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Resonant multiphoton ionization by finite-bandwidth chaotic fields

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Resonant multiphoton ionization by finite bandwidth multimode radiation is investigated without the restriction of either a "long" or a "short" correlation time of the field. By approximating the multimode radiation by a stochastic model of a chaotic field, the stochastic atomic-density-matrix equation is reduced to a tractable infinite set of differential equations. For a given bandwidth this set can be suitably truncated and numerically integrated. For large-bandwidth fields it turns out that these truncated equations constitute a systematic improvement of the method usually employed of decorrelating the atom-field variables. For zero-bandwidth fields the well-known result is recovered in that the statistical averaging of the ionization probability reduces to an average with respect to the Glauber P-distribution function of the chaotic field. Detailed results of numerical calculations for two-photon ionization are presented which reveal a number of new interesting features.

I. INTRODUCTION

Recently a number of authors have investigated theoretically resonant multiphoton ionization (RMPI) in intense multimode laser fields.[1-7] The multimode laser field is usually approximated by an ideal chaotic field (CF) which is known to be a good model for a multimode laser with a large number of uncorrelated modes.[8] But up to now only the limiting cases of long and short correlation time, i.e., small and large bandwidth, of the CF have been considered, where "long" and "short" corresponds to a comparison of the time scale of the atom as determined by the induced widths, shifts, and Rabi frequencies of the atom with the time scale of the fluctuations in the CF.[1-7] In the limit of long coherence time the laser can be assumed to be monochromatic and the averaging of the ionization probability (IP) with respect to the fluctuations in the CF reduces to a simple averaging with respect to the Glauber P-distribution function[9] of the CF.[1-5,10] The limit of short correlation time has been treated by statistical factorization assumptions,[6,7] because the fluctuations in the atomic populations are much slower than those in the CF. However, as has been pointed out by Georges and Lambropoulos,[11] theories employing decorrelation assumptions are necessarily weak-field theories. If the intensity is increased, the correlation time becomes comparable to the atomic time scale, and no rigorous solution exists, however urgently it may be called for by current experiments.[6] In this paper we attempt a solution of the problem of RMPI by intense finite bandwidth CF's.

To this end, a certain stochastic model for the CF is introduced. As a consequence, the atomic density-matrix equations describing the resonant interaction with this field—originally a system of stochastic differential equations—are reduced to a tractable infinite set of differential equations. For a given bandwidth of the field, this set can be suitably truncated and hence numerically integrated. It turns out that for large-bandwidth fields, these truncated equations constitute a systematic improvement of the decorrelation results of Agostini *et al.*[6] When the bandwidth of the field is small, their solutions are good approximations only for interaction times not much larger than a few inverse Rabi frequencies or ionization rates, which, however, is just the time interval most interesting in the RMPI process. In a series of figures, we present the time evolution of the IP and populations of the resonant bound states as a function of the bandwidth. We also study the dispersion curves and the dependence of the IP on the Rabi frequency and ionization rate from the exited state.

II. A MODEL FOR RESONANT ATOM-MULTIMODE-LASER INTERACTION

A multimode-laser field with a large number of uncorrelated modes can be represented to a good approximation by a nonmonochromatic CF.[8] More specifically, we assume a model of chaotic light[24] where the amplitude of the electric field $E(t)=x(t)+iy(t)$ obeys the Langevin equations[12,20]

$$\frac{dx}{dt}=-bx+F_x(t)\,,\quad \frac{dy}{dt}=-by+F_y(t)\,. \tag{1}$$

$F_x(t)$ and F_y are Gaussian random forces with

$$\langle F_x(t)F_x(t')\rangle=\langle F_y(t)F_y(t')\rangle=bI_0\delta(t-t')$$

and

$$\langle F_x(t)F_y(t')\rangle = 0 .$$

Therefore, $E(t)$ obeys a normal Markov process. The meaning of b and I_0 and the chaotic nature of the light described by (1) can be established by calculating the correlation functions. To do this, we describe the stochastic process (1) by its associated Fokker-Planck equation[12] $(\partial/\partial t + L)P = 0$ as this is the approach on which we will extensively rely in later sections. P is the distribution function and L is the Fokker-Planck operator which for our model assumes the form

$$L = 2b\left(-\frac{\partial}{\partial I}(I - I_0) - I_0\frac{\partial^2}{\partial I^2}I - \frac{I_0}{4I}\frac{\partial^2}{\partial\phi^2}\right) . \tag{2}$$

Here we eliminated x and y in favor of the intensity I and phase ϕ according to $x + iy = \sqrt{I}e^{-i\phi}$. Now we easily find with the help of the Green's function $P(I,\phi,t\,|I',\phi',t')$ of the Fokker-Planck equation that the stationary two time correlation function

$$G^{(\alpha,\alpha)}(t,t') = \langle[E^*(t)E(t')]^\alpha\rangle$$

is given by[20]

$$\begin{aligned} G^{(\alpha,\alpha)}(t,t') &= \int dI\,d\phi\,dI'\,d\phi'\,(\sqrt{I}e^{i\phi})^\alpha P(I,\phi,t\,|I',\phi',t') \\ &\quad\times(\sqrt{I'}e^{-i\phi'})^\alpha P(I',\phi',t') \\ &= \alpha!\,I_0^\alpha e^{-\alpha b|t-t'|} \\ &= \alpha!\,(G^{(1,1)}(t,t'))^\alpha , \end{aligned} \tag{3}$$

which is a typical property of CF's.[9] Thus it turns out that I_0 and b must be identified with the mean intensity and bandwidth of the field, respectively. The frequency spectrum has a Lorentzian shape. By comparison, the same correlation function for a single-mode laser with a diffusing phase would be

$$G^{(\alpha,\alpha)}(t,t') = I_0^\alpha e^{-\alpha^2 b|t-t'|}$$

(Ref. 23).

