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Strong-field autoionization by $2\pi n$ pulses and multipeak photoelectron spectrum

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The first explicit solutions for laser-induced autoionization by a strong, smooth pulse are reported. Under certain "quantization conditions" for the area of the hyperbolic secant pulse, the photoelectron spectrum exhibits a novel coherence property: a multipeak structure.

Autoionization resonances are intensively investigated both experimentally[1] and theoretically.[2-7] Much attention has been devoted recently to a theoretical study of the strong-field ionization to an autoionization resonance. However, attempting to approach physically realizable conditions by inclusion of a finite width of the phase fluctuating laser light,[4] inhomogeneous broadening,[5] spontaneous emission,[6] and the existence of other atomic levels and continua,[2,7] all these papers remain unrealistic in one important respect: They all assume a sudden switching on of the strong laser signal which otherwise remains time independent.

The purpose of this Communication is to present first results for the strong-field autoionization by smooth pulses.

Consider a model atom with only one bound state and a single narrow autoionization resonance of the simplest, symmetric type (Fano asymmetry parameter[8] $q \to \infty$). This atom is subjected to the influence of a strong laser pulse with the envelope $E(t)=\epsilon_0 f(t)$, where ϵ_0 is its typical field strength and $f(t)$ is its dimensionless shape. The Hamiltonian for our system reads

$$H = E_0|0\rangle\langle 0| + \int d\omega\,\hbar\omega|\omega\rangle\langle\omega| + \int d\omega[\hbar\Omega(\omega)f(t)e^{i\omega_L t}|0\rangle\langle\omega| + \text{H.c.}] \ , \tag{1}$$

where E_0 is the bound-state energy, the ω integrals extend from $-\infty$ to $+\infty$ (i.e., we neglect the ionization threshold), and the radiative matrix element $\Omega(\omega)$ is given by

$$\Omega(\omega) = \Omega_0\sqrt{\gamma_0/4\pi}\,(\omega-\omega_e+i\gamma_0)^{-1} \ , \tag{2}$$

where $\hbar\omega_e$ is the energy of the autoionization resonance, γ_0 its width, and Ω_0 the Rabi frequency containing ϵ_0, the typical strength of the laser electric field.

We can substitute the state vector $|\psi(t)\rangle$,

$$|\psi(t)\rangle = \left[\alpha(t)|0\rangle + \int d\omega\,\beta(\omega,t)|\omega\rangle\right]e^{-\omega_L t} \ , \tag{3}$$

into the time-dependent Schrödinger equation (we put $E_0=0$ for convenience) and obtain the following c-number equations:

$$\begin{aligned}\dot\alpha &= -i\int d\omega\,\Omega(\omega)f(t)\beta(\omega,t) \ ,\\ \dot\beta &= -i(\omega-\omega_L)\beta(\omega,t) - i\Omega^*(\omega)f(t)\alpha \ .\end{aligned} \tag{4}$$

We can eliminate the amplitude $\beta(\omega,t)$, assuming that $\beta(\omega,-\infty)=0$ and $\lim_{t\to-\infty} f(t)=0$:

$$\dot\alpha = -\int d\omega|\Omega(\omega)|^2 f(t)\int_{-\infty}^{t} f(\tau)\exp[i(\omega-\omega_L)(\tau-t)_{\alpha(t)}] . \tag{5}$$

The simple Lorentzian form (2) of our matrix element allows for converting the integral equation (5) into the following second-order differential equation[9] for the amplitude α:

$$\ddot\alpha + \left(\xi - \frac{\dot f}{f}\right)\dot\alpha + \frac{\Omega_0^2}{4}f^2\alpha = 0 \ , \tag{6}$$

where

$$\xi = \gamma_0 + i(\omega_e-\omega_L) = \gamma_0 + i\Delta \ .$$

Together with the appropriate boundary condition $\alpha(-\infty)=1$, $\dot\alpha(-\infty)=0$, Eq. (6) is a very convenient starting point for a numerical study.

