

# CONTINUUM-CONTINUUM AUTLER-TOWNES SPLITTING IN CALCIUM

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## 1. Introduction

Strong-field ionization of two-electron atoms can result in scenarios in which the electron-electron correlation plays an important role. It was recently suggested[1] that a splitting similar to the Autler-Townes effect[2] would occur when two ionization continua are resonantly coupled in two-electron atoms. This is obviously at variance with the case of one electron atoms where coupling between continua does not induce oscillations but instead leads to exponential decay of one continuum into another. The special case considered by Grobe and Eberly is that of a strong radiation field resonantly coupling two ionic states (i.e. a core transition). Formally, the states which are coupled are continuum states (two-electron states in which one electron is in a continuum state), but nevertheless the corresponding photoelectron peak is split. Physically, the reason for this is that the electron-electron interaction transfers the energy shift of the core electron to the outgoing electron and has been dubbed "coherence transfer" by Ref. [1].

One simple way to see this effect is to think that the final ionic state is split by the resonant (core)-interaction, thus the outgoing electron sees two asymptotic energy limits separated by the Rabi frequency  $\Omega = \mu_{\pm} \mathcal{E} / \hbar$ , where  $\mu_{\pm}$  is the ionic dipole and  $\mathcal{E}$  the electric field. To emphasize the fact that it is actually two continua that are coupled, one can talk about continuum-continuum Autler-Townes splitting. Dynamically, the outer elec-

tron is being ionized and, at the same time, the core-electron is driven by a Rabi oscillation. Note that the splitting would reflect directly on Rydberg states as well[3]. Actually, the time-evolution of a Rydberg wave-packet under strong coupling of the core-electron gives rise to very interesting effects as discussed by Hanson and Lambropoulos[4]. For the phenomenology of strong-field optical resonance for two-level systems, the reader is referred to the literature[5]. We just summarize the general behavior of the photoelectron energy spectrum "on" resonance. At low intensity, the spectrum would consist of a single energy peak, as the intensity increases the peak will be symmetrically split by  $\Omega$  and proportional to the square root of the intensity. One should also recall that "on" resonance, the states are actually linear superpositions of bare states, thus any labelling of the split components by bare state quantum numbers is arbitrary.

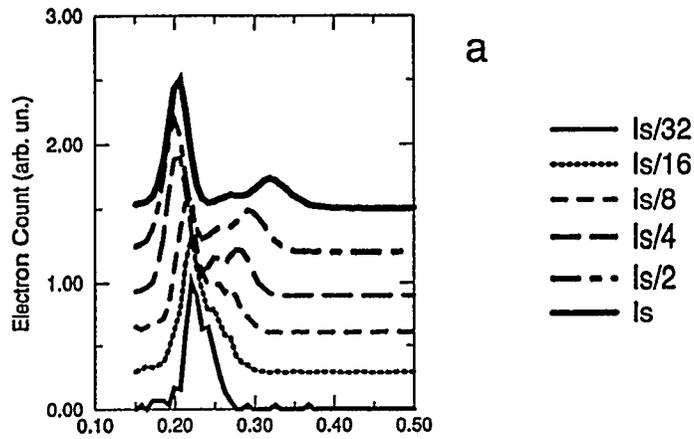
Experimentally, two-photon ionization of calcium around the core resonance  $4s-4p$  (393.5 nm) offers, in principle, an ideal realization of this situation. The strong ionic dipole moment (approximately 1.5 atomic units) yields, for an intensity of  $300 \text{ GW/cm}^2$ , easily observable Rabi splitting of about 120 meV. Furthermore, the wavelength needed conveniently corresponds to the second harmonic of a titanium sapphire laser. The first observation of such a splitting has been reported in a recent Letter[6]. Although a number of the observed features are in agreement with the simple prediction discussed above, even a superficial inspection of the data reveals a number of significant differences. The most conspicuous are an *asymmetric* splitting and the presence of extra peaks in the energy spectra. The former is easily understood if one takes into account more precisely the atomic structure of Ca. For instance, Hanson et al.[7] have shown that the *neutral atomic* resonance  $4s^2 - 4s4p$  modifies significantly the spectrum through the  $4s4p-(4s,\epsilon)$  coupling. A similar effect can be assigned to the interaction between the  $4p$  and  $5s$  continua. The presence of additional peaks may be traced to the influence of the fine structure for the  $4p$  ionic-state. We show that its role is large at low intensity and diminishes around saturation.

