Observation of Continuum-Continuum Autler-Townes Splitting

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Photoelectron energy spectra resulting from two-photon ionization of calcium atoms by 180 fs pulses have been studied as a function of wavelength (380–405 nm) and intensity (9–900 GW/cm^2). When the wavelength is tuned near the $4s \rightarrow 4p$ or $4s \rightarrow 5s$ *ionic* core transition, the photoelectron peaks display a characteristic splitting proportional to the field strength and assignable to a two-electron continuum-continuum Autler-Townes effect. Spectra obtained by an essential-state model involving three coupled continua are compared to the experiment.

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Two bound states strongly coupled by an ac-field manifest an energy splitting which is due to the oscillation of population between the states in the presence of the driving field. This phenomenon, well known as the Autler-Townes doublet [1] when probed by a transition to a third level, or the Mollow triplet [2] when probed by resonance fluorescence, is usually *not* observed between a single bound state coupled to the continuum or between two coupled continua. In general, there is then no population oscillation, as the breadth of the accessible phase space over which the coupling strength is distributed in the continuum makes excitation out of the initial state essentially irreversible. The initial state decays exponentially; saturation is reached without any splitting. For example, in multiphoton ionization, as the coupling between ionization continua is increased, only a broader distribution of the photoelectrons among those continua, separated by the photon energy, is achieved [above-threshold ionization (ATI)], with no splitting.

In two-electron atoms, coupling between continua can produce a final state splitting if the driving laser field is resonant with some ionic core transition [3–5]. During photoionization, the strong core coupling is reflected in the energy spectrum of the departing photoelectron as an intensity-dependent splitting *via* the electron-electron interaction and has been dubbed "coherence transfer" by Ref. [3].

Grobe and Eberly [3] determined that the minimum ingredients necessary to generate this effect are a two-electron bound state $|1, 1\rangle$, two continua ($|1, \epsilon\rangle$ and $|2, \epsilon\rangle$), and two couplings ($|1, 1\rangle \Leftrightarrow |1, \epsilon\rangle$ and $|1, \epsilon\rangle \Leftrightarrow |2, \epsilon\rangle$). This minimal model predicts an intensity-dependent, symmetric doublet in the outgoing electron's energy spectrum when the optical field is tuned "*on*" resonance with a core transition. The splitting is due to the $|1, \epsilon\rangle \Leftrightarrow |2, \epsilon\rangle$ continuum coupling, while the $|1, 1\rangle \Leftrightarrow |1, \epsilon\rangle$ coupling only transfers population (exponentially) to the continuum. Thus, unlike the case for a *single-electron* initial state splitting (Knight doublet) [6], the continuum-continuum Rabi oscillations are not reflected in the dynamics of the initial state probability.

In this Letter, we report on the first experimental realization of the coupled continuum model, and the observation of the predicted doublet in the photoelectron energy spectrum, using the two-photon ionization of calcium. The initial bound state is the calcium ground state $4s^2\,{}^1S_0$, and the first and second continua are the $|4s, \epsilon\rangle$ and $|4p_J, \epsilon\rangle$, respectively. As a first approximation, the continuum-continuum coupling can be estimated by using the known bound $4s \rightarrow 4p$ transition strength [7] in Ca^+. The dipole strength is approximately 1.5 atomic units (a.u.), which for a moderate field strength of 3×10^{-3} a.u. (intensity equal to 300 GW/cm^2) yields an easily observable Rabi splitting (Ω) of about 120 meV. The $4s \rightarrow 4p$ ionic transition frequency is approximately 25 300 cm^{-1} ($\sim$395 nm), and neutral calcium is ionized by absorbing two of the corresponding blue photons. Additional levels present in the calcium atom (Fig. 1) which are also coupled by the laser field complicate the minimal model described above. For instance, the $|4p, \epsilon\rangle \Leftrightarrow |5s, \epsilon\rangle$ and the two-photon $|4s, \epsilon\rangle \Leftrightarrow |5s, \epsilon\rangle$ couplings can be of the same order of magnitude as the $|4s, \epsilon\rangle \Leftrightarrow |4p, \epsilon\rangle$ coupling. Likewise, Hanson *et al.* [5] have shown that neutral resonance $4s^2 \rightarrow 4s4p$ transition will strongly modify the shifts and symmetry of the amplitudes of the doublet components. In addition, the role of the $4p$ fine structure can result in more intricate continuum effects.