In this paper, however, we will concentrate our attention on an exactly soluble case of the celebrated hyperbolic secant pulse[10]: $f(t)=1/\cosh\gamma t$. In this case, the solution of Eq. (6) reads

$$\alpha(t) = F\left(\frac{\Omega_0}{2\gamma}, -\frac{\Omega_0}{2\gamma}\left|\frac{\xi+\gamma}{2\gamma}\right|\frac{\tanh\gamma t+1}{2}\right) \ , \tag{7}$$

where $F(\ )$ denotes the hypergeometric function. This function reduces to a polynomial if $\Omega_0/2\gamma=n$ is an integer. This "quantization condition" translates into the $2\pi n$ area of the pulse:

$$A_n = \int_{-\infty}^{+\infty}\Omega_0 f(\tau)\,d\tau = \frac{\Omega_0\pi}{\gamma} = 2\pi n \ . \tag{8}$$

The polynomial in question is a Jacobi polynomial

$$\alpha_n = J_n\left(0, \frac{\xi+\gamma}{2\gamma}\left|\frac{\tanh\gamma t+1}{2}\right.\right) \ . \tag{9}$$

The general formula for the probability $P(\infty) = \lim_{t\to\infty}|\alpha(t)|^2$ of the atom to remain in its ground state after the pulse has passed,

$$P(\infty) = \left|\frac{\{\Gamma[(\xi+\gamma)/(2\gamma)]\}^2}{\Gamma[(\xi+\gamma-\Omega_0)/(2\gamma)]\Gamma[(\xi+\gamma+\Omega_0)/(2\gamma)]}\right|^2 \ , \tag{10}$$

shows that for the detuning $\Delta=0$, this probability equals zero for the $2\pi n$ pulses with sufficiently large n.

For example, for $\Delta=0$, $\gamma=\gamma_0$, the formula (10) simplifies to

$$P(\infty) = \frac{\sin^2\Omega_0\pi/2\gamma}{(\Omega_0\pi/2\gamma)^2} \ , \tag{11}$$

which is zero for *all* the $2\pi n$ pulses.

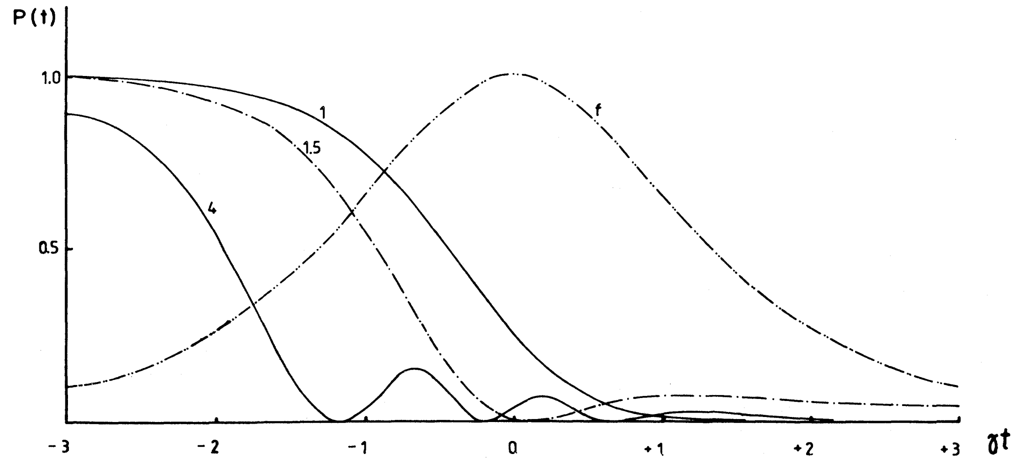

FIG. 1. Probability that the atom remain in its ground state vs dimensionless time plotted for various field strengths. Curves are labeled by the value of $\Omega_0/2\gamma$. The solid line curves correspond to 2π and 8π pulses. The curve labeled f is the hyperbolic secant pulse itself.

In Fig. 1 we have plotted the population of the ground state for the $\gamma=\gamma_0$, $\Delta=0$ case for hyperbolic secant pulses of different areas.

It is clear that our model contains as a limiting case the standard two-level Rosen-Zener[11] model of the kind discussed by Robiscoe.[12] Our Eq. (6), when $\gamma_0 \rightarrow 0$, reduces to the one recently derived by Bambini and Berman.[13] Also, our solution (9) reduces to a very familiar one in this limit. If $\Delta=0$, then we find

$$\alpha_n(t)=J_n\left(0,\frac{1}{2}\left|\frac{\tanh\gamma t+1}{2}\right.\right)=T_n(-\tanh\gamma t) \;, \tag{12}$$

where T_n is the Tschebyshev polynomial:

$$\alpha_n(t)=\cos[n\arccos(-\tanh\gamma t)]=\cos\left(n\gamma\int_{-\infty}^{t}\frac{d\tau}{\cosh\gamma\tau}\right) \;. \tag{13}$$

In other words, in this limit, the amplitude α is simply a cosine of the area of the pulse, which is the well-known result for the two-level atoms. In Fig. 2 we have plotted the probability $P(t)$ for the 8π pulse at resonance. Different curves correspond to the different ratios γ_0/γ. For $\gamma_0/\gamma \ll 1$, the pulse is too short to allow the atom to be ionized effectively, hence we are in the two-level atom regime. If $\gamma_0/\gamma \gg 1$, the pulse is so long that the Rabi oscillations are greatly reduced. When $\gamma_0/\gamma \rightarrow \infty$, even a multi-$\pi$ pulse is locally weak and acts on the atom as a small perturbation.

