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LETTER TO THE EDITOR

Spectral manifestation of core coherence in photoionization

R Grobe† and S L Haan‡

† Department of Physics and Astronomy, University of Rochester, Rochester, NY 14627, USA
‡ Department of Physics and Astronomy, Calvin College, Grand Rapids, MI 49546, USA

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Abstract. Photoionization processes are considered for situations in which the laser frequency is close to a resonant frequency of the final positive ion core. It is predicted that resonant coupling within the ionic core can split photoelectron spectral peaks into doublets, with components separated by the effective Rabi frequency of the core. The possibility of superposing this final-state effect with previously studied effects involving laser-induced autoionizing states is considered. Specific predictions are made for the splitting of photoelectron peaks in photoionization of strontium.

There are numerous processes which can influence the energy spectrum of outcoming electrons in photoionization and photodetachment. Even if there is only one optically active electron, there can be a series of above-threshold ionization (ATI) peaks (see review articles by Agostini and Petite 1988, Eberly *et al* 1991) separated by the photon energy. In addition, each ATI peak can exhibit substructure such as the doublet predicted by Knight (1977, 1978; see also Autler and Townes 1955, Knight and Milonni 1980, Knight *et al* 1991, Nicklich *et al* 1992). This doublet arises if two of the bound states of the ionizing atom are coupled by a strong, resonant laser field, so that the electron undergoes Rabi oscillations between the states prior to ionization. The components of the doublet are separated by the effective Rabi frequency of the oscillation, and the doublet can be explained as arising from the AC Stark shift or a 'dressing' of decaying discrete states by the external field.

In multielectron atoms, the interaction between electrons can also be important in determining the photoelectron spectrum. For example, in weak-field photoionization from a singly excited state to a continuum which contains an autoionizing resonance, electron–electron interaction is important in understanding the familiar Fano profile (Fano 1961). In strong fields (the case of laser-induced ionization or LIA), Rabi oscillations can occur between the bound state and the autoionizing state, giving rise to an asymmetric Knight doublet in the photoelectron spectrum (see for example Lambropoulos 1980, Lambropoulos and Zoller 1981, Rzazewski and Eberly 1981, 1983, Knight *et al* 1991). In the case of photodetachment, one can similarly tune the laser to an autodetaching state, thereby obtaining a doublet which is describable in terms of decaying dressed states of the negative ion (e.g. Haan *et al* 1990).

The existence of a different doublet in the photoelectron spectrum has recently been predicted (Grobe and Eberly 1993a, b) for photodetachment of negative ions. This predicted doublet does not arise from Rabi oscillations between resonant states in the *negative ion*, as would be the case to form the usual Knight doublet, but instead arises from a resonant coupling of states within the *neutral* atomic system. More specifically, Grobe and Eberly predicted that if the photodetaching laser is tuned to the ground–first-excited state transition

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frequency of the neutral atom, then the energy spectrum of photodetached electrons can exhibit a doublet. This doublet can be interpreted as arising from a dressing of the final states, and the splitting is the effective Rabi frequency for oscillations within the neutral atom.

In the present work we suggest that a similar doublet can arise in photoionization of a neutral atom if the laser is tuned to a resonant transition in the positive ion. We also present the possibility that by tuning to a frequency which is simultaneously close to resonance for both a bound state–autoionizing state transition in the non-ionized atom and a bound–bound transition within the final positive ion, one can superpose the two types of doublets.

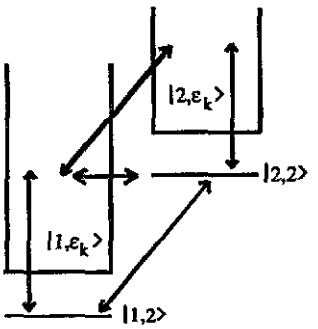

Figure 1. Energy level diagram of the atomic states included in the essential states analysis.

To present the new effect we consider first an essential-states model, shown in figure 1. The initial state of the system, denoted by $|1, 2\rangle$, has one electron in the ground state ('$|1\rangle$') and the other in the first excited state ('$|2\rangle$') of the positive ion. Of course, electron–electron interaction prevents the state from being a simple product state, yet the notation remains conceptually useful. Similarly, the lowest-energy doubly excited state of the system can be written as $|2, 2\rangle$, and is autoionizing. The continua are denoted by $\{|1, \varepsilon_k\rangle\}$ and $\{|2, \varepsilon_k\}$, where ε_k denotes the asymptotic kinetic energy of the outgoing electron. The $|1, 2\rangle$, $|2, 2\rangle$, $\{|1, \varepsilon_k\rangle\}$ subsystem corresponds to the simplest LIA system (e.g., Rzazewski and Eberly 1981). The frequency of light needed to examine the $|1, 2\rangle$–$|2, 2\rangle$ resonance can be comparable to the $|1\rangle$–$|2\rangle$ transition frequency of the ionic core system. How much the frequencies differ will depend upon the relative strengths of the electron–electron interaction. We include $|1\rangle$–$|2\rangle$ transitions in our model by introducing a continuum–continuum coupling

