Quantum electrodynamics of intense photon beams. New approximation method

Iwo Bialynicki-Birula*

Department of Physics, University of Pittsburgh, Pittsburgh, Pennsylvania 15260 and Institute of Theoretical Physics, Warsaw University, 00-681 Warsaw, Poland

Zofia Bialynicka-Birula Institute of Physics, Polish Academy of Sciences, 02-668 Warsaw, Poland (Received 17 March 1976)

We develop a new approximation method for determining resonance properties of atomic systems interacting with intense photon beams. This method, based on an improved iteration procedure, leads to analytic expressions for the atomic density matrix from which positions of the resonances are determined. Our iteration procedure is more rapidly convergent than the continued-fraction method, and already in its second step reproduces the Bloch-Siegert shift of the one-photon resonance within 1 ppm.

I. INTRODUCTION

The purpose of this paper is to derive and apply a new approximation method (based on an improved iteration procedure) of calculating resonance properties of atomic systems interacting with intense photon beams. Our approximation method can be most easily described with the use of the phase representation of state vectors and operators of the electromagnetic field. This representation, described in Sec. II, is very well suited for the analysis of all processes in which the number nof photons in a given mode is very large. It enables us to identify the leading terms in the largen asymptotic expansion of various physical quantities (transition amplitudes, density matrices, etc.) calculated in quantum electrodynamics and at the same time it gives precise relationships between those quantities and their semiclassical counterparts.

In Sec. III we use the phase representation to analyze, for large n, the simplest model of quantum electrodynamics: a two-level system interacting with a monochromatic mode of radiation. This analysis leads to a closed, although formal, expression for the "relative population" component p_3 of the reduced atomic density matrix in the resonance region.

In Sec. IV we propose an improved iteration procedure to evaluate p_3 and we calculate the Bloch-Siegert shift to test the convergence of our approximation scheme. From the analytic expression for p_3 , obtained in the second step of our iteration procedure, we get the Bloch-Siegert shift at the main resonance with accuracy better than 1 ppm. Numerical results given by our method coincide with those obtained by Stenholm¹ with his many-step computer iteration scheme, but we believe that our analytic expressions give a better insight into the dynamics of the system near resonance.

In Sec. V, our iteration procedure is extended to transition amplitudes and compared with the recent version of the continued-fraction method proposed by Gontier, Rahman, and Trahin.² It turns out that even though both methods sum similar classes of diagrams, our method is more rapidly convergent.

II. PHASE REPRESENTATION OF STATE VECTORS AND OPERATORS

Let us consider the quantum description of the electromagnetic radiation, decomposed into discrete modes. For each mode we will represent the state vectors by square-integrable wave functions $\Psi(\phi)$, where the phase ϕ varies from 0 to 2π ,

$$|\Psi\rangle \to \Psi(\phi). \tag{1}$$

The scalar product is defined by

$$\langle \Phi | \Psi \rangle = \int_{0}^{2\pi} \frac{d\phi}{2\pi} \Phi^{*}(\phi) \Psi(\phi).$$
 (2)

The photon number states will be represented by harmonic functions,

$$|n+m\rangle - e^{im\phi}, \qquad (3)$$

where the reference point n can be, in principle, arbitrary, but later will be assumed to be a very large number. The annihilation and creation operators, acting on functions $\Psi(\phi)$, can be represented in the following way:

$$a \rightarrow e^{-i\phi} \left(n + \frac{1}{i} \frac{\partial}{\partial \phi} \right)^{1/2},$$
 (4a)

$$a^{\dagger} - \left(n + \frac{1}{i} \frac{\partial}{\partial \phi}\right)^{1/2} e^{i\phi},$$
 (4b)

$$a^{\dagger}a \rightarrow n + \frac{1}{i} \frac{\partial}{\partial \phi}.$$
 (4c)

14

1101

Formulas (1)-(4) define what will be called the phase representation. Of course, our phase representation does not remove known difficulties connected with the nonexistence of the phase operator.³ In our representation, harmonic functions $e^{im\phi}$ with m < -n do not represent physical states and, as a result, the operation of multiplication by $e^{-ik\phi}$ is not defined on all wave functions. In our study we will consider only problems with a very large number of photons and the multiplication by $e^{-ik\phi}$ (for not too large k's) will be well defined.

More precisely, in what follows we will restrict ourselves to a subspace $M(n, m_0)$ spanned by only those basis vectors $|n+m\rangle$ for which $|m| \le m_0$, m_0 and n are fixed, and $m_0 \ll n$. This restriction is justified in most cases of physical interest, when an intense photon beam interacts with a microscopic system whose ability to deplete the beam is limited. The annihilation and creation operators in the phase representation, when acting in $M(n, m_0)$, can be approximated by the first few terms of the expansion with respect to 1/n,

$$a = e^{-i\phi}\sqrt{n}\left(1 + \frac{1}{2ni}\frac{\partial}{\partial\phi} + \cdots\right),$$
 (5a)

$$a^{\dagger} = \sqrt{n} \left(1 + \frac{1}{2ni} \frac{\partial}{\partial \phi} + \cdots \right) e^{i\phi}.$$
 (5b)

