Dipole interaction of a multilevel system with a continuous-wave or Gaussian-pulsed laser

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An algorithm [G. F. Thomas and W. J. Meath, J. Phys. B **16**, 951 (1983)] for evaluating the evolution operator of a multilevel system dipole interacting with either a continuous-wave or Gaussian-pulsed source, based on its Riemann product integral representation in conjunction with Frazer's method of mean coefficients, is appraised and extended. Our extension allows for the inclusion of spontaneous decay or a dissociation (or ionization) channel and useful relations are derived for the eigenvalues of the evolution operator for both the continuous wave and the Gaussian pulse. For the latter we now admit an arbitrary phase in the field and we derive a simple relation for cascading the state amplitudes of the system over the duration of an arbitrary number of identical phase-coherent pulses. As part of our appraisal, improvements in the implementation of the algorithm are provided. For a two-level system, the evolution operator is given as the chronologically ordered product of readily computed 2×2 matrices while for multilevel system it is conveniently evaluated using a resolvent method based on Leverrier's algorithm for computing the Bateman matrices. The asymptotic behavior of the evolution operator and the fluence in the direction of propagation for both the continuous-wave and the Gaussian-pulsed sources are explored. Applications to model two-level systems illustrate the influence of the source's phase and interaction duration on resonant and nonresonant transition probabilities, respectively. The saturation of a two-level system by a train of resonant phase-coherent Gaussian pulses is demonstrated.

I. INTRODUCTION

The dynamics of multiphoton processes induced in atomic and molecular systems through their interaction with intense laser-light sources continues to receive much attention from both theoretical and experimental perspectives.[1] It is usual to assume that the Hamiltonian operator is periodic in time, this periodicity being determined by the dipole interaction of the system with a classical oscillating laser field, operating in either the continuous-wave mode or as a pulsed source, with such long duration pulses as to validate a slowly-varying-amplitude approximation in which the field is taken to be monochromatic and of fixed amplitude. The periodicity of the interaction limits the time interval over which one has to integrate the dynamical equations of motion and, when coupled with Floquet analysis,[2,3] allows one, for instance, to investigate the time-resolved and long-time-averaged behavior of a diatomic[4] or an anharmonic bond mode[5] of a polyatomic subject to vibrational or rotational-vibrational multiple-photon excitation. With the assumption of a constant-amplitude field, such expedients as the rotating-wave approximation,[6] various perturbative approximations,[7] the Magnus approximation[6,8] and rotating-frame transformations[9] can be exploited to advantage. For nanosecond pulses, the number of optical cycles occurring throughout the pulse's duration is $\sim10^2$—10^5 in the infrared region and the assumed slowly varying amplitude approximation, with its consequent simplification in the description of the dynamics of the underlying processes, may be justified. However, with the quest for increasingly shorter pulses—which have now entered the femtosecond domain[10]—the number of optical cycles sustained during the pulse is accordingly smaller and the modulating envelope associated with the pulse will vary rapidly with the period of oscillation of the field and must therefore be explicitly considered in any attempt to simulate the dynamics on such ultrashort time scales.

Recently,[11] an algorithm for evaluating the evolution operator for a multilevel system dipole interacting with either a continuous-wave or Gaussian-pulsed source, based on its Riemann product integral representation[12] in conjunction with Frazer's method[13] of mean coefficients, was presented. After applying the algorithm to model two-level systems, it was apologetically concluded that while it represented a viable alternative to other methods of constructing the evolution operator for a continuous-wave source, it was restricted in pulsed-source applications to those cases where the number of optical cycles occurring during the pulse's lifetime is small. The objective of this paper is to reappraise and extend this algorithm.

In Sec. II A, the algorithm is extended to allow for the phenomenological inclusion of spontaneous decay or a dissociative channel by admitting a non-Hermitian Hamiltonian to describe the "isolated" system and, without invoking the notion of unitarity, useful relations are derived for the eigenvalues of the evolution operator for both continuous-wave and Gaussian-pulsed sources. We now include an arbitrary phase in the specification of the Gaussian-modulated field and a simple relation for cascading the state amplitudes of the system over the duration of an arbitrary number of identical phase-coherent pulses is derived. Following our appraisal of the algorithm in Sec. II B, improvements in its implementation are described for both the two-level and the multilevel system; for the former, the evolution operator is constructed

as a time-ordered product of 2×2 matrices while for the latter, multiple use of a resolvent scheme affords an efficient method of evaluation. The asymptotic behavior of the evolution operator and the fluence in the direction of propagation for both the continuous-wave and the Gaussian-pulsed sources are briefly explored in Sec. II C. Section III details the results and attendant discussion of some illustrative applications to model two-level systems. The paper closes with some concluding remarks in Sec. IV.

Unless otherwise indicated, atomic units are used throughout this paper.

II. RIEMANN PRODUCT INTEGRAL REPRESENTATION OF THE EVOLUTION OPERATOR

A. Theory

The evolution of the state amplitude matrix $\mathbf{a}(t)$ of a multilevel (N, for example) system is governed by

$$\dot{\mathbf{a}}(t)=\mathbf{A}(t)\mathbf{a}(t)\ . \tag{2.1}$$

Here $\mathbf{a}(t)$ is an N-vector whose components $a_k(t)$ are the probability amplitudes for the stationary eigenstates $|k\rangle$, $k=1,2,\ldots,N$ of the isolated system and $\mathbf{A}(t)=-i\mathbf{H}(t)$, where $\mathbf{H}(t)$ is an $N\times N$ matrix representation of the system's Hamiltonian operator. (In general, throughout the paper lower-case boldface letters refer to vectors, capital boldface letters to matrices.) For an N-level system dipole interacting with a classical electric field $E(t)$, arbitrarily polarized along the z axis,

$$\mathbf{A}(t)=-i[\mathbf{\Omega}-E(t)\boldsymbol{\mu}_z]$$

in the Schrödinger picture while

$$\mathbf{A}(t)=iE(t)\exp[i\mathbf{\Omega}(t-t_0)]\boldsymbol{\mu}_z\exp[-i\mathbf{\Omega}(t-t_0)]$$

in the Dirac picture, where t_0 is the time the field is switched on. The $N\times N$ matrices $\boldsymbol{\mu}_z$ and $\mathbf{\Omega}$ have components $\mu_{lm}=\langle l\,|\,\mu_z\,|\,m\rangle$ and

$$\Omega_{lm}=(\omega_m-i\gamma_m/2)\delta_{lm}\ ,$$

respectively, where μ_{lm} is the dipole transition matrix element coupling the levels $|l\rangle$ and $|m\rangle$, μ_z being the z component of the dipole moment operator, and ω_m is the eigenenergy of $|m\rangle$ with corresponding phenomenological natural width γ_m. If $\gamma_m=0$ for $m=1,2,\ldots,N-1$ and is nonzero for $m=N$ then the non-Hermitian Hamiltonian $\sum_{m=1}^{N}\Omega_{mm}\,|m\rangle\langle m|$ in the $|m\rangle$ basis admits a dissociation (or ionization) channel through the uppermost level. For convenience we take μ_{lm}, $l,m=1,2,\ldots,N$ to be real.

