Excitation of a multilevel system by a train of identical phase-coherent Gaussian-shaped laser pulses

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An algorithm for calculating the time-resolved evolution operator of a multilevel system dipole interacting with a Gaussian-shaped laser pulse is developed through use of a compound Magnus expansion in compliance with Maricq's convergence criterion. In conjunction with the Leverrier-Bateman resolvent method, the algorithm allows the frequency spectrum for the interaction of the multilevel system with a train of identical phase-coherent Gaussian-shaped laser pulses to be readily evaluated. Furthermore, the algorithm renders possible the expeditious averaging of operator expectation values over the spectral bandwidth of the laser field. A prescription for gauging the accuracy of the algorithm is provided.

I. INTRODUCTION

A recent investigation[1] of the dependence of multiphoton excitation of polyatomic molecules on laser intensity underscores the need to consider explicitly the shape of the pulse when comparing experimental data obtained using high-energy fluences with computer-simulation results which, however, are usually generated on the assumption that the pulses are rectangular in shape. In fact, real pulses are generally "bell shaped" and therefore do not have a constant intensity as when the pulses are rectangular. The expedience in taking the pulse to be of fixed intensity is generally justified on the basis of a slowly varying amplitude approximation[2] (SVAA) in which, for sufficiently long-duration pulses, the raising-on and switching-off times are so short relative to the pulse length that to all intents and purposes the pulse may be considered to be rectangular in shape. With ultrashort pulses[3] the number of optical cycles sustained throughout a pulse's tenure is small and the raising-on and switching-off times can represent a significant fraction of that epochal duration thereby invalidating the SVAA. At most, the absorbed energies calculated within the sudden approximation must be considered[1] an upper limit to their true values.

Elsewhere[4] I have promoted the usage of the Leverrier-Bateman resolvent method[5] (LBRM) in calculating the transition probabilities for the excitation of multilevel systems dipole interacting with a continuous-wave (cw) or Gaussian-pulsed laser field. In contrast to a previous treatment[4(b)] I now apply the rotating-wave approximation[6] (RWA)–in which one discards all rapidly oscillating contributions to the transition rates—in conjunction with the LBRM to provide a tractable algorithm for evaluating the evolution operator of a multilevel system interacting with a train of identical phase-coherent Gaussian-shaped laser pulses. The importance of a Gaussian pulse derives from the fact that, in principle and in practice, it represents[7] the idealized transform-limited output from a perfectly mode- and phase-locked laser. In formulating this algorithm in Sec. II, I also invoke the Magnus approximation,[8] a unitarity-preserving perturbative approach to the calculation of the evolution operator; the algorithm is contingent upon the convergence of the Magnus expansion in compliance with Maricq's criterion.[8(l)] Both the RWA and the Magnus approximation are valid for all but ultraintense fields and their joint use has proved useful in providing generalizations of the Rabi formula[6(a)] for the single- and multiple-photon excitation transition probabilities of an isolated and an aligned[9(a),9(b)] two-level system by a Gaussian-shaped pulse, and in the calculation[9(c)] of the single-photon fluorescence signal with attendant optical Ramsey fringes from a two-level system following its interaction with a train of phase-coherent Gaussian pulses. Also in Sec. II, a simple prescription for gauging the accuracy of the algorithm is provided and the requisite averaging of operator expectation values over the spectral bandwidth of the laser field is discussed. The paper closes with a concluding summary in Sec. III. Atomic units are used throughout the paper.

II. ALGORITHM

A. Asymptotic solution

Within the resonance-constrained global RWA,[6(c)] the interaction picture state amplitude matrix $c(t)$ of a multilevel (N, say) system evolves in accordance with the Schrödinger equation

$$\dot{\underline{c}}(t)=\underline{C}(t)\underline{c}(t)\ , \tag{1a}$$

where $\underline{c}(t)$ is an N-vector whose components $c_m(t)=\langle m|\Psi(t)\rangle$ are the probability amplitudes for the stationary energy eigenstates $|m\rangle$, $m=1,2,\ldots,N$ of the isolated system and the $N\times N$ proresonant coefficient matrix $\underline{C}(t)$ is given by

$$\underline{C}(t)=\frac{i}{2}E^0 f(t)\begin{bmatrix} 2\mu_{11}\cos(\omega_0 t) & \mu_{12}\exp(-i\tilde{\omega}_{21}t) & \cdots\ \mu_{1N}\exp(-i\tilde{\omega}_{N1}t) \\ \mu_{21}\exp(i\tilde{\omega}_{21}t) & 2\mu_{22}\cos(\omega_0 t) & \mu_{2N}\exp(-i\tilde{\omega}_{N2}t) \\ \vdots & & \vdots \\ \mu_{N1}\exp(i\tilde{\omega}_{N1}t) & \mu_{N2}\exp(i\tilde{\omega}_{N2}t) & 2\mu_{NN}\cos(\omega_0 t) \end{bmatrix} . \tag{1b}$$