This expansion will be shown to be most useful in the study of dynamical resonance properties, but it can also be applied to other problems (an example is given in Appendix A). In the study of the dynamical problems we will assume, for simplicity, that the interaction is linear in the field, and we will restrict ourselves to a single mode of radiation. The Hamiltonian for the atom-field system acting in the subspace $M(n, m_0)$ will be approximated by

$$H = \hbar \omega \left(n + \frac{1}{i} \frac{\partial}{\partial \phi} \right) + H_a + \mathbf{j} \cdot \mathbf{\vec{A}}(\phi), \tag{6}$$

where H_a is the Hamiltonian of the atomic system, \mathbf{j} is the current operator, $\mathbf{A}(\phi)$ is the potential operator in the phase representation,

$$\vec{\mathbf{A}}(\phi) = \sqrt{n} \left(\mathbf{\tilde{f}} e^{-i\phi} + \mathbf{\tilde{f}}^* e^{i\phi} \right), \tag{7}$$

and \tilde{f} is a mode function. In the formula (6) we have disregarded all terms which vanish when $n \rightarrow \infty$. The evolution operator in the Dirac picture has the standard form $(\hbar = 1)$,

$$U(t) = T \exp\left(-i \int_0^t dt' \,\mathbf{j}(t') \cdot \mathbf{\bar{A}}(t')\right), \tag{8}$$

where

$$\begin{aligned} \mathbf{j}(t) &= e^{iH_a t} \mathbf{j} e^{-iH_a t}, \\ \mathbf{\bar{A}}(t) &= \exp\left[i\omega t \left(n + \frac{1}{i} \frac{\partial}{\partial \phi}\right)\right] \mathbf{\bar{A}}(\phi) \\ &\times \exp\left[-i\omega t \left(n + \frac{1}{i} \frac{\partial}{\partial \phi}\right)\right] \\ &= \sqrt{n} (\mathbf{\bar{f}} e^{-i(\phi+\omega t)} + \mathbf{\bar{f}} * e^{i(\phi+\omega t)}) \\ &= \mathbf{\bar{A}}(\phi + \omega t). \end{aligned}$$
(9)

It is important to observe that as a result of our approximation in the Hamiltonian the evolution operator in the Dirac picture does not contain the differential operator $\partial/\partial\phi$.

The transition amplitude for an induced emission (m > 0) or absorption (m < 0) of |m| photons, while the atom undergoes the transition between states $|i\rangle$ and $|f\rangle$, is

$$\langle n+m | \langle f | U(t) | i \rangle | n \rangle$$

$$= \int_{0}^{2\pi} \frac{d\phi}{2\pi} e^{-im\phi} \langle f | U[t | A(\phi)] | i \rangle, \quad (10)$$

$$| m | \ll n,$$

where

$$U[t|A(\phi)] = T \exp\left(-i \int_0^t dt' \,\mathbf{j}(t') \cdot \mathbf{A}(\phi + \omega t)\right). \tag{11}$$

The operator $U[t|A(\phi)]$, treated as a given function of ϕ , has the form of the evolution operator of the atomic system interacting with an external field (the evolution operator in the semiclassical theory). The formula (10) for induced transition amplitudes coincides, therefore, with what we called in our earlier works⁴ the phase-average representation of transition amplitudes. Since formula (10) is valid for every pair $|i\rangle$ and $|f\rangle$ of atomic states, we can write it in the operator form in the space of atomic variables,

$$\langle (n+m) \left| U(t) \right| n \rangle = \int_{0}^{2\pi} \frac{d\phi}{2\pi} e^{-im\phi} U[t \left| A(\phi) \right], \quad (12)$$
$$|m| \ll n.$$

We will use this expression to study the time evolution of the density matrix reduced to the atomic system,

$$\rho_{a}(t) = \operatorname{Tr}_{f} \left[e^{-(i/\hbar)Ht} \rho(0) e^{(i/\hbar)Ht} \right]$$
$$= \sum_{m=-n}^{\infty} \langle (n+m) \left| e^{-(i/\hbar)Ht} \rho(0) e^{(i/\hbar)Ht} \right| (n+m) \rangle,$$
(13)

or equivalently

$$\rho_a(t) = e^{-(i/\hbar)H_a t} \rho_a^{int}(t) e^{(i/\hbar)H_a t},$$
(14)

where $\rho_a^{\mu\mu}(t)$ is the reduced atomic density operator in the Dirac picture,

$$\rho_a^{int}(t) = \operatorname{Tr}_f[U(t)\rho(0)U^{\dagger}(t)].$$
(15)

We will assume that at t=0 the state of the system is the following product state:

$$\rho(0) = \left| n \right\rangle \rho_a(0) \left\langle n \right| \,. \tag{16}$$