The solution of Eq. (2.1) for the initial stationary state population $\mathbf{a}(t_0)$ is

$$\mathbf{a}(t)=\mathbf{U}(t,t_0)\mathbf{a}(t_0)\ , \tag{2.2}$$

where the evolution operator $\mathbf{U}(t,t_0)$ is the integral matrix[14] for Eq. (2.1), i.e., the solution of Eq. (2.1) corresponding to the initial conditions $\mathbf{U}(t_0,t_0)=\mathbb{1}$. The integral matrix satisfies the identity

$$\det[\mathbf{U}(t,t_0)]=\exp\left[\int_{t_0}^{t}ds\ \mathrm{Tr}[\mathbf{A}(s)]\right]\ , \tag{2.3}$$

a consequence of which is that $\mathbf{U}(t,t_0)$ is nonsingular for all $t\geq t_0$. For $t_0\leq t'\leq t$, the integral matrix has the group property

$$\mathbf{U}(t,t_0)=\mathbf{U}(t,t')\mathbf{U}(t',t_0)\ . \tag{2.4}$$

It has been shown[15] that $\mathbf{U}(t,t_0)$ is bounded from above in the sense that

$$\hat{\mathbf{U}}(t,t_0)\leq\mathbb{1}+N^{-1}\mathbf{\Pi}\{\exp[N\Phi(t,t_0)]-1\}\ , \tag{2.5}$$

where $\mathbf{\Pi}$ is the Vandermonde matrix,

$$\Phi(t,t_0)=\int_{t_0}^{t}ds\,||\mathbf{A}(s)||\ ,$$

$||\mathbf{A}||$ is the Tschebyscheff norm of the operator $\mathbf{A}$, and the caret on $\mathbf{U}(t,t_0)$ signifies that we take the modulus of its matrix elements. The Tschebyscheff norm[16] of the N-vector $\mathbf{a}(t)$ is defined as $||\mathbf{a}(t)||=\max_{1\leq k\leq N}|a_k(t)|$. For the Hamiltonian operator of interest here, $\mathbf{A}(t)=-i\mathbf{H}(t)$ is continuous and hence bounded and integrable on $[t_0,t]$. The operator $\mathbf{A}(t)$ is bounded if for all $\mathbf{a}(t)$ in its domain there exists a constant K such that $||\mathbf{A}(t)\mathbf{a}(t)||\leq K$. The greatest lower bound of all possible K's is the norm of $\mathbf{A}(t)$, i.e.,

$$||\mathbf{A}(t)||=\sup||\mathbf{A}(t)\mathbf{a}(t)||/||\mathbf{a}(t)||\ .$$

The Tschebyscheff norm of the $N\times N$ matrix $\mathbf{A}(t)$ is

$$||\mathbf{A}(t)||=\max_{1\leq k\leq N}\sum_{l=1}^{N}|A_{kl}|\ .$$

For arbitrary $\mathbf{A}$ and $\mathbf{B}$, $||\mathbf{A}+\mathbf{B}||\leq||\mathbf{A}||+||\mathbf{B}||$, $||\mathbf{AB}||\leq||\mathbf{A}||||\mathbf{B}||$, and $\mathrm{Tr}(\mathbf{A})\leq||\mathbf{A}||$.

The field $E(t)$ is assumed to be of the form

$$E(t)=E^0f(t)\cos(\omega_0t+\phi)\ , \tag{2.6}$$

where E^0 is amplitude, ω_0 is the carrier frequency, and ϕ is the phase. The time-dependent pulse envelope $f(t)$ modulates the sinusoid. When $f(t)=1$, Eq. (2.6) is the ideal representation of a continuous-wave source of constant amplitude, fixed frequency, and fixed phase with a coherence time that is at least as large as the characteristic relaxation times for the N-level system. If, for example, one is interested in the multiple-photon excitation or dissociation of polyatomics[17] through use of intense fields or in the short-time-scale ($\lesssim 1$ ps) molecular dynamics and rate processes[18] in condensed phases, the light source must be pulsed. Both theoretical[19] and experimental[20] evidence suggest that, ideally, the output from a perfectly mode- and phase-locked pulsed laser consists of a train of Gaussian-amplitude sinusoids, each with a modulating pulse envelope $f(t)=\exp(-t^2/\tau_p^2)$, where τ_p is the approximate duration of the pulse. The pulse train originates from the beating of a fixed number of cavity modes (usually $\sim10^3$) having synchronized phases but oscillating at different frequencies and amplitudes. In practice, the individual pulses in the train, which are separated from each other by the cavity round-trip time (usually ~5 ns), vary in their characteristics (duration, spectral bandwidth, peak amplitude, and modulating envelope) from

the beginning to the end of the train. It is technically possible to isolate a suitable pulse from the train having sufficient power and well-defined parameters such as the peak amplitude E^0, mean carrier frequency ω_0, and a Gaussian modulating envelope $f(t)=\exp(-t^2/\tau_p^2)$ with an approximate epochal duration τ_p. The tradeoff in using pulsed lasers is the nonzero spectral bandwidth associated with the pulses. For frequencies $\omega\sim\omega_0$, the Gaussian pulse, which has a full width at half maximum (FWHM) temporal bandwidth $\Delta t=2(\ln 2)^{1/2}\tau_p$, transforms to a Gaussian profile

$$\mathcal{F}\{E(t)\}=\tfrac{1}{2}E^0\tau_p\pi^{1/2}\exp[-(\omega-\omega_0)^2\tau_p^2/4-i\phi]$$

with a FWHM spectral bandwidth $\Delta\omega=4(\ln 2)^{1/2}/\tau_p$ so that the bandwidth uncertainty product[21] for the pulse is $\Delta\omega\,\Delta t=8\ln 2$. In contrast, if $\tau_p\to\infty$ then $\Delta\omega\to 0$ and $\Delta t\to\infty$ so that the continuous-wave source sustains a monochromatic output at a constant amplitude over an arbitrarily long duration. For a continuous-wave source, the phase ϕ is not well defined and within the interval $0\le\phi\le\pi$ the field $E(t)$ initially ranges between adiabatic and sudden switchon. It is standard practice[15,22] to average computed physical properties of an N-level system interacting with a continuous-wave source over this arbitrary phase, a procedure that is equivalent to initial-time averaging.[3(b)] Pulsed light sources, however, by their epochal nature, are switched on suddenly, as in the case of a "rectangular-shaped" pulse, or are switched on adiabatically, as in the case of a "bell-shaped" pulse. In principle, the phase of a pulsed source is well defined and computed properties of the system interacting with the pulse(s) should not be averaged over the phase. Although the phase coherence between successive pulses in a train of time-delayed pulses can be important,[23] for excitation by a single isolated pulse one may define the time origin so that $\phi=0$.

To evaluate $\mathbf{a}(t)$ as given by Eq. (2.2) for arbitrary $\mathbf{a}(t_0)$, one may compute the evolution operator $\mathbf{U}(t,t_0)$ using an algorithm[11] based on its Riemann product integral representation[12] in conjunction with Frazer's method[13] of mean coefficients. This involves the discretization of the interval $[t_0,t]$ into a sufficient number n_p of subintervals such that

$$t_k=(t-t_0)/n_p+t_{k-1}$$

for $k=1,2,\ldots,n_p$ and $t_{n_p}=t$ and the evaluation of the time-ordered product of matrix exponentials

$$\mathbf{U}(t,t_0)=T\lim_{n_p\to\infty}\prod_{k=1}^{n_p}\exp\left[\int_{t_{k-1}}^{t_k}ds\,\mathbf{A}(s)\right] \tag{2.7}$$

with the implicit use of the group property of the evolution operator, Eq. (2.4), at adjacent subintervals. In Eq. (2.7), T is an operator that arranges the product in chronological order from right to left.

For a continuous-wave source, $E(t)$ is given by Eq. (2.6) with $f(t)=1$ and $\mathbf{A}(t)=\mathbf{A}(t+2\pi/\omega_0)$. Consequently, in the variable $0\le\theta=\omega_0 t+\phi\le 2\pi$, the solution of Eq. (2.2) for arbitrary initial conditions $\mathbf{a}(0)$ is given in Floquet form[2,3] by

$$\mathbf{a}(\theta+n\,2\pi)=\mathbf{L}(\theta)\exp[i\boldsymbol{\Delta}(\theta+n\,2\pi)]\mathbf{b}_0(\phi) \tag{2.8}$$

for $n=0,1,2,\ldots$. In Eq. (2.8), the Lyapunov matrix

$$\mathbf{L}(\theta)=\mathbf{U}(\theta,0)\mathbf{Z}\exp(-i\boldsymbol{\Delta}\theta)$$