The resonant interaction of an atom with the above chaotic light field is most simply described in terms of a two-state atom with levels $|0\rangle$ and $|1\rangle$ coupled to the continuum.[13-17] The nonresonant levels can be treated by perturbation theory leading to Stark shifts, ionization widths, and higher-order corrections to the Rabi frequency.[14] Of these nonresonant contributions, in the following only the ionization rate from the upper state $|1\rangle$ to the continuum is kept. Furthermore, we assume the lower state $|0\rangle$ to be resonantly coupled to $|1\rangle$ by the absorption of α photons while the ionization from $|1\rangle$ requires the absorption of β photons. Then we find for the slowly varying density-matrix elements[6]

$$\left(i\frac{d}{dt} + Q(I(t),\phi(t))\right)\begin{bmatrix}\rho_{10}(t)\\ \rho_{01}(t)\\ \rho_{11}(t)\\ \rho_{00}(t)\end{bmatrix} = 0 \tag{4}$$

with

$$Q(I,\phi) = \begin{bmatrix} \Delta + \frac{1}{2}i\gamma_1(I) & 0 & V(I)e^{-i\alpha\phi} & -V(I)e^{-i\alpha\phi} \\ 0 & -\Delta + \frac{1}{2}i\gamma_1(I) & -V(I)e^{i\alpha\phi} & V(I)e^{i\alpha\phi} \\ V(I)e^{i\alpha\phi} & -V(I)e^{-i\alpha\phi} & i\gamma_1(I) & 0 \\ -V(I)e^{i\alpha\phi} & V(I)e^{-i\alpha\phi} & 0 & 0 \end{bmatrix}$$

and $\Delta = \alpha\omega - \omega_{10}$. ω_{10} is the atomic transition frequency and ω is the mean frequency of the incident chaotic light. The coupling between $|0\rangle$ and $|1\rangle$ is determined by

$$V(I) = I^{\alpha/2}\sum_{i\cdots j\neq 1,0}\frac{(-\vec{\mu}_{0i}\vec{e})\cdots(-\vec{\mu}_{j1}\vec{e})}{(\omega_{0i}+\omega)\cdots[\omega_{0j}+(\alpha-1)\omega]} \tag{5}$$

with the atomic dipole matrix elements $\vec{\mu}_{ij}$ and the polarization vector of the radiation field $\vec{e}$.

$\gamma_1(I)$ is given by

$$\gamma_1(I) = 2\pi I^\beta\left|\sum_{i\cdots j\neq 1,0}\frac{(-\vec{\mu}_{1i}\vec{e})\cdots(-\vec{\mu}_{jE}\vec{e})}{(\omega_{1i}+\omega)\cdots[\omega_{1j}+(\beta-1)\omega]}\right|^2 . \tag{6}$$

Both V and γ_1 depend on $I(t)$ and are therefore stochastic functions of time. In principle, the IP $\langle P(t)\rangle$ can now be found by solving (4) for $\rho_{11}(t)$

and $\rho_{00}(t)$ and averaging with respect to the field fluctuations:

$$\langle P(t)\rangle = 1 - \langle\rho_{11}(t)\rangle - \langle\rho_{00}(t)\rangle . \quad (7)$$

The system of stochastic differential equations (4) is very difficult to solve. Only in the limits of large-bandwidth fields $V(I(t))$, $\gamma_1(I(t)) \ll b$ and, trivially, monochromatic radiation ($b=0$) general methods of solution have been given.[18] Instead of pursuing a direct integration of (4), we make use of a theorem[18] stating that the averaged populations $\langle\rho_{11}(t)\rangle$ and $\langle\rho_{00}(t)\rangle$ can be found by solving the system of partial differential equations

$$\left[i\left(\frac{\partial}{\partial t}+L\right)+Q(I,\phi)\right]\begin{bmatrix}\rho_{10}(I,\phi,t)\\ \rho_{01}(I,\phi,t)\\ \rho_{11}(I,\phi,t)\\ \rho_{00}(I,\phi,t)\end{bmatrix}=0 \quad (8)$$

with the intial condition

$$\rho_{ij}(I,\phi,t)=\langle\rho_{ij}(t=0)\rangle P_{00}(I,\phi),$$

where L is the Fokker-Planck operator (2) and

$$P_{00}(I,\phi)=(1/2\pi I_0)e^{-I/I_0}$$

is the stationary solution $LP_{00}=0$ of the Fokker-Planck equation. The statistical averages can then be found from

$$\langle\rho_{ij}(t)\rangle=\int_0^\infty dI\int_0^{2\pi}d\phi\,\rho_{ij}(I,\phi,t). \quad (9)$$

This formulation is usually more easily accessible to systematic analysis than the original equations (4). The same approach has been used previously in the context of interaction of a two-level system with a single-mode laser with a diffusing phase and has led in a simple way to exact solutions.[19]

III. RESONANT MULTIPHOTON IONIZATION BY MONOCHROMATIC CHAOTIC FIELDS

If the bandwidth of the CF can be neglected, i.e., the multimode laser has a large number of modes of the same frequency, the operator L drops out of (8), and (8) becomes formally identical with the usual density matrix equation depending only *parametrically* on I and ϕ. The corresponding solutions

$$\rho_{00}^{b=0}(I,\phi,t)=\left|\frac{q+p}{2q}e^{i(p-q)t}+\frac{q-p}{2q}e^{i(p+q)t}\right|^2, \quad (10a)$$

$$\rho_{11}^{b=0}(I,\phi,t)=\left|\frac{V(I)}{2q}\left(e^{i(p-q)t}-e^{i(p+q)t}\right)\right|^2, \quad (10b)$$

with

$$p=\tfrac{1}{2}[\Delta+\tfrac{1}{2}i\gamma_1(I)],$$

$$q=\{\tfrac{1}{4}[\Delta+\tfrac{1}{2}i\gamma_1(I)]^2+|V(I)|^2\}^{1/2},$$

and

$$\rho_{00}^{b=0}(I,\phi,t=0)=1,\quad \rho_{11}^{b=0}(I,\phi,t=0)=0$$

have been given by Beers and Armstrong[14] in their investigation of RMPI by monochromatic coherent single-mode laser fields. In view of (9) we find for the statistically averaged IP

$$\langle P(t)\rangle=1-\int_0^\infty dI\int_0^{2\pi}d\phi\,P_{00}(I,\phi)[\rho_{11}^{b=0}(I,\phi,t)+\rho_{00}^{b=0}(I,\phi,t)]. \quad (11)$$