## 2. Experiment

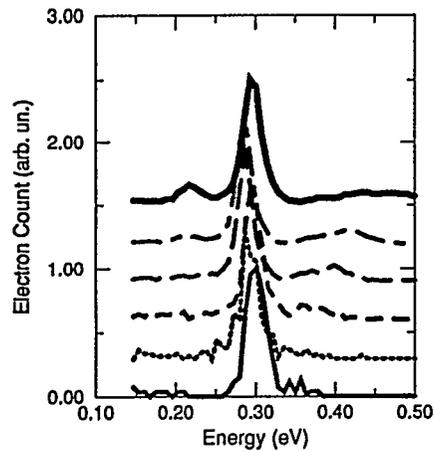
A frequency-doubled, regeneratively amplified titanium sapphire laser produces tunable (380-405 nm), 180 fs pulses. The pulse bandwidth ( $\sim 15 \text{ meV}$ ) is less than twice the transform limit and the intensity fluctuations are  $\leq 6\%$ . Spectral measurements were made on the fundamental light 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 an 775 K atomic beam and background contamination was less than 0.01%. Various lenses with  $f$ -numbers ranging from 7 to 25 focused

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a



b

Figure 1. Experimental spectra (a) “on resonance” ( $\lambda = 393.5\text{nm}$ ) and (b) “off resonance” ( $\lambda = 388.1\text{nm}$ ) for different laser intensities labelled in fraction of the saturation intensity  $I_s = 3 \times 10^{11} \text{ W/cm}^2$ .

the light into the atomic beam. The laser’s confocal length exceeded the atomic beam’s cross-sectional length, ensuring a flat intensity distribution in the interaction volume. Electron energy analysis was performed with a

time-of-flight spectrometer with  $2\pi$  solid angle collection.

Figure 1 shows the intensity dependence of the photoelectron energy spectra at constant wavelength. In Fig. 1a the laser is tuned "on resonance" with the ionic  $4s_{1/2}$ - $4p_{3/2}$  transition (393.5 nm) for intensities ranging from about  $10^{10}$  to  $3 \times 10^{11}$  W/cm<sup>2</sup>. At the lowest intensity only one peak emerges at the expected energy for the two-photon ionization, with a small shoulder evident on the high energy side. As the intensity increases, the main feature is red-shifted while the shoulder develops into new structures on the high energy side becoming progressively blue-shifted. In fact, the blue shifted structure resolves into a clear doublet, whose relative amplitude switches depending upon the intensity.

The laser is tuned "off resonance" in Fig. 1b. Besides the trivial shift due to the change in photon energy, the intensity dependence of the spectra is somewhat different: the main peak is basically unshifted, a weak component is increasingly blue-shifted and at the highest intensity, a new feature appears on the red side of the main peak. The qualitative behavior of the spectra as a function of the wavelength and intensity are certainly reminiscent of the predictions of the Autler-Townes model. However, other couplings could give rise to analogous splittings. For instance, the observed splitting could be related to a Rabi coupling of atomic *bound* states rather than continua[8]. This is ruled out immediately by inspection of the electron peak leaving the ion in the 3d states: this peak is *not* split at any intensity or wavelength. Furthermore, only a quantitative comparison may distinguish between the resonant splitting and "ordinary" Stark shifts. A simple plot of the energy difference between the two main peaks versus intensity shows a square root dependence which signifies a resonant shift. However, a more detailed theory is necessary in order to account for the obvious differences between the observed spectra and the predictions of the simple Rabi model.

