sufficient, however, to assign the observed hyperfine effects to specific lattice sites.

Because of the large number of presently unknown parameters, a detailed interpretation of the Knight shifts and spin-lattice relaxation rates in lanthanum, as well as the other two Group-IIIB transition metals, scandium and yttrium, is not possible at this time. Nevertheless, the data suggest strongly that the dominant magnetic-hyperfine mechanism in these metals is the direct s-contact interaction. It is also possible to conclude that the electron-phonon enhancement of the electronic specific heat is large, as indicated by the available band-structure calculations, and that the effective d-spin core-polarization hyperfine fields, at least in yttrium and lanthanum, are significantly smaller than in the corresponding Group-VIII metals, palladium and platinum.

As in the case of scandium, the lanthanum quadrupole coupling constant is approximately 2.5 times larger than can be accounted for by the point-ion model. Thus, the relative importance of conduction-electron contributions to the EFG is approximately the same in both metals.

Note added in proof. F. Y. Fradin, Phys. Letters **28A**, 441 (1968), has recently determined the field-orientation dependence of the spin-lattice relaxation rate in a single-crystal specimen of scandium. An analysis of the observed anisotropy based on the extreme tight-binding model[21] led to the conclusion that the orbital hyperfine interaction is the dominant relaxation mechanism in scandium metal. Because of the many approximations inherent in the model the accuracy of this conclusion is difficult to assess.

ACKNOWLEDGMENTS

The author is indebted to Professor R. G. Barnes for his interest in this work as well as for the loan of his lanthanum sample. Several stimulating discussions with Dr. D. B. McWhan were most helpful. The author is especially grateful to D. C. Barham and Dr. H. T. Weaver for their expert experimental assistance.

PHYSICAL REVIEW VOLUME 179, NUMBER 2 10 MARCH 1969

Quantum Theory of an Optical Maser. III. Theory of Photoelectron Counting Statistics*

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(Received 31 January 1968)

In this paper we determine the photoelectron counting statistics produced by the fully quantum-mechanical laser considered in the first paper of this series. The problem of obtaining the photocount distribution from the now known photon statistics is solved in a completely quantum-mechanical fashion. The time evolution of the combined photodetector-laser system is derived. The techniques developed for the solution of this problem are of general interest in the area of nonequilibrium quantum statistical dynamics.

I. INTRODUCTION

IN the first paper[1] of this series the steady-state photon distribution in a laser cavity (above, at, or below threshold) was found to be

$$\rho_{n,n}=Z^{-1}\left\{\frac{(A^2/BC)^{n+A/B}}{(n+A/B)!}\right\}. \quad (1)$$

The photon statistical distribution is inferred in practice by photoelectron counting techniques. The photoelectron counting statistics produced by laser radiation

* A preliminary account of this work was presented at the Second Rochester Conference on Coherence and Quantum Optics.

† Work supported by the Advanced Research Projects Agency under Contract No. SD-90.

‡ Work supported in part by the National Aeronautics and Space Administration and in part by the U. S. Air Force Office of Scientific Research.

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has been the subject of recent experimental[2–9] and theoretical[10–16] investigations. An object of the present paper is to obtain the distribution of photoelectrons produced by a fully quantum-mechanical laser.

The usual procedure for obtaining the photocount distribution breaks the time T of observation into many small time intervals. In each of these small time intervals a quantum-mechanical calculation is carried out in low-order perturbation theory to obtain the probability of obtaining a count. The calculation is then completed by classical probabilistic arguments for the number of counts observed in the larger time interval T. In this type of analysis one is "looking" at the system at the end of each of the small time intervals. That is, since the usual experiments are so complicated and microscopically disruptive,[17] it seems reasonable to assume that each count is equivalent to "looking" at the system. As is well known, upon looking at a system we destroy its wave function, e.g., when we trace over the laser coordinates, we produce a statistical mixture. Hence, one might well ask, "Does the procedure of looking after every count give the same counting distribution as would be observed if the system were not interrupted until a large number of potential counts had accumulated?" The latter problem will here be solved in a fully quantum-mechanical fashion, whereas the former requires the use of classical probability theory. In order to accomplish this we consider the following detection scheme:

(i) Having established the steady-state photon distribution (1), remove all of the "lasing" and "damping"[18] atoms from the cavity. The radiation density matrix is now time-independent.

