad m \geq 2 \end{cases} \tag{130}$$

where we have made use of the notation

$$\epsilon_q \equiv E_{2q} - E_{1q}, \quad \delta\epsilon_q \equiv \delta E_{2q} - \delta E_{1q} \,, \qquad \Omega_q \equiv E_{2q} + \delta E_{2q} + E_{1q} + \delta E_{1q} \,. \tag{131}$$

Henceforth q will label only those atoms whose spins are equal to $\frac{1}{2}$.

In many cases it is highly convenient to use total spin operators, often referred to as "collective dipole operators." We proceed to give an example of this. First, we assume homogeneous line broadening, i.e., we assume that E_{2q}, δE_{2q}, E_{1q}, and δE_{1q} do not depend on q. At the same time we introduce the notation

$$\epsilon \equiv \epsilon_q, \quad \delta\epsilon \equiv \delta\epsilon_q, \quad \Omega \equiv \Omega_q \,. \tag{132}$$

If the dimensions of the atomic system are small compared to the lengths of the waves participating in the mode interaction, then the dependence on q can be neglected in the coupling constants:

$$\chi^{n}_{\kappa_1\cdots\kappa_m,\mu\nu} \equiv \chi^{n}_{\kappa_1\cdots\kappa_m,\mu\nu q} \,. \tag{133}$$

Now, defining operators

$$S_3 \equiv \sum_{q} \sigma_{3q} \,, \qquad S^{\pm} \equiv \sum_{q} \sigma_q^{\pm} \,, \qquad S \equiv \sum_{q} \sigma_q \,, \tag{134}$$

having the meaning of total spin operators,

$$[S^{+},S^{-}] = 2S_3, \quad [S_3,S^{\pm}] = \pm S^{\pm} \,, \tag{135}$$

we obtain the following expressions:

$$\tilde{\mathscr{K}}_0 = \sum_{\kappa} \omega_{\kappa}a_{\kappa}^{\dagger}a_{\kappa} + \Omega S + (\epsilon + \delta\epsilon)S_3 \,, \tag{136}$$

$$\tilde{\mathscr{K}}_m = \begin{cases} \displaystyle\sum_{\kappa} (\chi^{0}_{\kappa,21}a_{\kappa}S^{+} + \chi^{1}_{\kappa,12}a_{\kappa}^{\dagger}S^{-}), \quad m=1 \\ \displaystyle\sum_{n=0}^{m-1}\sum_{\kappa_1,\ldots,\kappa_m} (\chi^{n}_{\kappa_1\cdots\kappa_m,21}a_{\kappa_1}^{\dagger}\cdots a_{\kappa_n}^{\dagger}a_{\kappa_{n+1}}\cdots a_{\kappa_m}S^{+} + \text{H.c}) \\ \displaystyle+\sum_{n=1}^{m-1}\sum_{\kappa_1,\ldots,\kappa_m} [\chi^{n}_{\kappa_1\cdots\kappa_m,22}(S + S_3) + \chi^{n}_{\kappa_1\cdots\kappa_m,11}(S - S_3)]a_{\kappa_1}^{\dagger}\cdots a_{\kappa_n}^{\dagger}a_{\kappa_{n+1}}\cdots a_{\kappa_m}, \quad m \geq 2 \,. \end{cases} \tag{137}$$

The total spin S has to satisfy the relation $S \leq N/2$.

If the dimensions of the system are large it is, in general, no longer sufficient to introduce a single operator of total spin. A set of such operators has to be introduced (see, e.g., Ref. 12). Thus, e.g., for a system of atoms in a box of volume V we define (see Ref. 13)

$$S_3(\vec{k}) \equiv \sum_q e^{i\vec{\Theta}_q \cdot \vec{k}} \sigma_{3q}, \quad S^{\pm}(\vec{k}) \equiv \sum_q e^{\pm i\vec{\Theta}_q \cdot \vec{k}} \sigma_q^{\pm}, \quad S \equiv \sum_q \sigma_q \ . \tag{138}$$

Moreover, we shall also apply the notation

$$S_3 \equiv S_3(\vec{0}) \ . \tag{139}$$

The above operators fulfill the following commutation rules:

$$[S^+(\vec{k}), S^-(\vec{k}\,')] = 2S_3(\vec{k}-\vec{k}\,'), \quad [S_3(\vec{k}), S^{\pm}(\vec{k}\,')] = \pm S^{\pm}(\vec{k}\pm\vec{k}\,') \ . \tag{140}$$