and is of period 2π in θ,

$$\mathbf{b}_0(\phi)=\mathbf{Z}^{-1}\mathbf{U}(0,\phi)\mathbf{a}(0)\ ,$$

and $\mathbf{U}(2\pi,0)=\mathbf{Z}\boldsymbol{\Lambda}\mathbf{Z}^{-1}$, where $\boldsymbol{\Lambda}$ in a diagonal matrix containing the eigenvalues of $\mathbf{U}(2\pi,0)$ and whose corresponding orthonormal eigenvectors constitute the columns of $\mathbf{Z}$. The eigenvalues of $\mathbf{U}(2\pi,0)$ may be written as

$$\lambda_{kk}=|\lambda_{kk}|\exp(i\Delta_{kk}2\pi),\quad k=1,2,\ldots,N \tag{2.9}$$

where the Δ_{kk}'s are the entries in the diagonal characteristic exponent matrix $\boldsymbol{\Delta}$. Using Eqs. (2.3) and (2.9) one can easily show that

$$\sum_{k=1}^{N}(\omega_k+\omega_0\Delta_{kk})=0 \tag{2.10a}$$

and

$$\sum_{k=1}^{N}(\pi\gamma_k+\omega_0\ln|\lambda_{kk}|)=0\ , \tag{2.10b}$$

where ω_k and γ_k are the eigenenergy and natural width of $|k\rangle$, respectively. The time-resolved expectation value of an operator $\mathcal{O}$ of the system over an arbitrary number $n=0,1,2,\ldots$ of the optical cycles of the field is given by the usual trace prescription $\langle\mathcal{O}\rangle=\mathrm{Tr}[\mathcal{O}\boldsymbol{\rho}]$, where

$$\boldsymbol{\rho}(\theta+n\,2\pi)=\mathbf{a}(\theta+n\,2\pi)\mathbf{a}^{\dagger}(\theta+n\,2\pi)$$

is the density matrix. The argument of the matrix exponential in Eq. (2.7) has entries[24(a)]

$$A_{lm}^{(k)}=-\frac{i}{\omega_0}\{[\omega_l-(i/2)\gamma_l]\delta_{lm}(\theta_k-\theta_{k-1})-2\mu_{lm}E^0\sin[\tfrac{1}{2}(\theta_k-\theta_{k-1})]\times\cos[\tfrac{1}{2}(\theta_k+\theta_{k-1})]\} \tag{2.11}$$

for $l,m=1,2,\ldots,N$.

In the case of a Gaussian pulse, $E(t)$ is given by Eq. (2.6) with $f(t)=\exp(-t^2/\tau_p^2)$. The solution of Eq. (2.2), in the variable $\theta=\omega_0 t+\phi$, for arbitrary initial conditions $\mathbf{a}(\theta_0)$ is formally

$$\mathbf{a}(\theta)=\mathbf{U}(\theta,\theta_0)\mathbf{a}(\theta_0)\ .$$

If the system interacts with $n=1,2,\ldots$ identical phase-coherent pulses, then its state amplitude throughout the duration of the nth pulse is

$$\mathbf{a}^{(n)}(\theta)=\mathbf{U}(\theta,\theta_0)\mathbf{U}^{n-1}(+\infty,-\infty)\mathbf{a}(\theta_0) \tag{2.12}$$

which is the pulse's analog of Eq. (2.8). Using Eq. (2.3) one can easily show that

$$\sum_{k=1}^{N}\Delta_{kk}=\tfrac{1}{2}E^0\tau_p\pi^{-1/2}\exp(-\omega_0^2\tau_p^2/4)\cos\phi\sum_{k=1}^{N}\mu_{kk} \tag{2.13a}$$

and

$$\sum_{k=1}^{N} \ln |\lambda_{kk}| = 0 , \tag{2.13b}$$

where λ_{kk} and Δ_{kk} are related through Eq. (2.9) but here the λ's are the eigenvalues of $\mathbf{U}(+\infty,-\infty)$. As before, $\langle \mathcal{O} \rangle = \mathrm{Tr}[\mathcal{O}\rho]$ where

$$\rho(\theta,\theta_0) = \mathbf{a}(\theta)\mathbf{a}^{\dagger}(\theta)$$
$$= \mathbf{U}(\theta,\theta_0)\rho(\theta_0,\theta_0)\mathbf{U}^{\dagger}(\theta,\theta_0) .$$

The argument of the matrix exponential in Eq. (2.7) has entries[24(b)]

$$A_{lm}^{(k)} = \frac{i}{4}\mu_{lm}E^0\tau_p\pi^{1/2}(a_+ + a_-)$$
$$\times \exp[-(\phi^2 + i\omega_{lm}\omega_0\tau_p^2\theta_0)/\omega_0^2\tau_p^2] \tag{2.14}$$

for $l,m = 1,2,\ldots,N$, where

$$a_{\pm} = \exp(b_{\pm}^2/4\omega_0^2\tau_p^2)\{\mathrm{erf}[(\theta_k - \tfrac{1}{2}b_{\pm})/\omega_0\tau_p]$$
$$- \mathrm{erf}[(\theta_{k-1} - \tfrac{1}{2}b_{\pm})/\omega_0\tau_p]\} , \tag{2.15a}$$

$$b_{\pm} = 2\phi \pm i\omega_0\tau_p^2(\omega_0 \pm \omega_{lm}) , \tag{2.15b}$$

$\omega_{lm} = \omega_l - \omega_m - (i/2)(\gamma_l - \gamma_m)$ and $\mathrm{erf}(z)$ is the error function at an arbitrary point z in the complex plane.

As is well known,[25] the *ad hoc* ascription of the width γ_k to $|k\rangle$ in an attempt to include spontaneous decay continues to ignore the fact that the "isolated" N-level system is continually interacting with the surrounding quantized electromagnetic field. Further, the basis which diagonalizes the "unperturbed" Hamiltonian $\sum_{k=1}^{N}\omega_k |k\rangle\langle k|$ may no longer be appropriate.[26] If the time-varying field $E(t)$ is of such short duration that spontaneous emission can be ignored by setting $\gamma_k = 0$ for $k = 1,2,\ldots,N$ then the Euclidean norm[16] $||\mathbf{a}(t)|| = 1$ since $\mathbf{H}(t) = \mathbf{H}^{\dagger}(t)$ and from Eq. (2.2) it follows that $\mathbf{U}(t,t_0)$ is unitary for all $t \geq t_0$.

B. Constructive criticism

In previous applications[4(i),11,15] of Eq. (2.7) to two- and three-level systems, typical single-precision execution times for evaluating the evolution operator over one optical cycle with 180 subintervals was ~ 3 s on the Cyber 7300. For multilevel systems, execution times increase with the number of levels. The consumption of processor time is attributable to the calculation of $\exp(\mathbf{A}^{(k)})$ over $k = 1,2,\ldots,n_p$ subintervals, the underlying rate-determining stage in each subinterval being the diagonalization of $\mathbf{A}^{(k)}$, in particular, the evaluation of the matrix $\mathbf{Z}^{(k)}$ of eigenvectors (and its inverse) corresponding to the matrix $\mathbf{\Lambda}^{(k)}$ of eigenvalues of $\mathbf{A}^{(k)} = \mathbf{Z}^{(k)}\mathbf{\Lambda}^{(k)}(\mathbf{Z}^{(k)})^{-1}$. The choice of n_p is dictated on the one hand by the fulfillment at any local time $t \geq t_0$ of the inequality in Eq. (2.5) and, if one ignores spontaneous decay, by the unitarity of $\mathbf{U}(t,t_0)$, and on the other hand by the extent to which the relations in Eqs. (2.10) and (2.13) are satisfied for the continuous-wave and Gaussian-pulsed sources, respectively. Of course, for a continuous-wave source, the evolution operator is required only over the period of the Hamiltonian. However, for a Gaussian-pulsed source with an effective duration of $|t| \lesssim 4\tau_p$, one must employ the algorithm over each of $\sim 8\tau_p(\omega_0/2\pi)$ optical cycles occurring throughout the pulse's tenure. This severely restricts the algorithm's usefulness to values of τ_p and ω_0 such that $8\tau_p(\omega_0/2\pi)$ is not too large, particularly for $N \gg 2$. To further exacerbate matters, one is required to average[27] the computed expectation values of the N-level system interacting with a pulse over the spectral bandwidth, so that the evolution operator must be evaluated over the frequency range $|\omega - \omega_0| \leq \frac{1}{4}\Delta t^{-1}$. In previous applications, the exponential of $\mathbf{A}^{(k)}$ was achieved through

$$\exp(\mathbf{A}^{(k)}) = \mathbf{Z}^{(k)}\exp(\mathbf{\Lambda}^{(k)})(\mathbf{Z}^{(k)})^{-1}$$

using the highly efficient diagonalization sequence available in the EISPACK library,[28(a)] although the equivalent routines in the IMSL (International Mathematical and Statistical Library) package[28(b)] could also have been used. Fortunately, one can significantly expedite the use of Eq. (2.7) by adopting different approaches to the calculation of $\exp(\mathbf{A}^{(k)})$, $k = 1,2,\ldots,n_p$ for $N = 2$ and $N > 2$.