A frequency-doubled, regeneratively amplified titanium sapphire laser produces tunable (380–405 nm), 180 fs pulses. The pulse bandwidth ($\sim$15 meV) is less than twice the transform limit, and the intensity fluctuations are $\leq$6%. Spectral measurements were made on the fundamental with a monochromator and an optical multichannel analyzer calibrated with a krypton arc lamp. The spectral resolution was 0.5 nm. The calcium was produced in a 775 K atomic beam, and background contamination was less than 0.01%. Various lenses with f numbers ranging from 7 to 25 focused the light into the atomic beam. The

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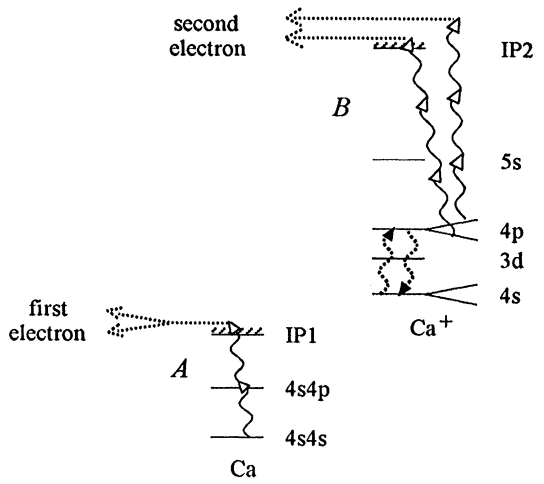

FIG. 1. Simplified level diagram of calcium showing the 2-photon ionization of the neutral (process A) and the $4s \Leftrightarrow 4p_J$ coupled (split) core transition. The splitting of the photoelectron energy due to the continuum-continuum coupling is illustrated by the dotted line. Also shown is the four photon ionization (process B) of Ca^+ and the splitting of the energy spectrum due to the Rabi split ground state.

laser's confocal length exceeded the atomic beam's cross-sectional length, ensuring a flat intensity distribution [8] in the interaction volume. Electron energy analysis was performed with a time-of-flight spectrometer with 2π solid angle collection.

In Fig. 2, the change in the photoelectron energy spectrum (PES) with increasing photon energy (bottom to top) is displayed in two intensity limits. In the low intensity perturbative limit [Fig. 2(a)], the spectra reflect 2-photon excitation to an unperturbed $4s$ ion ground state. The electron emission is confined in a narrow peak (~30 meV width) centered at $2E_\omega$ − IP, where IP is the neutral calcium ionization potential (6.11 eV). The high intensity spectra shown in Fig. 2(b) are taken at the 2-photon saturation intensity (300 GW/cm^2) for neutral ionization [9]. The fraction of Ca^{2+} present at this intensity is negligible ($<10^{-3}$). Several new peaks appear for wavelengths shorter than the $4s \rightarrow 4p_{1/2}$ core resonance (397 nm), whose relative amplitudes evolve with the laser wavelength. The most prominent features appear at wavelengths near the 1-photon $4s \rightarrow 4p_{3/2}$ (393.5 nm) and 2-photon $4s \rightarrow 5s$ (383.4 nm) core transitions. The same features are reproduced in the ATI peaks, with amplitudes which are a few percent of those of the main peak for wavelengths shorter than 400 nm.

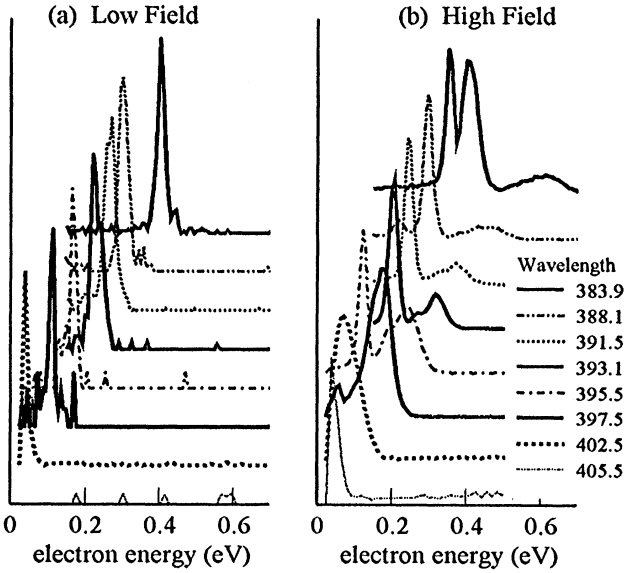

FIG. 2. PES for calcium at several wavelengths at intensities of (a) 10 and (b) 300 GW/cm^2. The solid lines correspond to "on" resonance spectra.