Since the coupling constants are given by the formulas (119) we obtain

$$\tilde{\mathcal{H}}_0 = \sum_\kappa \omega_\kappa a_\kappa^\dagger a_\kappa + \Omega S + (\epsilon+\delta\epsilon) S_3 \ , \tag{141}$$

$$\tilde{\mathcal{H}}_m = \begin{cases} \sum_\kappa [\chi^0_{\kappa,21} a_\kappa S^+(\vec{k}_\kappa) + \chi^1_{\kappa,12} a_\kappa^\dagger S^-(\vec{k}_\kappa)], \quad m=1 \\ \sum_{n=0}^{m-1} \sum_{\kappa_1,\ldots,\kappa_m} [\chi^n_{\kappa_1\cdots\kappa_m,21} a^\dagger_{\kappa_1} \cdots a^\dagger_{\kappa_n} a_{\kappa_{n+1}} \cdots a_{\kappa_m} S^+(\vec{k}_{\kappa_1} + \cdots + \vec{k}_{\kappa_n} - \vec{k}_{\kappa_{n+1}} - \cdots - \vec{k}_{\kappa_m}) + \mathrm{H.c.}] \\ + \sum_{n=1}^{m-1} \sum_{\kappa_1,\ldots,\kappa_m} [\chi^n_{\kappa_1\cdots\kappa_m,22}(S+S_3) + \chi^n_{\kappa_1\cdots\kappa_m,11}(S-S_3)] a^\dagger_{\kappa_1} \cdots a^\dagger_{\kappa_n} a_{\kappa_{n+1}} \cdots a_{\kappa_m}, \quad m \geq 2 \ . \end{cases} \tag{142}$$

Here, we have come to the crucial point of our considerations. The Hamiltonian $\tilde{\mathcal{H}}$ consists, in general, of an infinite number of terms. It is well known, however, that in the description of a given physical system not all of them are equally essential. Some are zero, whereas others may well be neglected. Regrettably, there are as yet no general criteria for a good approximation, i.e., for deciding as to which effects can be dealt with independently and which have to be considered jointly. Hence, essentially, each physical system has to be analyzed individually and the decision lies with experiment. In quantum optics it is most usual to apply a Hamiltonian of the form of Eqs. (136) and (137) in which all coupling constants, with the exception of several, are omitted. Two types of Hamiltonians are thus obtained. The one describes processes in which the state of the atom changes. A typical example is the Hamiltonian

$$\tilde{\mathcal{H}} = \sum_\kappa \omega_\kappa a_\kappa^\dagger a_\kappa + \Omega S + (\epsilon+\delta\epsilon) S_3 + (\lambda^* a^\dagger_{\kappa_1} \cdots a^\dagger_{\kappa_n} a_{\kappa_{n+1}} \cdots a_{\kappa_m} S^+ + \mathrm{H.c.}) \ , \tag{143}$$

where (for atoms in a box)

$$S^{\pm} \equiv S^{\pm}(\vec{k}_{\kappa_1} + \cdots + \vec{k}_{\kappa_n} - \vec{k}_{\kappa_{n+1}} - \cdots - \vec{k}_{\kappa_m}) \ ,$$

$$\lambda^* \equiv (n!)(m!) \chi^n_{\kappa_1\cdots\kappa_m,21} \ . \tag{144}$$

In the other type of process the atoms are passive; here, a typical example of the Hamiltonian is as follows:

$$\tilde{\mathcal{H}} = \sum_\kappa \omega_\kappa a_\kappa^\dagger a_\kappa + \Omega S + (\epsilon+\delta\epsilon) S_3 + \{[\lambda_2(S+S_3) + \lambda_1(S-S_3)] \times a^\dagger_{\kappa_1} \cdots a^\dagger_{\kappa_n} a_{\kappa_{n+1}} \cdots a_{\kappa_m} + \mathrm{H.c.}\}, \quad m \geq 2 \tag{145}$$

where

$$\lambda_\mu \equiv (n!)(m!) \chi^n_{\kappa_1\cdots\kappa_m,\mu\mu}, \quad \mu = 1,2 \ . \tag{146}$$

In this case, averaging can be performed over the spin states and constants neglected as irrelevant to the dynamics of the system. This leads to the Hamiltonian

$$\tilde{\mathcal{H}} = \sum_\kappa \omega_\kappa a_\kappa^\dagger a_\kappa + (\lambda a^\dagger_{\kappa_1} \cdots a^\dagger_{\kappa_n} a_{\kappa_{n+1}} \cdots a_{\kappa_m} + \mathrm{H.c.}) \ . \tag{147}$$

Obviously, here the summation is carried only over those $\kappa_1, \ldots, \kappa_m$ which are distinct from one another.

We proceed to applications. First, we shall derive effective Hamiltonians in the two-level approximation for the case of many-photon resonance.