For a two-level system the conjugate eigenvalues $\alpha_k \pm \beta_k$ of $\mathbf{A}^{(k)}$ are readily determined as the roots of the characteristic equation, and use of the Sylvester-Lagrange theorem[13(b),14(b)] gives the exponential of $\mathbf{A}^{(k)}$ as

$$\exp(\mathbf{A}^{(k)}) = \begin{bmatrix} (A_{11}^{(k)} - \alpha_k)\sinh\beta_k/\beta_k + \cosh\beta_k & A_{12}^{(k)}\sinh\beta_k/\beta_k \\ A_{21}^{(k)}\sinh\beta_k/\beta_k & (A_{22}^{(k)} - \alpha_k)\sinh\beta_k/\beta_k + \cosh\beta_k \end{bmatrix} \exp(\alpha_k) . \tag{2.16}$$

Thus, for $N = 2$, Eqs. (2.7) and (2.16) give the evolution operator explicitly as a chronological product of n_p 2×2 matrices. Evaluation of $\mathrm{erf}(z) = 1 - \exp(-z^2)\omega(iz)$, for complex z, occurring in Eq. (2.14a) for Gaussian pulses, can be accomplished using the Gautschi algorithm[29] for the probability function ω which is available as subroutine MERRCZ of the IMSL package.[28(b)] Because of its relationship to the Voigt function, there are a number of efficient nonproprietary rational approximants available[30] for the evaluation of $\omega(z)$ over the entire z plane. Thus, for a two-level system dipole interacting with either a continuous-wave or Gaussian-pulsed source, the Riemann product integral algorithm for the evaluation of the evolution operator is readily implemented on any of the popular 16-bit microprocessors equipped with adequate memory and storage capacities.

When $N > 2$ one can conveniently compute[13(b),14(b)] the exponential of $\mathbf{A}^{(k)}$ as

$$\exp(\mathbf{A}^{(k)})=\sum_{l=1}^{N}\exp(\lambda_l^{(k)})\mathbf{N}_l^{(k)}\ , \tag{2.17}$$

where the $\lambda^{(k)}$'s are the nondegenerate eigenvalues of $\mathbf{A}^{(k)}$ and the $\mathbf{N}^{(k)}$'s are $N\times N$ Bateman coefficient matrices[31] with the projection, idempotant, and sum-rule properties

$$\mathbf{N}_l^{(k)}\mathbf{N}_m^{(k)}=0 \text{ if } l\neq m\ , \tag{2.18a}$$

$$(\mathbf{N}_l^{(k)})^m=\mathbf{N}_l^{(k)} \text{ for } m \text{ any positive integer}\ , \tag{2.18b}$$

and

$$\sum_{l=1}^{N}\mathbf{N}_l^{(k)}=\mathbb{1}\ , \tag{2.18c}$$

respectively. The eigenvalues of $\mathbf{A}^{(k)}$ can be computed using proprietary software available on main-frame machines. The Bateman matrices are expeditiously evaluated using Faddeev's modification[16(a)] of Laverrier's algorithm[32] as

$$\mathbf{N}_l^{(k)}=\sum_{m=1}^{N}(\lambda_l^{(k)})^{(N-m)}\mathbf{Z}_m^{(k)}\Big/\prod_{m\neq l}^{N}(\lambda_m^{(k)}-\lambda_l^{(k)})\ , \tag{2.19a}$$

where the $N\times N$ matrices $\mathbf{Z}_m^{(k)}$ are obtained recursively for $m=2,\ldots,N$ as

$$\mathbf{Z}_m^{(k)}=\mathbf{A}^{(k)}\mathbf{Z}_{m-1}^{(k)}+\theta_{m-1}^{(k)}\mathbb{1}\ , \tag{2.19b}$$

with $\mathbf{Z}_1^{(k)}=\mathbb{1}$, and the scalar $\theta^{(k)}$'s are given for $m=1,2,\ldots,N$ as

$$m\theta_m^{(k)}=-\mathrm{Tr}(\mathbf{A}^{(k)}\mathbf{Z}_m^{(k)})\ . \tag{2.19c}$$

One recognizes Eq. (2.17) as deriving from the residue theorem, where the λ's are the unique poles and the Bateman matrices are the corresponding residues of the resolvent of $\mathbf{A}^{(k)}$.

In particular applications it is unlikely that $\mathbf{A}^{(k)}$ will exhibit degeneracies in its eigenvalues; thus, the generalization of the foregoing scheme to allow for such degeneracies, which results in formulas that while trivial to derive are problematic to code, will probably not be required. The method is inherently faster[33] than that based on spectral decomposition for computing the exponential of $\mathbf{A}^{(k)}$ and although both methods are computationally laborious, the resolvent scheme based on Laverrier's algorithm avoids the time consuming tasks of generating the matrix of eigenvectors of $\mathbf{A}^{(k)}$ *and* its inverse. Further, the extent to which the conditions on the Bateman matrices given in Eq. (2.18) and the relation

$$\mathbf{A}^{(k)}\mathbf{Z}_N^{(k)}+\theta_N\mathbb{1}=0 \tag{2.19d}$$

are fulfilled serve to gauge the numerical performance of the method. In an unrelated context, the method has been applied[34] to the point nuclide first-order kinetics of the three species system ^{2}H, ^{3}H, and ^{3}He in a thermal neutron flux. One can readily implement the Leverrier algorithm within the memory and storage constraints of a personal 16-bit microprocessor for a system with $N\sim 3$–5 levels, provided the eigenvalues of $\mathbf{A}^{(k)}$ can be computed. The symmetry of the dipole-transition matrix μ_z for a given (2–5)-level system is usually such that one can exhaustively provide a convenient expression for the characteristic equation of $\mathbf{A}^{(k)}$ from which the eigenvalues can be computed using the efficient algorithm due to Jenkins and co-worker.[35]

C. Fluence and the asymptotic behavior of the evolution operator

The Poynting vector[36] of the classical field $E(t)$ propagating in the z direction is

$$\mathbf{S}=c\epsilon_0E^2(t)\hat{\mathbf{k}}\ , \tag{2.20}$$

where c and ϵ_0 are the velocity of light and permittivity of free space, respectively. If $E(t)$ is viewed as characterizing the statistical properties of an underlying stationary and ergodic stochastic process,[37] one may formally define the average value of $\mathbf{S}$ as

$$\langle\mathbf{S}\rangle=c\epsilon_0\frac{1}{\tau}\int_0^\infty dt\exp(-t/\tau)E^2(t)\hat{\mathbf{k}}\ , \tag{2.21}$$

where the integral is the Laplace time average of $E^2(t)$. Time averages of this type were introduced by Golden and Longuet-Higgins[38] in studies of quantum ergodicity and more recently Kay,[39] in connection with the formulation of a semiclassical ergodic theory, has shown the equivalence of time averages

$$\lim_{\tau\to\infty}\frac{1}{\tau}\int_0^\infty dt\exp(-t/\tau)E^2(t)=\lim_{T\to\infty}\frac{1}{T}\int_0^T dt\,E^2(t) \tag{2.22}$$

whenever the limit on the right exists and $T^{-1}\int_0^T dt\,E^2(t)$ is finite for all $0<T<\infty$. Using either of the averaging processes in Eq. (2.22) for the continuous-wave source gives the well-known result[36]