Red detuned from any core resonances, neutral calcium is observed to ionize in a nonresonant perturbative 2-photon process, as verified by a measured I^2 dependence of the photoelectron yield [10]. Figure 3 shows the evolution of the spectrum as a function of intensity for two wavelengths. In Fig. 3(a) the laser is tuned exactly on the ion core resonance ($4s \rightarrow 4p_{3/2}$) and the splitting and amplitude of the peaks show a strong intensity dependence. The splitting scales approximately linearly with the electric field strength ($I^{1/2}$) and has a magnitude consistent with the simple Rabi splitting estimate discussed above. In Fig. 3(b), the laser's detuning [Δ = 40 meV (5 nm)] from resonance is large enough to show the reduced amplitude of the $4p$ component and the decreased intensity dependence of the $4s$ shift expected in the simple Rabi picture, but still below the limit $\Delta/\Omega \gg 1$ where the $4p$ shift would become linear in intensity.

The origin of this structure in specific core resonances is established by varying the frequency, intensity, and polarization. A detailed account of the observations will be given elsewhere. Here we briefly summarize the general behavior. In agreement with the simple Rabi model, (1) there are two wavelength regions where significant field dependent structures are assignable to the 1-photon, $4s \rightarrow 4p_{1/2,3/2}$ and the 2-photon, $4s \rightarrow 5s$ core transitions. (2) Tuning increasingly "*off*" resonance reduces the amplitude (population) of one of the components of the Rabi doublet and eventually [Fig. 3(b)], a single peak with a weakly intensity-dependent energy becomes the dominant feature. (3) *On* resonance the two Rabi components have equal population but, in disagreement with the minimal model, asymmetric amplitudes, shifts, and broadenings. Finally, (4) photoelectron peaks assignable to ion final states which are not resonantly coupled by the laser field, i.e., the $3d$ level, show *no splitting* (unperturbed) at the core resonance frequencies for all intensities examined.

Use of circularly polarized (CP) light further elucidates the nature of some of the couplings. Figure 4 shows the PES resulting from excitation of calcium with 381.3 nm

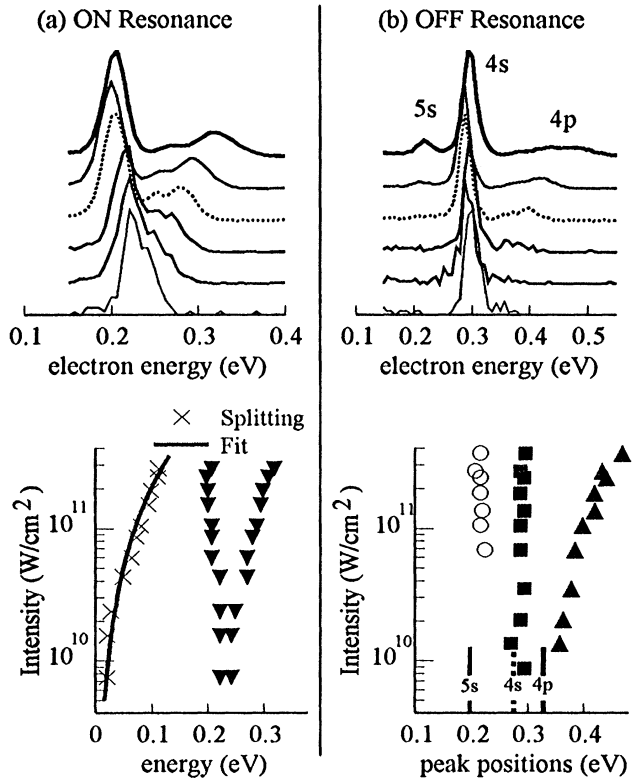

FIG. 3. PES for calcium (a) resonant with the $4s_{1/2} \rightarrow 4p_{3/2}$ transition at 393.5 nm and (b) detuned from all ion core resonances at 388.5 nm. A plot of the peak positions as a function of intensity accompanies the PES, as well as an $I^{1/2}$ fit in (a) and field free state assignments in (b). The intensities are from top to bottom: I_s, $I_s/2$, $I_s/4$, $I_s/8$, $I_s/16$, and $I_s/32$, where I_s is the saturation intensity (300 GW/cm^2).