This result is well known: for infinite correlation time the statistical averaging reduces to an average with respect to the Glauber P-distribution function[9] of the CF $P_{00}(I,\phi)$.[1-5,10]

In general, (11) can only be integrated numerically. We readily verify, however, that many of the analytical formulas that have so far been published on RMPI by monochromatic CF's are special cases of (11). For example, for $\Omega\ll\gamma_1$, $\gamma_1 t\gg 1$, and $\alpha\geq\beta$, we find[1,2,4,5]

$$\frac{d}{dt}\langle P(t)\rangle=\int_0^\infty dx\,e^{-x}\,\frac{1}{4}\,\frac{\Omega^2\gamma_1 x^{\alpha+\beta}}{\Delta^2+(\frac{1}{2}\gamma_1 x^\beta)^2} \quad (12)$$

with $\Omega=|2V(I_0)|$ and $\gamma_1=\gamma_1(I_0)$ while for $\Omega\ll\gamma_1$ and $\gamma_1 t\ll 1$ (11) reduces to[3-5]

$$\frac{d}{dt}\langle P(t)\rangle=\int_0^\infty dx\,e^{-x}\frac{1}{2}\frac{\Omega^2\gamma_1 x^{\alpha+\beta}}{\Delta^2+\Omega^2 x^\alpha}. \quad (13)$$

Ω and γ_1 denote the Rabi frequency and ionization rate at $I=I_0$. Both expressions yield the well-known off-resonance factor $(\alpha+\beta)!$ which on-resonance reduces to $(\alpha-\beta)!$ and $\beta!$, respectively. However, the range of validity of these analytical expressions is so restricted that hardly more than a qualitative insight can be gained from them in the general case. Moreover, there is little more numerical labor involved in evaluating (11) directly. Results of such calculations will be presented in Sec. V.

IV. RESONANT MULTIPHOTON IONIZATION BY FINITE-BANDWIDTH FIELDS

For finite-bandwidth fields we solve the system of partial differential equations (8) by expanding $\rho_{ij}(I,\phi,t)$ in the complete biorthogonal set of eigenfunctions $\varphi_{\alpha n}$ and $P_{\alpha n}$ of L,[12,20]

$$\rho_{ij}(I,\phi,t)=\sum_{\alpha n}P_{\alpha n}(I,\phi)\rho_{ij}^{\alpha n}(t)$$

with

$$\rho_{ij}^{\alpha n}(t)=\int dI\,d\phi\,\varphi_{\alpha n}^{*}(I,\phi)\rho_{ij}(I,\phi,t)\,. \tag{14}$$

The functions $\varphi_{\alpha n}$ and $P_{\alpha n}$ are solutions of $LP_{\alpha n} = \Lambda_{\alpha n}P_{\alpha n}$ and $L^{+}\varphi_{\alpha n}=\Lambda_{\alpha n}\varphi_{\alpha n}$, respectively. Explicitly, they are given by

$$P_{\alpha n}(I,\phi)=P_{\alpha n}(I)\frac{e^{i\alpha\phi}}{2\pi}$$

with

$$P_{\alpha n}(I)=\frac{1}{I_0}\left(\frac{n!}{(n+|\alpha|)!}\right)^{1/2}e^{-x}x^{|\alpha|/2}L_n^{|\alpha|}(x)$$

$$(\alpha=0,\pm1,\ldots;n=0,1)\,,$$

and (15)

$$\varphi_{\alpha n}(I,\phi)=\varphi_{\alpha n}(I)e^{+i\alpha\phi}$$

with

$$\varphi_{\alpha n}(I)=\left(\frac{n!}{(n+|\alpha|)!}\right)^{1/2}x^{|\alpha|/2}L_n^{|\alpha|}(x)\,,$$

where $x=I/I_0$. The eigenvalues are $\Lambda_{\alpha n}=b(2n+|\alpha|)$. $L_n^{\alpha}(x)$ are Laguerre polynomials as defined by Gradsteyn and Ryzhik.[21] In view of $\varphi_{00}=1$, we have

$$\langle\rho_{ij}(t)\rangle=\rho_{ij}^{00}(t)\,. \tag{16}$$

By inserting the ansatz (14) into (8) we are led to an infinite system of differential equations for the unknown coefficients $\rho_{ij}^{\alpha n}(t)$:

$$\begin{aligned}
&\left(i\frac{d}{dt}+\Delta+i\Lambda_{-\alpha n}\right)\rho_{10}^{-\alpha n}+\sum_m\left[\tfrac{1}{2}i(\varphi_{-\alpha n},\gamma_1(I)P_{-\alpha m})\rho_{10}^{-\alpha m}+(\varphi_{-\alpha n},V(I)P_{0m})\rho_{11}^{0m}-(_{-\alpha n},V(I)P_{0m})\rho_{00}^{0m}\right]=0\,,\\
&\left(i\frac{d}{dt}-\Delta+i\Lambda_{\alpha n}\right)\rho_{01}^{\alpha n}+\sum_m\left[\tfrac{1}{2}i(\varphi_{\alpha n},\gamma_1(I)P_{\alpha m})\rho_{01}^{\alpha m}-(\varphi_{\alpha n},V(I)P_{0m})\rho_{11}^{0m}+(\varphi_{\alpha n},V(I)P_{0m})\rho_{00}^{0m}\right]=0\,,\\
&\left(i\frac{d}{dt}+i\Lambda_{0n}\right)\rho_{11}^{0n}+\sum_m\left[i(\varphi_{0n},\gamma_1(I)P_{0m})\rho_{11}^{0m}+(\varphi_{0n},V(I)P_{-\alpha m})\rho_{10}^{-\alpha m}-(\varphi_{0n},V(I)P_{\alpha m})\rho_{01}^{\alpha m}=0\,,\right.\\
&\left(i\frac{d}{dt}+i\Lambda_{0n}\right)\rho_{00}^{0n}-\sum_m\left[(\varphi_{0n},V(I)P_{-\alpha m})\rho_{10}^{-\alpha m}-(\varphi_{0n},V(I)P_{\alpha m})\rho_{01}^{\alpha m}\right]=0\,.
\end{aligned} \tag{17}$$

The matrix elements are easily calculated with the help of

$$\begin{aligned}
&(\varphi_{\alpha n},\sqrt{I}P_{\alpha+1m})=\sqrt{I_0}\begin{cases}(n+\alpha+1)^{1/2}\delta_{n,m}-n^{1/2}\delta_{n,m+1}\,, & \alpha\geq0\\ (n+|\alpha|)^{1/2}\delta_{n,m}-(n+1)^{1/2}\delta_{n,m-1}\,, & \alpha<0\,,\end{cases}\\
&(\varphi_{\alpha n},\sqrt{I}P_{\alpha-1m})=\sqrt{I_0}\begin{cases}(n+\alpha)^{1/2}\delta_{n,m}-(n+1)^{1/2}\delta_{n,m-1}\,, & \alpha>0\\ (n+|\alpha|+1)^{1/2}\delta_{n,m}-n^{1/2}\delta_{n,m+1}\,, & \alpha\leq0\,,\end{cases}\\
&(\varphi_{\alpha n},IP_{\alpha m})=I_0\left\{-[(n+1)(n+|\alpha|+1)]^{1/2}\delta_{n,m-1}+\delta_{n,m}-[n(n+|\alpha|)]^{1/2}\delta_{n,m+1}\right\}.
\end{aligned} \tag{18}$$

For an atom initially in the ground state, these equations must be solved subject to the initial condition

$$\rho_{ij}^{\alpha n}(t=0)=\delta_{i0}\delta_{j0}\delta_{\alpha 0}\delta_{n0}\,. \tag{19}$$

The usefulness of this expansion method depends on the convergence of the series (14), since in practical calculations only a few terms can be kept. We first investigate the possibility of such a truncation for large-bandwidth fields ($\Omega,\gamma_1\ll b$) in the near-resonance region. For these, we readily verify that the truncated set of differential equations yield solutions which are approximations in the sense of a power series expansion in $\Omega/b\ll1$ and $\gamma_1/b\ll1$. To lowest order, if only the first term in expansion (14) is kept, it turns out that (17) becomes identical with the decorrelation results of Agostini *et al.*[6] If, in this approximation, we adiabatically eliminate the nondiagonal elements, i.e., neglect the time derivatives $(d/dt)\rho_{10}^{-10}=0$, $(d/dt)\rho_{01}^{10}=0$ according to Ω, $\gamma_1\ll b$,[22] we find a rate equation for the populations of the resonant states:

$$\begin{aligned}
&\frac{d}{dt}\langle\rho_{00}\rangle=-W\langle\rho_{00}\rangle+W\langle\rho_{11}\rangle\,,\\
&\frac{d}{dt}\langle\rho_{11}\rangle=-(W+\beta!\gamma_1)\langle\rho_{11}\rangle+W\langle\rho_{00}\rangle\,;
\end{aligned} \tag{20}$$

$$W=\alpha!\,\frac{1}{2}\Omega^2\frac{\alpha b+[(\alpha+\beta)!/2\alpha!]\gamma_1}{\Delta^2+\{\alpha b+[(\alpha+\beta)!/2\alpha!]\gamma_1\}^2} \quad (21)$$

may be interpreted as the transition probability per unit time $|0\rangle-|1\rangle$ while $\beta!\gamma_1$ gives the ionization rate from $|1\rangle$ to the continuum. The modifications as compared with the finite-bandwidth single-mode case[7,25] are the factorial enhancement factors $\alpha!$ and $\beta!$ for the transition probabilities which are typical of CF's.[23] Note that also the width of the Lorentzian in (21) due to ionization is increased by $(\alpha+\beta)!/\alpha!$. Similarly, a second-order Stark shift, which has not been included in (21) would have been increased by a factor $\alpha+1$. The solution for $\langle P(t)\rangle$ is found to be

$$\langle P(t)\rangle=1-\frac{1}{2}\frac{A+B}{B}e^{-(A-B)t}+\frac{1}{2}\frac{A-B}{B}e^{-(A+B)t} \quad (22)$$

with

$$A=W+\tfrac{1}{2}\beta!\gamma_1 \text{ and } B=[W^2+(\tfrac{1}{2}\beta!\gamma_1)^2]^{1/2}.$$

Depending on $W\ll\frac{1}{2}\beta!\gamma_1$ or $W\gg\frac{1}{2}\beta!\gamma_1$, a "bottleneck" for the ionization process occurs in the first or second step and the atom ionizes with the rate

$$\tfrac{1}{2}W\beta!\gamma_1/(W+\tfrac{1}{2}\beta!\gamma_1), \quad (23)$$

which closely parallels the corresponding result for the nonmonochromatic single-mode field.[7,25] The off-resonance factor $(\alpha+\beta)!$ is correctly reproduced by the rate equations for large detuning $\Delta\gg\Omega,\ \gamma_1,\ b$

$$\frac{d\langle P(t)\rangle}{dt}=(\alpha+\beta)!\,\frac{1}{4}\frac{\Omega^2\gamma_1}{\Delta^2}+\alpha!\,\frac{1}{2}\frac{\Omega^2\alpha b}{\Delta^2}. \quad (24)$$

The factor proportional to b stems from the on-resonance component of the Lorentzian spectrum of the CF[7] and does not vanish for large detunings due to the long tail of the Lorentzian.[11] Since a realistic laser spectrum has a cutoff,[13] this is an unphysical feature of models with Lorentzian frequency spectra[6,7,25] and restricts their application to the resonance region.