In Eq. (1b), E^0 is the field strength of the semiclassical laser field which is polarized in the z direction and is of carrier frequency ω_0 and has a Gaussian pulse envelope $f(t)=\exp(-\pi t^2/\tau_p^2)$ of epochal duration τ_p and full-width-at-half-maximum (FWHM) spectral bandwidth $\Delta\omega=4(\pi\ln 2)^{1/2}/\tau_p$; $\tilde{\omega}_{mn}=\omega_{mn}-\omega_0$ is the frequency detuning from the level separation $\omega_{mn}=\omega_m-\omega_n$, where the eigenstate $|m\rangle$ has energy ω_m, and $\mu_{mn}=\langle m|\mu_z|n\rangle=\mu_{nm}$ is the dipole transition matrix element coupling the states $|m\rangle$ and $|n\rangle$, μ_z being the z component of the dipole moment vector. One may assume the μ's to be real. If the states are of definite parity the diagonal entries in $\underline{C}(t)$ vanish since μ_z is an odd operator. Also, if $|m\rangle$ and $|n\rangle$, say, are degenerate then the phase factors $\exp(-i\tilde{\omega}_{mn}t)$ and $\exp(i\tilde{\omega}_{mn}t)$ are both replaced by $2\cos(\omega_0 t)$, with the retention of the counter-rotating phasor of the field, as in the case of the diagonal entries in $\underline{C}(t)$.

The Magnus[8] asymptotic solution to Eq. (1a) for the initial conditions $\underline{c}(-\infty)$ is

$$\underline{c}(\infty)=\underline{U}(\infty)\underline{c}(-\infty) , \tag{2}$$

where the evolution operator $\underline{U}(\infty)$ is given by the cumulant expansion

$$\underline{U}(\infty)=\exp[\underline{M}(\infty)] \tag{3a}$$

and with

$$\underline{M}(\infty)=\sum_{k=1}^{\infty}\underline{M}_k(\infty) , \tag{3b}$$

$$\underline{M}_1(\infty)=\int_{-\infty}^{+\infty}dt\,\underline{C}(t) \tag{3c}$$

and

$$\underline{M}_2(\infty)=-\tfrac{1}{2}\int_{-\infty}^{+\infty}dt\int_{-\infty}^{t}dt'[\underline{C}(t),\underline{C}(t')] , \tag{3d}$$

while for $k\geq 3$, $\underline{M}_k(\infty)$ is generally a sum of integrals of k-fold nested commutators of $\underline{C}(t)$.

Using Eq. (1b) in (3c) and (3d), it is straightforward to evaluate the entries in $\underline{M}_1(\infty)=(M_{1,kl})$ and $\underline{M}_2(\infty)=(M_{2,kl})$ as

$$M_{1,kl}(\infty)=i\tau_p E^0\mu_{kl}\delta_{kl}\exp[-(\omega_0\tau_p)^2/4\pi]+\frac{i}{2}\tau_p E^0\mu_{kl}(1-\delta_{kl})\exp\{-[\mathrm{sgn}\tilde{\omega}_{kl}\tau_p]^2/4\pi\} \tag{4a}$$

and

$$\begin{aligned} M_{2,kl}(\infty)=\tfrac{1}{2}(\tau_p E^0/2)^2\sum_{m=1}^{N} & \mu_{km}\delta_{km}\mu_{ml}(1-\delta_{ml})\exp\{-[\omega_0^2+(\mathrm{sgn}\tilde{\omega}_{ml})^2]\tau_p^2/4\pi\} \\ & \times\{\mathrm{erf}[i(\omega_0-\mathrm{sgn}\tilde{\omega}_{ml})\tau_p/2(2\pi)^{1/2}]-\mathrm{erf}[i(\omega_0+\mathrm{sgn}\tilde{\omega}_{ml})\tau_p/2(2\pi)^{1/2}]\} \\ & +\mu_{km}(1-\delta_{km})\mu_{ml}\delta_{ml}\exp\{-[\omega_0^2+(\mathrm{sgn}\tilde{\omega}_{km})^2]\tau_p^2/4\pi\} \\ & \times\{\mathrm{erf}[i(\omega_0+\mathrm{sgn}\tilde{\omega}_{km})\tau_p/2(2\pi)^{1/2}]-\mathrm{erf}[i(\omega_0-\mathrm{sgn}\tilde{\omega}_{km})\tau_p/2(2\pi)^{1/2}]\} \\ & +\mu_{km}(1-\delta_{km})\mu_{ml}(1-\delta_{ml})\exp\{-[(\mathrm{sgn}\tilde{\omega}_{km}\tau_p)^2+(\mathrm{sgn}\tilde{\omega}_{ml}\tau_p)^2]/4\pi\} \\ & \times\mathrm{erf}[i(\mathrm{sgn}\tilde{\omega}_{km}-\mathrm{sgn}\tilde{\omega}_{ml})\tau_p/2(2\pi)^{1/2}] , \end{aligned} \tag{4b}$$