### 3. "Essential states" calculation

The theory, beyond the simple model[1], has been worked out by several groups[7, 9, 10]. All calculations rely on the "essential states" approximation. We follow here an equivalent approach: by using projection operators, the eigenstate space is partitioned into the essential states subspace and the complementary space. The choice of the "essential states" is guided by the neutral and ionic calcium energy level scheme and the photon energy. We have retained the following states:  $4s^2$   $^1S_0$ ,  $4s4p$   $^1P_1$  bound states and  $4s^2S_{1/2}$ ,  $4p^2P_{3/2}$ ,  $4p^2P_{1/2}$  and  $5s^2S_{1/2}$  continua in CaI. Projecting the time-dependent Schrödinger equation on the essential states yields a set of coupled integro-differential equations which read:

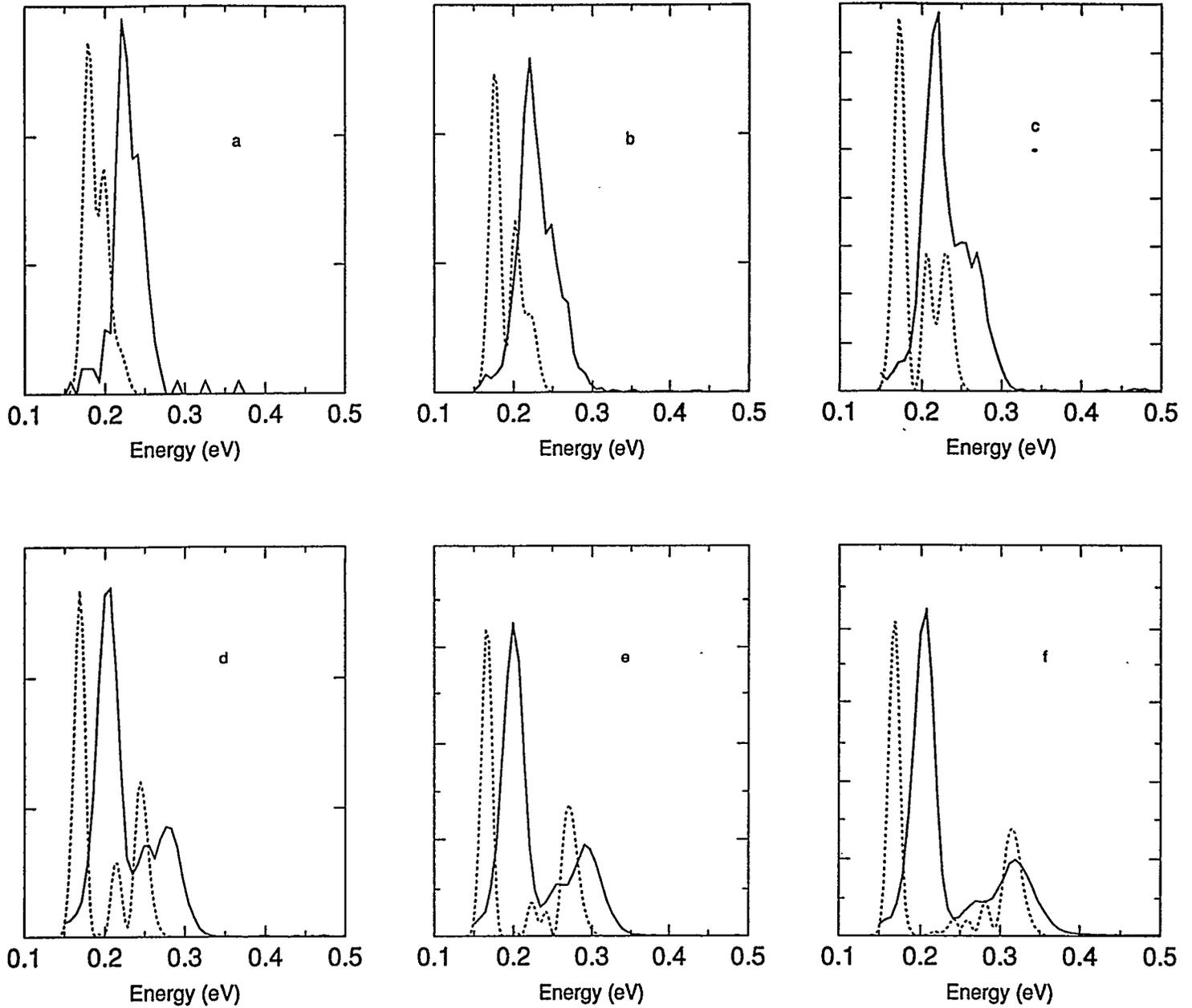


Figure 2. Calculated (dashed) and observed (solid) electron energy spectra at 393.5 nm for different intensities: (a)  $10^{10}$  W/cm<sup>2</sup>, (b)  $2 \times 10^{10}$  W/cm<sup>2</sup>, (c)  $4 \times 10^{10}$  W/cm<sup>2</sup>, (d)  $8 \times 10^{10}$  W/cm<sup>2</sup>, (e)  $1.5 \times 10^{11}$  W/cm<sup>2</sup> and (f)  $3 \times 10^{11}$  W/cm<sup>2</sup>.

$$i\dot{a}_0(t) = E_0 a_0(t) + \Omega_{01}(t)a_1(t) + \int d\epsilon' g_{02}(\epsilon', t) a_2(\epsilon', t) \quad (1)$$

$$i\dot{a}_1(t) = (E_1 - \omega) a_1(t) + \Omega_{01}(t)a_0(t) + \int d\epsilon g_{12}(\epsilon', t) a_2(\epsilon', t) \quad (2)$$

$$i\dot{a}_2(\epsilon, t) = (E_2 + \epsilon - 2\omega) a_2(\epsilon, t) + \Omega_{02}(t) a_0(t) + \Omega_{12}(t) a_1(t) + \Omega_{23}(t) a_3(\epsilon, t) + \Omega_{24}(t) a_4(\epsilon, t) + \Omega_{25}(t) a_5(\epsilon, t) \quad (3)$$