Many-photon resonance. We start by considering a system of one atom and one mode of the radiation field. When possible, we shall omit the index q labeling the atom and κ labeling the field mode. Let the (usual) p-photon resonance condition be fulfilled for two states of the atom:

$$\omega_{21} \equiv E_2 - E_1 = p\omega \ . \tag{148}$$

Through (124) and (125) we easily obtain the following expression:

$$\tilde{\mathcal{H}} = \omega a^\dagger a + \mathcal{E}_2(a^\dagger a) b_2^\dagger b_2 + \mathcal{E}_1(a^\dagger a) b_1^\dagger b_1 + [\lambda_p(a^\dagger a) a^p b_2^\dagger b_1 + \mathrm{H.c.}] \ , \tag{149}$$

where

$$\mathscr{E}_\mu(a^\dagger a)=E_\mu+\delta E_\mu+\sum_{r=1}^{\infty}\chi^{r}_{(2r)\kappa,\mu\mu}a^\dagger a(a^\dagger a-1)\cdots(a^\dagger a-r+1),\quad \mu=1,2$$
$$\lambda_p(a^\dagger a)=\sum_{r=0}^{\infty}\chi^{r}_{(p+r)\kappa,21}a^\dagger a(a^\dagger a-1)\cdots(a^\dagger a-r+1) \tag{150}$$

and use was made of the identity

$$(a^\dagger)^r a^r=a^\dagger a(a^\dagger a-1)\cdots(a^\dagger a-r+1)\,, \tag{151}$$

and $(2r)\kappa$ and $(p+r)\kappa$ are shorthand of $2r$ and $p+r$ identical subscripts κ. Above, an essential difference by comparison with the usually applied Hamiltonians resides in the fact that, beside the usual shift of the levels, there appears a shift dependent on the number of photons $n=a^\dagger a$ and, moreover, the coupling constants are also functions of n. On performing an approximation consisting in the omission of all terms but those of lowest order in the expansions (150), i.e., in the assumption of

$$\mathscr{E}_\mu(a^\dagger a)\approx E_\mu+\delta E_\mu,\quad \lambda_p(a^\dagger a)\approx\chi^{0}_{(p)\kappa,21}\equiv\lambda_p\,, \tag{152}$$

we obtain the Hamiltonian

$$\tilde{\mathscr{H}}=\omega a^\dagger a+(E_2+\delta E_2)b_2^\dagger b_2+(E_1+\delta E_1)b_1^\dagger b_1+\lambda_p a^p b_2^\dagger b_1+\lambda_p^*(a^\dagger)^p b_1^\dagger b_2\,. \tag{153}$$

If the transition of the electron occurs by way of virtual levels only, i.e., if no intermediate resonance level is present, the explicit form of the constants λ_p can be given easily (on neglecting the $\vec{\mathrm{A}}^2$-type interaction):

$$\lambda_p=\sum_{\mu_1,\mu_2,\ldots,\mu_{p-1}}\left|\left(\cdots\left[\left(\begin{matrix}-\kappa\\2\mu_1\end{matrix}\right]^{\{n,\omega\}}\begin{matrix}-\kappa\\\mu_1\mu_2\end{matrix}\right]^{\{n,\omega\}}\cdots\begin{matrix}-\kappa\\\mu_{p-2}\mu_{p-1}\end{matrix}\right)^{\{n,\omega\}}\begin{matrix}-\kappa\\\mu_{p-1}1\end{matrix}\right|^{\{d\}}$$
$$=\left[\sum_{\mu_1,\mu_2,\ldots,\mu_{p-1}}\left|\left(\cdots\left[\left(\begin{matrix}+\kappa\\1\mu_1\end{matrix}\right]^{\{n,\omega\}}\begin{matrix}+\kappa\\\mu_1\mu_2\end{matrix}\right]^{\{n,\omega\}}\cdots\begin{matrix}+\kappa\\\mu_{p-2}\mu_{p-1}\end{matrix}\right)^{\{,\omega\}}\begin{matrix}+\kappa\\\mu_{p-2}2\end{matrix}\right|^{\{d\}}\right]^*\,. \tag{154}$$

On rewriting these formulas in the equivalent form

$$\lambda_p=\sum_{\mu_1,\mu_2,\ldots,\mu_{p-1}}\frac{\left|\begin{matrix}-\kappa\\2\mu_1\end{matrix}\right|^{\{d\}}\left|\begin{matrix}-\kappa\\\mu_1\mu_2\end{matrix}\right|^{\{d\}}\cdots\left|\begin{matrix}-\kappa\\\mu_{p-2}\mu_{p-1}\end{matrix}\right|^{\{d\}}\left|\begin{matrix}-\kappa\\\mu_{p-1}1\end{matrix}\right|^{\{d\}}}{(\omega_{2\mu_1}-\omega)(\omega_{2\mu_2}-2\omega)\cdots[\omega_{2\mu_{p-1}}-(p-1)\omega]}$$
$$=\left[\sum_{\mu_1,\mu_2,\ldots,\mu_{p-1}}\frac{\left|\begin{matrix}+\kappa\\1\mu_1\end{matrix}\right|^{\{d\}}\left|\begin{matrix}+\kappa\\\mu_1\mu_2\end{matrix}\right|^{\{d\}}\cdots\left|\begin{matrix}+\kappa\\\mu_{p-2}\mu_{p-1}\end{matrix}\right|^{\{d\}}\left|\begin{matrix}+\kappa\\\mu_{p-1}2\end{matrix}\right|^{\{d\}}}{(\omega_{1\mu_1}+\omega)(\omega_{1\mu_2}+2\omega)\cdots[\omega_{1\mu_{p-1}}+(p-1)\omega]}\right]^*, \tag{155}$$

one immediately sees that they are in agreement with ones derived earlier.[14]