In addition we will assume that the changes in the

intensity of the photon beam, resulting from the interaction with the atomic system, are not large. It means that the transition amplitudes $\langle n+m | \langle f | U(t) | i \rangle | n \rangle$ are negligible, unless $|m| \ll n$. We expect also that the Fourier components of $\langle f | U[t | A(\phi)] | i \rangle$, as defined by the right-hand side of Eq. (10), fall off rapidly with the increase of |m|. Under these assumptions, we obtain

$$\rho_{a}^{int}(t) = \int_{0}^{2\pi} \frac{d\phi_{1}}{2\pi} \int_{0}^{2\pi} \frac{d\phi_{2}}{2\pi} \sum_{m=-n}^{\infty} e^{-im(\phi_{1}-\phi_{2})} U[t|A(\phi_{1})]\rho_{a}(0)U^{\dagger}[t|A(\phi_{2})], \qquad (17)$$

which, after the summation over m, gives, finally,

$$\rho_{a}(t) = \int_{0}^{2\pi} \frac{d\phi}{2\pi} \rho(\phi, t),$$
(18)

where

$$\rho(\phi, t) = e^{-(i/\hbar)H_a t} U[t|A(\phi)]\rho_a(0)U^{\dagger}[t|A(\phi)]e^{(i/\hbar)H_a t}.$$
(19)

The right-hand side of Eq. (18) has the form of the average over the phase of the field of the atomic density matrix, whose time evolution is calculated according to the semiclassical theory.

The averaging over the phase of the field (or equivalently over the initial time) has been often introduced in the past⁵ as an independent additional postulate, in order to recover the connection between the results of the semiclassical theory and the quantum theory. Here, the averaging over the phase follows, as a mathematical consequence, from our choice of the *n*-photon state as the initial state.

III. RESONANCE PROPERTIES OF A TWO-LEVEL SYSTEM

The Hamiltonian of the two-level system interacting with a single mode of radiation will be assumed to have the form

$$H = \frac{1}{2}\hbar\omega_0\sigma_3 + \hbar\omega a^{\dagger}a + \hbar\kappa\sigma_1(a+a^{\dagger}).$$
(20)

Such a Hamiltonian can be used, for example, in describing experiments⁶ on the magnetic interaction between two Zeeman levels of an atom and the radiation field. In order to use the results of Sec. II. we will write the Hamiltonian (20) in the phase representation, retaining only the leading terms in 1/n,

$$H = \frac{1}{2}\hbar\omega_0\sigma_3 + \hbar\omega\left(n + \frac{1}{i}\frac{\partial}{\partial\phi}\right) + 2\hbar\lambda\sigma_1\cos\phi, \qquad (21)$$

where $\lambda = \kappa \sqrt{n}$.

The density operator $\rho(\phi, t)$ given by Eq. (19) is a 2×2 matrix and can be written as a combination of Pauli matrices,

$$\rho(\phi, t) = \frac{1}{2} + \vec{\mathbf{p}}(\phi, t) \cdot \vec{\sigma}.$$
(22)

By differentiation of Eq. (19) with respect to t, we obtain the differential equations for the coefficients $p_i(\phi, t)$,

$$\dot{p}_1 = -\omega_0 p_2, \tag{23a}$$

$$\dot{p}_2 = \omega_0 p_1 - 4\lambda \cos(\phi + \omega t) p_3, \qquad (23b)$$

$$\dot{p}_3 = 4\lambda \cos(\phi + \omega t)p_2, \qquad (23c)$$

which are equivalent to the following equations for the Laplace transforms of p_i 's:

$$z\tilde{p}_1 = p_1(t=0) - \omega_0 \tilde{p}_2,$$
 (24a)

$$z\tilde{p}_2 = p_2(t=0) + \omega_0 \tilde{p}_1 - 2\lambda (e^{-i\phi}\Delta_+ + e^{i\phi}\Delta_-)\tilde{p}_3, \quad (24b)$$

$$z\tilde{p}_3 = p_3(t=0) + 2\lambda(e^{-i\phi}\Delta_+ + e^{i\phi}\Delta_-)\tilde{p}_2, \qquad (24c)$$

where

$$\tilde{p}_i(\phi, z) = \int_0^\infty dt \, e^{-zt} \, p_i(\phi, t), \tag{25}$$

and where the operators Δ_{+} and Δ_{-} shift the argument z by $+i\omega$ and $-i\omega$, respectively, according to the formula

$$\Delta_{\pm}f(z) = f(z \pm i\omega). \tag{26}$$

We will be interested in the resonance properties of the two-level system. Such properties were extensively studied both theoretically and experimentally. We begin with a new definition of the resonance, which gives the same results as those obtained by other authors. We say that the atomic system is in resonance with the radiation field if the energy fluctuations in the atomic system reach their maximal value. For a twolevel atom the energy operator is proportional to σ_3 , and therefore our resonance condition can be written in the form

$$\langle (\sigma_3 - \langle \sigma_3 \rangle)^2 \rangle = 1 - \langle \sigma_3 \rangle^2 = \max,$$
 (27)

where the averaging symbol denotes both the en-

semble and the time averaging,

$$\langle \sigma_3 \rangle \equiv \langle \operatorname{Tr}[\rho_a(t)\sigma_3] \rangle_{\text{time av.}}$$
$$\equiv \lim_{T \to \infty} \frac{1}{T} \int_0^T dt \operatorname{Tr}[\rho_a(t)\sigma_3].$$
(28)

From Eqs. (18) and (22) we obtain

$$\langle \sigma_3 \rangle = \overline{p}_3 = \lim_{T \to \infty} \frac{1}{T} \int_0^T dt \int_0^{2\pi} \frac{d\phi}{2\pi} \, p_3(\phi, t), \tag{29}$$

and therefore condition (27) means that at the resonance, $(\overline{p}_3)^2$ attains its lowest value. We will show in Sec. IV that this minimal value is equal to zero. The condition $\overline{p}_3 = 0$ has been used by other authors⁷ to determine the position of the resonance.

