Effect of Coherent Continuum-Continuum Relaxation and Saturation in Multiphoton Ionization

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Atomic continuum-continuum transitions appear to be saturable at realistic light intensities. We discuss some consequences in multiphoton ionization.

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The atomic unit of electromagnetic radiation intensity is $I_{\mathrm{at}} = ce^2/2\pi a_0^4 \sim 10^{17}$ W/cm^2. This intensity corresponds to an electric field strength equal to the Coulomb field of a proton at the distance of one Bohr radius. In such radiation fields one expects the bound-state structure of the atom to begin to break down. Continuum-continuum (C-C) transitions can become dominant, and a range of new photon absorption phenomena may be opened.

It turns out that this estimate is surprisingly far off the mark. As we indicate below, C-C transitions have not only been observed in atomic multiphoton absorption experiments[1,2] carried out over the past five years, but most probably have been observed to be *saturated* (i.e., having upper and lower states nearly equally populated), as has been speculated.[2] The theoretical support for this speculation is provided by a reformulation of Fermi's "golden rule" (FGR) for transitions between a discrete state and several continua.

First we introduce (see Fig. 1) the simplest model of C-C photoabsorption.[3] There is a discrete state $|0\rangle$ connected by the matrix element V_{01} to states in a continuum labeled 1. These states, which carry the index p, are coupled by another one-photon matrix element V_{12} to states in continuum 2, which carry the index d (for example, one can think of p and d as angular momentum labels). That is, V_{12} is the C-C coupling strength in this model. Schrödinger's equation gives the following state-amplitude equations:

$$\dot{C}_0 = -iV_{01}\sum C_p, \tag{1a}$$

$$\dot{C}_p = -i\Delta_p C_p - iV_{10}C_0 - iV_{12}\sum C_d, \tag{1b}$$

$$\dot{C}_d = -i\Delta_d C_d - iV_{21}\sum C_p. \tag{1c}$$

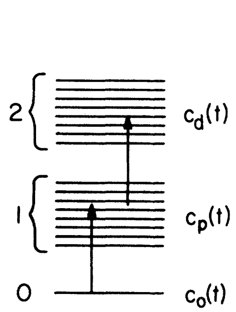

FIG. 1. Model for two-continuum absorption.

The effect of continuum 2 is to act as an absorptive reservoir for continuum 1. What is surprising is that the reverse is also true, and the reverse leads to the possibility of C-C saturation. In our simplified model this can be shown by solving (1c) adiabatically (with use of the Weisskopf-Wigner or pole approximation) and substituting it into (1b), thereby eliminating C_d. Then the last term in (1b) is replaced by the amplitude-coherent sum $-\gamma\sum C_p$, where $\gamma = \pi|V_{12}|^2\rho_2$ is the FGR linewidth for a transition from a typical p state into continuum 2.

The solution of (1a) and (1b) is easily carried out, by use of Laplace transforms for example, and one finds for the amplitude of the discrete state the Laplace domain solution $\tilde{C}_0(z) = 1/(z+\gamma_0)$, where the "coherent" amplitude decay rate is given by

$$\gamma_0 = \frac{|V_{01}|^2 P(z;\Delta)}{1+\gamma P(z;\Delta)}. \tag{2}$$

Here $P(x;y) = \sum_m (x+imy)^{-1} = (\pi/y)\coth(\pi x/y)$, and Δ is the uniform level spacing in (quasi-) continuum 1. We must take the limit $\Delta \to 0$, and we recognize that $1/\Delta = \rho_1$, the (uniform) density of states of continuum 1.

In the numerator of (2) we have the usual FGR result for transitions from discrete state $|0\rangle$. The full result is easily found[3]:

$$\gamma_0 = \frac{\pi|V_{01}|^2\rho_1}{1+\pi^2|V_{12}|^2\rho_1\rho_2}. \tag{3}$$

Equation (3) shows that the C-C matrix elements V_{12} affect the FGR expression for the discrete-continuum V_{10} transition through a dimensionless parameter $Z_{12} = \pi^2|V_{12}|^2\rho_1\rho_2$.

Through Z we can define a new saturation intensity $I_{\rm sat}$ via the relation $Z = I/I_{\rm sat}$. Note that as I *increases* above $I_{\rm sat}$ the decay rate $2\gamma_0$ from the ground level *decreases.* This is because continuum 1 has a coherence time or "linewidth" that can be power broadened as a result of transitions into continuum 2, as we discuss below. Some C-C matrix elements are available in the literature.[4] They show that $I_{\rm sat} \sim 10^{11}$–10^{14} W/cm^2, much lower than 10^{17} W/cm^2, the intensity level at which one might first expect novel C-C effects.

A qualitative interpretation of (3) must be made in order to understand how the saturation described here can be expected to work in situations more complex than our model. In order to do this we identify the role of the transition's "Rabi frequency" $2V_{01}$. From (3) the population transition rate $2\gamma_0$ out of $|0\rangle$ has the value

$$2\gamma_0 = |2V_{01}|^2/[2(\pi\rho_1)^{-1} + 2\pi|V_{12}|^2\rho_2].$$

That is, the transition rate is given by the ratio of the square of the transition's "Rabi frequency" $|2V_{01}|^2$ to the total transition "linewidth."