$$i\dot{a}_3(\epsilon, t) = (E_3 + \epsilon - 3\omega) a_3(t) + \Omega_{03}(t) a_0(\epsilon, t) + \Omega_{23}(t) a_2(\epsilon, t) + \Omega_{35}(t) a_5(\epsilon, t) \quad (4)$$

$$i\dot{a}_4(\epsilon, t) = (E_4 + \epsilon - 3\omega) a_4(\epsilon, t) + \Omega_{04}(t) a_0(t) + \Omega_{24}(t) a_2(t) + \Omega_{45}(t) a_5(\epsilon, t) \quad (5)$$

$$i\dot{a}_5(\epsilon, t) = (E_5 + \epsilon - 4\omega) a_5(\epsilon, t) + \Omega_{25}(t) a_2(\epsilon, t) + \Omega_{35}(t) a_3(\epsilon, t) + \Omega_{45}(t) a_4(\epsilon, t) \quad (6)$$

Where the labels "0", "1", ..., "5" refer to the 6 essential states in the above order,  $\epsilon$  is the photoelectron energy, and  $t$  the time.

The continuum-continuum couplings  $\Omega_{ij}$  are approximated by the corresponding single-electron ionic dipoles using the independent-electron coupling ansatz[1]:

$$\langle i, \epsilon | H'(r_1, r_2, t) | j, \epsilon' \rangle = \Omega_{ij}(t) \delta(\epsilon - \epsilon') \quad (7)$$

and the bound-free couplings  $g_{ij}(\epsilon, t)$  are defined as:

$$g_{i,j}(\epsilon, t) = \langle a, a | H(r_1, r_2, t) | a, \epsilon \rangle. \quad (8)$$

The dependence of the  $\Omega$ 's on  $\epsilon$  has been dropped by making the usual assumption of a "flat" continua. Eqns. (1) and (2) are reduced to differential equations by standard methods. The CaI bound-bound dipoles are taken from the tables[11] and the bound-free matrix elements are those used by Hanson et al.[7, 12]. The CaII dipoles were re-calculated using a non-relativistic approximation of the core polarization[13]. The resulting differential equations are numerically integrated using a fourth-order Runge-Kutta method for a gaussian pulse with FWHM of 180 fs.