For the case of the $2\pi n$ pulse it is possible to derive an explicit formula for the photoelectron spectrum, defined as

$$W_n(\omega)=\lim_{t\rightarrow\infty}|\beta(\omega,t)|^2 \;. \tag{14}$$

The main feature of this spectrum is its multipeaked struc-

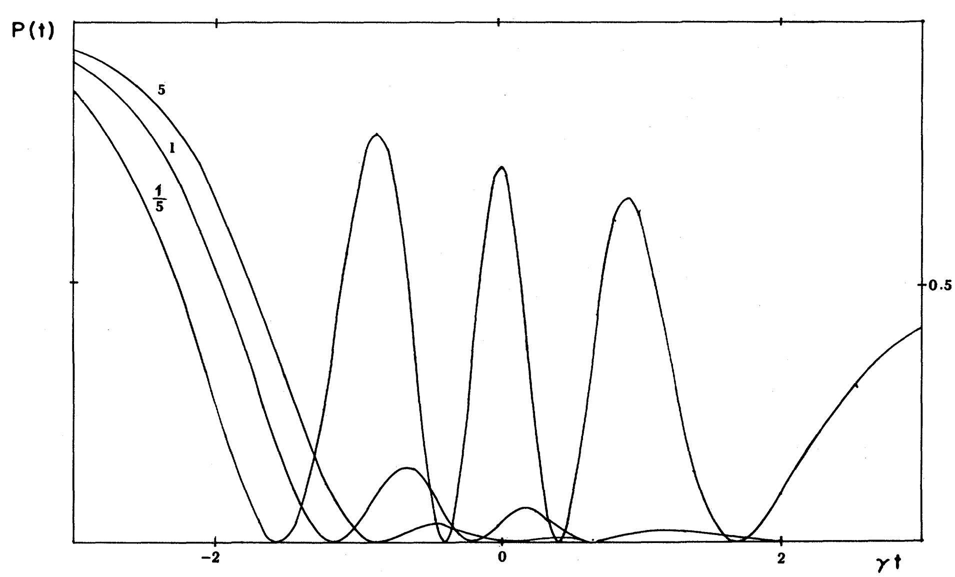

FIG. 2. Probability that the atom remain in its ground state $P(t)$ vs dimensionless time for the 8π pulse, is plotted for various ratios γ_0/γ. They are labeled by the value of γ_0/γ.

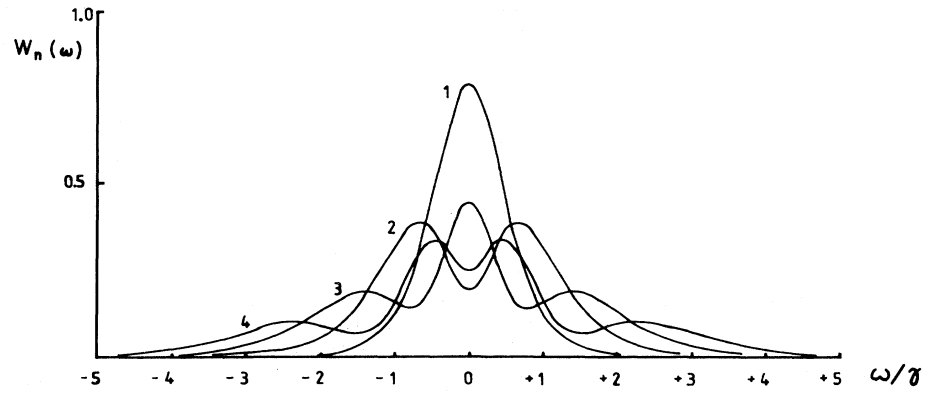

FIG. 3. Spectrum $W_n(\omega)$ of photoelectrons emitted by the atom irradiated by the $2\pi n$ hyperbolic secant pulses. The curves are labeled by the value of n. The number of the maxima is equal to n.

ture. In Fig. 3 this spectrum is plotted for several lowest $2\pi n$ pulses for $\gamma_0=\gamma$, $\Delta=0$.