respectively, where $\delta_{mn}\equiv\delta(\omega_m,\omega_n)=1$ if $\omega_m=\omega_n$ and $\delta_{mn}\equiv\delta(\omega_m,\omega_n)=0$ if $\omega_m\neq\omega_n$, $\mathrm{sgn}\tilde{\omega}_{mn}=\tilde{\omega}_{mn}$ if $m>n$ and $\mathrm{sgn}\tilde{\omega}_{mn}=-\tilde{\omega}_{nm}$ if $m<n$ for $m,n=1,2,\ldots,N$, and $\mathrm{erf}(z)$ is the error function at an arbitrary point in the complex plane. The δ factors in Eq. (4) account for the possible presence of state-specific permanent dipole moments and/or level degeneracies in the system, both of which require the retention of the counter-rotating phasor of the field.

The LBRM [Refs. 4, 5(a), and 5(b)] gives $\underline{U}(\infty)$ in Eq. (3a) as

$$\underline{U}(\infty)=\sum_k \exp(\lambda_k)\underline{N}_k , \tag{5}$$

where the λ's are the unique eigenvalues of $\underline{M}(\infty)$ and the $\underline{N}$'s are $N\times N$ generalized[4(a)] Bateman coefficient matrices[5(c)] with the projection, idempotent, and sum-rule properties

$$\underline{N}_m\underline{N}_n=0 \quad \text{if } m\neq n , \tag{6a}$$

$$\underline{N}_m^n=\underline{N}_m \quad \text{for } n \text{ any positive integer} , \tag{6b}$$

and

$$\sum_m \lambda_m^n\underline{N}_m=\underline{M}^n(\infty) \text{ for } n \text{ any integer} , \tag{6c}$$

respectively. The summations in Eqs. (5) and (6c) are over the distinct eigenvalues of $\underline{M}(\infty)$ which must be calculated numerically and the corresponding $\underline{N}$'s are expeditiously evaluated using Leverrier's algorithm.[4(a),5(a),5(b)] The properties given in Eq. (6) serve to gauge the numerical performance of this scheme for evaluating $\underline{U}(\infty)$ as Eq. (5), subject to the supply of a suitable approximation to $\underline{M}(\infty)$. Formally, as the integral matrix for Eq. (1a), $\underline{U}(\infty)=\sum_{k=1}^{N}\underline{c}_k(\infty)\underline{e}_k^T$, where the $\underline{c}_k$'s are the linearly independent solutions of Eq. (1a) corresponding to the initial conditions $\underline{e}_k=(\delta_{jk})$, $j=1,2,\ldots,N$. The LB decomposition of $\underline{U}(\infty)$ in Eq. (5) gives the $\underline{c}_k$'s as $\underline{c}_k(\infty)=\sum_m \exp(\lambda_m)\underline{N}_m\underline{e}_k$, i.e., the resultant of the nonunitary rephasing of the Bateman projections of $\underline{e}_k$ over all the unique eigenvalues of $\underline{M}(\infty)$.

