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Pulse Area Effects in Resonant Multiphoton Ionization

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Abstract

A theory of resonant multiphoton ionization by smooth pulses is developed. It is shown that the spectrum of the effect may exhibit structure associated with time domain correlations.

Among the effects of nonlinear optics, multiphoton ionization and detachment were among the earliest to be investigated after the invention of the laser (Geltman 1963, 1965, Hall et al 1965, Hall 1966, Robinson and Geltman 1967, Zernik 1964, Zernik and Klopfenstein 1965, Bebb 1966, 1967, Bebb and Gold 1966, Chin and Isenor 1967, Chin et al 1968, Young et al 1969, Fox et al 1971, Kogan et al 1971, Agostini et al 1971, Luger and Robinson 1970, Luvain and Mainfray 1972). These are bound-free electronic transitions in atomic systems generated by the simultaneous absorption of more than one quantum. The calculation of rates for these processes typically requires the evaluation of summations over intermediate states. In the crudest approximation, the theory assumes that the radiation field is monochromatic and is switched on adiabatically. If the intensity is low and there are no nearly resonant intermediate states, the transition rate is given by the lowest order of time-dependent perturbation theory consistent with the number of photons of energy $h\nu$ needed to compensate for the binding energy. Much work has been done on this problem, and it remains an active area of research (Flank et al 1976, Flank and Rachman 1975, Teague et al 1976.)

In recent times, with the availability of tunable lasers, the study of multiphoton ionization where one or more intermediate states are close to resonance has come to the fore (Beers and Armstrong 1975, Geltman 1979, Dixit and Lambropoulous 1979, Gontier and Trahin 1979, Granneman and Van der Wiel 1975, Eberly and O'Neil

1979). (Negative ions have few, if any, bound excited states, so that these systems are excluded.) The simple perturbation theory, acceptable in the nonresonant case, becomes invalid when the population of the initial level is depleted and/or its phase is significantly affected by coupling to the intermediate states. Theories which take this into account have been developed and predict interesting effects such as structure in the photoionization spectrum due to AC Stark effect, the departure of the transition probability from a simple linear function of time and bandwidth effects. The richness of this field has stimulated great activity.

The purpose of the present paper is to point out that under special, but potentially achievable circumstances, the combined temporal behavior of a pulsed laser and an atomic system undergoing population oscillations can conspire to produce a spectrum (transition probability versus frequency) in multiple photon ionization which has a minimum at exact intermediate state resonance (ISR), exhibits maxima as the laser is detuned above and below, and then falls off rapidly. This superficially resembles the AC Stark effect in experiments where there are three resonantly coupled bound levels, but has a very different origin. It involves the radiation from only one pulsed laser, not two. In addition, the power dependence of the effect differs markedly from that associated with Stark structure, being present only for pulses whose areas are close to $2\pi n$, n integral. (Our "pulse area" is twice the area used in the sense of photon echoes.) In principle, n may be any integer, but it is unlikely that the effect could

readily be seen for any value but n=1. This is discussed in detail below. We designate this splitting as "mock structure" because of its misleading resemblence to Stark structure.

This structure occurs because the probability of inducing the final state is a function of the product of the pulse envelope and the intermediate state population. The latter oscillates in time, and if it should pass through a minimum coincidentally with the maximum in the envelope function of the field strength, the transition probability will be lower than at neighboring frequencies where this anticorrelation does not occur.

We consider an isolated atom interacting with a pulsed laser, undergoing multiple photon ionization from the ground state via a single resonant intermediate state. For simplicity, effects due to the other, nonresonant intermediate states are ignored in this article. The ground and intermediate states are strongly coupled, with the latter connected also to the final states in the continuum. The coupling to the continuum is weak, and is treated perturbatively, with the effect of the continuum on the populations and phases of the intermediate and ground states ignored. Final state amplitudes are to be computed via a quadrature over the intermediate state amplitude.

Let the pulse coupling the two level bound complex have the form $f(t) \cos \Omega t$, where $f(t)$ is a smooth function of time, vanishing at $t = \pm \infty$. No assumption about the "adiabaticity" or "suddenness" is made. The time-dependent Schroedinger equation for the amplitudes of this pair of states becomes, in rotating wave approximation,

$$i\dot{a}_1 = V_{12}(t)\, a_2 \exp(i\Delta t), \tag{1a}$$

$$i\dot{a}_2 = V_{12}(t)\, a_1 \exp(-i\Delta t), \tag{1b}$$

where Δ is the detuning of the central frequency of the pulse from exact ISR. The amplitude of a particular continuum state is given perturbatively by

$$a_k(t) = -i \int_{-\infty}^{t} a_2(t')\, V_{2k}(t')\, \exp(-i\Delta_k t')\, dt', \tag{2a}$$

so that the probability of finding the system is state $|k\rangle$ as $t \to +\infty$ becomes

$$P_k = |a_k(t=+\infty)|^2 =$$

$$\int_{-\infty}^{\infty} a_2(t)\, V_{2k}(t)\, \exp(-i\Delta_k t)\, dt \int_{-\infty}^{\infty} V_{2k}(t') \exp(i\Delta_k t')\, a_2^*(t')\, dt'. \tag{2b}$$