A comparison between the calculated and experimental spectra is shown in Fig. 2 and Fig. 3 for the "on" and "off" resonant cases, respectively. The

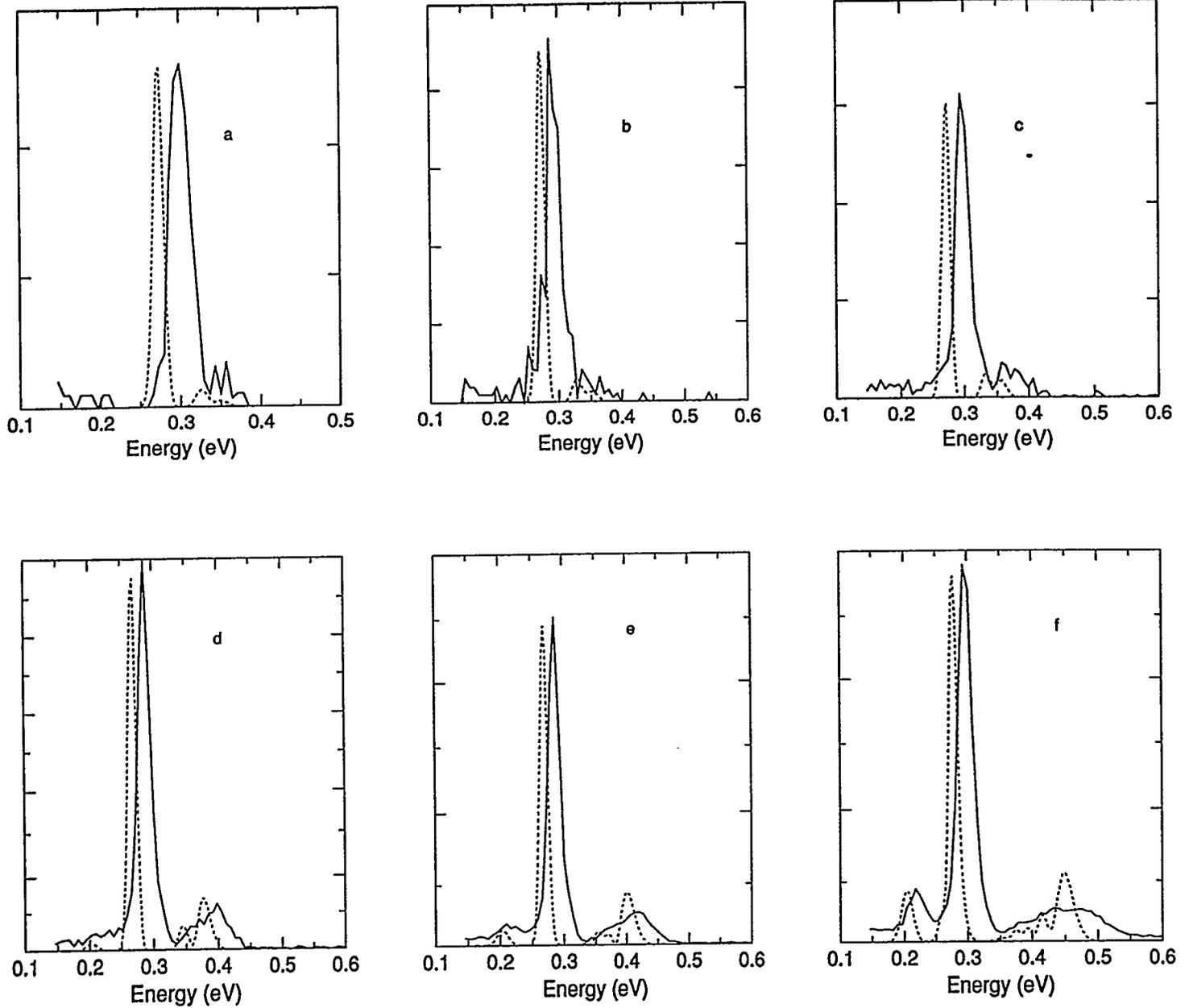


Figure 3. Same as Fig. 2 except at 388.3 nm.