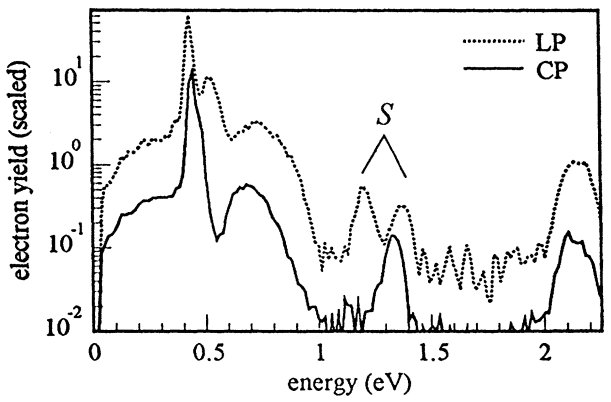

FIG. 4. The polarization dependence of the PES near (381.3 nm) the 2-photon $4s \rightarrow 5s$ core transition. The peaks labeled S are due to ionization of the ion, process B in Fig. 1. The spectra are recorded with an intensity greater than the neutral and ionic saturation intensities. The ionic saturation intensity is 2–3 times greater than the neutral.

photons, above the neutral saturation intensity, for linear and circular polarization. This frequency is blue-detuned off all core resonances but nearest to the 2-photon, $5s$ resonance. The peaks labeled S correspond to the sequential ionization of the Ca^+ ground state; $Ca^+ + Nh\nu \rightarrow Ca^{2+} + e$, all other peaks originate from neutral ionization. The disappearance of specific spectral features caused by the CP light is evident for both the Ca (near 0.5 eV) and Ca^+ (label S) ionization and suggests a 2-photon transition which is forbidden with CP light, i.e., $s \rightarrow s$. Furthermore, the energy dependence of the structures supports the assignment as a result of the 2-photon $4s \Leftrightarrow 5s$ coupling. This coupling influences the ionization of both the neutral and ion, but with distinctly different dynamics. In the neutral, it behaves similar to the Rabi oscillating ion core effect discussed above for the $4s \Leftrightarrow 4p$ coupling, while for the ion it is probably an initial-state bound-bound coupling [6,11] (process B in Fig. 1). In addition, the 2.2 eV electron shown in Fig. 4, which is assignable to neutral ionization leaving the ion in the $3d$ excited state, shows no splitting or polarization dependence, since it is *not coupled* at these wavelengths. This fact further substantiates that the observed doublets are indeed a continuum-continuum final state effect.

The minimal model's failure in reproducing some of the experimental behavior is not surprising considering the extent of additional couplings in calcium. A more exact treatment should incorporate both the bound and core couplings illustrated in Fig. 1. For instance, the neutral resonance ($4s^2 \rightarrow 4s4p$) could induce an Autler-Townes doublet in the initial state [11], which would be observable as a splitting in all photoelectron peaks. This can be eliminated as the source of the observed doublets since no splitting is seen in the $3d$ photoelectron. However, it was shown [5] that the neutral $4s^2 \rightarrow 4s4p$ coupling can significantly influence the shift and amplitudes (asymmetry) of the individual peaks, due to the ac-Stark shift of the neutral's ground state. This is particularly relevant near the saturation intensity in the current experiment. In order to examine how the inclusion of the continuum's fine structure affects the predictions of the minimal model, we have solved the time-dependent Schrödinger equation for an effective two-electron Hamiltonian involving three continua using known atomic parameters [7]. Each of the three continua ($|4s_{1/2}, \epsilon\rangle$, $|4p_{1/2}, \epsilon\rangle$, and $|4p_{3/2}, \epsilon\rangle$) is modeled as an energy band comprised of N_c equidistant states with a spacing of ΔE. A bandwidth of $0.1E_{4s-4p_{3/2}}$ and $N_c = 80$ are sufficient to obtain convergence. The dipole interaction part of the Hamiltonian is modeled in analogy to Ref. [3]. The ground state to $|4s_{1/2}\rangle$ continuum dipole transition matrix element, $\mu_0\sqrt{\Delta E}$ ($\mu_0 = 5.2$ a.u.), was adjusted to provide saturation in the model at the experimentally determined value. The excitation of the ground state by a pulse with

a $\sin^2$ intensity envelope of 180 fs FWHM is followed using a Lanczos time propagator. The model spectrum is produced by summing the probability distributions in the individual continua at the end of the pulse.