The preceding formulas dealt with the case of a single atom. For a system of many atoms, albeit neglecting cooperative interactions, we obtain, similarly,

$$\tilde{\mathscr{H}}=\omega a^\dagger a+\sum_q[\mathscr{E}_{2q}(a^\dagger a)b_{2q}^\dagger b_{2q}+\mathscr{E}_{1q}(a^\dagger a)b_{1q}^\dagger b_{1q}]+[\lambda_{pq}(a^\dagger a)a^p b_{2q}^\dagger b_{1q}+\text{H.c.}]\,, \tag{156}$$

where

$$\mathscr{E}_{\mu q}(a^\dagger a)=E_{\mu q}+\delta E_{\mu q}+\sum_{r=0}^{\infty}\chi^{r}_{(2r)\kappa,\mu\mu q}a^\dagger a(a^\dagger a-1)\cdots(a^\dagger a-p+1),\quad \mu=1,2$$
$$\lambda_{pq}(a^\dagger a)=\sum_{r=0}^{\infty}\chi^{r}_{(p+r)\kappa,21q}a^\dagger a(a^\dagger a-1)\cdots(a^\dagger a-r+1)\,. \tag{157}$$

On making the approximation

$$\mathscr{E}_{\mu q}(a^\dagger a)\approx E_{\mu q}+\delta E_{\mu q},\quad \lambda_{pq}(a^\dagger a)\approx\chi^{0}_{(p)\kappa,21}\equiv\lambda_p\,, \tag{158}$$

we obtain the Hamiltonian

$$\tilde{\mathscr{H}}=\omega a^\dagger a+\sum_q[(E_{2q}+\delta E_{2q})b_{2q}^\dagger b_{2q}+(E_{1q}+\delta E_{1q})b_{1q}^\dagger b_{1q}]+\lambda_{pq}a^p b_{2q}^\dagger b_{1q}+\lambda_{pq}^*(a^\dagger)^p b_{1q}^\dagger b_{2q}\,. \tag{159}$$

The coupling constants λ_{pq}, calculated on the same assumptions as those made for the single atom, are of the form

$$\lambda_{pq} = \sum_{\mu_1,\mu_2,\ldots,\mu_{p-1}} \Bigg| \Bigg[\cdots \Bigg[\Bigg[\begin{pmatrix} -\kappa \\ 2\mu_1 q \end{pmatrix}^{\{n,\omega\}} \begin{matrix} -\kappa \\ \mu_1\mu_2 q \end{matrix} \Bigg]^{\{n,\omega\}} \cdots \begin{matrix} -\kappa \\ \mu_{p-2}\mu_{p-1} q \end{matrix} \Bigg]^{\{n,\omega\}} \begin{matrix} -\kappa \\ \mu_{p-1} 1 q \end{matrix} \Bigg|^{\{d\}}$$

$$= \Bigg[\sum_{\mu_1,\mu_2,\ldots,\mu_{p-1}} \Bigg| \Bigg[\cdots \Bigg[\Bigg[\begin{pmatrix} +\kappa \\ 1\mu_1 q \end{pmatrix}^{\{n,\omega\}} \begin{matrix} +\kappa \\ \mu_1\mu_2 q \end{matrix} \Bigg]^{\{n,\omega\}} \cdots \begin{matrix} +\kappa \\ \mu_{p-2}\mu_{p-1} q \end{matrix} \Bigg]^{\{n,\omega\}} \begin{matrix} +\kappa \\ \mu_{p-1} 2 q \end{matrix} \Bigg|^{\{d\}} \Bigg]^{*} . \tag{160}$$

In the above system we can go over to a description in terms of spin operators. We obtain

$$\mathscr{K}^{\sim} = \omega a^{\dagger} a + \sum_{q} [\Omega_q \sigma_q + (\epsilon_q + \delta\epsilon_q)\sigma_{3q} + \lambda_{pq} a^{p} \sigma_q^{+} + \lambda_{pq}^{*} (a^{\dagger})^{p} \sigma_q^{-}] . \tag{161}$$

For a system of atoms with a homogeneously broadened line, the preceding Hamiltonian can be reduced to the form

$$\mathscr{K}^{\sim\prime} = \omega a^{\dagger} a + \Omega S + (\epsilon + \delta\epsilon) S_3 + \lambda_p a^{p} S^{+} + \lambda_p^{*} (a^{\dagger})^{p} S^{-} , \tag{162}$$

which is a particular case of formula (143). In this Hamiltonian, particularly if $p=1$, we recognize the well-known Hamiltonian of Dicke.