We will assume that initially there is no coherence in the atomic system,

$$p_1(t=0) = 0 = p_2(t=0),$$
 (30)

which corresponds to a typical experimental situation. With the use of the initial condition (30), we can write the solution of Eqs. (24) for $\tilde{p}_3(\phi, z)$ in the form

$$\tilde{p}_{3}(\phi, z) = R(\phi, z)p_{3}(t=0),$$
 (31)

where

$$R(\phi, z) = [A(z) + B_{+}(\phi, z) + B_{-}(\phi, z)]^{-1}$$
(32a)

$$A(z) = z + \Delta_{+}F\Delta_{-} + \Delta_{-}F\Delta_{+}, \qquad (32b)$$

$$B_{+}(\phi, z) = e^{-2i\phi} \Delta_{+} F \Delta_{+} , \qquad (32c)$$

$$B_{-}(\phi, z) = e^{2i\phi} \Delta_{-} F \Delta_{-}, \qquad (32d)$$

$$F(z) = 4\lambda^2 z / (z^2 + \omega_0^2).$$
 (32e)

Since $R(\phi, z)$ contains the shifting operators Δ_{\pm} , the expression (31) can be regarded only as a formal solution. In Sec. IV we will develop a systematic approximation procedure to evaluate $R(\phi, z)$. Once we know $R(\phi, z)$, we can determine the position of the resonance $[p_3(t=0) \text{ is just a number}]$ from the formula

$$\operatorname{Res}_{z=0} \int_{0}^{2\pi} \frac{d\phi}{2\pi} R(\phi, z) = 0.$$
 (33)

In deriving Eq. (33) we have used the fact⁸ that the time average of the function (i.e., its timeindependent component) is given as the residue of its Laplace transform at the point z = 0. The frequencies of the time-dependent components of $p_3(\phi, t)$ are determined by the positions of the poles of $R(\phi, z)$ (different than z = 0) and their amplitudes are given by the corresponding residues.

IV. NEW APPROXIMATION METHOD AND THE BLOCH-SIEGERT SHIFT

From the definition (32a) of $R(\phi, z)$ we obtain the following operator equation:

$$R = A^{-1} - A^{-1}(B_{+} + B_{-})R.$$
(34)

The solution of this equation, in the form of a finite number of terms obtained by the simple iteration

$$R = A^{-1} - A^{-1}(B_{+} + B_{-})A^{-1} + A^{-1}(B_{+} + B_{-})A^{-1} - \cdots, \qquad (35)$$

diverges at the resonance. We propose to replace the simple iteration by an improved iteration procedure (IIP) which overcomes this difficulty. This procedure is suggested by the observation that only terms independent of ϕ contribute to the integral in Eq. (33). In the IIP, in order to obtain successive approximate expressions R_n for $R(\phi, z)$, we alternately iterate the operator equation for R, separate the part explicitly independent of ϕ , and use it as a new "free propagation" term.

In the zeroth approximation we obtain simply $R_0 = A^{-1}$ as the only part of Eq. (34) which is explicitly independent of ϕ . Next, we iterate Eq. (34) once and write it in the form

$$R = R_1 - R_1 (B_+ + B_-) R_0 + R_1 (B_+ R_0 B_+ + B_- R_0 B_-) R, \qquad (36)$$

where

$$R_1 = (R_0^{-1} - B_+ R_0 B_- - B_- R_0 B_+)^{-1}.$$
(37)

If we now retain only terms which are explicitly ϕ independent, we obtain the first approximation R_1 to the operator R. The second term on the right-hand side of Eq. (36) can be dropped because it will never lead to nonvanishing contributions to the integral over ϕ in Eq. (33). The third term, however, will lead to ϕ -independent contributions to R in higher approximations. To see this, we iterate Eq. (36) once more and write it in the form analogous to Eq. (36),

$$R = R_2 + R_2 (B_* R_0 B_* R_1 B_* R_0 B_* + B_R B_R B_R B_R B_R B_R)R,$$
(38)

where

$$R_{2} = (R_{1}^{-1} - B_{*}R_{0}B_{*}R_{1}B_{*}R_{0}B_{-} - B_{*}R_{0}B_{*}R_{1}B_{*}R_{0}B_{*})^{-1},$$
(39)

and we have again dropped the irrelevant ϕ -dependent term. The only explicitly ϕ -independent term in Eq. (38) is R_2 , which in the IIP serves as the second approximation to R. The third approximation to R is

$$R_{3} = (R_{2}^{-1} - B_{*}R_{0}B_{*}R_{1}B_{*}R_{0}B_{*}R_{2}B_{*}R_{0}B_{*}R_{1}B_{*}R_{0}B_{*}$$
$$- B_{*}R_{0}B_{*}R_{1}B_{*}R_{0}B_{*}R_{2}B_{*}R_{0}B_{*}R_{1}B_{*}R_{0}B_{*})^{-1}.$$
(40)