The total probability of ionization is then obtained by integrating over the continuum

$$P = \int_0^\infty P_k\, dE_k = \int_0^\infty dE_k \int_{-\infty}^{\infty} V_{2k}(t) \exp(-i\Delta_k t)\, a_2(t)\, dt \int_{-\infty}^{\infty} V_{2k}(t') \exp(i\Delta_k t')\, a_2^*(t')\, dt', \tag{3}$$

where we have incorporated the density of continuum states into the definition of V_{2k}. If the overall process is a two quantum effect, V_{2k} has the same time dependence as V_{12}. If states $|1\rangle$ and $|2\rangle$

differ by one photon, and states $|2\rangle$ and $|k\rangle$ by N-1 photons (for a total absorption of N photons in ionizing the ground state), V_{2k} is an effective operator whose time dependence is $f^{(N-1)}(t)$. Making the usual mild approximations of neglecting the variation of $V_{2k} = V_o^e f^{N-1}(t)$ with final state energy and extending the lower limit of the energy integration to $-\infty$ (Geltman 1979), the integrated transition probability becomes

$$P = \int_{-\infty}^{\infty} d\Delta_k (V_o^e)^2 \int_{-\infty}^{\infty} f^{N-1}(t) \exp(-i\Delta_k t)\, a_2(t)\, dt \int_{-\infty}^{\infty} f^{N-1}(t') \exp(i\Delta_k t')\, a_2^*(t')\, dt', \tag{4}$$

or, since the detuning integral is proportional to $\delta(t-t')$,

$$P = 2\pi (V_o^e)^2 \int_{-\infty}^{\infty} f^{2N-2}(t)\, |a_2(t)|^2\, dt. \tag{5}$$

We note that while $a_2(t)$ is a function of the detuning between states $|1\rangle$ and $|2\rangle$, the integral in equation (5) is independent of Δ_k, the detuning between the intermediate and final states. The population $|a_2(t)|^2$ undergoes Rabi-type oscillations, whose amplitude in the strong field region is roughly independent of detuning, but where the positions of extrema do depend on Δ, the first detuning. Hence, if the time duration of $f^{(2N-2)}(t)$ is short, one may expect to see an oscillatory variation of P with laser frequency. This is most clearly demonstrated in the limiting case of large N, where f^{2N-2} is approximated by the delta function $C\delta(t)$, C constant, so that

$$P = 2\pi C^2 |a_2(0)|^2 . \tag{6}$$

We see that in this extreme situation, the photoionization spectrum is simply the population variations of level $|2\rangle$ transformed into the frequency domain.

We now examine the question of where and whether one may find a sufficiently narrow f^{2N-2} to be possibly observable in actual systems with realistic pulses. We require the coupling between levels $|1\rangle$ and $|2\rangle$ to be sufficiently strong to render the amplitude of the population oscillations at least roughly independent of detuning, without causing the oscillation frequency to be rapid enough to wash out the effect. The details will, of course, depend upon the pulse shape. To simplify the work, we resort to a model in which f(t) is a hyperbolic secant in time. This provides a smooth envelope which might well resemble true pulses. It also enables one to solve the two level part of the problem analytically (Rosen and Zener 1932, Robiscoe 1978). Equations (1) become

$$i\dot{a}_1 = V_o \operatorname{sech} \frac{\pi t}{T} \exp(i\Delta t)\, a_2, \qquad (7a)$$

$$i\dot{a}_2 = V_o \operatorname{sech} \frac{\pi t}{T} \exp(-i\Delta t)\, a_1 . \qquad (7b)$$

In terms of the parameters a, b, c, Z, given by

$$a = \frac{V_o T}{\pi},\ b = -\frac{V_o T}{\pi},\ c = \frac{1}{2} + \frac{i\Delta T}{2\pi},$$

$$Z = \frac{1}{2}\left\{\tanh \frac{\pi t}{T} + 1\right\},$$

with the initial conditions $a_1 = 1$ and $a_2 = 0$ at $t = -\infty$, equation

(5) becomes

$$P = 4T(V_e^0)^2 \int_0^1 \left\{4Z(1-Z)\right\}^{N-2} |a_2(Z)|^2\, dZ, \qquad (8)$$

with a_2 given in terms of the variable Z by (Robiscoe, Rosen and Zener)

$$a_2 = \frac{V_o T}{\pi(1-c)}\, Z^{1-c}\, {}_2F_1(c-a+1,\ c-b+1,\ 2-c,\ Z), \qquad (9)$$

where ${}_2F_1$ is a hypergeometric function.