(ii) Insert into the cavity a group of photosensitive atoms (for example, a block of film containing silver nitrate in some host matrix).

(iii) After a time T remove the film and count the number of excited atoms.

(iv) Repeat the procedure a large number of times (each time starting from the same photon distribution $\rho_{n,n}$ and fresh film) in order to determine the probability of finding m excited atoms.

This theoretical model where the combined laser-detector system is left undisturbed until time T does not correspond to the familiar experimental situation. However, it may be solved quantum-mechanically and it is of interest to see how closely the results correspond to those obtained in the usual analysis. Furthermore, the problem has an intrinsic interest in its own right in the field of nonequilibrium quantum statistical mechanics. That is, we must solve the problem of a quantized laser field interacting with many atoms. Clearly simple perturbation theory is not appropriate for this situation. We develop a differential equation describing the laser-detector system which is then solved to obtain the time dependence of the laser-detector system. This approach is equivalent to summing an infinite set of diagrams.

In Sec. II we present our model and outline the method of calculation. In Sec. III the analysis of this model is developed. The photoelectron distribution implied by our quantum theory of a laser is found in Sec. IV. A concluding discussion is given in Sec. V.

II. MODEL OF A PHOTODETECTOR

We take the photodetector to consist of N-independent, equivalent but distinguishable atoms[19] each of which has a ground state $|g\rangle$ and a continuum of excited

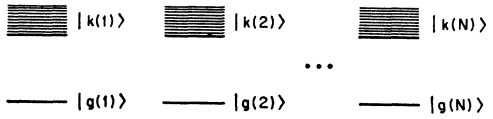

FIG. 1. Pictorial representation of photodetector consisting of N-independent atoms. Each atom in detector has a ground state $|g\rangle$ and continuum of excited states $|k\rangle$. Atoms are labeled by indexing atomic state with particle number, e.g., $|k(m)\rangle$.

[2] A summary of experimental results is given in *Proceedings of the International Conference on the Physics of Quantum Electronics, Puerto Rico, 1965*, edited by P. Kelley, B. Lax, and P. Tannenwald (McGraw-Hill Book Co., New York, 1965). See especially J. A. Armstrong and A. W. Smith, p. 701; F. Johnson, T. McLean, and E. Pike, p. 706; C. Freed and H. A. Haus, p. 715.

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[15] G. Bédard, thesis, University of Rochester (unpublished); J. Opt. Soc. Am. **57**, 1201 (1967).

[16] B. R. Mollow, Phys. Rev. **168**, 1896 (1968).

[17] When the electrons are liberated they are accelerated by an external electric field and "banged" into a photosensitive surface.

[18] The damping atoms are nonresonant atoms injected into the laser cavity in their lower state in order to provide our cavity with a finite Q.

[19] The photodetecting medium will be taken to be optically thin so that each atom sees the same radiation field as any other atom.

states $|k\rangle$, see Fig. 1. The atoms are placed in the cavity at time $t=0$ in their ground state, while the radiation field is initially described by the diagonal density matrix $\rho_{n,n}(0)$. The combined laser-detector system will be described at later times by the density matrix

$$\rho_{n,D;n,D'}(t),$$

where D and D' denote the states of the detector. For example, initially, the only nonvanishing elements of the density matrix are those given by

$$D=g(1),g(2),\cdots,g(N), \tag{2a}$$

while at some later time elements of the density matrix might exist for

$$D=k(1),k(2),g(3),\cdots,k(N). \tag{2b}$$

After a time T, the N atoms are removed from the cavity and the number of ionized atoms is determined (equal, of course, to the number of photoelectrons). This process is repeated several times in order to obtain the relative probability $P_m(T)$ for observing m photoelectrons irrespective of the state of the laser field. This is

$$P_m(T)=\sum_n P_{n,m}(T), \tag{3}$$

where $P_{n,m}(T)$ is the probability of finding the field in the state $|n\rangle$ with m photoelectrons ejected.