The (n+1)th approximation to R can be obtained with the use of the following recurrence formulas,

$$R_{n+1}^{-1} = R_n^{-1} - B_n^+ R_n B_n^- - B_n^- R_n B_n^+,$$
(41a)

$$B_{n+1}^{+} = B_n^{+} R_n B_n^{+}, \tag{41b}$$

$$B_{n+1}^{-} = B_n^{-} R_n B_n^{-}, \tag{41c}$$

where

14

$$R_0 = A^{-1}, \quad B_0^+ = B_+, \quad B_0^- = B_-.$$
 (42)

It is important to observe that all R_n are ordinary functions of z, because of the pairwise cancellation of the shifting operators Δ_+ and Δ_- , appearing in equal number in operators B_n^+ and B_n^- . Obviously, all R_n are ϕ independent, $R_n(\phi, z) = R_n(z)$, whereas the B_n^\pm are proportional to $e^{\pi 2^{n+1}i\phi}$.

To obtain the *n*th approximation to R we must set $B_{n+1}^{\pm}=0$. This means that the *n*th step of the IIP consists in restricting the space of relevant states to the subspace $M(n, 2^{n+1} - 1)$, defined in Sec. II. The position of the resonance is determined by locating the pole in ω_0 of the expression $dR^{-1}(z)/dz$ evaluated at z=0. It may seem at first, when we look at Eq. (41a), that the (n+1)th approximation will still have poles characteristic of the *n*th approximation, coming from R_n^{-1} . In actual fact (this is confirmed by a detailed calculation below), the terms $-B_n^*R_nB_n^* - B_n^*R_nB_n^*$ not only introduce new poles at corrected positions, but also cancel exactly the old poles.

Now we will study in detail the results obtained by the IIP. In the zeroth step of the IIP we obtain

$$R_0^{-1}(z) = z + F(z + i\omega) + F(z - i\omega)$$

= $z \{ 1 + 8\lambda^2 (z^2 + \omega_0^2 + \omega^2) \times [z^4 + 2z^2 (\omega_0^2 + \omega^2) + (\omega_0^2 - \omega^2)^2]^{-1} \}, (43)$

and the resonance condition (33) has the form

$$[1+8\lambda^2(\omega_0^2+\omega^2)(\omega_0^2-\omega^2)^{-2}]^{-1}=0.$$
(44)

Equation (44) gives the correct position of the resonance $\omega_0^2 = \omega^2$ in the zeroth order in λ . In the first step of the IIP we obtain

$$R_{1}^{-1}(z) = z + F(z + i\omega) + F(z - i\omega) - F^{2}(z + i\omega)[z + 2i\omega + F(z + i\omega) + F(z + 3i\omega)]^{-1}$$

- $F^{2}(z - i\omega)[z - 2i\omega + F(z - i\omega) + F(z - 3i\omega)]^{-1},$ (45)

which leads to the following expression for the time-independent component of $p_3(t)$:

$$\langle p_{3}(t) \rangle_{\text{time av.}} = (W_{1})^{2} \{ [(\omega_{0}^{2} - \omega^{2})(\omega_{0}^{2} - 9\omega^{2} + 6\lambda^{2}) + 2\lambda^{2}(\omega_{0}^{2} - 9\omega^{2})]^{2} \\ + 8\lambda^{2} [(\omega_{0}^{2} + \omega^{2})(\omega_{0}^{2} - 9\omega^{2} + 6\lambda^{2})^{2} + \lambda^{2}(\omega_{0}^{2} - 9\omega^{2})^{2} + 4\lambda^{4}(\omega_{0}^{2} + 9\omega^{2})]^{-1} p_{3}(t=0).$$

$$(46)$$

The resonance condition (33) reads

$$W_1 \equiv (\omega_0^2 - \omega^2)(\omega_0^2 - 9\omega^2 + 6\lambda^2) + 2\lambda^2(\omega_0^2 - 9\omega^2) = 0.$$

(47)

The solution of Eq. (47),

$$\omega_0^2 = 5\omega^2 - 4\lambda^2 - 4(\omega^4 - \lambda^2\omega^2 + \lambda^4)^{1/2}, \qquad (48)$$

reproduces correctly the shift of the position of the main resonance (the Bloch-Siegert shift) up to the fourth order in λ .