Next, we comment on the dependence of the spectrum on γ_0/γ. We quote here the formula for the 6π pulse:

$$W_3(\omega)=36\pi\frac{\gamma_0}{\gamma^2}\left\{\left[5\frac{(\omega-\omega_L)^2}{\gamma^2}-\frac{1}{2}\left(\frac{\gamma_0}{\gamma}\right)^2-\frac{5}{2}\right]^2+16\frac{(\omega-\omega_L)^2\gamma_0^2}{\gamma^4}\right\}\Bigg/\left[\left(\frac{\gamma_0}{\gamma}+1\right)\left(\frac{\gamma_0}{\gamma}+3\right)\left(\frac{\gamma_0}{\gamma}+5\right)\right]^2\cosh^2\frac{(\omega-\omega_L)\pi}{2\gamma} \quad . \quad (15)$$

The result is illustrated in Fig. 4. We show here the spectrum W_3 for three different values of γ_0/γ. If the pulse is long enough ($\gamma_0/\gamma >> 1$) it is locally too weak to produce any splitting. Conversely, short pulses allow the multipeaked structure of our spectrum to be more easily resolved.

This fact comes as a surprise. For a step turn-on of the constant laser signal one gets[3] a single-peaked spectrum for a weak field and a two-peaked spectrum above a certain threshold Rabi frequency. The distance between the two peaks increases with the Rabi frequency. In the case of the smooth pulse of the kind discussed here, an instantaneous Rabi frequency varies between zero and its maximal value denoted by Ω. One could therefore expect simply a smearing of the two-peak structure known from the square pulse theory. A dramatically different result derived here is the consequence of the coherent character of the interaction described by our Hamiltonian (1).

The exact solution presented in this Communication, while predicting the existence of new features of the photoelectron spectrum, pertains to a very simplified model. We ignored the existance of the sponteneous radiative decay, fluctuations of the laser, and the coupling to other atomic levels. All these complications, although present in the realistic experiment, are expected not to alter our picture in a significant way. Their influence on the strong-field

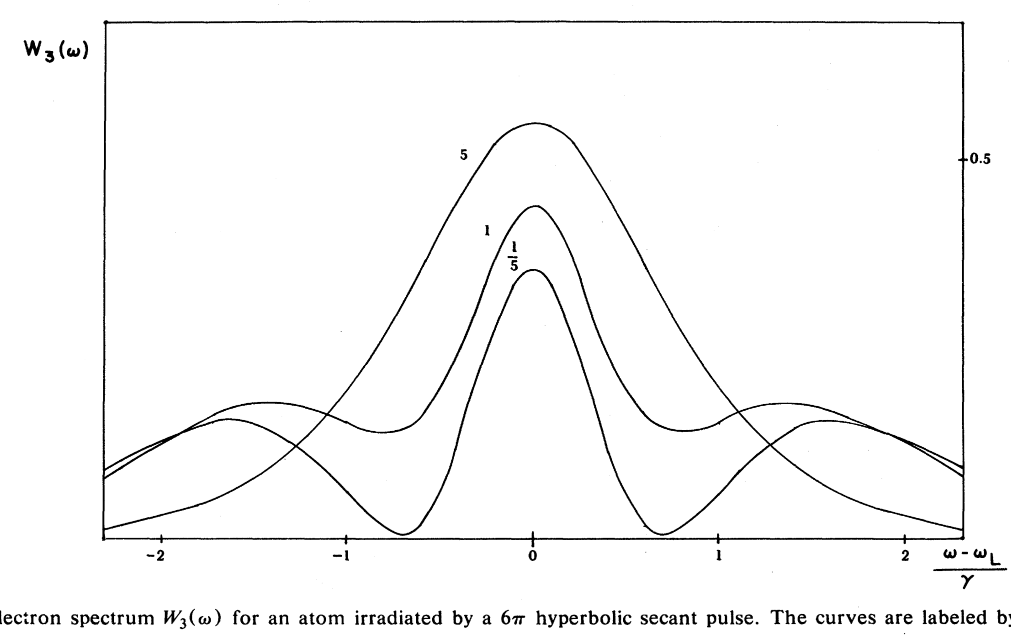

FIG. 4. Photoelectron spectrum $W_3(\omega)$ for an atom irradiated by a 6π hyperbolic secant pulse. The curves are labeled by the value of γ_0/γ.

autoionization by a step turn-on cw signal has been carefully studied[4–7] and proven to yield quantitative corrections only.

We conclude with a remark on possible ways of observing the regularities described above. The most direct observation of the photoelectron spectrum will remain difficult for some time, due to the insufficient spectral resolution of electrons.

The properties of the photoelectron spectrum, however, manifest themselves in a somewhat more complicated configuration of the double-resonance-type experiment[2,7(a)] or, more directly, in the photon spectrum of the spontaneous relaxation accompanying the ionization process.[6]

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