We now proceed with the calculation of $P_{n,m}(T)$. Since in our model, the probability of exciting any given configuration of m atoms is equal to the probability of exciting any other group of m atoms, it is clear that we need consider only the probability of exciting a specified group of m atoms (such as the first m). It is convenient to denote such a specific excited state by $D(m,\{k\})$ where m indicates that the first m atoms are excited and $\{k\}$ is a state of the photodetector, for example,

$$\{k\}=k(1)\cdots k(m),g(m+1)\cdots g(N), \tag{4}$$

where each of the one-electron states $k(i)$, $i=1\cdots m$, is allowed to range over the continuous spectrum of the ith atom. In order to obtain the total probability of these atoms being ionized we must sum over the probabilities of finding each atom in any of its continuum states, that is,

$$\begin{aligned}\mathcal{P}_{n,m}(T)&=P_{n,K(1)\cdots K(m),g(m+1)\cdots g(N)}(T)\\&=\sum_{\{k\}}\rho_{n,D(m,\{k\});n,D(m,\{k\})}(T),\end{aligned} \tag{5}$$

where the subscript $K(m)$ on P means that the mth atom is in any of its excited states. Explicitly this sum is

$$\sum_{k(1)}\cdots\sum_{k(m)}\rho_{n,k(1)\cdots k(m)\cdots g(N);n,k(1)\cdots k(m)\cdots g(N)}(T).$$

Once the probability $\mathcal{P}_{n,m}(T)$ of having the first m atoms ionized (while the field is in state $|n\rangle$) is known, the corresponding probability of any m atoms being excited is obtained by multiplying by the combinatorial factor $\binom{N}{m}$ representing the number of ways m atoms may be chosen from N, i.e.,

$$P_{n,m}(T)=\binom{N}{m}\mathcal{P}_{n,m}(T). \tag{6}$$

We now proceed to obtain a differential equation for $P_{n,m}(T)$. The interaction Hamiltonian $\hbar V(t)$ for the combined laser-detector system is (in the interaction picture)

$$\hbar V(t)=-e\sum_{i=1}^{N}\hbar x_i(t)E(t)\equiv\sum_i\hbar V_i(t), \tag{7a}$$

where e is the electronic charge and $E(t)$ is the quantized electric-field operator[20]

$$E(t)=e^{+i\nu a^\dagger at}\mathcal{E}(a+a^\dagger)e^{-i\nu a^\dagger at}, \tag{7b}$$

while $x_i(t)$ is the position operator for the atomic electron of the ith atom,

$$x_i(t)=\exp\{iH_{\text{atom}}t\}x_i\exp\{-iH_{\text{atom}}t\}. \tag{7c}$$

We will take the only nonvanishing matrix elements of (7a) to be those connecting the ground state with an arbitrary excited state, and restrict ourselves to energy-conserving transitions (work in the rotating-wave approximation). It will be useful to denote positive and negative frequency parts of $V(t)$ by $V(t)^{(-)}$ and $V(t)^{(+)}$, respectively, so that

$$V(t)=\sum_i[V_i(t)^{(+)}+V_i(t)^{(-)}]. \tag{7d}$$

The relevant matrix elements of (7d) are

$$\begin{aligned}&\langle k(i),n|\,V_i(t)^{(+)}\,|g(i),n+1\rangle\\&\qquad=g(n+1)^{1/2}\exp\{i(\omega_{k(i)}-\nu)t\}\end{aligned} \tag{8a}$$

and

$$\begin{aligned}&\langle g,n|\,V_i(t)^{(-)}\,|k,n-1\rangle\\&\qquad=g\sqrt{n}\exp\{-i(\omega_{k(i)}-\nu)t\},\end{aligned} \tag{8b}$$

where the atomic frequency $\omega_{k(i)}$ is

$$\omega_{k(i)}=(\epsilon_{k(i)}-\epsilon_g)/\hbar, \tag{9}$$

and the coupling constant g is given by

$$g=-ex_{k,m}\mathcal{E}/\hbar.$$

III. ANALYSIS

In order to obtain a differential equation for $P_{n,m}$ as used in Eq. (3), we consider the equation of motion for the density matrix $\rho(t)$ of the detector-laser system

[20] In the notation of Ref. 16, ν is the frequency of the optical radiation, $a^\dagger$ and a are the creation and annihilation operators, and $\mathcal{E}$ is a constant having the units of an electric field.