Use of the RWA and the Magnus approximation in providing $\underline{M}(\infty)$ must now be justified. While criteria for validating the RWA are available[6(b)–6(d)] in the case of the cw excitation of multilevel systems, it is not until recently that the convergence of the Magnus expansion for periodically perturbed two-state systems received much attention,[8(e)–8(l)] although earlier comparative studies[10] of particularized systems against corresponding perturbative treatments have been made. Maricq's criterion—Eq. (5) of Ref. 8(l)—for the convergence of the Magnus expansion is equally valid when the perturbing field is modulated by a pulse envelope. Joint use of the RWA and the Magnus approximation amounts to a compound Magnus expansion since the former is in fact[8(l)] equivalent to a first-order Magnus approximation. One must first justify use of the RWA and further justify the use of a subsequent Magnus approximation. If any two-level subsystem, $|m\rangle$ and $|n\rangle$, say, of the multilevel system violates the convergence criterion for application of the Magnus expansion, Maricq conjectures[8(l)] that the expansion should not hold for the full system either. Applying Maricq's criterion to Eq. (1a) for the two-state system, but with retention of the counter rotating phasor of the Gaussian-pulsed laser field in $\underline{C}(t)$, requires that $|\mu_{mn}E^0\tau_p|<\pi$ for the RWA to hold throughout the entire duration of the pulse. Again applying Maricq's criterion to Eq. (1a), but this time with $\underline{C}(t)$ being given within the RWA by Eq. (1b), yields the requirement $|\mu_{mn}E^0\tau_p|<2\pi$ so that if use of the RWA is justified, then so also is a subsequent Magnus approximation. Implicit in the practical applications of the Magnus expansion is its assumed rapid convergence whereby only a few low-order terms have a nonnegligible contribution to the sum. The conservation of probability is not a suitable indicator of such convergence since if $\underline{C}^\dagger(t)=-\underline{C}(t)$, a truncated Magnus expansion assures a unitary estimate of $\underline{U}(\infty)$, while if $\underline{C}^\dagger(t)\neq-\underline{C}(t)$, as when one requires to admit a dissociation (or ionization) channel through the uppermost level by ascribing to it the phenomenological natural width γ_N so that its energy is $\omega_N-i\gamma_N/2$, one does not wish to forego use of the Magnus approximation. I have previously shown[4(b)] that the eigenvalues $\lambda=|\lambda|\exp(i2\pi\Delta)$, where

$$\Delta=(1/2\pi)\tan^{-1}[\mathrm{Im}(\lambda)/\mathrm{Re}(\lambda)]\ ,$$

of the *exact* $\underline{U}(\infty)$, irrespective of whether or not it is unitary, satisfy the relations

$$\prod_{k=1}^{N}|\lambda_k(\infty)|=1 \tag{7a}$$

and

$$\sum_{k=1}^{N}\Delta_k(\infty)=(\tau_pE^0/2\pi)\exp[-(\omega_0\tau_p)^2/4\pi]\sum_{k=1}^{N}\mu_{kk}\ . \tag{7b}$$

Thus for the additional effort required to evaluate the eigenvalues of the *approximate* $\underline{U}(\infty)$, the extent to which these eigenvalues fulfill the conditions in Eq. (7) serves as an indicator of the quality of the second-order Magnus approximation and may suggest the need to proceed to even higher orders in the expansion. One can explicitly show that Eq. (7) holds exactly for the two-state system[9(a)–9(c)] within both the first- and second-order Magnus approximations.

If the system interacts with a train of $n=1,2,\ldots,$ identical phase-coherent pulses,[11] then its state amplitude matrix is given through use of Eqs. (5) and (6b) in Eq. (2) as

$$\underline{c}^{(n)}(\infty)=\sum_k \exp(n\lambda_k)\underline{N}_k\underline{c}(-\infty)\ . \tag{8a}$$

Equation (8a) cascades the state amplitude matrix of the multilevel system over the duration of n identical pulses using the Leverrier-Bateman decomposition of $\underline{U}(\infty)$, as given in Eq. (5), for the *first* pulse in the train. Indeed, one may view Eq. (8a) as the pulse train's analog of the Floquet form[12–14] of the state amplitude matrix for a multilevel system interacting with a cw laser field in terms of the Lyapunov and characteristic exponent matrices, both of which are obtained by evaluating $\underline{U}(\omega_0t)$ over the *initial* $2\pi/\omega_0$ period of the Hamiltonian operator. Essentially, the idempotency of the Bateman matrices for the initial (and successive) pulse(s) in the train is the counterpart of the periodicity of the Lyapunov matrix over the initial (and successive) optical cycle(s) of a rectangular-shaped pulse.