When the bandwidth of the field is small ($\Omega,\gamma_1\gtrsim b$) in the near-resonance region, the truncated expansion can be expected to converge only for interaction times of the order of a few inverse Rabi frequencies and ionization rates.[18] However, this condition is far less restricting than it may seem since this time suffices to ionize most of the atoms. The accuracy of the result so obtained can be easily assessed for $b=0$, where one would expect the convergence to be worse, since the exact solution [cf. Eq. (11)] is known in this case. This is done in Sec. V, where the numerical solution of (17) for various values of b is graphically represented together with the exact solution for $b=0$.

V. NUMERICAL RESULTS AND DISCUSSION

In general, the truncated set of differential equations (17) must be solved numerically. Results of such calculations are presented in Figs. 1–6 for $\langle P(t)\rangle$, $\langle\rho_{11}(t)\rangle$, and $\langle\rho_{00}(t)\rangle$ for the case of two-photon ionization, i.e., $\alpha=\beta=1$. In all of these figures, the results for the single-mode field with $b=0$, i.e., a monochromatic coherent radiation field with constant amplitude and phase, are compared with those for the CF, whose amplitude is distributed according to a Gaussian function,[9] with $b=0$, 0.5, 1, 2, and 5, respectively. The case of zero-bandwidth field has been calculated both with nine terms (Figs. 1–4) or five terms (Figs. 5 and 6) of the expansion method and the exact expression (10) in order to assess the accuracy of the former.

A. On-resonance time evolution

The nine figures 1(a)–3(c) show the on-resonance time evolution ($\Delta=0$). In all of them, the letters (a), (b), and (c) stand for $\langle P(t)\rangle$, $\langle\rho_{11}(t)\rangle$, and $\langle\rho_{00}(t)\rangle$, respectively, while the first figure index denotes a specific value of the ionization rate γ_1. In particular, in the three sets of figures 1, 2, and 3 γ_1 takes on the values 0.2, 2, and 5, respectively. The Rabi frequency is kept constant at $\Omega=1$ throughout.

In Fig. 1 the condition $\Omega=1\gg\frac{1}{2}\gamma_1=0.1$ is satisfied. In the single-mode case well-resolved Rabi oscillations appear in the population of the ground and excited states [Figs. 1(b) and 1(c)]. The behavior of the ionization process for times $\gamma_1 t<1$ is well described by formula (18) of Beers and Armstrong.[14] The corresponding IP for monochromatic CF's is given by (13). As described by these equations, the on-resonance IP is the same for the single mode and CF for $t<1/\gamma_1$. This behavior is clearly brought out in Fig. 1(a). For large interaction times $t<1/\gamma_1$, however, the on-resonance single-mode laser ionizes more efficiently according to $P(t)=1-\exp(-\frac{1}{2}\gamma_1 t)$ as compared with $\langle P(t)\rangle=\frac{1}{2}\gamma_1 t/(1+\frac{1}{2}\gamma_1 t)$ for the CF. This different time behavior is explained by the CF distribution function $P_{00}(I,\phi)$ favoring small intensities which ionize on a slower time scale. The difference becomes even more pronounced for the bound-state populations where instead of Rabi oscillations the atomic occupation numbers equalize in times of the order $1/\Omega$ and afterwards slowly decay due to ionization [Figs. 1(b) and 1(c)]. If the bandwidth of the CF is increased, the on-resonance IP decreases, thus smoothing the time

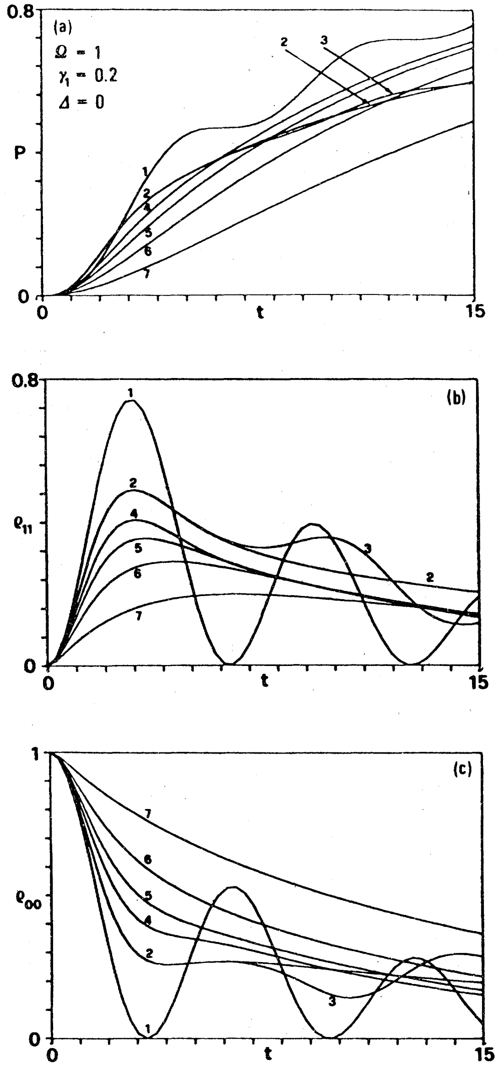

FIG. 1. On-resonance ($\Delta = 0$) time evolution of (a) the ionization probability $\langle P(t)\rangle$, (b) the excited-state population $\langle \rho_{11}(t)\rangle$, and the ground-state population $\langle \rho_{00}(t)\rangle$, with $\Omega = 1$ and $\gamma_1 = 0.2$. The curve index running from 1–7 denotes results for 1, monochromatic single-mode radiation (Ref. 14); 2, the CF with $b = 0$ as given by the exact expression (11); and 3–7 the CF with $b = 0$, 0.5, 1, 2, and 5, respectively, employing a nine-term expansion.