$$\langle\mathbf{S}\rangle=c\epsilon_0\tfrac{1}{2}(E^0)^2\hat{\mathbf{k}}\ , \tag{2.23}$$

where $\frac{1}{2}(E^0)^2$ is the mean value of $E^2(t)$ over many optical cycles, and one notes that $\langle\mathbf{S}\rangle$ is independent of the phase of the field. In the last section we remarked that, like any quasimonochromatic source,[40] the Gaussian pulse has an epochal existence only during a finite time interval, $|t|\lesssim 4\tau_p$, say. For the Gaussian pulse, Eq. (2.21) yields

$$\langle\mathbf{S}\rangle=c\epsilon_0(E^0)^2\frac{1}{2\tau}\sum_{l=0}^{\infty}\frac{\Gamma(2l+1)}{l!}(-2/\tau_p^2)^l\left[\tau^{2l+1}+\tfrac{1}{2}\left[\frac{\exp(i2\phi)}{(\tau^{-1}-i2\omega_0)^{2l+1}}+\text{c.c.}\right]\right]\hat{\mathbf{k}}\ , \tag{2.24}$$

which vanishes as $\tau\to\infty$ unless one also admits $\tau_p\to\infty$. This is not an artifice but is a result of the finite pulse duration τ_p; if τ_p becomes arbitrarily large so that the pulse evolves into a continuous-wave source then $\langle\mathbf{S}\rangle$ is given by Eq. (2.23) as $\tau\to\infty$. As $\tau\to 0^+$ it readily follows from Eq. (2.24) that

$$\langle\mathbf{S}\rangle=c\epsilon_0(E^0)^2\cos^2\phi\hat{\mathbf{k}}\ , \tag{2.25}$$

and one can eliminate the explicit dependence on the arbi-

trary phase of the field by averaging over the interval $0 \le \phi \le \pi$ to yield Eq. (2.23). One usually rationalizes the applicability of Eq. (2.23) to Gaussian pulses by invoking a slowly-varying-envelope approximation. The fluence or energy output per unit cross-sectional area in the direction of the beam is $\tau_I \langle \mathbf{S} \rangle$, where τ_I is the duration of irradiation at intensity $\langle \mathbf{S} \rangle$.

In the foregoing use of the Laplace time average, and the equivalent long-time average in Eq. (2.22), implicit allowance has been made for the fact that, for the fields of interest here, $E(t)=E(-t)$. More generally, for the density operator $\boldsymbol{\rho}(t,t_0)$ we define[41] its Laplace time-average by

$$\boldsymbol{\rho}^{\tau}(t_0)=\frac{1}{\tau}\int_{t_0}^{\infty} dt \exp[-(t-t_0)/\tau]\boldsymbol{\rho}(t,t_0) , \tag{2.26}$$

where

$$\lim_{\tau \to 0^+} \boldsymbol{\rho}^{\tau}(t_0)=\boldsymbol{\rho}(t_0,t_0) \tag{2.27a}$$

and

$$\lim_{\tau \to \infty} \boldsymbol{\rho}^{\tau}(t_0)=\boldsymbol{\rho}(\infty,t_0) . \tag{2.27b}$$

Physically, τ may be viewed as a uniform relaxation time for processes that effectively interrupt the dipole interaction of the N-level system with the field $E(t)$, for example, through the collisions between members in an ensemble of N-level systems at reasonable densities. For a short relaxation time, Eq. (2.27a) expresses the fact that the system is essentially unperturbed from its initial state $\boldsymbol{\rho}(t_0,t_0)$ by the field while Eq. (2.27b) formally defines the asymptotic state $\boldsymbol{\rho}(\infty,t_0)$ of the system.

The Laplace time-averaged value of the transition probability

$$P_{kk}(\omega_0,\phi,t,t_0) \equiv \rho_{kk}(t,t_0)$$

for excitation to $|k\rangle$ of an N-level system prepared initially in the state $\mathbf{a}(t_0)$ is

$$P_{kk}^{\tau}(\omega_0,\phi,t_0)=\frac{1}{\tau}\int_0^{\infty} dt \exp[-(t-t_0)/\tau] \times P_{kk}(\omega_0,\phi,t,t_0) , \tag{2.28a}$$

or from Eq. (2.2),

$$P_{kk}^{\tau}(\omega_0,\phi,t_0)=\sum_{l,m=1}^{N} a_l^*(t_0)a_m(t_0) \times \frac{1}{\tau}\int_0^{\infty} dt \exp[-(t-t_0)/\tau] \times U_{kl}^*(t,t_0)U_{km}(t,t_0) . \tag{2.28b}$$

For a continuous-wave source we have previously[11,15] dealt in detail with the evaluation of the various transition probabilities: time-resolved $P_{kk}(\omega_0,\phi,t,0)$ and $P_{kk}(\omega_0,t,0)$, time-averaged $P_{kk}^{\tau}(\omega_0,\phi,0)$ and $P_{kk}^{\tau}(\omega_0,0)$, and the asymptotic long-time-averaged $P_{kk}^{\infty}(\omega_0,\phi)$ and $P_{kk}^{\infty}(\omega_0)$. These are evaluated through use of the Floquet form of $\mathbf{a}(t)$ as given in Eq. (2.8) and require $\mathbf{U}(t,0)$ only over the period of the Hamiltonian, $0 \le t \le 2\pi/\omega_0$. To obtain the time-resolved, time-averaged, and long-time-averaged transition probabilities $P_{kk}(\omega_0,\phi,\tau,-\infty)$, $P_{kk}^{\tau}(\omega_0,\phi,-\infty)$, and $P_{kk}^{\infty}(\omega_0,\phi,-\infty)$, respectively, for the Gaussian-pulsed source requires the evolution operator $\mathbf{U}(t,-\infty)$ over the entire temporal range and this ultimately raises the question as to the existence of the limit

$$\mathbf{U}(+\infty,-\infty)=\lim_{t \to \infty} \mathbf{U}(t,-\infty) . \tag{2.29}$$

Following Dollard and Friedman,[12(a)] one can establish the existence of the limit in Eq. (2.29) by using the Riemann product integral representation of the evolution operator to show that $\mathbf{U}(t,t_0)$ fulfills the Cauchy criterion[16(b)] on $[t_0,t]$. For $t_2 > t_1$,

$$||\mathbf{U}(t_2,t_0)-U(t_1,t_0)|| \le ||\mathbf{U}(t_2,t_1)-\mathbb{1}||\,||\mathbf{U}(t_1,t_0)|| , \tag{2.30}$$

where we use the group property given in Eq. (2.4). Now, from Eq. (2.7),

$$\begin{aligned} ||\mathbf{U}(t,t_0)|| &= T \lim_{n_p \to \infty} \left|\left| \prod_{k=1}^{n_p} \exp\left[\int_{t_{k-1}}^{t_k} ds\, \mathbf{A}(s)\right] \right|\right| , \\ &\le T \lim_{n_p \to \infty} \exp\left[\sum_{k=1}^{n_p} \int_{t_{k-1}}^{t_k} ds\, ||\mathbf{A}(s)||\right] \\ &= \exp\left[\int_{t_0}^{t} ds\, ||\mathbf{A}(s)||\right] . \end{aligned} \tag{2.31a}$$

Formally integrating the first-order system

$$\dot{\mathbf{U}}(t,t_0)=\mathbf{A}(t)\mathbf{U}(t,t_0)$$

subject to the initial conditions $\mathbf{U}(t_0,t_0)=\mathbb{1}$ gives

$$\begin{aligned} ||\mathbf{U}(t,t_0)-\mathbb{1}|| &= \left|\left| \int_{t_0}^{t} ds\, \mathbf{A}(s)\mathbf{U}(s,t_0) \right|\right| \\ &\le \int_{t_0}^{t} ds\, ||\mathbf{A}(s)||\,||\mathbf{U}(s,t_0)|| \\ &\le \exp\left[\int_{t_0}^{t} ds\, ||\mathbf{A}(s)||\right]-1 , \end{aligned} \tag{2.31b}$$

where we have used Eq. (2.31a). Using Eq. (2.31) in Eq. (2.30) gives

$$||\mathbf{U}(t_2,t_0)-\mathbf{U}(t_1,t_0)|| \le \left[\exp\left[\int_{t_1}^{t_2} ds\, ||\mathbf{A}(s)||\right]-1\right] \times \exp\left[\int_{t_0}^{t_1} ds\, ||\mathbf{A}(s)||\right] . \tag{2.32}$$