On using Eq. (6a) in (8a) it follows that for all unique eigenvalues λ_m of the Magnus operator

$$\underline{N}_m\underline{c}^{(n)}(\infty)=\exp(\lambda_m)\underline{N}_m\underline{c}^{(n-1)}(\infty),\ n=1,2,\ldots \tag{8b}$$

where $\underline{c}^{(0)}(\infty)\equiv\underline{c}(-\infty)$. Thus the nth pulse causes a Bateman projection of $\underline{c}^{(n-1)}(\infty)$ to evolve to its current value through the action of the corresponding rephasing transformation. Equation (8a) gives $\underline{c}^{(n)}(\infty)$ as the resultant of n-fold rephased Bateman projections of $\underline{c}(-\infty)$. Similarly, a summation of Eq. (8b) over all the unique eigenvalues of $\underline{M}(\infty)$ and use of Eq. (6c) gives $\underline{c}^{(n)}(\infty)$ as the resultant of the rephased projections of $\underline{c}^{(n-1)}(\infty)$.

The induced transition probability $P_{kk}^{(n)}(\omega_0,\infty)=|c_k^{(n)}(\infty)|^2$ for excitation to the state $|k\rangle$, following the interaction of the system with the train of n identical phase-coherent pulses and for the most interesting case in which the ground state is initially fully populated, is given by

$$P_{kk}^{(n)}(\omega_0,\infty)=\sum_{m,m'}\exp[n(\lambda_m+\lambda_{m'}^*)]N_{k1,m}N_{k1,m'}^* \tag{9}$$

and the dissociation probability in the event that the uppermost level has energy $\omega_N-i\gamma_N/2$, where γ_N is the phenomenological radiative width, is given by

$$P_{\text{diss}}^{(n)}(\omega_0,\infty)=1-\sum_{k=1}^{N}P_{kk}^{(n)}(\omega_0,\infty)\ . \tag{10}$$

The mean number of photons of energy ω_0 absorbed by the system following its interaction with the train of pulses is

$$\langle\omega_0(\infty)\rangle^{(n)}=\sum_{k=1}^{N}\omega_k P_{kk}^{(n)}(\omega_0,\infty)/\omega_0\ . \tag{11}$$

B. Time-resolved solution

So far it has been assumed that only the value of the evolution operator at the completion of the initial pulse is of interest. Actually the time-resolved entries in $\underline{M}_1(t)$ and $\underline{M}_2(t)$ are given by

$$\begin{aligned}M_{1,kl}(t)=&\frac{i}{4}\tau_p E^0\mu_{kl}\delta_{kl}\exp[-(\omega_0\tau_p)^2/4\pi]\\&\times\{2+\text{erf}[\pi^{1/2}(t-i\omega_0\tau_p^2/2\pi)/\tau_p]+\text{erf}[\pi^{1/2}(t+i\omega_0\tau_p^2/2\pi)/\tau_p]\}\\&+\frac{i}{4}\tau_p E^0\mu_{kl}(1-\delta_{kl})\exp[-(\text{sgn}\tilde{\omega}_{kl}\tau_p)^2/4\pi]\{1+\text{erf}[\pi^{1/2}(t-i\,\text{sgn}\tilde{\omega}_{kl}\tau_p^2/2\pi)/\tau_p]\}\end{aligned} \tag{12a}$$

and

$$\begin{aligned}M_{2,kl}(t)=&\tfrac{1}{2}(E^0/2)^2\sum_{m=1}^{N}\mu_{km}\delta_{km}\mu_{ml}(1-\delta_{ml})\{[I(\omega_0,\text{sgn}\tilde{\omega}_{ml},t)+I(-\omega_0,\text{sgn}\tilde{\omega}_{ml},t)]\\&-[I(\text{sgn}\tilde{\omega}_{ml},\omega_0,t)+I(\text{sgn}\tilde{\omega}_{ml},-\omega_0,t)]\}\\&+\mu_{km}(1-\delta_{km})\mu_{ml}\delta_{ml}\{[I(\text{sgn}\tilde{\omega}_{km},\omega_0,t)+I(\text{sgn}\tilde{\omega}_{km},-\omega_0,t)]-[I(\omega_0,\text{sgn}\tilde{\omega}_{km},t)+I(-\omega_0,\text{sgn}\tilde{\omega}_{km},t)]\}\\&+\mu_{km}(1-\delta_{km})\mu_{ml}(1-\delta_{ml})[I(\text{sgn}\tilde{\omega}_{km},\text{sgn}\tilde{\omega}_{ml},t)-I(\text{sgn}\tilde{\omega}_{ml},\text{sgn}\tilde{\omega}_{km},t)]\ ,\end{aligned} \tag{12b}$$

respectively, for $k,l=1,2,\ldots,N$ and they reduce to Eq. (4) in the long-time limit. The I's appearing in Eq. (12b) are defined by