(in the interaction picture)

$$\dot{\rho}(t)=-i[V(t),\rho(t)]. \tag{10}$$

The solution of Eq. (10) is formally

$$\rho(t)-\rho(0)=-i\int_0^t dt'[V(t'),\rho(t')]. \tag{11}$$

For reasons which will become apparent it is more convenient to substitute Eq. (11) into Eq. (10) to obtain the alternative form[21] for the time rate of change of $\rho(t)$

$$\dot{\rho}(t)=(-i)^2\left[V(t),\int_0^t dt'[V(t'),\rho(t')]\right]. \tag{12}$$

Taking diagonal matrix elements of Eq. (12) for states $|n,D(m,\{k\})\rangle$ and summing over all excited states, we obtain an equation for $\mathcal{P}_{n,m}(t)$

$$d\mathcal{P}_{n,m}(t)/dt=-\sum_{\{k\}}\langle n,D(m,\{k\})|$$
$$\times\left[V(t),\int_0^t dt'[V(t'),\rho(t')]\right]|n,D(m,\{k\})\rangle. \tag{13}$$

After a rather involved calculation, assuming only that the number of excited states is large, Eq. (13) reduces to

$$d\mathcal{P}_{n,m}(t)/dt=-2r(N-m)n\mathcal{P}_{n,m}(t)$$
$$+2rm(n+1)\mathcal{P}_{n+1,m-1}(t), \tag{14}$$

where r is the transition rate given by Eq. (A13′). A detailed account of the transition from Eq. (13) to (14) is given in the Appendix.

Equation (14) refers to the probability of exciting the first m atoms. We proceed to find an equation for any m atoms by multiplying (14) by $\binom{N}{m}$ as required by Eq. (6):

$$\dot{P}_{n,m}=-2r(N-m)nP_{n,m}+2r\binom{N}{m}m(n+1)\mathcal{P}_{n+1,m-1} \tag{15}$$

and, since

$$m\binom{N}{m}=[N-(m-1)]\binom{N}{m-1},$$

Eq. (15) becomes

$$\dot{P}_{n,m}=-2r(N-m)nP_{n,m}(t)$$
$$+2r(N-m+1)(n+1)P_{n+1,m-1}(t). \tag{16}$$

Since normally $N\gg m$, we write (16) as

$$\dot{P}_{n,m}=-\gamma nP_{n,m}(t)+\gamma(n+1)P_{n+1,m-1}(t), \tag{17}$$

where

$$\gamma=2rN. \tag{18}$$

[21] A contribution involving $\rho(0)$ is omitted in (12) as it does not give any matrix elements of interest to us.

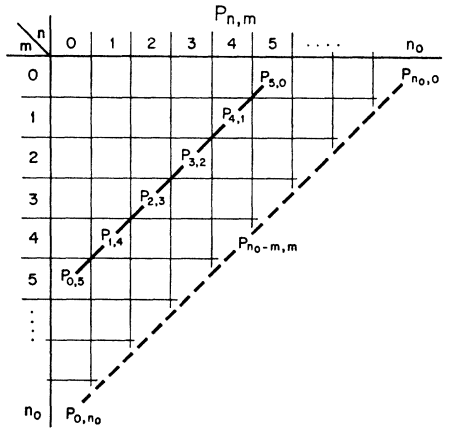

FIG. 2. Grouping of Eq. (17) according to value of n_0. First example is for $n_0=5$. Dashed line is for arbitrary n_0.

It is clear from the form of Eq. (17) that this system of equations breaks up into an infinite set of equations, as depicted in Fig. 2, one set for each value of n_0, where

$$n_0=n+m. \tag{19}$$

This is in accord with the obvious physical fact that the appearance of a photoelectron is associated with the disappearance of a photon. We denote the solution of one of these sets by

$$P_{n,m}(n_0,t)=P_{n_0-m,m}(n_0,t)\equiv P_m(n_0,t). \tag{20}$$

For any one of these sets, characterized by n_0, Eq. (17) becomes

$$\dot{P}_m(n_0,t)=-\gamma(n_0-m)P_m(n_0,t)$$
$$+\gamma[n_0-(m-1)]P_{m-1}(n_0,t). \tag{21}$$