Such an expression for a transition rate is universal in resonance physics.[5] It shows that in this model discrete *and continuum* transitions are similarly "resonant." As in the discrete case, part of the total linewidth is the inverse of the coherence time of the level and the other ("dynamic") part contributes power broadening and is exactly equal to the stimulated rate for population to leave the level, in this case to go from continuum 1 to continuum 2. Note carefully that the static "linewidth" $2/\pi\rho_1$ is *completely different* from the total width of the continuum (which is infinite in this model anyway).

The traditional role of power broadening has been almost completely unexpected in ionization or in above threshold ionization (ATI), although Kruit *et al.*,[2] mention an apparent similarity with bound-bound saturation effects in discussing their data. This may be the key to interpreting ATI effects reported since 1979,[1,2] as we now show.

One need not restrict one's attention to situations with only two continua.[6] If one adds an infinite ladder of continua beyond continuum 2, the model equations remain tractable as long as the continua are perfectly flat. Actual physical continua studied so far[1,2] appear to have no structural features that are important, and so a flat continuum should not be a bad working hypothesis for a first study of C-C saturation effects. Equations (1a) and (1b) remain valid, and in place of (1c) one has (allowing j to index the second and higher continua):

$$\dot{C}_j = -i\Delta_j C_j - iV_{j,j-1}\sum C_{j-1} - iV_{j,j+1}\sum C_{j+1}. \tag{4}$$

These equations can be solved as before, both for the bound state decay rate and for the individual continuum populations, by introducing the convenient quantity $K_j(t) = \sum C_j(t)$, and the Laplace transforms $\tilde{K}_j$ and $\tilde{C}_j$. We always take the limit $\Delta \to 0$ in interpreting the $P(z;\Delta)$ functions that arise from the detuning sums over the individual continua.

For ease in writing we will assume that the C-C matrix elements between various continua are not only independent of energy, but also independent of the continuum index j. This mean that only one V and one saturation parameter Z arise for the C-C transitions in the theory. One finds, for example,

$$\begin{aligned}\tilde{C}_j(z) &= -iV(z+i\Delta_j)^{-1}(\tilde{K}_{j-1} + \tilde{K}_{j+1})\\ &= -iV(z+i\Delta_j)^{-1}\tilde{C}_0(z)G_j,\end{aligned} \tag{5}$$

where the $\tilde{K}_j$ are easily shown to satisfy $\tilde{K}_j/\tilde{K}_{j-1} = -i\pi V/(1 + i\pi V\tilde{K}_{j+1}/\tilde{K}_j)$, and $\tilde{K}_1 = -i\pi \times V\tilde{C}_0/(1+F)$. Here F is the infinite continued fraction implied by the $\tilde{K}$'s, i.e., $F = Z/(1 + Z/(1 + Z/(1 + \ldots)))$.

Several consequences are immediate. Our model gives γ_0 as an infinite continued fraction:

$$\gamma_0 = \pi|V_{01}|^2\rho_1/(1 + Z_{12}/(1 + Z_{23}/(1 + \ldots))), \tag{6}$$

where we have temporarily resupplied the C-C indices dropped above. We must remember that V_{01} is quite distinct from the C-C V's. We note that Eq. (6) provides a derivation and an all-orders summation of the empirical Gontier-Trahin[7] power series for ATI rates, and if $I >> I_{\rm sat}$ for all transitions we obtain the new result that $\gamma_0 \sim I^{M-1/2}$ if $|V_{01}|^2 \sim I^M$, where M is the number of photons required to exceed the ionization threshold.

Now consider short-time (undepleted, $\gamma_0 t << 1$) ionization, when we can take $\tilde{C}_0 = 1/z$, and invert the transform given in (5) and compute the jth peak population rate: $R_j = d|C_j|^2/dt$. One finds easily $R_j = 2\pi|G_j|^2$, where G_j can be obtained from (5) and the relations among the $\tilde{K}_j$ given below (5). It can be thought of as an effective matrix element from the ground state to the j continuum which contains all orders of C-C transitions. Note that R_j is an important quantity for the model because its multiphoton index $k_j = \partial \ln R_j/\partial \ln I$ can be compared with experimental ATI observations already made.[2] We find $k_j = M-1$ ($M=11$ for xenon and 1.06-μm photons). This result has been obtained in a different way by Edwards, Pan, and Armstrong,[8] and is in remarkably good agreement with one experiment.[2]

In addition, we determine $|C_j|^2$ at long times, after the ground-state population is completely depleted ($\gamma_0 t >> 1$). In this case we must use the solution $\tilde{C}_0 = 1/(z+\gamma_0)$ in (5) when inverting the transforms, and we find a photoelectron peak at every multiple of photon energy above the first ("normal") photoelectron peak. These peaks were first found numerically in a similarly simple model by Białynicka-Birula.[9] Our photoelectron spectra are shown in Fig. 2 for three different values of Z (i.e., of I). We also give here the first compact expression for the photoelectron peak line shape:

$$|C_j|^2 \sim |V_{10}/V|^2 (\Delta_j^2 + \gamma_0^2)^{-1}. \tag{7}$$

From (7) two more distinct predictions are also made. First, the peak's line strength (integral over Δ_j) scales with intensity as $|V_{10}/V|^2 \gamma_0^{-1}$, i.e., independent of V_{10}, and the peak width is proportional to γ_0. According to the continued fraction above, and if Z is roughly independent of j, the width should then scale with intensity as $I^{M-1/2}$, for xenon an extremely sensitive I dependence for peak width. This might be tested experimentally, but it is not clear that experiments have been carried out entirely in the depletion time regime yet. The far wings of (7) obey the short-time predictions given above.