$$I(a,b,t)=\int_{-\infty}^{t}dt'\exp(-\pi t'^2/\tau_p^2+iat')\int_{-\infty}^{t'}dt''\exp(-\pi t''^2/\tau_p^2+ibt'')\ , \tag{12c}$$

where a and b are constants, and while they do not have obvious closed-form representations they are easily evaluated using standard Gaussian quadratures when expressed in the form

$$\begin{aligned}I(a,b,t)=&\tfrac{1}{4}\tau_p^2\exp[-(a^2+b^2)\tau_p^2/4\pi]\\&\times\left\{1+\text{erf}[\pi^{1/2}(t-ia\tau_p^2/2\pi)/\tau_p]+(2/\tau_p)\int_{-\infty}^{t}dt'\exp[-\pi(t'-ia\tau_p^2/2\pi)^2/\tau_p^2]\right.\\&\left.\times\text{erf}[\pi^{1/2}(t'-ib\tau_p^2/2\pi)/\tau_p]\right\}\ ,\end{aligned} \tag{12d}$$

which in the long-time limit reduces to

$$I(a,b,\infty)=\tfrac{1}{2}\tau_p^2\exp[-(a^2+b^2)\tau_p^2/4\pi]\{1+\text{erf}[i(a-b)\tau_p/2(2\pi)^{1/2}]\}\ . \tag{12e}$$

The time-resolved $\underline{U}(t)$ is evaluated through its Leverrier-Bateman decomposition in the form given by Eq. (5) where the λ's and the $\underline{N}$'s are now implicit functions of time. The accuracy of the computed $\underline{U}(t)$ can be assessed by the extent to which it fulfills the inequality[14(b)]

$$\hat{\underline{U}}(t)\le\underline{I}+N^{-1}\Pi\{\exp[N\Phi(t)]-1\}\ , \tag{13a}$$

where the caret on $\underline{U}(t)$ signifies that one takes the modulus of its matrix elements, Π is the Vandermonde matrix, and the scalar function $\Phi(t)$ is defined by

$$\Phi(t)=\int_{-\infty}^{t}dt\|\underline{C}(t)\|\le\tfrac{1}{2}\tau_p E^0[1+\text{erf}(\pi^{1/2}t/\tau_p)]\|\mu_z\|\ . \tag{13b}$$

Additionally, since the eigenvalues of the exact $\underline{U}(t)$ also satisfy the generalization of Eq. (7) as

$$\prod_{k=1}^{N} |\lambda_k(t)|=1 \tag{14a}$$

and

$$\sum_{k=1}^{N} \Delta_k(t)=(\tau_p E^0/8\pi)\exp[-(\omega_0\tau_p)^2/4\pi]\{2+\mathrm{erf}[\pi^{1/2}(t-i\omega_0\tau_p^2/2\pi)/\tau_p]+\mathrm{erf}[\pi^{1/2}(t+i\omega_0\tau_p^2/2\pi)/\tau_p]\} \sum_{k=1}^{N} \mu_{kk} \tag{14b}$$

for all t, the degree to which the approximate $\underline{U}(t)$ fulfills these relations is an indicator of its quality.

The time-resolved transition probabilities $P_{kk}^{(1)}(\omega_0,t)$, the dissociation probability $P_{\mathrm{diss}}^{(1)}(\omega_0,t)$, and the mean number of photons absorbed $\langle\omega_0(t)\rangle^{(1)}$ throughout the duration of a single pulse are given by the analog of Eqs. (9)–(11), respectively.

C. Spectral bandwidth averaging

The time-resolved and asymptotic operator expectation values, while they fully include the temporal variation in the field strength throughout the pulse duration, should strictly be further averaged over the pulse's spectral bandwidth. Thus, for example, the bandwidth-averaged asymptotic transition probability for excitation to $|k\rangle$ at the mean carrier frequency ω_0 is

$$P_{kk}^{(n)}(\omega_0,\infty)=\int_{-\infty}^{+\infty}d\omega\, F(E(t))P_{kk}^{(n)}(\omega,\infty)\Big/\int_{-\infty}^{+\infty}d\omega\, F(E(t)) , \tag{15a}$$

where the Fourier spectrum of $E(t)$ is given by

$$F(E(t))=\tfrac{1}{2}\tau_p E^0\{\exp[-(\omega-\omega_0)^2\tau_p^2/4\pi]+\exp[-(\omega+\omega_0)^2\tau_p^2/4\pi]\}\exp\left[\frac{i}{2}(n-1)\omega T\right]\csc(\tfrac{1}{2}\omega T)\sin(\tfrac{1}{2}n\omega T) , \tag{15b}$$