$$\langle 1, \varepsilon_k | V | 2, \varepsilon_k' \rangle = V_{12} e^{i\omega t} \delta(\varepsilon_k - \varepsilon_k') \tag{1}$$

where $V_{12} = \frac{1}{2} F \langle 1|x|2\rangle$, with F the electric field strength. This coupling is analogous to that used by Grobe and Eberly in their study of photodetachment. We also include in our analysis the dipole coupling between $|2, 2\rangle$ and $\{|2, \varepsilon_k\rangle\}$ which allows direct photoionization from the autoionizing state.

In our essential-states analysis, we assume instantaneous turn on of a monochromatic laser of frequency ω and we make the rotating-wave approximation. The solution is straightforward using Laplace transforms and neglecting all principal value integrals over the $\{|1, \varepsilon_k\rangle\}$ and $\{|2, \varepsilon_k\rangle\}$ continua. Rather than presenting lengthy algebraic expressions in the present work, we shall simply summarize our primary results concerning effects of non-zero V_{12} and present graphical results. Our first major conclusion is that the V_{12} (core) coupling has no effect on the time development of the populations of the discrete states of the system. This result is what one would expect for a delta function coupling between final-state continua (e.g. Fontana 1982), and contrasts with the results for a separable coupling

between final-state continua, since the latter coupling can influence the decay rate of the discrete states (Lefebvre and Beswick 1972, Armstrong *et al* 1978, Haan and Cooper 1983, Deng and Eberly 1984).

Our second major conclusion is that V_{12} can indeed have a dramatic effect on the photoelectron spectrum, splitting each peak of the long-time photoelectron spectra in each of the $\{|1,\varepsilon_k\rangle\}$ and $\{|2,\varepsilon_k\rangle\}$ continua into a doublet. For example, a peak in the $\{|1,\varepsilon_k\rangle\}$ continuum which would lie at ε_j in the absence of the core coupling is shifted and split to $\varepsilon_j - \frac{1}{2}\Delta \pm \frac{1}{2}\Omega$, where $\Delta = E_1 + \omega - E_2$ is the detuning from the core resonance frequency and $\Omega = (\Delta^2 + 4V_{12}^2)^{1/2}$ is the effective Rabi frequency.

In order to describe the effect on the photoelectron spectrum more completely, we begin by noting that in the limit $V_{12} = 0$ and $\langle 2,2|V|2,\varepsilon_k\rangle = 0$, the only continuum which is accessible is $\{|1,\varepsilon_k\rangle\}$. For this situation, our essential-states reduces to the standard LIA model and the kinetic energy spectrum at long times can be written

$$S(\varepsilon_k) = \frac{|A(\varepsilon_k)|^2}{|P(\varepsilon_k)|^2}. \tag{2}$$

Here $A(\varepsilon_k)$ is linear in ε_k and $P(\varepsilon_k) = (\varepsilon_k - z_1)(\varepsilon_k - z_2)$ is quadratic because our system contains two discrete states. The precise forms of A and P need not concern us here. The roots z_n can be interpreted as corresponding to 'decaying dressed states', with $\mathrm{Re}(z_n)$ giving the energy of the nth dressed state (relative to the continuum threshold) and $|\mathrm{Im}(z_n)|$ being one half its decay rate. The area under each peak in $S(\varepsilon_k)$ corresponds to the initial population of the corresponding dressed state.

If V_{12} is allowed to be non-zero the long-time kinetic energy spectrum within the $\{|1,\varepsilon_k\rangle\}$ continuum is

$$S_1(\varepsilon_k) = \left| \frac{\Omega+\Delta}{2\Omega} \frac{A(\varepsilon_k - \frac{1}{2}\Delta + \frac{1}{2}\Omega)}{P(\varepsilon_k - \frac{1}{2}\Delta + \frac{1}{2}\Omega)} + \frac{\Omega-\Delta}{2\Omega} \frac{A(\varepsilon_k - \frac{1}{2}\Delta - \frac{1}{2}\Omega)}{P(\varepsilon_k - \frac{1}{2}\Delta - \frac{1}{2}\Omega)} e^{i\Omega t} \right|^2. \tag{3}$$