If one assumes the existence of

$$\int_{t_0}^{\infty} ds\, ||\mathbf{A}(s)|| = \lim_{t \to \infty} \int_{t_0}^{t} ds\, ||\mathbf{A}(s)|| < \infty \tag{2.33}$$

then $\int_{t_1}^{t_2} ds\, ||\mathbf{A}(s)||$ can be made arbitrarily small by choosing t_3 to be large and $t_1, t_2 > t_3$. Now since the first factor on the right in Eq. (2.32) can be made arbitrarily small and the second factor is finite by virtue of Eq. (2.33), it follows that $\mathbf{U}(t,t_0)$ is Cauchy on $[t_0,t_1]$, provided Eq. (2.33) is true. Consequently, the limit on the right in Eq. (2.29) exists. Since

$$\mathbf{A}(t)=iE(t)\exp[i\Omega(t-t_0)]\mu_z\exp[-i\Omega(t-t_0)]$$

then $||\mathbf{A}(t)|| \leq |E(t)|\,||\mu_z||$ so that for a Gaussian-modulated sinusoidal pulse, Eq. (2.33) is valid. Further, since $\mathrm{Tr}[\mathbf{A}(t)] \leq ||\mathbf{A}(t)||$ it follows from Eq. (2.3) that the validity of Eq. (2.33) ensures that $\mathbf{U}(+\infty,-\infty)$ is nonsingular. Note that the use of the Riemann product integral representation of the evolution operator in Eq. (2.31) allows one to define $\mathbf{U}(t,t_0)$ for all t, subject to the condition in Eq. (2.33), without recourse to the usual adiabatic switching of interactions.[42]

III. APPLICATIONS AND DISCUSSION

The two-level system considered in this section is characterized by the level separation $\omega_{21}=10^{-3}$ and dipole transition matrix elements $\mu_{ij}=\mu_{ji}=1-\delta_{ij}$ for $i,j=1,2$. The system is assumed to be initially in the ground state $|1\rangle$ and interacts with a field of strength $E^0=5\times10^{-4}\simeq3\times10^{8}$ V m^{-1} which is representative of the large field strengths produced in focused lasers. The radiative width of each level was set to zero. For both the continuous-wave and Gaussian-pulsed sources, the choice of $n_p=180$ for each optical cycle ensured the fulfillment of the local time inequality in Eq. (2.5) and the asymptotic time relations in Eq. (2.10) or Eq. (2.13) with the evolution operator being generated on a 16-bit microprocessor, using Eq. (2.16) in Eq. (2.7), to within optimal single precision accuracy.

We have previously[11] examined the effects of pulse duration $\Delta t=2\tau_p(\ln2)^{1/2}$ on the induced transition probability $P_{22}^{\infty}(\omega_0,0,-\infty)$ at the near-resonant frequency $\omega_0=\omega_{21}$ for $\tau_p=0.125$, 0.625, 1.25, and 2.5 ps. These results demonstrate that at the on-resonance frequency, the value assumed by $P_{22}^{\infty}(\omega_0,0,-\infty)$ is highly sensitive to the number of optical cycles sustained throughout the pulse duration; if the behavior of the time-resolved transition probability $P_{22}(\omega_0,0,t,-\infty)$ is such that it assumes a large (small) value when the pulse decays then $P_{22}^{\infty}(\omega_0,0,-\infty)$ will be large (small). This contrasts with the continuous-wave source where at the Bloch-Siegert shifted one-photon resonance $\omega_0=1.0626\omega_{21}$, $P_{22}^{\infty}(\omega_0,0,0)=0.5$, and the two-level system saturates. Figure 1 displays the time-resolved induced transition probability $P_{22}(\omega_0,0,t,-\infty)$ at the off-resonance frequency $\omega_0=0.5\omega_{21}$ for $\tau_p=0.125$, 0.625, 1.25, and 2.5 ps, as well as the laser field $E(t)$ scaled by the peak amplitude E^0, i.e., $E(t)/E^0$. Irrespective of the ultrashort duration, the efficiency of the pulse in inducing transitions to $|2\rangle$ is significantly diminished. In all cases the transition probability is at a maximum $\ll1$ when the pulse peaks but it vanishes as the off-resonance pulse decays and the system reverts to $|1\rangle$. Figure 2 gives

$$P_{11}(\omega_0,0,t,0)=1-P_{22}(\omega_0,0,t,0)$$

over ten optical cycles of the continuous-wave source at the off-resonance frequency $\omega_0=0.55\omega_{21}$ which, when averaged over phase and time, assumes a steady-state value of ~0.25 and is consequently more efficient at maintaining population in $|2\rangle$, albeit at subsaturation levels. The exhibition of multiphoton resonances in a two-level system interacting with a constant-amplitude

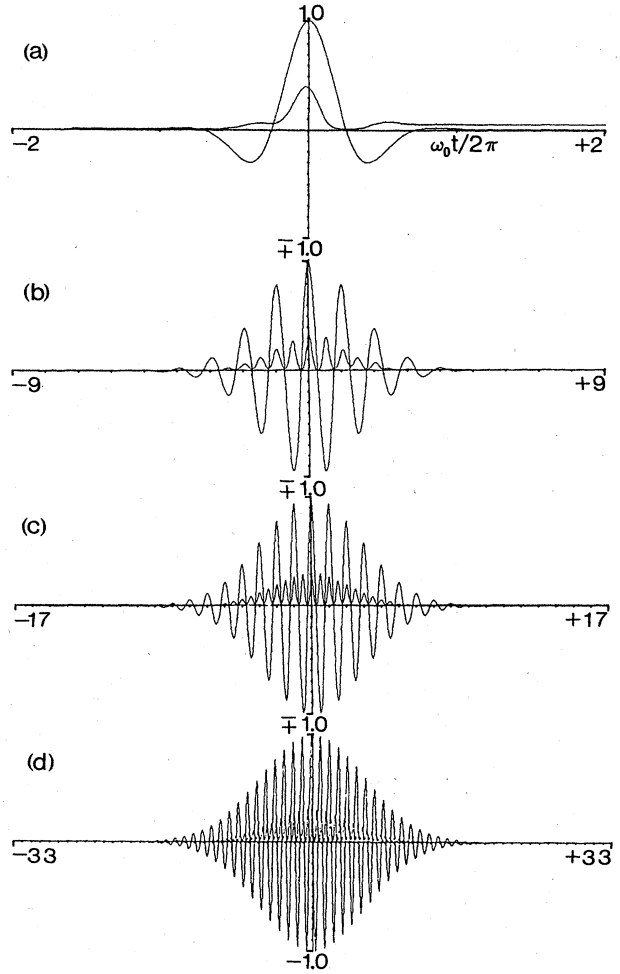

FIG. 1. The time-resolved induced transition probability $P_{22}(\omega_0,0,t,-\infty)$ at the off-resonance frequency $\omega_0=0.5\omega_{21}$ for (a) $\tau_p=0.125$ ps, (b) $\tau_p=0.625$ ps, (c) $\tau_p=1.25$ ps, and (d) $\tau_p=2.5$ ps. The curves displaying both positive and negative values represent the oscillating field scaled by the peak field strength, i.e., $-1\leq\exp(-t^2/\tau_p^2)\cos(\omega_0 t)\leq+1$.