T being the time delay between contiguous pulses. George and co-workers[15] have addressed the necessity of properly including the time variation of the field and their calculations indicate that single-photon collision cross sections in a multimode laser field can be considerably different from those in a single-mode field of the same average intensity. The choice of N_Q Gauss-Hermite quadrature points $\{u_m\}$ and weights $\{a_m\}$ for the bandwidth averaging amounts operationally to convoluting $P_{kk}^{(n)}(\omega,\infty)$, say, with an N_Q-mode laser field;[15(a),15(b),16] these N_Q modes are adjacent axial modes of a single transverse mode and have nonfluctuating amplitudes and phases whose photons are Gaussian distributed about ω_0 at frequencies $\omega_0+\Delta\omega u_m$ containing fractional photon inventories $a_m/\pi^{1/2}$, $m=1,2,\ldots,N_Q$, respectively, delivered over a linewidth $\Delta\omega=4(\pi\ln 2)^{1/2}/\tau_p$. In the single-mode limit that $\Delta\omega\to 0$, $\tau_p\to\infty$ with which the cw laser field is monochromatic and bandwidth averaging is unnecessary.

III. DISCUSSION AND CONCLUSIONS

The last two decades have yielded a substantive literature[14] on the application of Floquet theory[12,13] to the dynamics of multiphoton processes in atomic and molecular systems through their interaction with intense laser-light sources. With the laser operating in either the cw mode or as a pulsed source having such long-duration pulses as to validate the SVAA,[2] the field is taken to be monochromatic and of constant intensity and the periodicity of the interaction limits the time interval over which one has to integrate the dynamical equations of motion to the initial optical cycle (2π in the variable $\omega_0 t$), following which the Floquet theorem permits one to cascade the evolution operator over an arbitrary number of optical cycles of the field. If the Hamiltonian operator is also self-adjoint and the quasienergies are discrete and bounded, Hogg and Huberman[17] have shown that the state amplitudes, or any bounded operator, are almost periodic and return arbitrarily close to their initial values infinitely often. Recent significant advances in the practical utility of the Floquet formalism are Tietz and Chu's most probable path approximation[18(a)] and Chang and Wyatt's use of artificial intelligence techniques[18(b),18(c)] for pruning the order of the Floquet matrix, each employing selection criteria that are dependent upon both resonance detunings and dipole coupling strength parameters, and the introduction of the recursive residue generation method (RRGM) by Wyatt and co-workers[19] for the evaluation of transition amplitudes of systems with a large number ($N\geq 10^3$) of levels. I have recently introduced[4(a)] an algebraic prescription for the construction of a Kubo-type transformation to a representation wherein the Hamiltonian operator for a multilevel system dipole interacting with a cw field is time independent, subject to the imposition of the RWA and the appeal to transition dipole selection rules that validate the neglect of unimportant couplings; the resultant sparse Hamiltonian operator is arbitrarily constructed on the basis of the near-resonant laser coupling of the ground state to the excited states, but without direct appeal to the magnitude of the dipole coupling strength parameters.

The Hamiltonian operator of a multilevel system under excitation by an ultrashort-duration laser pulse is not periodic and the entire arsenal of Floquet theory[12–14] and transformation procedure[4(a),6(c),20] is inapplicable. In previous papers[4(b),21] the strategy of computing the evolution operator, for both periodic and aperiodic Hamiltonian operators, through use of an algorithm based on its Riemann product integral representation[22] in conjunction with Frazer's method of mean coefficients,[23] has been advocated. Basically this involves the discretization of the interval $(-\infty, t]$ into a sufficient number of serial subintervals and evaluating a chronologically ordered product of matrix exponentials of the subinterval-averaged Hamiltonian operator. The optimum number of subintervals per optical cycle ensures the fulfillment at any time t of the inequality in Eq. (13) and the condition in Eq. (14) on the eigenvalues of $\underline{U}(t)$. The matrix exponentials are evaluated either by a complete spectral decomposition of their arguments[21] or, more efficiently, through use[4(b)] of the LBRM.[5] The more recent so-called "time-slicer" method proposed by Hirschfelder and co-workers[24] is identical to that algorithm[4(b),21] whose essential approach appears to have been first used[25] in an engineering context and whose rigorous basis is provided by the Riemann product integral formalism.[22] When one considers the requirement to bandwidth average operator expectation values, this approach is clearly prohibitively costly in processor and storage resources when N is large, say, $N \geq 100$.