evolution of the bound-state populations. This is to be expected, since the intensity of the CF is now distributed with a bandwidth b over the resonance. The limit of large-bandwidth fields is well described by the rate equations (20). It is interesting to note that for large interaction times the finite-bandwidth IP may exceed the corresponding value for monochromatic light. Therefore, an optimum bandwidth exists as a function of time which is not a peculiarity of our approximation scheme, as may be seen from the excellent agreement of the approximate solution with the exact one for $b = 0$ [Fig. 1(a)].

Increasing the rate γ_1 to $\gamma_1 = 2$ (Fig. 2), we reach the critical parameter regime $\Omega \approx \frac{1}{2}\gamma_1$ where no single rate approximation is available for the IP in the single-mode laser field (case 3 of Beers and Armstrong[14]). Similar to Fig. 1(a), the IP for the zero bandwidth CF is initially very near the single-mode value, but later falls behind the IP in the nonmonochromatic CF as we noted before [Fig. 2(a)]. After the transient time $t > 1/\Omega$, $1/\gamma_1$ the ground- and excited-state populations take on values which on the average are much

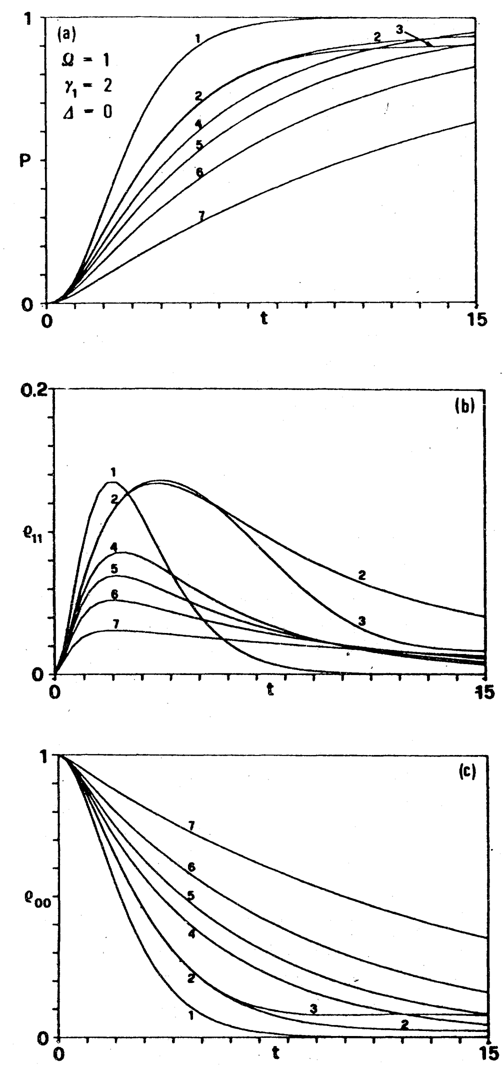

FIG. 2. On-resonance time evolution of (a) $\langle P(t)\rangle$, (b) $\langle \rho_{11}(t)\rangle$, and (c) $\langle \rho_{00}(t)\rangle$ with $\Omega = 1$ and $\gamma_1 = 2$. The meaning of the curve index is explained in the caption for Fig. 1.

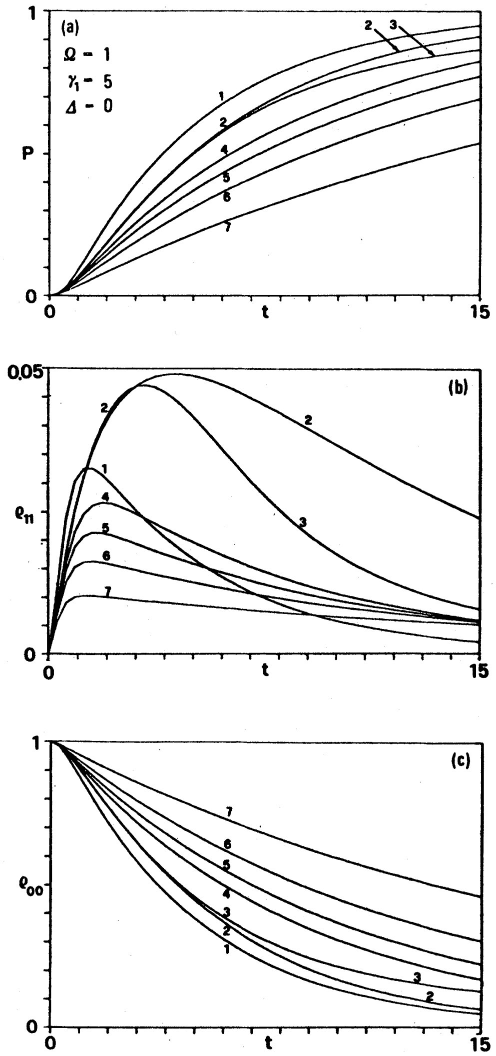

FIG. 3. On-resonance time evolution of (a) $\langle P(t)\rangle$, (b) $\langle \rho_{11}(t)\rangle$, and (c) $\langle \rho_{00}(t)\rangle$, with $\Omega=1$ and $\gamma_1=5$. The meaning of the curve index is explained in the caption of Fig. 1.

larger than those in the single-mode field [Figs. 2(b) and 2(c)]. With increasing bandwidth the IP is again reduced except for large interaction times, where the most efficient ionization again occurs for a small but nonzero bandwidth [Fig. 2(a)].