The appropriate parameters to characterize the problem in this regime of small V_e^0 are T, N, and V_oT, where the pulse area V_oT determines whether the transition from level $|1\rangle$ to level $|2\rangle$ is saturated, and, if so, the number of oscillations that the population of the intermediate state undergoes for all times.

To gain a quantitative understanding of how the frequency spectrum for multiphoton ionization varies with the parameters of the problem, we integrated equation (8) numerically, varying N between 3 and 13, and let V_oT assume values in the vicinity of $2\pi n$, with $n = 1, 2, 3$. (At exact resonance, this condition assures that the amplitude a_2 passes through zero at $t=0$.) We also explored the regions where the pulse area was very different from an integral multiple of 2π. We found the following results. For $V_oT < 5$, the spectra peaked at $\Delta=0$ and diminished monotonically with increasing detuning for all values of N in the range investigated, i.e., no structure is predicted. For $V_oT = 2\pi$, and $N \leq 4$, no structure was perceived, but for $N>4$, we found that the calculated ionization probability had a local minimum at $\Delta = 0$, rose to a maximum for small but finite detuning, and then decreased monotonically with still greater detuning. As N increased, the value of Δ for which the maximum occurred tended to increase slowly. The ratio, $1+R$, of the maximum ionization probability to the value at $\Delta=0$, increased more rapidly with increasing N. The excess probability R varied from a few percent at $N=5$ to nearly unity at $N=13$, i.e., for high order processes, as one might expect, the effect is more pronounced. It was not necessary that the pulse area be exactly 2π to see the mock structure. We found that the splitting was perceivable in the range from about $V_oT = 5.5$ to $V_oT = 7$, peaking, of course, at 2π. For $V_oT > 7$, the effect washes out. It reappears in the limit of large N for pulse areas near 4π. (See below.) Table I shows, for $V_oT = 2\pi$, the position of the maximum and the ratio $1+R$ of the maximum ionization probability to the resonant probability, for increasing N.

For $N=13$, we found that some small structure (R = a few percent) could be seen in the vicinity of $V_oT = 4\pi$. It appears that this "second harmonic" will be obtainable experimentally only with great difficulty.

While our quantitative predictions are strictly valid only for the hyperbolic secant pulse, we believe that qualitatively similar results should obtain for general pulse shapes and be observable with mode-locked lasers.

The influence of pulse areas on the spectrum of multiphoton ionization has previously been considered by Crance and Feneuille (1977a, 1977b). These authors also include the effect of non-resonant (NR) intermediate states via an effective operator approach. Interference between the NR amplitude and that of the ISR has the effect of shifting the maximum of the spectrum, and removing its symmetry with respect to detuning. It should also distort, but not eliminate, any mock structure effects that might be present. Crance and Feneuille analyzed not only rectangular pulses, for which they solved the two level part of the problem in closed form, and where the doubling predicted in the present article cannot occur, but also several different smooth pulses (not hyperbolic secants), for which they used numerical methods in the two level portion. Their results for smoothly varying pulses do not predict mock structure, apparently because the N values chosen were below the required minimum, according to the present calculation.

That the effect is appreciable only for pulses whose area is near 2π suggests that it may be useful in measuring the integrated intensity of focused short pulses, provided the resonant dipole matrix element is accurately known. The presence of this doublet would then establish the product V_oT to within rather narrow limits. One could determine whether the effect was indeed mock structure by observing if the splitting disappeared with increased and decreased intensity.

To summarize, we have formulated a theory of resonant multiphoton ionization by pulses which predicts a spectrum whose maximum is split into a doublet by purely temporal factors. It appears similar to the AC Stark effect, but occurs under conditions where the latter is absent, and exhibits a totally different intensity dependence.

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TABLE I

Position and amplitude of mock structure maxima

N	$(\Delta T)_{max}$	1+R
5	2.0	1.02
6	3.0	1.10
7	3.5	1.20
8	4.0	1.31
9	4.0	1.43
10	4.3	1.55
11	4.3	1.66
12	4.5	1.79
13	4.5	1.91

N = number of photons needed to ionize the ground state. $(\Delta T)_{max}$ = position of mock structure maximum in dimensionless units. T is the characteristic time that appears in the hyperbolic secant envelope function. 1+R = ratio of transition probability at maximum to probability at exact resonance.

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