sinusoidal field owes its origin to the attainment of almost periodic[43] nutations in $P_{22}(\omega_0,t,0)$ as a function of $\omega_0 t$ with the approximate Rabi "period" increasing with the number of photons of the resonance and being determined in part by the magnitude of the coupling strength $\mu_{21}E^0/\omega_{21}$. The presence of a relaxation mechanism with characteristic time τ effectively interrupts the absorption of photons to the extent that $P_{22}(\omega_0,t,0)$ does not attain almost periodic behavior as a function of $\omega_0 t$ and in accordance with Eq. (2.27a),

$$P_{22}^{\tau}(\omega_0,0)\rightarrow|a_2(0)|^2$$

as $\tau\rightarrow0^+$. In the other extreme, $P_{22}^{\infty}(\omega_0,0)$ exhibits multiphoton resonances at $\omega_0\lesssim\omega_{21}$ which increasingly encroach upon each other as the number of photons in the resonances increase. The steady-state value of $P_{22}^{\infty}(\omega_0,0)\simeq0.25$ at $\omega_0=0.55\omega_{21}$ is determined in part by the one-photon and three-photon resonances at $1.0626\omega_{21}$

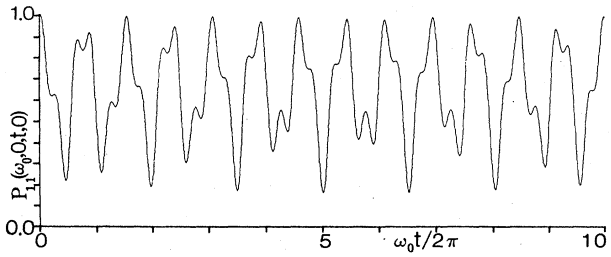

FIG. 2. The time-resolved induced transition probability $P_{11}(\omega_0,0,t,0)$ over ten optical cycles at the off-resonance frequency $\omega_0=0.55\omega_{21}$.

and $0.4129\omega_{21}$, respectively. Hogg and Huberman[44] have recently shown that for a multilevel system described by a self-adjoint periodic Hamiltonian, the state amplitudes and expectation values of bounded operators are almost periodic and return arbitrarily close to their initial values infinitely often. However, in contrast to a continuous-wave source, the epochal nature of the Gaussian pulse provides an inherent deterrent to the establishment of almost periodic oscillations in $P_{22}(\omega_0,0,t,-\infty)$ as a function of $\omega_0 t$, especially for very short duration pulses. Unless the duration of the pulse is long enough to sustain many optical cycles $\sim 8\tau_p(\omega_0/2\pi)$ throughout its tenure, then at frequencies $\omega_0 \lesssim \omega_{21}$ the number of optical cycles occurring will be even fewer and consequently $P_{22}(\omega_0,0,t,-\infty)$ will not exhibit almost periodic nutations at frequencies corresponding to the absorption of several photons. Further, the nonappearance of multiphoton resonances for short duration pulses will be enhanced by any relaxation mechanism with a short characteristic time in accordance with Eq. (2.27a). For the two-level system considered here and in an earlier[11] paper, the pulse durations are sufficiently short that only the single-photon resonances appear. Consequently, it is of little surprise to observe $P_{22}^{\infty}(\omega_0,0,-\infty)$ vanish in Fig. 1 at the off-resonance frequency $\omega_0=0.5\omega_{21}$ since it is not flanked at some $\omega_0<\omega_{21}$ by a multiphoton resonance for any of the pulse durations considered.

The effects of the phase of the Gaussian-pulsed field on the time-resolved transition probability $P_{22}(\omega_0,\phi,t,-\infty)$ are displayed in Fig. 3 at the on-resonance frequency $\omega_0=\omega_{21}$ for $\phi=0,\pi/3$, and $\pi/2$. Figure 4 shows the phase-averaged

$$P_{11}(\omega_0,t,0)=1-P_{22}(\omega_0,t,0)$$

and phase-dependent

$$P_{11}(\omega_0,\phi,t,0)=1-P_{22}(\omega_0,\phi,t,0)$$

transition probabilities for $\phi=0$, $\pi/3$, and $\pi/2$ at the one-photon resonance $\omega_0=1.0626\omega_{21}$ over the period of the Hamiltonian in the case of continuous-wave excitation. For both sources, the time-resolved and phase-dependent transition probabilities exhibit recurring undulations at frequencies

$$\omega_0 t=(k+\tfrac{1}{2})\pi-\phi, \quad k=0,1,2,\ldots. \tag{3.1}$$

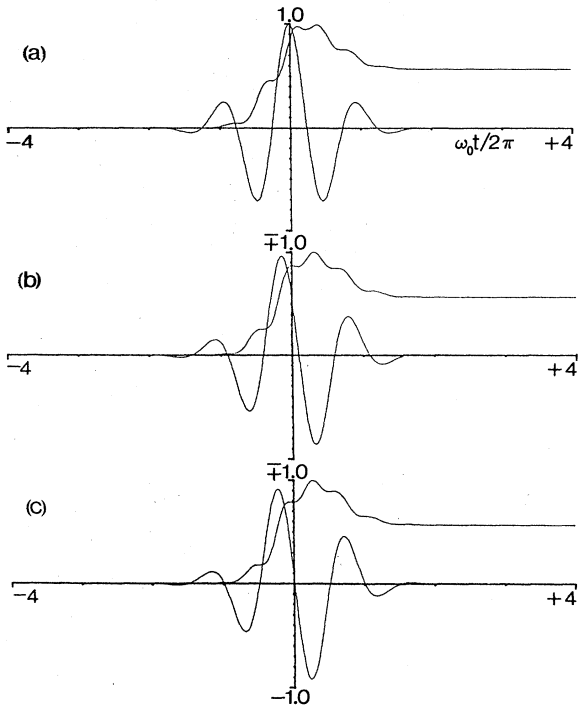

FIG. 3. The time-resolved and phase-dependent induced transition probability $P_{22}(\omega_0,\phi,t,-\infty)$ at the on-resonance frequency $\omega_0=\omega_{21}$ with $\tau_p=0.125$ ps for (a) $\phi=0$, (b) $\phi=\pi/3$, and (c) $\phi=\pi/2$. The curves displaying both positive and negative values represent the oscillating field scaled by the peak field strength, i.e., $-1\le\exp(-t^2/\tau_p^2)\cos(\omega_0 t)\le+1$.

At these frequencies the interaction-picture transition rates vanish, as follows from Eq. (2.1). The effect of the phase ϕ is to shift the undulations without affecting the asymptotic behavior of the transition probability, at least for on-resonance frequencies. In a realistic pulsed source, the phase ϕ will generally vary with time but in the mode- and phase-locked operation, it is ostensibly constant. As remarked in Sec. II A, one may take $\phi=0$ for an isolated pulse or maintain it at a fixed value for a phase-coherent train of pulses. However, for a continuous-wave source, one averages computed properties over the phase. As is

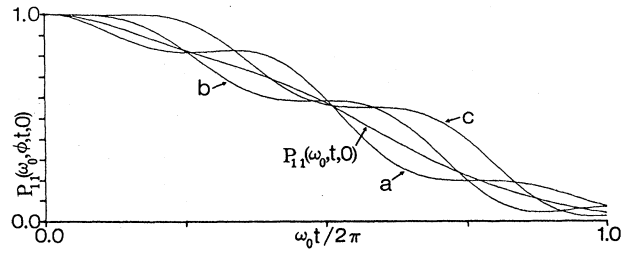

FIG. 4. The time-resolved and phase-dependent induced transition probability $P_{11}(\omega_0,\phi,t,0)$ at the one-photon resonance $\omega_0=1.0626\omega_{21}$ over one optical cycle for (a) $\phi=0$, (b) $\phi=\pi/3$, and (c) $\phi=\pi/2$. Included is the phase-averaged induced transition probability $P_{11}(\omega_0,t,0)$.

evident from Fig. 4, such phase averaging smooths out the undulations and when further time averaged, the transition probability is insensitive to phase at near-resonance frequencies but off resonance, the phase dependence becomes more pronounced.[15] Similar considerations apply to the Gaussian pulse, as indicated in Fig. 3 for the on-resonance interaction. At off-resonance frequencies the influence of the phase of the pulse on the transition probability is somewhat academic in view of the fact that at such frequencies the Gaussian-pulsed source is not very efficient at creating a desirable population inversion, especially at ultrashort interaction times.