We can also compare (48) with the known exact result⁹ for the Bloch-Siegert shift obtained for the vanishing level-splitting field ($\omega_0 = 0$). In this case, the position of the resonance is found from the condition

$$J_{0}(4\lambda/\omega) = 0, \qquad (49)$$

and therefore the resonance value of λ/ω for the main resonance (one-photon resonance) is

$$\lambda/\omega = 0.601\ 206\ 389,\tag{50}$$

the exact value. Setting $\omega_0 = 0$ in Eq. (48) we obtain

$$\lambda/\omega = 0.612,\tag{51}$$

in the first step in the IIP. The second solution of Eq. (47),

$$\omega_0^2 = 5\omega^2 - 4\lambda^2 + 4(\omega^4 - \lambda^2\omega^2 + \lambda^4)^{1/2},$$
 (52)

reproduces the position of the three-photon resonance in the zeroth order in $\lambda.$

In the second step of the IIP we obtain

$$R_{2}^{-1}(z) = R_{0}^{-1}(z) - F^{2}(z + i\omega)R_{0}(z + 2i\omega) - F^{2}(z - i\omega)R_{0}(z - 2i\omega) - F^{2}(z + i\omega)F^{2}(z + 3i\omega)R_{0}^{2}(z + 2i\omega)[R_{0}^{-1}(z + 4i\omega) - F^{2}(z + 3i\omega)R_{0}(z + 2i\omega) - F^{2}(z + 5i\omega)R_{0}(z + 6i\omega)]^{-1} - F^{2}(z - i\omega)F^{2}(z - 3i\omega)R_{0}^{2}(z - 2i\omega)[R_{0}^{-1}(z - 4i\omega) - F^{2}(z - 3i\omega)R_{0}(z - 2i\omega) - F^{2}(z - 5i\omega)R_{0}(z - 6i\omega)]^{-1},$$
(53)

which leads to

$$\langle p_3(t) \rangle_{\text{time av.}} = (W_2)^2 p_3(t=0) / D(\omega_0, \omega, \lambda),$$
 (54)

where $D(\omega_0, \omega, \lambda)$ is a positive function of ω_0 , ω , and λ whose explicit form is given in Appendix B. The resonance condition reads

$$W_{2} = \left\{ \left[1 + 5\lambda^{2}(\omega_{0}^{2} - 25\omega^{2})^{-1} - 50\lambda^{4}(\omega_{0}^{2} - 25\omega^{2})^{-2} \left(3 + \frac{14\lambda^{2}}{\omega_{0}^{2} - 49\omega^{2}} + \frac{10\lambda^{2}}{\omega_{0}^{2} - 25\omega^{2}} \right)^{-1} \right] (\omega_{0}^{2} - 9\omega^{2}) + 3\lambda^{2} \right\} W_{1} - 18\lambda^{4}(\omega_{0}^{2} - \omega^{2}) = 0.$$

$$(55)$$

As we have previously pointed out,¹⁰ the positions of the one- and three-photon resonances, determined from our Eq. (55), agree very well with the numerical results of Stenholm¹ and also with the analytic result of Swain.¹⁴ To test the accuracy of the second step in our approximation scheme, we will again compare the resonance value of λ/ω at $\omega_0 = 0$ with the exact result,

$$\lambda/\omega = 0.601\ 206\ 4,\tag{56}$$

in the second step in the IIP. Since this value differs from the exact value (50) only by less than 1 ppm, there is no point of going beyond the second step in the IIP in the study of the main resonance.

We can also derive from (55) the perturbative formula for the Bloch-Siegert shift up to the 12th order in λ ,

$$\frac{\omega_0^2}{\omega^2} = 1 - 2\frac{\lambda^2}{\omega^2} - \frac{3}{2}\frac{\lambda^4}{\omega^4} - \frac{21}{16}\frac{\lambda^6}{\omega^6} - \frac{63}{64}\frac{\lambda^8}{\omega^8} - \frac{193}{384}\frac{\lambda^{10}}{\omega^{10}} + \frac{4237}{110592}\frac{\lambda^{12}}{\omega^{12}}.$$
(57)

The first six terms in this expansion coincide with the result of Ahmad and Bullough,¹¹ but the 12th-order term, to our knowledge, is obtained here for the first time.

One can also use our approximate analytic formulas for R(z) to determine the time-dependent components of $p_3(t)$. Already from the zerothstep expression (43) for R(z) we get four frequencies obtained recently by Ahmad¹² by a different method.

V. NEW APPROXIMATION METHOD FOR THE TRANSITION AMPLITUDES

In this section we will show how to extend our improved iteration procedure to the calculation of the transition amplitudes, and we compare it with the recent version of the continued-fraction method proposed by Gontier, Rahman, and Trahin.¹³ This method uses the resolvent operator G(E). The basic equation obeyed by G(E), in the notation of Ref. 2, is

$$G(E) = G_0(E) + G_0(E)(V^* + V^*)G(E),$$
(58)

and it has clearly the same general form as our Eq. (34) for R(z). Therefore we can apply the IIP to calculate the diagonal part of the resolvent operator in the photon space. In complete analogy with formulas (37), (39), and (40) we obtain now successive approximate expressions, G_1 , G_2 , and G_3 , for the diagonal part of the resolvent operator:

$$G_1 = (G_0^{-1} - V^+ G_0 V^- - V^- G_0 V^+)^{-1},$$
(59a)

$$G_2 = (G_1^{-1} - V^+G_0V^+G_1V^-G_0V^- - V^-G_0V^-G_1V^+G_0V^+)^{-1},$$
(59b)

$$G_{3} = (G_{2}^{-1} - V^{*}G_{0}V^{*}G_{1}V^{*}G_{0}V^{*}G_{2}V^{-}G_{0}V^{-}G_{1}V^{-}G_{0}V^{-}$$
$$- V^{-}G_{0}V^{-}G_{1}V^{-}G_{0}V^{-}G_{2}V^{*}G_{0}V^{*}G_{1}V^{*}G_{0}V^{*})^{-1}.$$
(59c)