Figure 5 shows the result of this calculation for a photon frequency resonant with the ionic $4s \rightarrow 4p_{3/2}$ transition at the saturation intensity. The dotted line is calculated neglecting the $4p_{1/2}$ continuum. The two symmetric peaks are consistent with the minimal two-continuum model [3], with each peak containing equal contributions from the $4s$ and $4p_{3/2}$ continua. However, symmetric peaks are not observed in the experiment. The solid line is obtained by setting the coupling to the third continuum ($4p_{1/2}$) to its bare ion value. The calculated curve clearly shows an asymmetry in the peak amplitudes, a broadening of the higher energy component, and a splitting consistent with the saturated spectrum presented in Fig. 3(a) [12].

The experiment fully confirms that, in a two-electron atom, ionization involving continua strongly coupled via a core resonance results in a split photoelectron energy spectrum [3,5]. The intensity dependence is in good agreement with the Rabi splitting estimated using calcium ion transition strengths. The inclusion of fine structure in the minimal model gives a marked improvement in experimental agreement, but a complete theory will have to include the neutral resonance as well as other coupled ion levels. Most importantly, the influence of the electron-electron correlation on the dynamics of the ionizing electron is clearly demonstrated in this experiment, further illustrating the way correlation strongly modifies the independent electron picture of multiphoton ionization.

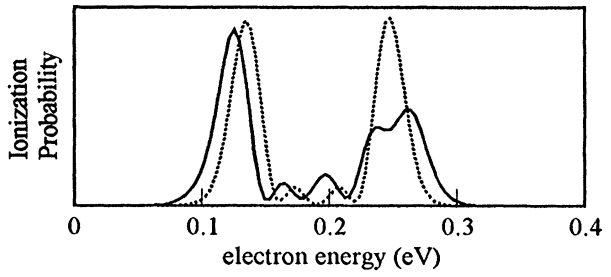

FIG. 5. Theoretical PES corresponding to the excitation with a 283 GW/cm^2 pulse and a frequency resonant with the ionic $4s \rightarrow 4p_{3/2}$ transition. PES in the case of (a) zero and (b) realistic $4s \Leftrightarrow 4p_{1/2}$ coupling.

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[1] S. L. Autler and C. H. Townes, Phys. Rev. **100**, 703 (1955).
[2] B. R. Mollow, Phys. Rev. A **5**, 2217 (1972).
[3] R. Grobe and J. H. Eberly, Phys. Rev. A **48**, 623 (1993); R. Grobe and S. L. Haan, J. Phys. B **27**, L735 (1994).
[4] F. Robicheaux, Phys. Rev. A **47**, 1391 (1993).
[5] L. G. Hanson, J. Zhang, and P. Lambropoulos, Europhysics Lett. **30**, 81 (1995).
[6] P. L. Knight, J. Phys. B **11**, L511 (1978).
[7] C. E. Moore, *Atomic Transition Probabilities,* National Standards Reference Data Series-22 (U.S. GPO, Washington, DC, 1969), Vol. II.
[8] Our intensity is accurate to a factor of 2, as limited by the laser spot size measurement.
[9] The experimentally determined saturation intensity is assigned to be the intensity where the single atom ionization probability is approximately e^{-1}.
[10] In this study, the closest the wavelengths came to a neutral resonance was 1000 cm^{-1}. The quadratic scaling was observed for larger detunings from this point up until ion resonances make the analysis more complex.
[11] W. Nicklich *et al.,* Phys. Rev. Lett. **69**, 3455 (1992); D. G. Papaioannou and T. F. Gallagher, Phys. Rev. Lett. **69**, 3161 (1992).
[12] Further work has shown that the spectral manifestations of the fine structure is diminished at intensities near saturation but it plays a role in the low to intermediate intensity range of our experiment and is responsible for the doublet structure in the high energy $4p$ component of Fig. 3.