Subject to the condition that initially

$$P_m(n_0,0)=\rho_{n_0,n_0}\rho_{(1)\cdots g(N),g(1)\cdots g(N)}=\rho_{n_0,n_0}, \tag{22}$$

the solution to Eq. (21) is

$$P_m(n_0,t)=\binom{n_0}{m}e^{-\gamma(n_0-m)t}(1-e^{-\gamma t})^m\rho_{n_0,n_0}(0) \tag{23}$$

as may be verified by direct differentiation. Equation (23) may be written as

$$P_m(n_0,t)=\binom{n_0}{m}\eta^m(1-\eta)^{n_0-m}\rho_{n_0,n_0}(0), \tag{24}$$

where the "quantum efficiency" η is

$$\eta=(1-e^{-\gamma t}). \tag{25}$$

The solution of Eq. (17) is obtained from (24) by summing over $n_0=1, 2, 3, \cdots$, recognizing the constraint that $n+m=n_0$,

$$P_{n,m}(t)=\sum_{n_0}P_m(n_0,t)\delta_{n+m,n_0}. \tag{26}$$

Finally we sum over all possible quantum states of the field (trace over the field) to obtain $P_m(t)$ as given in (3):

$$P_m(t)=\sum_n \sum_{n_0} \binom{n_0}{m}\eta^m(1-\eta)^{n_0-m}\rho_{n_0,n_0}(0)\delta_{n+m,n_0}$$

$$=\sum_{n_0} \binom{n_0}{m}\eta^m(1-\eta)^{n_0-m}\rho_{n_0,n_0}(0)\,, \qquad (27)$$

which is the probability of detecting m photoelectrons if the quantum efficiency is η and the incident photon distribution is given by ρ_{n_0,n_0}.

IV. PHOTOSTATISTICS IMPLIED BY A FULLY QUANTIZED LASER

We may now calculate the photocount distribution for a fully quantized laser by inserting $\rho_{n,n}$ as given by Eq. (1) into (27). The probability for finding m photoelectrons is

$$P_m=\sum_n \binom{n}{m}\eta^m(1-\eta)^{n-m}\left\{Z^{-1}\frac{(A^2/BC)^{n+A/B}}{(n+A/B)!}\right\}. \qquad (28)$$

Summing the series, we find the basic relation

$$P_m=Z^{-1}\eta^m\frac{(A/BC)^{m+A/B}}{(m+A/B)!}$$

$$\times {}_1F_1\left(m+1,m+\frac{A}{B}+1,(1-\eta)\frac{A^2}{BC}\right), \qquad (29)$$

where ${}_1F_1$ is the confluent hypergeometric function. We summarize the notation as follows: A = linear gain, B = nonlinear parameter, $C=\nu/Q$, η = detector parameter, and Z^{-1} = normalization factor which is given by

$$Z=\sum_n \frac{(A^2/BC)^{n+A/B}}{(n+A/B)!}$$

$$=\left[\frac{(A^2/BC)}{(A/B)!}\right]{}_1F_1\left(1;1+\frac{A}{B};\frac{A^2}{BC}\right).$$

V. DISCUSSION

Equation (27) may be understood as follows: Consider a state of the field having just one photon $|1\rangle$. Let the probability of having a photoelectron ejected from a detector interacting with this field for a certain time be given by η. Now if the state of the radiation field is $|n\rangle$, the probability of observing m photoelectrons should be proportional to

$$P_m{}^{(n)}\propto \eta^m, \qquad (30)$$

which is to be multiplied by the probability that $n-m$ quanta were not absorbed, i.e., $(1-\eta)^{n-m}$,

$$P_m{}^{(n)}\propto \eta^m(1-\eta)^{n-m}, \qquad (31)$$

but we, of course, do not know which m photons of the original number n were absorbed so we must include a combinational factor

$$P_m{}^{(n)}=\binom{n}{m}\eta^m(1-\eta)^{n-m}. \qquad (32)$$

This is Bernoulli's distribution for m successful events (counts) and $n-m$ failures, each event having a probability η. Now if we have a distribution of n values, we must multiply (32) by $\rho_{n,n}$ and sum on n,

$$P_m=\sum_n P_m{}^{(n)}\rho_{n,n}$$

$$=\sum_n \binom{n}{m}\eta^m(1-\eta)^{n-m}\rho_{n,n}\,, \qquad (33)$$

which is the result of (27).