The first term indicates that for $\Delta > 0$ a peak which was at $\varepsilon_k = \mathrm{Re}(z_n)$ for $V_{12} = 0$ shifts to $\varepsilon_k = \mathrm{Re}(z_n) + \frac{1}{2}\Delta - \frac{1}{2}\Omega$. The height of the peak is diminished because of the multiplicative factor $(\Omega + \Delta)/(2\Omega)$, which is nearly 1 for $4V_{12}^2 \ll \Delta$ but approaches $\frac{1}{2}$ for strong couplings $4V_{12}^2 \gg \Delta$. The second term in (3) indicates that a new peak appears at kinetic energy $\varepsilon_k = \mathrm{Re}(z_n) + \frac{1}{2}\Delta + \frac{1}{2}\Omega$ (an energy Ω greater than the other peak), with multiplicative factor $(\Omega - \Delta)/(2\Omega)$. For $\Delta = 0$, both peaks are shifted equally from $\mathrm{Re}(z_n)$, and the multiplicative factors $(\Omega \pm \Delta)/(2\Omega)$ both equal $\frac{1}{2}$. Regardless of Δ or V_{12}, the widths of the two peaks are equal and determined by $\mathrm{Im}(z_n)$.

Physically the V_{12} core coupling allows population to reach the $\{|2,\varepsilon_k\rangle\}$ continuum. Thus equation (3) gives only part of the full kinetic energy spectrum. We find that the kinetic energy spectrum in the second continuum has (in the time average) two equal-height peaks centred about the same kinetic energies as in the first continuum.

Our results are easily explained by dressing the final states of the system: Positive-ion states $|1\rangle$ and $|2\rangle$ are coupled by a laser of detuning Δ, and together form dressed states $|+\rangle$ and $|-\rangle$ which are separated in energy by Ω. The ionized system lies in either of the two dressed continua $\{|+,\varepsilon_k\rangle\}$ or $\{|-,\varepsilon_k\rangle\}$. The resonant energies $\varepsilon_k = \mathrm{Re}(z_n) + \frac{1}{2}\Delta \mp \frac{1}{2}\Omega$ are simply the energies in the two dressed continua which conserve energy.

If one allows for photoionization from state $|2,2\rangle$ into the $\{|2,\varepsilon_k\rangle\}$ continuum, then the analysis becomes slightly more complicated because of the existence of an additional pathway into the $\{|2,\varepsilon_k\rangle\}$ continuum, which affects the couplings between the 'decaying

dressed states' and the 'dressed continua' $\{|+,\varepsilon_k\rangle\}$ and $\{|-,\varepsilon_k\rangle\}$. Changes in the coupling can affect the branching ratios and peak heights, but the basic peak-splitting effect remains.

We shall defer a full discussion of the algebraic form of our essential states studies to a subsequent communication. We turn our attention here to numerical results. We first choose matrix elements and energies which are appropriate for 1D helium, a model system in which the particles interact through the soft-core Coulomb interaction (e.g., Su and Eberly 1991, Pindzola *et al* 1991, Grobe and Eberly 1992). Using this model system allows us to compare predictions of our essential states model with results of an exact numerical solution of the time-dependent Schrödinger equation. The structure of this atom and the split-operator methods employed to obtain a full numerical solution on a spatial grid have been described elsewhere (Grobe and Eberly 1993, Haan *et al* 1994, Feit *et al* 1982). Figure 2(a) shows the predictions of our essential states model both with and without the V_{12} core coupling for a laser field of amplitude $F = 0.075$ au, frequency $\omega = 0.711$ au (the resonant frequency for $|1\rangle$–$|2\rangle$ core transitions), and duration 40 cycles. The broken curve shows the kinetic-energy spectrum for $V_{12} = 0$, and exhibits an asymmetric doublet. The solid curve shows the spectrum when $V_{12} \neq 0$. Each peak is split into a doublet, as described above. The asymmetry of the larger frequency doublet arises primarily because of the inclusion of photoionization from $|2, 2\rangle$. Figure 2(b) shows the results of a kinetic-energy analysis of the exact numerical solution for the same laser parameters, but with a two-cycle linearly ramped turn-on and turn-on of the laser field. The similarity of the spectrum to the full curve of figure 2(a) gives credence to our essential states results. The difference in scale between figures 2(a) and (b) is easily explained by noting that figure 2(b) shows only one component of a sequence of ATI peaks, while our essential states analysis did not include continuum–continuum couplings needed to obtain ATI peaks.