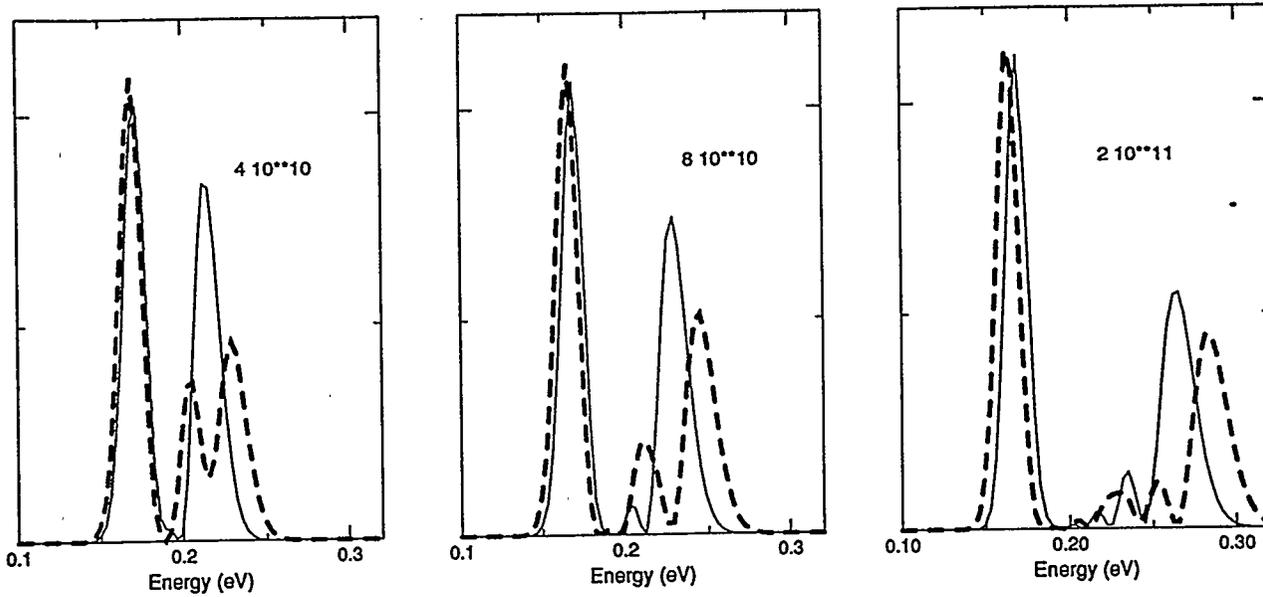


Figure 4. Calculated electron energy spectra for three peak intensities, with (dashed lines) and without (solid lines) the  $p_{1/2}$  continuum (fine structure). The intensity is indicated in each plot in  $\text{W}/\text{cm}^2$ .

overall agreement is excellent (the slight systematic shift of the experimental spectra with respect to the calculated ones is probably due to a contact potential inside the spectrometer). In order to check in more details the role of the fine structure, the  $(4p_{1/2}, \epsilon)$  continuum has been removed from the above equations. The result is shown in Fig. 4 for three intensities. For intensities below  $10^{11} \text{ W}/\text{cm}^2$ , the fine structure is responsible for the splitting of the high energy peak into two components. Note also the different shifts. Above  $10^{11} \text{ W}/\text{cm}^2$ , the fine structure manifests itself only in the splitting. This confirms our previous calculation[6] and is further illustrated in Fig. 5 which shows the peak energies versus intensity for the "on resonance" case.

The  $(5s_{1/2}, \epsilon)$  influences the system through the generalized Rabi frequencies  $\Omega_{i5}$ , ( $i = 2, 3, 4$ ). The two-photon Rabi frequency  $\Omega_{25}$  is taken as the non-resonant part of the two-photon coupling. This coupling is weak and at the two-photon resonance wavelength ( $\lambda = 388.3 \text{ nm}$ ) produces basically no effect. At  $393.5 \text{ nm}$  (the  $4s$ - $4p$  resonance), on the other hand,