These expressions are to be compared with the corresponding expressions (denoted by us by G_1^F , G_2^F , and G_3^F) obtained in the first, second, and third step of the continued-fraction method of Ref. 2:

$$G_1^F = [1 - T]^{-1} G_0, (60a)$$

$$G_{2}^{F} = \left(1 - T - Y\frac{1}{1 - T}X - X\frac{1}{1 - T}Y\right)^{-1}G_{0}, \quad (60b)$$

$$G_{3}^{F} = \left[1 - T - Y\left(1 - T - Y\frac{1}{1 - T}X\right)^{-1}X - X\left(1 - T - X\frac{1}{1 - T}Y\right)^{-1}Y\right]^{-1}G_{0}, \quad (60c)$$

where

1106

$$T = G_0 V^* G_0 V^- + G_0 V^- G_0 V^+, (61a)$$

$$X = G_0 V^* G_0 V^*, (61b)$$

$$Y = G_0 V G_0 V. \tag{61c}$$

One can easily check that our expressions G_1 and G_2 coincide with their counterparts G_1^F and G_2^F , but beginning with the third step the two methods differ. Using the notation (61) we write G_3 in the form

$$G_{3} = \left[1 - T - X\frac{1}{1 - T}Y - Y\frac{1}{1 - T}X - X\frac{1}{1 - T}X\left(1 - T - X\frac{1}{1 - T}Y - Y\frac{1}{1 - T}X\right)^{-1}Y\frac{1}{1 - T}Y - Y\frac{1}{1 - T}Y\right]$$
$$- Y\frac{1}{1 - T}Y\left(1 - T - X\frac{1}{1 - T}Y - Y\frac{1}{1 - T}X\right)^{-1}X\frac{1}{1 - T}X^{-1}G_{0}.$$
 (62)

This is clearly different from G_3^F , but by simple algebraic manipulation it can be shown to coincide with G_4^F obtained in the fourth step of the continuedfraction method. In general, the *n*th step of our improved iteration procedure coincides with the 2^{n-1} th step of the continued-fraction method of Ref. 2.

Our procedure therefore is more rapidly convergent than the continued-fraction method, by which we mean that the *n*th step of the IIP automatically includes a summation over many terms obtained by the continued-fraction method; the number of these terms grows exponentially with *n*. There is a price to be paid for this improved convergence. For example, in order to calculate the residue of $R_n(z)$ at z = 0 we must know all the functions $R_0(z), \ldots, R_{n-1}(z)$, and not only their residues at z = 0. This complication may render our method inconvenient for numerical calculations, but with its use we were able in Sec. IV to evaluate analytically the Bloch-Siegert shift with an accuracy exceeding that of all previous authors.

Note added in proof. Our improved iteration procedure, when applied to transition amplitudes in the simplest case of the two-level system, is equivalent to the method developed recently by P. R. Fontana and P. Thomann [Phys. Rev. A <u>13</u>, 1512 (1976)].

ACKNOWLEDGMENTS

The authors would like to thank Professor R. Bonifacio, Professor P. Stehle, and Professor M. Trahin for valuable discussions and suggestions.

APPENDIX A

Here, as an illustration of the phase representation method, we will find the wave function $\Psi_{\alpha}(\phi)$ of the coherent state in the subspace $M(n, m_0)$. To this end we solve the differential equation

$$e^{-i\phi}\sqrt{n}\left(1+\frac{1}{2ni}\frac{\partial}{\partial\phi}\right)\Psi_{\alpha}(\phi)=\alpha\Psi_{\alpha}(\phi).$$
 (A1)

The normalized solution of this equation is

$$\Psi_{\alpha}(\phi) = I_0(4n |\alpha|)^{-1/2} \exp(2\sqrt{n} \alpha e^{i\phi} - 2in\phi). \quad (A2)$$

This wave function will be a good approximation of the exact wave function for the coherent state if and only if

 $|\alpha| \simeq \sqrt{n}$,

because only then are contributions from higher terms in the expansion (5a) of a small. Since n is assumed to be large, the wave function $\Psi_{\alpha}(\phi)$ is effectively a Gaussian, centered around the phase of α ($\alpha = |\alpha| e^{-i\psi}$),

$$\Psi_{\alpha}(\phi) \simeq (8\pi\sqrt{n} |\alpha|)^{1/8} e^{-2in\psi} e^{-(\psi-\phi)^2 \sqrt{n} |\alpha|}.$$
 (A3)