Hamiltonians of the form (143) and (144) are well adapted to the visualization of the highly interesting difference between "active" and "passive" processes with regard to an atomic system. We shall discuss the matter further on.

Supereffects (in the meaning of Dicke). Consider a system of many atoms ($N \gg 1$) at interaction with a radiation field. We now assume the following m-photon processes: (A) absorption of m-l photons (indices $\kappa_{l+1}, \kappa_{l+2}, \ldots, \kappa_m$) and emission of l photons (indices $\kappa_1, \kappa_2, \ldots, \kappa_l$) with simultaneous transition of the atomic system from the upper to the lower state; (B) the inverse process, i.e., absorption of l photons (indices $\kappa_1, \kappa_2, \ldots, \kappa_l$) and emission of m-l photons (indices $\kappa_{l+1}, \kappa_{l+2}, \ldots, \kappa_m$) with simultaneous transition of the atomic system from the lower to the upper level; (C) absorption of m-l photons (indices $\kappa_{l+1}, \kappa_{l+2}, \ldots, \kappa_m$) and emission of l photons (indices $\kappa_1, \kappa_2, \ldots, \kappa_l$) with no change in the energy of the atomic system.

Applying Fermi's golden rule, we calculate the probabilities per unit time of the occurrence of the above processes as follows:

$$P_{|i\rangle \to |f\rangle} = 2\pi \, | \langle f \, | \mathscr{H} | \, i \rangle |^2 \rho(| f \rangle) , \tag{163}$$

where $| i \rangle$ and $| f \rangle$ denote the initial and final states of the system as a whole, respectively. The Hamiltonian $\mathscr{H}$ can be expressed by the formula

$$\mathscr{H} = \mathscr{U}^{\dagger} \mathscr{K} \mathscr{U} , \tag{164}$$

where $\mathscr{K}$ is the diagonal operator (13). We derive the following exact result:

$$\begin{aligned} P_{|i\rangle \to |f\rangle} &= 2\pi \, | \langle f \, | \, \mathscr{U}^{\dagger} \mathscr{K} \mathscr{U} \, | \, i \rangle |^2 \rho(| f \rangle) \\ &= 2\pi \, | \langle f \, | \, \mathscr{U}^{\dagger} Q \mathscr{K} Q \mathscr{U} \, | \, i \rangle |^2 \rho(| f \rangle) \\ &= 2\pi \, | \langle f \, | \, \mathscr{U}^{\dagger} Q \mathscr{K}^{\sim} Q \mathscr{U} \, | \, i \rangle |^2 \rho(| f \rangle . \end{aligned} \tag{165}$$

Considering the probabilities per unit time, it is thus not necessary to take into account the whole Hamiltonian $\mathscr{K}$—it can be replaced by $\mathscr{K}^{\sim}$. In particular, the processes (A), (B), and (C) are described by terms of the operator $\mathscr{K}^{\sim}$ as follows:

$$\text{(A)} \quad \lambda a_{\kappa_1}^{\dagger} a_{\kappa_2}^{\dagger} \cdots a_{\kappa_l}^{\dagger} a_{\kappa_{l+1}} \cdots a_{\kappa_m} S^{-} , \tag{166}$$

$$\text{(B)} \quad \lambda^{*} a_{\kappa_m}^{\dagger} a_{\kappa_{m-1}}^{\dagger} \cdots a_{\kappa_{l+1}}^{\dagger} a_{\kappa_l} \cdots a_{\kappa_1} S^{+} , \tag{167}$$

$$\text{(C)} \quad [\lambda_2 (S + S_3) + \lambda_1 (S - S_3)] \times a_{\kappa_1}^{\dagger} a_{\kappa_2}^{\dagger} \cdots a_{\kappa_l}^{\dagger} a_{\kappa_{l+1}} \cdots a_{\kappa_m} , \tag{168}$$

respectively.

Let us assume the system to have been, at the initial moment of time, in a state $| i \rangle$ such that

$$Q \mathscr{U} \, | \, i \rangle = | \ldots, n_{\kappa_r}, \ldots \rangle \otimes | s, s_3 \rangle . \tag{169}$$

Here, we describe the atomic system in terms of the total spin and its third component:

$$\begin{aligned} S^2 \, | s, s_3 \rangle &= s(s+1) \, | s, s_3 \rangle , \\ S_3 \, | s, s_3 \rangle &= s_3 \, | s, s_3 \rangle , \\ S^{\pm} \, | s, s_3 \rangle &= [(s \mp s_3)(s \pm s_3 + 1)]^{1/2} \, | s, s_3 \pm 1 \rangle . \end{aligned} \tag{170}$$