Finally, we consider the case in which the two-level system interacts with a train of phase coherent identical Gaussian pulses whose time delays are sufficiently short to arrive before spontaneous decay becomes established. Figure 5 shows the variation in $P_{22}^{\infty}(\omega_0,0,-\infty)$ as a function of n, the number of pulses in the train, within the range $1-10^3$ at the near-resonance frequency $\omega_0=\omega_{21}$ and with $\tau_p=0.625$ ps, $E^0=5\times10^{-4}$, and $\phi=0$ for each pulse. Initially, $P_{22}(\omega_0,0,-\infty)=0$ but following the first pulse, $P_{22}^{\infty}(\omega_0,0,-\infty)\simeq0.55$ as shown in Fig. 3(a). This now becomes the initial condition for the incoming second pulse and continuing in this fashion one cascades the transition probability over an arbitrary number of identical pulses in accordance with Eq. (2.12) once the evolution operator has been determined for the first pulse in the train. As depicted in Fig. 5, the system saturates over the duration of the train of pulses at the near-resonance frequency with the transition probability averaging to 0.5, exactly as happens when the system absorbs a single photon from a near-resonant continuous-wave source. However, in contrast to the continuous-wave case, the transition probability oscillates near zero at the off-resonance frequency $\omega=0.5\omega_{21}$ no matter how many pulses there are in the train. The dynamics of the interaction of a two-level system with a train of pulses will reflect the ability of a single pulse's duration to encompass sustained Rabi nutations at specific frequencies.

IV. CONCLUSIONS

In this paper some generalizations and improvements in implementation of an algorithm[11] for evaluating the evolution operator of a multilevel system dipole interacting with either a continuous-wave or Gaussian-pulsed laser were presented.

The generalizations in the algorithm include allowance for spontaneous decay or a dissociation (or ionization) channel, the inclusion of an arbitrary phase in the specification of the Gaussian pulse, the provision of useful relations for the eigenvalues of the evolution operator, and a simple scheme for cascading the state amplitudes over the duration of an arbitrary number of identical phase coherent pulses. The dependence of the laser fluence on the phase of the field was briefly explored as was the asymptotic behavior of the evolution operator for a Gaussian-modulated source.

The improvements in implementation of the algorithm accrue from avoiding the repetitive complete spectral decomposition of potentially large $N\times N$ matrices. For the two-level system, the evolution operator is evaluated as the product of 2×2 matrices while for a multilevel system one may apply a resolvent scheme in conjunction with Leverrier's algorithm for evaluating the Bateman coefficient matrices. We have not belabored the computational aspects of the algorithm as these are essentially straightforward. The main difficulty one encounters is exponential underflow or overflow in the evaluation of Eq. (2.14) using Eq. (2.15), but this is easily overcome by due attention to how the various factors are combined in affecting the evaluation. For both two-level and multilevel applications, the algorithm is easily implemented on either a main-frame or personal computer. Of course, for personal microprocessor implementation one is restricted to $N\sim2-5$, for example, and one must provide the code for evaluating the error function in the complex plane when considering Gaussian pulses, in addition to the code to generate the roots of the characteristic polynomial. In this paper the numerical demonstration of the algorithm

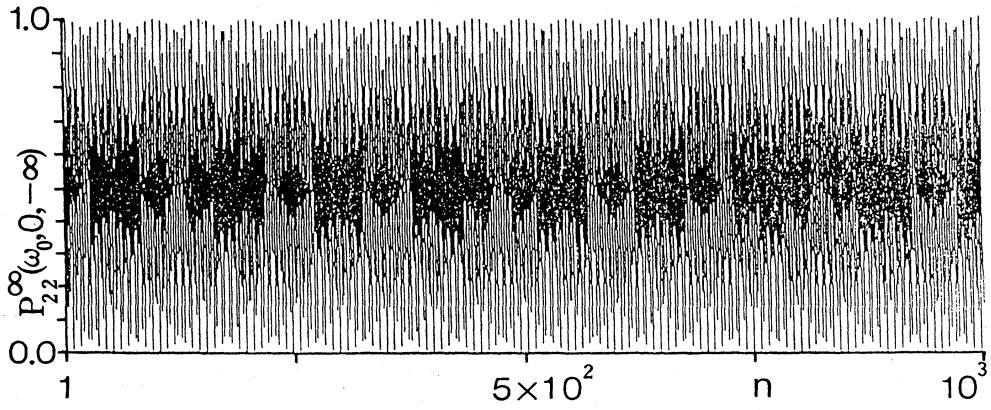

FIG. 5. The variation of the steady-state induced transition probability $P_{22}^{\infty}(\omega_0,0,-\infty)$ with the number (n) of pulses in a train of $1\leq n\leq10^3$ phase-coherent identical pulses, each at the on-resonance frequency $\omega_0=\omega_{21}$ and with $\tau_p=0.625$ ps.

has been restricted to a two-level system. The calculations were performed on a 16-bit microprocessor and they complement previous calculations[11] carried out on a mainframe machine on the same model two-level system. In the case of the Gaussian-pulsed source calculations, a comparison between the computed results and those given analytically[45(a)] within the rotating wave and second order Magnus approximations will be reported elsewhere.[45(b)] Like the recursive-residue-generation method given recently by Nauts and Wyatt,[46] the resolvent scheme proposed here for tackling multilevel systems should be highly efficient and enjoys the additional advantage of allowing one to gauge its numerical performance through Eqs. (2.18) and (2.19d). We reserve discussion of the application of the scheme to particular multilevel systems for a forthcoming paper in which such issues as the inclusion of spontaneous decay or a dissociation (or ionization) channel through a non-Hermitian Hamiltonian operator,[47(a)] the averaging of induced transition probabilities over the laser-pulse bandwidth, and the effects of including an additional static electric field will be addressed. Baker[47(b)] has generalized the procedure of Armstrong and Baker[47(c)] for the systematic construction of an appropriate effective Hamiltonian for an ionizing and/or decaying multilevel system. George and coworkers[27,48] have emphasized the importance of the pulse's time-varying envelope with the finding that the collision cross sections in a multimode laser field can be considerably different from those in a single-mode field with the same average high intensity. Any investigation of bandwidth averaging on the transition probabilities should incorporate a description of the laser statistics, fluctuations, and mode structure, such as the classical field treatments given by George[49(a)] and Mittleman.[49(b),49(c)] Both theoretical[15,50(a)] and experimental[50(b)] evidence suggest that an additional static electric field enhances multiphoton processes in atoms and molecules. An upper limit on the number of states to be included in a simulation of the dynamics of a multilevel system with an intense laser field must be set, especially as the density of states generally increases quite rapidly with energy. Thus, use of the resolvent scheme we have proposed (or indeed any equivalent algorithm) may become impractical,[51] particularly if the temporal variation in the field amplitude has also to be considered.

The illustrative applications presented reveal some of the differences in the dynamics of interaction of two-level systems with a constant amplitude field or a Gaussian time-varying-amplitude field. The most dramatic difference between the two is the failure of the latter, for the short interaction times considered here, to allow the establishment of steady state Rabi oscillations of the induced transition probabilities at off-resonance or multiphoton frequencies. Only with the advent of sources delivering ultrashort pulses have the effects of amplitude variation on multiphoton processes begun to receive attention.[52] Thus, Lompre *et al.*[53] have reported the absence of multiphotoionization in rare gases irradiated by picosecond pulses although such phenomena were observed with nanosecond pulses at the same carrier frequency, the difference being attributable to a decrease in the time available for the absorption of the requisite number of photons. With the availability of pulses now extending down to the femtosecond domain,[10] the nature of the rapidly varying pulse envelope and the consequences for the dynamics of the interaction of atomic and molecular systems with intense ultrashort-duration sources will undoubtedly be of importance. On resonance, the phase of the field shifts recurring undulations in the time-resolved induced transition probability without affecting its asymptotic behavior. Under the action of a train of phase-coherent on-resonance pulses, the two-level system saturates although the train is less successful in affecting a population inversion at off-resonance frequencies.

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