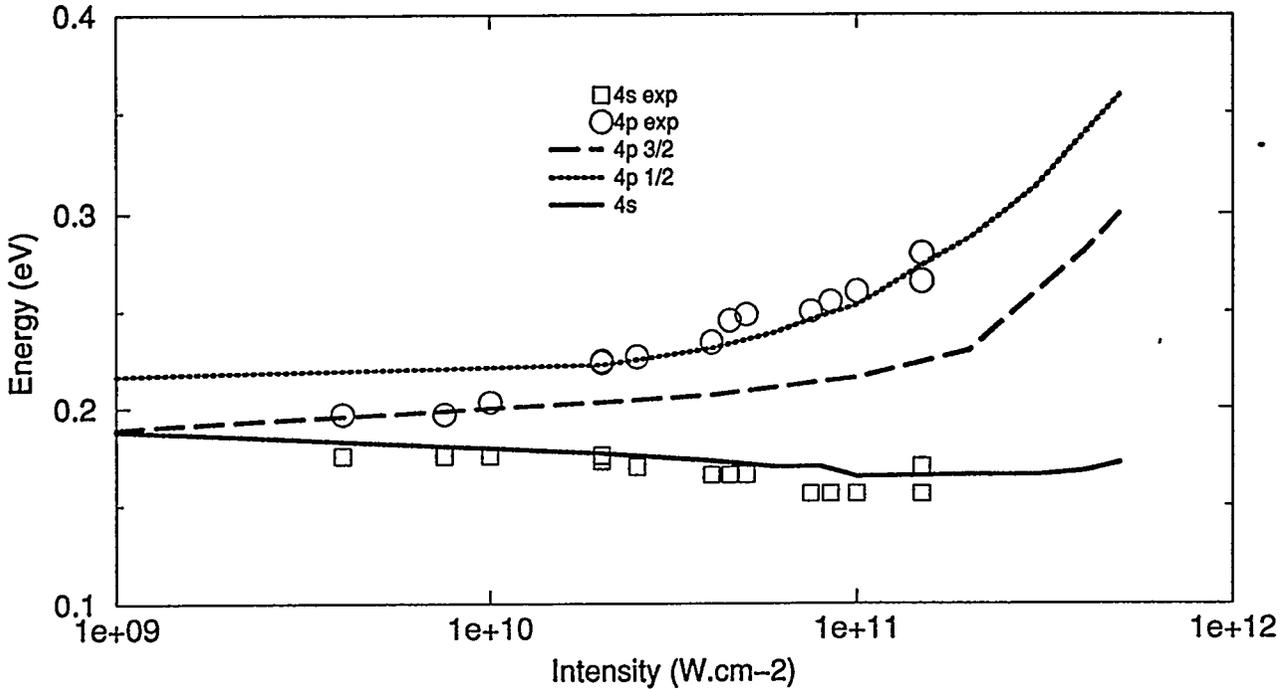


Figure 5. Calculated and experimental peak positions versus intensity at  $\lambda = 393.5$  nm. Note that the experiment for the highest energy peak first agrees better with the calculated  $4p_{3/2}$  then with the  $4p_{1/2}$  positions.

the  $5s_{1/2}$  strongly repels the  $4p_{3/2}$  through  $\Omega_{35}$ , therefore the corresponding electron peak has *more* energy. This effect combined with the influence of the neutral resonance produces the observed splitting. The role of the neutral resonance was first stressed by Hanson et al.[7].

#### 4. Conclusions

In conclusion, the continuum-continuum Autler-Townes splitting[1] occurring in two-electron atom when the radiation frequency is close to a core-

resonance has been experimentally observed in the two-photon ionization of calcium. However, the experimental observations cannot be described by a model involving only one discrete state coupled to two coupled continua. A reasonable description is obtained by including the neutral resonance and four continua. Finally, let us remark that the effect of the core resonance on the outgoing electron energy relies entirely on the electron-electron correlation[1]. As pointed out by Hanson et al.[7]: "if the two electrons were non-interacting particles, nothing unusual would be expected to happen". Interestingly enough, the most important coupling in this problem, namely  $\Omega_{23}$ , is obtained through an independent-electron approximation.

#### Acknowledgments

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