The physical meaning of these has been discussed in Ref. 15. We only note that

$$| s_3 | \le s \le N/2 . \tag{171}$$

Applying the formula (171) we consider the probabilities per unit time P_A, P_B, P_C for the occurrence of the processes (A), (B), and (C), respectively. (i) Incoherently pumped atomic system: $s \approx s_3 \approx N/2$. We now have

$$\begin{aligned} P_{\mathrm{A}} &\approx \eta (n_{\kappa_1} + 1) \cdots (n_{\kappa_l} + 1) n_{\kappa_{l+1}} \cdots n_{\kappa_m} N , \\ P_{\mathrm{B}} &\approx 0 , \\ P_{\mathrm{C}} &\approx \xi_1 (n_{\kappa_1} + 1) \cdots (n_{\kappa_l} + 1) n_{\kappa_{l+1}} \cdots n_{\kappa_m} N . \end{aligned} \tag{172}$$

Hence, $P_{\mathrm{A}}, P_{\mathrm{C}} \propto N$ and $P_{\mathrm{B}} \approx 0$. (ii) Thermal equlibrium of the atomic system: $s \approx -s_3 \approx N/2$. Here,

$$P_{\mathrm{A}} \approx 0 \,,$$
$$P_{\mathrm{B}} \approx \eta(n_{\kappa_m}+1)\cdots(n_{\kappa_{l+1}}+1)n_{\kappa_1}\cdots n_{\kappa_1}N \,, \qquad (173)$$
$$P_{\mathrm{C}} \approx \xi_2(n_{\kappa_1}+1)\cdots(n_{\kappa_l}+1)n_{\kappa_{l+1}}\cdots n_{\kappa_m}N \,.$$

Accordingly, $P_{\mathrm{A}} \approx 0$ and $P_{\mathrm{B}}, P_{\mathrm{C}} \propto N$. (iii) Generalized super-radiant state: $s \approx N/2$, $s_3 \approx 0$. Now,

$$P_{\mathrm{A}} \approx \eta(n_{\kappa_1}+1)\cdots(n_{\kappa_l}+1)n_{\kappa_{l+1}}\cdots n_{\kappa_m}\frac{N}{2}\left[\frac{N}{2}+1\right] ,$$
$$P_{\mathrm{B}} \approx \eta(n_{\kappa_m}+1)\cdots(n_{\kappa_{l+1}}+1)n_{\kappa_l}\cdots n_{\kappa_1}\frac{N}{2}\left[\frac{N}{2}+1\right] ,$$
$$P_{\mathrm{C}} \approx \tfrac{1}{2}(\xi_1+\xi_2)(n_{\kappa_1}+1)\cdots(n_{\kappa_l}+1)n_{\kappa_{l+1}}\cdots n_{\kappa_m}N \,. \qquad (174)$$

Since $N \gg 1$, we now have $P_{\mathrm{A}}, P_{\mathrm{B}} \propto N^2$ and $P_{\mathrm{C}} \propto N$.

In the literature it has hitherto been usual to speak of super-radiance and superfluorescence, the prefix "super" meaning that the probability per unit time was dependent on N^2 rather than on N. In this sense—which is that of Dicke[15]—all effects of the types (A) and (B) can be "supereffects," provided that the initial state $|i\rangle$ has been prepared adequately [see case (iii) above]. The conclusion to be drawn is that all effects in which the atomic system plays an active role can be supereffects in the meaning of Dicke. It suffices that the atomic system be prepared in a state $|i\rangle$ such that

$$Q\mathscr{U}\,|i\rangle = |\ldots,n_{\kappa_r},\ldots\rangle \otimes |s,s_3\rangle \,, \qquad (175)$$
$$s \approx N/2, \quad s_3 \approx 0 \,.$$

Hence the definition of this state as a generalized super-radiant state. When expanding the operator U in a series and cutting off at the first term, i.e., replacing U by the unit operator I, we derive the "usual" super-radiant state. On the other hand, effects in which the atoms are passive in the sense that they do not participate in the effect but are its catalysts—their presence being necessary for its occurrence—cannot be super-radiant. For example, Raman supereffects as well as one- and many-photon absorption or emission supereffects can exist, whereas speaking of a Rayleigh or frequency mixing (up-conversion) supereffect would make no sense.