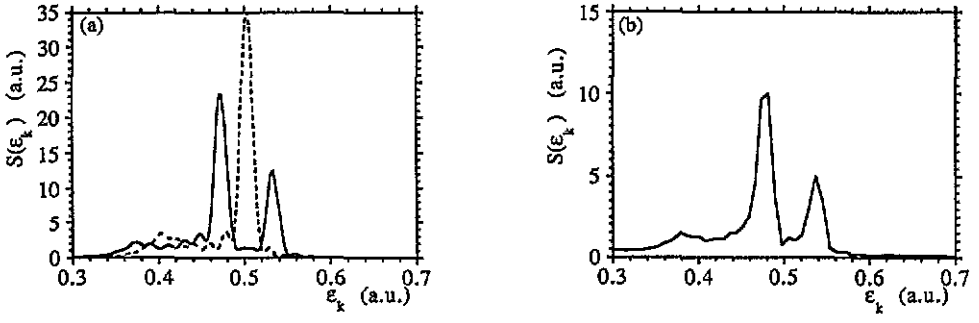

Figure 2. (a) Essential-states kinetic energy spectrum for 1D helium for laser field strength $F = 0.075$ au, $\omega = 0.711$ au. Broken curve: $\langle 1|V|2\rangle = 0$. Full curve: $\langle 1|V|2\rangle = 0.404$ F. Laser pulse duration is 40 cycles. (b) Kinetic energy analysis of exact numerical solution for same laser parameters, except for ramped turn-on and turn-off of the laser. The three-peaked structure is repeated at higher energies as a series of ATI peaks.

Strontium is a very good laboratory candidate to study the core coherence as well as the outer-electron coherence. We choose the initial state to be the 5s5p state, which is coupled to the 4d5d autoionizing state and the Sr^+5s continuum (Boller *et al* 1991, Newsom *et al* 1973). The resonance frequency for the 5s–6p core transition in Sr^+ is 0.1117 au (24 520 cm^{-1}) and for the initial state—autoionizing state resonance is 0.1352 au (29 660 cm^{-1}). Our essential states model predicts that tuning to the core 5s–6p Sr^+ resonance gives rise to a doublet in the kinetic energy spectrum shown in figure 3(a). (Photoionization from the

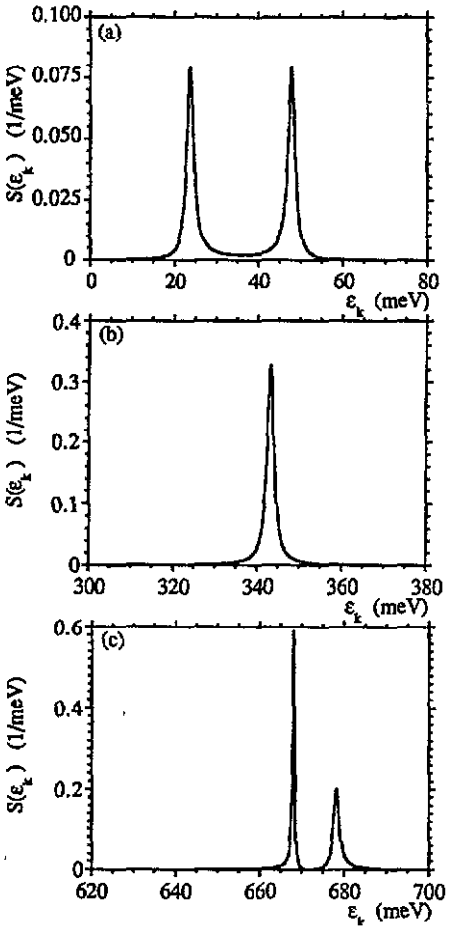

Figure 3. Kinetic energy spectrum from essential states analysis applied to strontium atom for $F = 2.5 \times 10^{-4}$ au ($I = 2.2 \times 10^9$ W cm^{-2}) and a laser pulse duration of 10 ps for laser frequencies (a) 24 520 cm^{-1}, (b) 27 000 cm^{-1}, and (c) 29 660 cm^{-1}.

autoionizing state has not been included. It can be expected to alter the relative heights of the two peaks.) A Knight doublet arises for tuning to the autoionizing resonance frequency and is shown in figure 3(c). For laser frequencies intermediate between these two resonant frequencies, the photoelectron spectrum (figure 3(b)) is a singlet. For strontium the relevant tuning for exciting the core transitions is only slightly larger than the threshold frequency for photoionization, and thus in stronger fields one component of the doublet would be shifted below threshold and interesting new effects could occur. We do not consider here field strengths sufficient for such shifts or for superposing the two resonance effects.

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