As a direct consequence of our model, (27) contains not only the small η limit ($\eta\ll 1$), but is valid for all $\eta(0\leq\eta\leq 1)$. Clearly, if we wish to obtain the *photon* statistics by counting photoelectrons we must require $\eta=1$, for then, as we see from Eq. (27),

$$P_m=\rho_{m,m}.$$

In all other cases ($\eta<1$) we are measuring the *photoelectron* statistics which in general will be very different, e.g., Eq. (29) for the laser is

$$P_m=\rho_{m,m}\{\eta^m{}_1F_1[m+1,m+(A/B)+1,(1-\eta)(A^2/BC)]\}.$$

To cast (33) into another form we write ρ in the $P(\alpha)$ representation[13]:

$$\rho_{n,n}=\int d^2\alpha\, P(\alpha)[(\alpha^*\alpha)^n/n!]\exp\{-\alpha^*\alpha\}\,, \qquad (34)$$

so that (33) becomes

$$P_m=\int d^2\alpha \sum_{n=m}\binom{n}{m}P(\alpha)[(\alpha^*\alpha)^n/n!]$$

$$\times\exp\{-\alpha^*\alpha\}\eta^m(1-\eta)^{n-m}, \quad n=l+m$$

$$=\int d^2\alpha\exp\{-\alpha^*\alpha\}[(\alpha^*\alpha\eta)^m/m!]P(\alpha)$$

$$\times\sum_{l=0}[(\alpha^*\alpha)^l/l!](1-\eta)^l$$

$$=\int d^2\alpha P(\alpha)[(\alpha^*\alpha\eta)^m/m!]\exp\{-\alpha^*\alpha\eta\}. \qquad (35)$$

Expression (35) [or, equivalently, (27)] agrees with that obtained in the usual treatment. Equation (27) is a more convenient formulation of the photocounting distribution for our purpose as we most easily find the photon statistical distribution in the n representation. We obtain $P(I)$ only after an auxiliary calculation of some complexity. We remark that the expression for the parameter η obtained in the present analysis (see also

Ref. 15),

$$\eta=(1-e^{-\gamma t}),$$

has the correct limiting value $\eta\rightarrow 1$ for $\gamma t\rightarrow\infty$. This is not the case in calculations giving $\eta=\gamma t$.

It should perhaps be pointed out that if the number of photoatoms is not much larger than the number of ejected photoelectrons then we should use Eq. (16) instead of (17). For example, if we only have 10 atoms we could never observe more than 10 photoelectrons. Further, we have neglected the fact that atoms deeper inside the photodetector would see a weaker field. It might also be noted that the presence of the photodetector would slightly load the laser in a way which could be described by giving the cavity a smaller Q value. These points could be included in the theory but are of secondary interest.

ACKNOWLEDGMENT

One of us (M.S.) wishes to thank Professor R. Glauber for several helpful and stimulating discussions.

APPENDIX

This Appendix justifies Eq. (14) beginning from (13):

$$d\mathcal{P}_{n,m}(t)/dt=-\sum_{\{k\}}\langle n,D(m,\{k\})|\Big[V(t),\int_0^t dt'\times[V(t'),\rho(t')]\Big]|n,D(m,\{k\})\rangle. \quad (13)$$

Inserting $V(t)$ as given by (9), we have

$$d\mathcal{P}_{n,m}(t)/dt=-\int_0^t dt'\sum_{k(i)}\sum_{l(j)}\sum_{i,j}\langle n,k(i),l(j)|\times[(V_i(t)^{(+)}+V_i(t)^{(-)}),[(V_j(t')^{(+)}+V_j(t')^{(-)}),\sum_{\{k\}}{}'\langle\mathfrak{D}|\rho(t')|\mathfrak{D}\rangle]]|n,k(i),l(j)\rangle, \quad (A1)$$

where the primed sum means no sum over $k(i)$ and $l(j)$ and $|\mathfrak{D}\rangle$ denotes the set of detector states for $N-2$ atoms omitting the ith and jth atoms. It is convenient to introduce the notation

$$\sum_{\{k\}}{}'\langle\mathfrak{D}|\rho(t)|\mathfrak{D}\rangle=R(t). \quad (A2)$$

It will be shown that only those terms in (A1) with $i=j$ and $k=l$ contribute significantly. Consider a typical term in (A1) with $i\neq j$:

$$\sum_{i\neq j}\sum_{k(i)}\sum_{l(j)}\langle n,k(i),l(j)|V_i^{(-)}\exp-i(\omega_{k(i)}-\nu)t\times V_j^{(+)}\exp i(\omega_{l(j)}-\nu)t'R(t')|k(i),l(j),n\rangle. \quad (A3)$$