VI. GENERALIZED MAXWELL-BLOCH EQUATIONS

We have only one step to make in order to arrive at the generalized Maxwell-Bloch equations. It suffices to write the Heisenberg equations, derived by using the Hamiltonian in Eqs. (129) and (130) or one of the Hamiltonians originating therein and to adjoin terms describing the influence of the surroundings (damping of the field, relaxation of the atomic system, Langevin forces, etc.). The problem of this adjustment to physical reality has been considered in various approaches, e.g., by Haken,[16] Senitzky,[17,18] Louisell,[19] and Lax.[20] We derive

$$\dot{a}_\kappa = -(\Gamma_\kappa + i\omega_\kappa)a_\kappa - i\sum_{m=1}^{\infty}\sum_{n=0}^{m-1}\sum_{\kappa_1,\ldots,\kappa_m}\sum_{q}\Bigg[\chi^{n}_{\kappa_1}\cdots{}_{\chi_m,21q}\frac{\partial}{\partial a^\dagger_\kappa}a^\dagger_{\kappa_1}\cdots a^\dagger_{\kappa_n}a_{\kappa_{n+1}}\cdots a_{\kappa_m}\sigma^-_q$$
$$+\chi^{m-n}_{\kappa_1}\cdots{}_{\kappa_m,12q}\frac{\partial}{\partial a^\dagger_\kappa}a^\dagger_{\kappa_m}\cdots a^\dagger_{\kappa_{n+1}}a_{\kappa_n}\cdots a_{\kappa_1}\sigma^\dagger_q\Bigg] + \mathscr{F}_{a_\kappa}(t) \,, \qquad (176a)$$

$$\sigma^-_q = -\left[\gamma_{\perp q} + i\left[\epsilon_q + \delta\epsilon_q + \sum_{m=2}^{\infty}\sum_{n=1}^{m-2}\sum_{\kappa_1,\ldots,\kappa_m}(\chi^{n}_{\kappa_1\cdots\kappa_m,22q} - \chi^{n}_{\kappa_1\cdots\kappa_m,11q})a^\dagger_{\kappa_1}\cdots a^\dagger_{\kappa_n}a_{\kappa_{n+1}}\cdots a_{\kappa_m}\right]\right]\sigma^-_q$$
$$+2i\sum_{m=1}^{\infty}\sum_{n=0}^{m-1}\sum_{\kappa_1,\ldots,\kappa_m}\chi^{n}_{\kappa_1\cdots\kappa_m,21q}a^\dagger_{\kappa_1}\cdots a^\dagger_{\kappa_n}a_{\kappa_{n+1}}\cdots a_{\kappa_m}\sigma_{3q} + \mathscr{F}_{\sigma^-_q}(t) \,, \qquad (176b)$$

$$\dot{\sigma}_{3q} = -\gamma_{\|q}(\sigma_{3q} - \eta_q) - i\sum_{m=1}^{\infty}\sum_{n=0}^{m-1}\sum_{\kappa_1,\ldots,\kappa_m}(\chi^{n}_{\kappa_1\cdots\kappa_m,21q}a^\dagger_{\kappa_1}\cdots a^\dagger_{\kappa_n}a_{\kappa_{n+1}}\cdots a_{\kappa_m}\sigma^+_q + \mathrm{H.c.}) + \mathscr{F}_{\sigma_{3q}}(t) \,. \qquad (176c)$$

Γ_κ describes the field damping; $\gamma_{\perp q}$, $\gamma_{\|q}$, and η_q denote the transversal and longitudinal constants and the population inversion parameter of the qth two-level atom. $\mathscr{F}_{a_\kappa}(t)$, $\mathscr{F}_{\sigma^-_q}(t)$, and $\mathscr{F}_{\sigma_{3q}}(t)$ are the respective Langevin forces for the quantities a_κ, σ^-_q, σ^-_q, and σ_{3q}.

From the most general form of the Maxwell-Bloch equations as proposed above, special cases can be derived. Suppose, for example, that we deal with the case of a system of atoms at p-photon resonance with the radiation field. We will neglect cooperative effects. Then, by applying the same approximations as when deriving the Hamiltonian (159), we obtain the following generalized Maxwell-Bloch equations (we drop the redundant mode index κ):

$$\dot{a} = -(\Gamma + i\omega)a - ip(a^\dagger)^{p-1}\sum_q \lambda^*_{pq}\sigma^-_q + \mathscr{F}_a(t) \,, \qquad (177a)$$

$$\sigma^-_q = -[\gamma_{\perp q} + i(\epsilon + \delta\epsilon)]\sigma^-_q + 2i\lambda_{pq}a^p\sigma_{3q} + \mathscr{F}_{\sigma^-_q}(t) \,, \qquad (177b)$$

$$\dot{\sigma}_{3q} = -\gamma_{\|q}(\sigma_{3q} - \eta_q) + i[\lambda^*_{pq}(a^\dagger)^p \sigma^-_q - \lambda_{pq} a^p \sigma^+_q] + \mathscr{F}_{\sigma_{3q}}(t) \;. \tag{177c}$$

In deriving the generalized Maxwell-Bloch equations (176), we assumed yet another approximation. In fact, the Hamiltonian part of the Heisenberg equation for an arbitrary operator X (in two-level approximation) has the form

$$\dot{X} = i[\mathscr{H}^{\sim}, X] \;. \tag{178}$$

However, we used the formula

$$\dot{X} = i[\mathscr{K}^{\sim}, X] \tag{179}$$

thus performing the replacement

$$\mathscr{H}^{\sim} = (\mathscr{U}^\dagger \mathscr{K} \mathscr{U})^{\sim} \rightarrow \mathscr{K}^{\sim} \;. \tag{180}$$