APPENDIX B

In this Appendix we give in full detail the function $D(\omega_0, \omega, \lambda)$, which appears in Eq. (54):

$$D(\omega_{0}, \omega, \lambda^{2}) \equiv (W_{2})^{2} + 8\lambda^{2}(\omega_{0}^{2} + \omega^{2})[18\lambda^{4} + C(\omega_{0}^{2} - 9\omega^{2} + 6\lambda^{2}]^{2} + 8\lambda^{4}(\omega_{0}^{2} - 9\omega^{2})C^{2}(\omega_{0}^{2} - 9\omega^{2} + 4\lambda^{2}) - 192\lambda^{8}(\omega_{0}^{2} + 9\omega^{2})C + 72\lambda^{8}(\omega_{0}^{2} - 9\omega^{2})^{2} + 288\lambda^{10}(\omega_{0}^{2} - 9\omega^{2})[1 + (\omega_{0}^{2} - 9\omega^{2})(\omega_{0}^{2} + 25\omega^{2})(\omega_{0}^{2} - 25\omega^{2})^{-2}] + 72\lambda^{12} \bigg[-80\lambda^{4}(\omega_{0}^{2} + 25\omega^{2})(\omega_{0}^{2} - 9\omega^{2})^{2}(\omega_{0}^{2} - 25\omega^{2})^{3} \bigg(3 + \frac{14\lambda^{2}}{\omega_{0}^{2} - 49\omega^{2}} + \frac{10\lambda^{2}}{\omega_{0}^{2} - 25\omega^{2}} \bigg)^{-1} + 100\lambda^{4}(\omega_{0}^{2} - 9\omega^{2})^{2}(\omega_{0}^{2} - 25\omega^{2})^{-2} \bigg(1 + \frac{4\lambda^{2}(\omega_{0}^{2} + 49\omega^{2})}{(\omega_{0}^{2} - 49\omega^{2})^{2}} + \frac{4\lambda^{2}(\omega_{0}^{2} + 25\omega^{2})}{(\omega_{0}^{2} - 25\omega^{2})^{2}} \bigg) \times \bigg(3 + \frac{14\lambda^{2}}{\omega_{0}^{2} - 49\omega^{2}} + \frac{10\lambda^{2}}{\omega_{0}^{2} - 25\omega^{2}} \bigg)^{-2} \bigg],$$
(B1)

14

2

(B3)

where

$$C = \left[1 + 5\lambda^2(\omega_0^2 - 25\omega^2)^{-1} - 50\lambda^4(\omega_0^2 - 25\omega^2)^{-2}\left(3 + \frac{14\lambda^2}{\omega_0^2 - 49\omega^2} + \frac{10\lambda^2}{\omega_0^2 - 25\omega^2}\right)\right](\omega_0^2 - 9\omega^2) + 3\lambda^2.$$
(B2)

The function C is related to W_2 by

 $W_2 = W_1 C - 18\lambda^4 (\omega_0^2 - \omega^2).$

- ⁸G. Doetsch, Handbuch der Laplace-Transformation
- ¹S. Stenholm, J. Phys. B <u>5</u>, 878 (1972); <u>5</u>, 890 (1972). ²Y. Gontier, N. K. Rahman, and M. Trahin, Phys. Lett. 54A, 341 (1975).

*Research partly supported by NSF Grant No. GF 36217.

- ³P. A. M. Dirac, Proc. R. Soc. A <u>114</u>, 243 (1927); P. Carruthers and M. M. Nieto, Phys. Rev. Lett. 14, 387 (1965); Rev. Mod. Phys. 40, 411 (1968). See also a recent review article by H. Paul, Fortschr. Phys. 22, 657 (1974).
- ⁴I. Bialynicki-Birula and Z. Bialynicka-Birula, Phys. Rev. A 8, 3146 (1973); and Quantum Electrodynamics (Pergamon, Oxford, 1975), p. 213.
- ⁵J. H. Shirley, Phys. Rev. <u>138</u>, B979 (1965). See also Ref. 1.
- ⁶E. Arimondo and G. Moruzzi, J. Phys. B <u>6</u>, 2382 (1973).
- ⁷F. Ahmad and R. K. Bullough, J. Phys. B7, L275 (1974).

- (Birkhäuser, Basel, 1950), Vol. 1, p. 462. ⁹C. Cohen-Tannoudji, J. Dupont-Roc, and C. Fabre,
- J. Phys. B 6, L218 (1973); P. Hannaford, D. T. Pegg, and G. W. Series, *ibid*. <u>6</u>, L222 (1973).
- ¹⁰Z. Bialynicka-Birula and I. Bialynicki-Birula, Nuovo Cimento Lett. 15, 627 (1976).
- ¹¹See Ref. 7, and also F. T. Hioe and J. H. Eberly, Phys. Rev. A 11, 1358 (1975).
- ¹²F. Ahmad, Phys. Rev. A <u>12</u>, 1539 (1975).
- ¹³See Ref. 2. An earlier application of the continuedfraction method to transition amplitudes can be found in the paper by R. Gush and H. P. Gush, Phys. Rev. A 6, 129 (1972), and by S. Swain, J. Phys. A 8, 1277 (1975).
- ¹⁴S. Swain, J. Phys. B 7, 2363 (1974).