Writing Eq. (A3) in matrix element form, we find

$$\int dt'\sum_{i\neq j}\sum_{k(i)}\sum_{l(j)}\{\langle n,k(i),g(j)|V_i^{(-)}|n-1,g(i),g(j)\rangle\times\exp-i(\omega_{k(i)}-\nu)t\langle n-1,g(i),g(j)|V_j^{(+)}|n,g(i),l(j)\rangle\times\exp i(\omega_{l(j)}-\nu)t'\langle n,g(i)l(j)|R(t')|n,k(i),g(j)\rangle\}. \quad (A4)$$

We proceed by considering the density of excited states to be so large that we may replace the sum over excited states by integrations over atomic frequencies with the appropriate density of states $\sigma(\omega)$. Expression (A4) then becomes

$$\int_0^t dt'\sum_{i\neq j}\int d\omega_i\sigma(\omega_i)\int d\omega_j\sigma(\omega_j)\langle n,k(i)|V_i^{(-)}|n-1,g(i)\rangle\times\exp-i(\omega_{k(i)}-\nu)t\langle n-1,g(j)|V_j^{(+)}|n,l(j)\rangle\times\exp i(\omega_{l(j)}-\nu)t'\langle n,g(i)l(j)|R(t')|n,k(i),g(j)\rangle. \quad (A5)$$

Now, in general, all the factors except the exponential appearing in (A5) will be slowly varying functions of atomic frequency, so we take them outside the integral sign and perform the integration over ω_i and ω_j. When these integrations are carried out, we obtain a δ function in time from each integration:

$$\int d\omega_i\exp[-i(\omega_i-\nu)t]\rightarrow\delta(t),$$

$$\int d\omega_j\exp i(\omega_j-\nu)t'\rightarrow\delta(t').$$

Hence, when we integrate over t' we find that Eq. (13) is proportional to the elements of the density matrix which are off-diagonal in the atomic states, and vanish at time $t=0$ as the system is prepared with all the "photoatoms" in their ground states:

$$\langle n,g(i),l(j)|R(0)|n,k(i),g(j)\rangle=\langle n,g(i),l(j)|\rho(0)|n,k(i),g(j)\rangle=0. \quad (A6)$$

Similarly, terms with $j=i$ will vanish, unless $k(i)=l(i)$, i.e., in Eq. (A1) we need keep only the terms with $i=j$ and $k=l$. Equation (A1) now reads

$$d\mathcal{P}_{n,m}(t)/dt=(-i)^2\sum_i\sum_{k(i)}\int_0^t dt'\langle n,D(m,\{k\})|\times\{(V_i(t)^{(+)}+V_i(t)^{(-)})(V_i(t')^{(+)}+V_i(t')^{(-)})\rho(t')+\rho(t')(V_i(t')^{(+)}+V_i(t')^{(-)})(V_i(t)^{(+)}+V_i(t)^{(-)})-(V_i(t)^{(+)}+V_i(t)^{(-)})\rho(t')(V_i(t')^{(+)}+V_i(t')^{(-)})-(V_i(t')^{(+)}+V_i(t')^{(-)})\rho(t')(V_i(t)^{(+)}+V_i(t)^{(-)})\}\times|n,D(m,\{k\})\rangle. \quad (A7)$$

In view of the relations (8a) and (8b) the only nonvanishing terms in Eq. (A1) are