When γ_1 takes on the large value $\gamma_1=5$ (Fig. 3), the single-mode-laser IP is described by standard perturbation theory [see Eq. (16) of Beers and Armstrong[14]]. The IP for the monochromatic CF, as given by (12) for $\gamma_1 t \gg 1$ and $\langle P(t)\rangle \ll 1$, is now always only slightly below the single-mode value contrary to our earlier findings [Fig. 3(a)]. The number of atoms in the excited state, which is rather small due to the large value of γ_1, is strongly increased by the CF as compared with the single-mode laser. A larger bandwidth again results in a lowering of the IP and a rapid decrease of the population in the resonant excited state. However, within the range of interaction times covered by Fig. 3, the finite-bandwidth IP no longer dominates the monochromatic CF value [Fig. 3(a)].

From the above discussion it is obvious that in

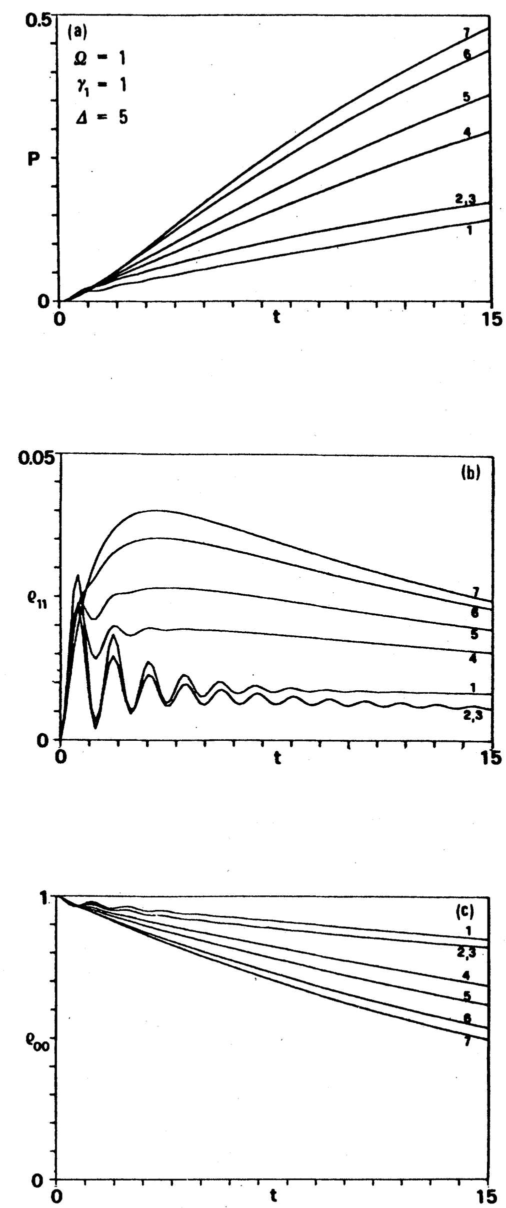

FIG. 4. Off-resonance ($\Delta=5$) time evolution of (a) $\langle P(t)\rangle$, (b) $\langle \rho_{11}(t)\rangle$, and (c) $\langle \rho_{00}(t)\rangle$, with $\Omega=1$ and $\gamma_1=1$. The meaning of the curve index is explained in the caption of Fig. 1.

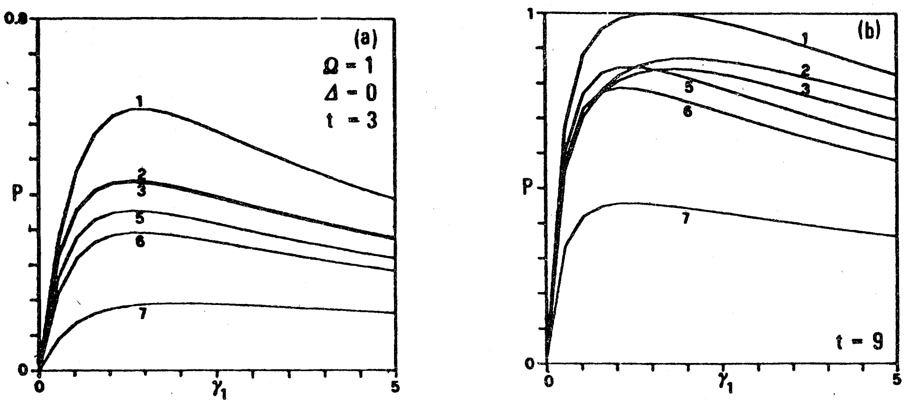

FIG. 5. γ_1 dependence of $\langle P(t)\rangle$ with $\Delta=0$, $\Omega=1$, $t=3$, and $t=9$ in (a) and (b), respectively, employing a five-term expansion. The meaning of the curve index is explained in the caption of Fig. 1.

general the on-resonance IP cannot be described by a single time-independent rate γ, implying $\langle P(t)\rangle = 1-\exp(-\gamma t)$. An analogous conclusion was drawn by Beers and Armstrong[14] in the context of RMPI by monochromatic single-mode lasers, who suggested that a transition probability per unit time need not exist as a meaningful quantity.

As for the accuracy of the nine-term expansion, which has been used in the calculation of Figs. 1–3, we find that in all three cases the zero-bandwidth IP agrees with the exact values not only for some inverse Rabi frequencies Ω and ionization rates γ_1, as expected from the general discussion in Sec. IV, but is remarkably accurate for the whole time interval considered. For the bound-state populations the deviations from the exact value are larger, but they mainly occur after most of the atoms are ionized.