In this paper, as a result of adopting the compound Magnus expansion in compliance with Maricq's convergence criterion,[8(l)] I have succeeded in dispensing with the computationally taxing need to evaluate matrix exponentials at a potentially large number of subintervals of each optical cycle sustained throughout a single Gaussian-shaped pulse's tenure in favor of a much smaller number of such evaluations when the time-resolved evolution operator is required or to just a single evaluation when one seeks the asymptotic value of the evolution operator. As before, Eqs. (13) and (14) serve to gauge the accuracy of the approximate $\underline{U}(t)$ for $|t| < \infty$. Since application of Eq. (7) requires the eigenvalues of $\underline{U}(\infty)$, one could, as an alternative to the use of Eq. (8a), also compute the corresponding eigenvectors and evaluate $\underline{U}^n(\infty)$ as $\underline{Z}\exp(n\underline{\Lambda})\underline{Z}^{-1}$ for $n = 1, 2, \ldots$, where $\underline{\Lambda}$ is the diagonal matrix containing the eigenvalues of $\underline{U}(\infty)$ and $\underline{Z}$ has the corresponding eigenvectors as columns. However, the computational time required to evaluate the requisite Bateman $\underline{N}$ matrices is much shorter than that required to evaluate $\underline{Z}$ and $\underline{Z}^{-1}$. The LBRM and the RRGM require only the eigenvalues of $\underline{M}(t)$ and obviate the need for the corresponding eigenvectors in constructing $\underline{U}(t) = \exp[\underline{M}(t)]$ for $|t| < \infty$; this common feature partly accounts for the efficiencies of both of these schemes.[26] Indeed, the independent Bateman matrices can be evaluated concurrently by parallel array processors and the fulfillment of the properties in Eq. (6) provides a cross-check on their accuracies. Furthermore, their idempotency renders Eq. (8a) as the analog of the cw Floquet result for a multilevel system interacting with a train of identical phase-coherent laser pulses.

The joint use of the RWA and the Magnus exponential expansion warrants some commentary. Appeal to the RWA was made solely on physical grounds and not as an expedient to yield Eqs. (4) and (12). Maricq's conjecture[8(l)] provides the basis for use of the compound Magnus expansion. Extension to higher orders in the expansion is straightforward although, whatever about MACSYMA, the MuMath and Maple Leaf symbolic manipulators are not helpful in this regard.

One notes that the approach adopted herein for solving the state amplitude equation of motion for a multilevel system interacting with a Gaussian-shaped laser pulse is formally analogous to Hyman's procedure[27] for solving the many-channel, close-coupled symmetrized impact-parameter equations description[28] of electron-atom collisions. In the electron-atom scattering context the Hamiltonian operator originates with the coupling of many internal states of the target atom by the incident electron via their long-range Coulombic forces and is given in terms of the asymptotic straight-line trajectory velocities and wave numbers for each channel, the impact parameter, the position vector of the electron with respect to the atom as origin, and the matrix elements coupling different channels through the dipolar term of the potential. In both cases the Magnus matrix is available through first- and second-order terms as closed-form analytic expressions and the numerical work is essentially reduced to a matrix exponentiation. In the present context this exponentiation is required over the spectral bandwidth of the exciting pulse, while in the electron-atom collision context it is required for each value of the impact parameter of the projectile. To accomplish this task I advocate use of the LBRM, while Hyman, following Mandelberg,[29] chooses the aforementioned diagonalization procedure.

In conclusion, the computational effort required to evaluate the evolution operator for a multilevel system interacting with a realistically shaped short-duration pulse is not appreciably greater than that needed to apply either Floquet analysis[14] or transformation procedures[4(a),6(c),20] and mitigates reliance on the assumption that the SVAA is valid. In return for the effort expended in evaluating the evolution operator over the tenure of a single pulse, Eq. (8a) permits one to efficiently cascade the state amplitude matrix over the total duration of a train of such phase-coherent pulses, predicated on the convergence of the Magnus cumulant expansion of the evolution operator. Obviously the entire approach presented here lends itself to pulse shapes other than Gaussian; indeed, only Eqs. (4), (7b), (12), (13), (14b), and (15b) are affected by a change in the pulse's shape as embodied in the modulating envelope $f(t)$. Also, Maricq's radius of convergence of the compound Magnus expansion depends on $\int_{-\infty}^{+\infty} dt\, f(t)$, the epochal duration of the pulse. The suitability[26] of the proposed algorithm for implementation on emerging multiprocessor machines[30] is under investigation in the context of the multiphoton excitation of molecules by pulsed high-intensity lasers. The results of these applications will be reported elsewhere.

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