$$d\mathcal{P}_{n,m}(t)/dt=\sum_{k(1)}\cdots\sum_{k(m)}(-i)^2\int_0^t dt'\langle n,D(m,\{k\})|\{\sum_{i=1}^{m}\sum_{k(i)}(V_i(t)^{(-)}V_i(t')^{(+)}\rho(t'))$$
$$+\sum_{i=m+1}^{N}\sum_{k(i)}(V_i(t)^{(+)}V_i(t')^{(-)}\rho(t'))+\sum_{i=1}^{m}\sum_{k(i)}(\rho(t')V_i(t)^{(-)}V_i(t')^{(+)})+\sum_{i=m+1}^{N}\sum_{k(i)}(\rho(t')V_i(t')^{(+)}V_i(t)^{(-)})$$
$$-\sum_{i=1}^{m}\sum_{k(i)}(V_i(t)^{(-)}\rho(t')V_i(t')^{(+)})-\sum_{i=m+1}^{N}\sum_{k(i)}(V_i(t)^{(+)}\rho(t')V_i(t')^{(-)})-\sum_{i=1}^{m}\sum_{k(i)}(V_i(t')^{(-)}\rho(t')V_i(t)^{(+)})$$
$$-\sum_{i=m+1}^{N}\sum_{k(i)}(V_i(t')^{(+)}\rho(t')V_i(t)^{(-)})\}|n,D(m,\{k\})\rangle. \quad \text{(A8)}$$

As we are from now on only interested in diagonal elements of ρ, let us introduce the notation

$$\langle n,D|\rho|n,D\rangle\equiv\rho(t,D,n). \quad \text{(A9)}$$

Certain terms in Eq. (A8) may be neglected. These terms are those involving integrals over the excited-state frequencies such as

$$\int d\omega_i\exp\{i(\omega_i-\nu)(t-t')\}\rho(t,k(i),n), \quad \text{(A10)}$$

where the density matrix corresponds to the excited state $k(i)$. In this case we note that since there are a large numbero f states $\mathfrak{N}$ to which the electron may be excited, $\rho(t',k(i),n)$ is to a good approximation

$$\rho[t',k(i),n]\cong\sum_{k(i)}\frac{\rho(t',k(i),n)}{\mathfrak{N}}\equiv\frac{\rho(t',K(i),n)}{\mathfrak{N}}, \quad \text{(A11)}$$

which vanishes as $\mathfrak{N}$ becomes very large. The argument $K(i)$ in (A11) means that we have summed over the excited states of the ith atom. Finally, the dominant terms in (A1) are

$$d\mathcal{P}_{n,m}(t)/dt$$
$$=-\int_0^t dt'\sum_{i=m+1}^{N}\sum_{k(i)}\langle n,g(i)|V_i(t)^{(+)}V_i(t')^{(-)}|n,g(i)\rangle$$
$$\times\rho(n,K(1)\cdots K(m)\cdots g(N),t')-\text{c.c.}$$
$$+\int_0^t dt'\sum_{i=1}^{m}\sum_{k(i)}\langle n,k(i)|V_i(t)^{(-)}V_i(t')^{(+)}|n,k(i)\rangle$$
$$\times\rho((n+1),K(1)\cdots g(i)$$
$$\cdots K(m)\cdots g(N),t')+\text{c.c.}, \quad \text{(A12)}$$

where, as in (A11), the symbol $K(m)$, for example, denotes the fact that the mth atom is in any of its excited states. If we replace the sum on $k(i)$ as it appears in (A12) by an integral over ω_i

$$\sum_{k(i)}\rightarrow\int d\omega_i\sigma(\omega_i),$$

and note that $\rho(t',D(m,\{k\}),n)$ does not depend on $k(i)$ (i.e., the ith atom is in its ground state since $i>m$), the integral over ω_i leads to a δ function $\delta(t-t')$. The integration over t' may then be done and yields

$$d\mathcal{P}_{n,m}(t)/dt=-\sum_{i=m+1}^{N}2rn\rho(n,K(1)\cdots K(m)\cdots g(N),t)$$
$$+\sum_{i=1}^{m}2r(n+1)\rho((n+1),K(1)\cdots g(i)\cdots K(m)\cdots g(N),t), \quad \text{(A13)}$$

where

$$r=\pi g_{k,i}^2\sigma(\nu). \quad \text{(A13')}$$

Recalling that the probability of exciting any given atom is the same as any other, we write

$$\rho(n,K(1)\cdots g(i)\cdots K(m),g(m+1)\cdots g(N))=\mathcal{P}_{n+1,n-1}, \quad \text{(A14)}$$

and noting that the first sum in (A13) ($m+1$ to N) leads to a factor $N-m$, while the second sum (1 to m) is replaced by m, (A13) becomes

$$d\mathcal{P}_{n,m}(t)/dt=-2r(N-m)n\mathcal{P}_{n,m}(t)$$
$$+2rm(n+1)\mathcal{P}_{n+1,m-1}(t).$$