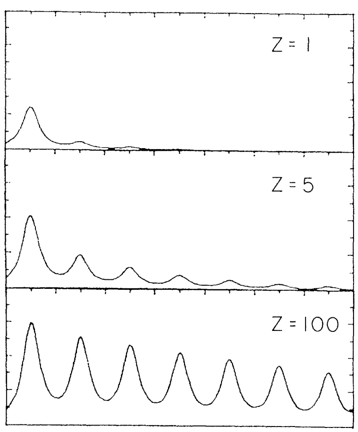

FIG. 2. Photoelectron energy spectra predicted by the theory. The dependence on $Z = I/I_{\text{sat}}$ agrees qualitatively with the data of Kruit *et al.* (Ref. 2), and the form of the top curve is similar to the observation of Agostini *et al.* (Ref. 1). The first peak is highest because V has been chosen independent of continuum energy in these examples. The peak widths are appropriate to the short-time undepleted regime.

The effect on spectra of V's (and therefore Z's) that change from one C-C transition to the next is illustrated in Fig. 3. We show the short-time growth rate of population in the first several continua and also indicate the intercontinuum Z values by vertical bars. The evident prediction is that $Z=1$ acts as an effective cutoff. That is, only continua connected to the lower continua by Z values greater than unity will be appreciably excited. This suggests a "fluid dynamic" interpretation of the excitation process which we discuss elsewhere.[10]

In summary, we have shown that, in multiphoton absorption, the saturation of the usual FGR transition rate formula due to rapid C-C transitions has very broad and largely unexpected consequences. We have shown that this breakdown can be interpreted as a dynamic two-level-type resonance effect, even though the continuum is perfectly flat and featureless. We have identified the saturation intensity I_{sat}, and Eq. (6) indicates the crucial roles played by the C-C dimensionless parameter Z. We have also obtained the first indication of photoelectron behavior in two important time regimes, both before and long after ground-state depletion occurs.

Estimated values of I_{sat} indicate that $Z > 1$ can be expected in a variety of realizable situations. We point out that *all* absorptive transitions in atoms and molecules are connected to C-C transitions via one or a few further photon absorptions, which are often dominant in multiphoton experiments. Thus some effects of Z on bound-bound transitions, for example modifications of k values, might be observable. There are also evident consequences for

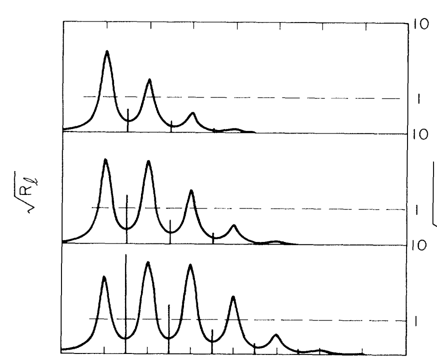

FIG. 3. Photoelectron energy spectra showing the effect of a decreasing sequence of Z values from one continuum to the next. The intercontinuum Z values are given on the right axis. One sees the cutoff character of the value $Z=1$.

laser pumping of regions deep in the continuum, which suggest a new inversion mechanism for short-wavelength laser action. The wavelength of the emission would be tunable with the pump wavelength. Investigations of these effects, as well as of C-C time dependences and other behavior under more realistic assumptions about the energy dependence of C-C matrix elements, will have to be presented elsewhere.[10]

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Note added.—Since submission of this note we have learned of a recent nondynamic, purely statistical theory of similar phenomena which is also of interest [M. Crance, J. Phys. B **17**, L355 (1984)].

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[4]Y. Gontier and M. Trahin, J. Phys. B **13**, 4383 (1980); M. Aymar and M. Crance, J. Phys. B **14**, 3585 (1981).

[5]J. R. Ackerhalt and J. H. Eberly, Phys. Rev. A **14**, 1705 (1976). In the C-C context one should say "linewidth per unit volume," etc., of course.

[6]The dynamical behavior of a multiple-continuum model of atomic ionization was studied by M. Crance and M. Aymar, J. Phys. B **13**, L421 (1980). Their model was more complex than the one described here, and their conclusions were quite different.

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[8]M. Edwards, L. Pan, and L. Armstrong, Jr., J. Phys. B **17**, L515 (1984).

[9]Z. Białynicka-Birula, J. Phys. B **17**, 3091 (1984).

[10]Z. Deng and J. H. Eberly, to be published.