B. Off-resonance time evolution

In Fig. 4 the time evolution of $\langle P(t)\rangle$, $\langle \rho_{11}(t)\rangle$, and $\langle \rho_{00}(t)\rangle$ is given when the laser is tuned slightly off resonance with $\Delta=5$, $\Omega=1$, and $\gamma_1=1$. Now, the IP due to the monochromatic CF already approaches twice the single-mode value as predicted by perturbation theory for nonresonant two-photon ionization [Fig. 4(a)]. With increasing bandwidth, the on-resonance components of the spectrum populate the excited state more effectively and, therefore, the IP strongly grows. Contrary to our on-resonance findings, Rabi oscillations appear in the bound-state population [Figs. 4(b) and 4(c)]. For the parameter values in Fig. 4 their frequency is approximately the same as in the single-mode field, but they are smaller in amplitude and become progressively damped for increasing bandwidth. For large detuning, these oscillations are the usual transient effects which are negligible because of their large frequency and small amplitude.

C. γ_1-dependence of the ionization probability

A comparison of Eqs. (12) and (13) shows that on resonance, the growing rate γ_1 leads to an increase of the IP for $\Omega \gg \gamma_1$, but to a decrease for $\Omega \ll \gamma_1$. This dependence of $\langle P(t)\rangle$ on γ_1 is shown for $\Omega=1$, $t=3$, and $t=9$ in Figs. 5(a) and 5(b). There it can be seen that the ionization is

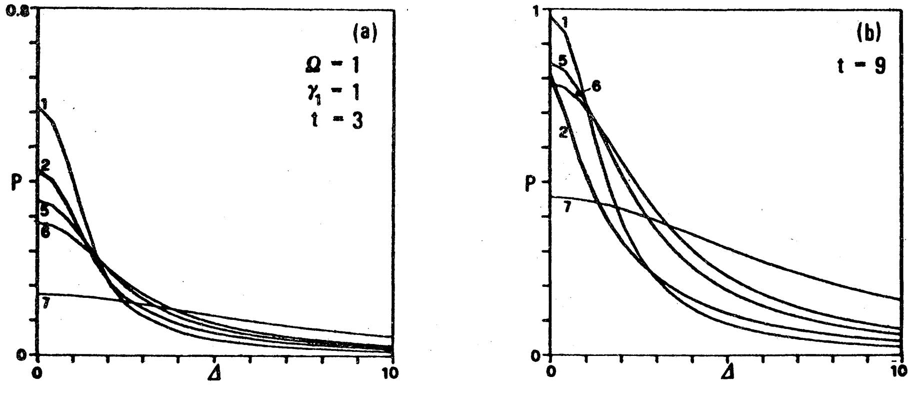

FIG. 6. Dispersion curve of $\langle P(t)\rangle$ with $\Omega=1$, $\gamma_1=1$, $t=3$, and $t=9$ in (a) and (b), respectively, employing a five-term expansion. The meaning of the curve index is explained in the caption of Fig. 1. Curves 2 and 3 are indistinguishable. The dispersion curve is symmetric about $\Delta=0$.

most efficient for $\gamma_1 \approx 1.5\Omega$ for single-mode laser fields. For the CF, the maximum of the curve is shifted to slightly larger values of γ_1 [Fig. 5(a)]. The curve becomes rather flat for $\gamma_1 > \Omega$ and even more so for finite bandwidth fields. Calculations for large interaction times $t = 9$ again indicate the existence on an optimum bandwidth. Furthermore, in this limit the maximum is shifted to $\gamma_1 \gg \Omega$ for $b = 0$ [Fig. 5(b)].

D. Dispersion curves

Figures 6(a) and 6(b) shows the dependence of the IP on the detuning Δ of the laser for $\Omega = 1$, $\gamma_1 = 1$, $t = 3$, and $t = 9$. Here we clearly recover the features already noted in Figs. 1–4. The dispersion curve of the IP in the single-mode laser field is higher on-resonance for $t = 3$ with more strongly dropping wings than for the monochromatic CF. Off-resonance, the zero-bandwidth CF yields twice the IP of the single-mode field as predicted by perturbation theory. A nonzero-bandwidth field tends to blur the resonance [Fig. 6(a)]. For large interaction times, when most of the atoms are ionized, the finite-bandwidth field may ionize more efficiently in the near-resonance region than the monochromatic CF [Fig. 6(b)].

VI. CONCLUSIONS

The present investigation of resonant two-photon ionization has revealed a number of interesting points which can be summarized as follows: On resonance the monochromatic CF and the single-mode laser ionize equally efficiently so long as the number of ionized atoms is small. As this number increases the CF IP may fall behind if the relevant parameters are suitably chosen. The resonant finite-bandwidth field is usually less effective in the ionization process than the limiting case of the monochromatic CF. However, if we consider long interaction times, there may exist a nonzero optimum bandwidth, for which the ionization probability reaches a maximum and then drops off sharply towards the limit of the monochromatic CF. This complex time evolution of the system may thus render the notion of an IP per unit time a physically meaningless quantity.

For the monochromatic on-resonance CF the average bound-state populations are higher than those corresponding to the single mode field. As the field is tuned slightly off resonance, Rabi oscillations appear in the bound-state populations which are absent on-resonance.

The resonant IP as a function of γ_1 and Ω exhibits a maximum for $\gamma_1 \approx 1.5\Omega$ which is rather independent of the bandwidth except for long interaction times where it is shifted to $\gamma_1 \gg \Omega$ for monochromatic CF's.

Clearly, the present work consitutes but a first step towards a more complete investigation of RMPI by finite bandwidth CF's. A variety of new effects is to be expected if the present model is extended to include, e.g., nonresonant contributions to the ionization process.[4,10]

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