Replacing $(\mathscr{U}^\dagger \mathscr{K} \mathscr{U})^{\sim}$ by $\mathscr{K}^{\sim}$ in the equation of motion is, in fact, the mathematical expression of the rotating-wave approximation (RWA). For this interpretation we refer to the Dicke Hamiltonian of the form

$$\begin{aligned} H &= H_0 + H_{\text{int}} \;, \\ H_0 &= \epsilon S_3 + \omega a^\dagger a, \quad \epsilon \approx \omega \\ H_{\text{int}} &= \lambda(a^\dagger + a)(S^+ + S^-) \;. \end{aligned} \tag{181}$$

The commonly used rotating-wave approximation gives

$$H \underset{\text{RWA}}{\rightarrow} \epsilon S_3 + a^\dagger a + \lambda(a^\dagger S^- + a S^+) \;. \tag{182}$$

The application of the procedures described in the previous sections neglecting the terms with powers of the coupling constant λ higher than 1 leads to the same result.

The preceding generalized Maxwell-Bloch equations (176) stand in direct connection with practice. These equations, when specified for a concrete physical system, can be used to calculate many effects of quantum optics. At this point we conclude the considerations of the present paper.

VII. FINAL REMARK

An advantage of our method of obtaining effective Hamiltonians resides in the fact that it requires no equation of motion. Certain steps of the method still need to be refined; e.g., the use of the two-level approximation. Nonetheless, the effective Hamiltonians are derived on a strict mathematical basis and can serve for further analyses and evaluations. The new Hamiltonians derived strictly in this paper, e.g., those given by Eqs. (113), (149), and (156), are more general than those used until now and reduce to the latter if rougher approximations are applied.

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APPENDIX: PROOF OF FORMULA (26)

We have

$$\begin{aligned} \mathscr{U}_l &= \frac{1}{l!}\left[\frac{d^l}{d\epsilon^l}\mathscr{U}(\epsilon)\right]_{\epsilon=0} \\ &= \frac{1}{l!}\left[\mathscr{D}\left\{\frac{d^{l-1}}{d\epsilon^{l-1}}\sum_{m=0}^{\infty}(m+1)\epsilon^m X_{m+1}\mathscr{U}(\epsilon)\right\}\right]_{\epsilon=0} \\ &= \frac{1}{l!}\left[\mathscr{D}\left\{\frac{d^{l-1}}{d\epsilon^{l-1}}\sum_{m=0}^{l-1}(m+1)\epsilon^m X_{m+1}\mathscr{U}(\epsilon)\right\}\right]_{\epsilon=0} \end{aligned}$$

By equality

$$\frac{1}{r!}\frac{d^r}{d\epsilon^r}\epsilon^s = \sum_{t=0}^{s}\begin{bmatrix} s \\ t \end{bmatrix}\epsilon^{s-t}\frac{1}{(r-t)!}\frac{d^{r-t}}{d\epsilon^{r-t}}, \quad s \le r$$

we obtain

$$\begin{aligned} \mathscr{U}_l &= \frac{1}{l}\left[\mathscr{D}\left\{\sum_{m=0}^{l-1}(m+1)X_{m+1}\sum_{n=0}^{m}\begin{bmatrix} m \\ n \end{bmatrix}\frac{1}{(l-1-n)!}\frac{d^{l-1-m}}{d\epsilon^{l-1-m}}\mathscr{U}(\epsilon)\right\}\right]_{\epsilon=0} = \frac{1}{l}\mathscr{D}\left\{\sum_{m=0}^{l-1}(m+1)X_{m+1}\mathscr{U}_{l-m+1}\right\} \\ &= \sum_{m=1}^{l}\frac{m}{l}\mathscr{D}\{X_m \mathscr{U}_{l-m}\} \\ &= X_l + \sum_{m=1}^{l-1}\frac{m}{l}\mathscr{D}\{X_m \mathscr{U}_{l-m}\} \;. \end{aligned}$$

Applying this theorem, we easily derive formulas for the successive operators $\mathscr{U}_l$:

$$\mathcal{U}_0=I\,,$$

$$\mathcal{U}_1=X_1\,,$$

$$\mathcal{U}_2=X_2+\frac{1}{2!}X_1^2\,,$$

$$\mathcal{U}_3=X_3+X_2X_1+\frac{1}{3!}X_1^3\,,$$

$$\mathcal{U}_4=X_4+X_3X_1+\frac{1}{2!}X_2^2+\frac{1}{1!2!}X_2X_1^2+\frac{1}{4!}X_1^4\,,$$

etc. Obviously, these formulas are explicit expressions of the relations (24).

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