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CALCULATIONS OF POPULATION TRANSFER DURING INTENSE LASER PULSES

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1. Introduction

Recent experiments by several groups have examined the question of population transfer to resonantly excited states during intense short laser pulses, in particular the amount of population that remains "trapped" in excited states at the end of a laser pulse.[1-5] In this chapter we present calculations of population transfer and resonant ionization in xenon at both 660 and 620 nm. At the longer wavelength, the seven photon channel closes at 2.5×10^{13} W/cm^2. Pulses with peak intensities higher than this result in "Rydberg trapping", the resonant transfer of population to a broad range of high-lying states. The amount of population transferred depends on both the peak intensity and pulse duration. At 620 nm there are numerous possible six photon resonances to states with *p* or *f* angular momentum. We have done a large number of calculations for 40 fs pulses at different peak intensities and have examined the population transferred to these low-lying resonant states as a function of the peak laser intensity. We do not have room to comment upon the resonantly enhanced ionized electron energy spectra that we also determine in the same calculations.

Our calculations involve the direct numerical integration of the time-dependent Schrödinger equation for an atom interacting with a strong laser field.[6] The time-dependent wave function of a given valence electron is calculated on a spatial grid using a one-electron pseudo potential. This single active electron approximation (SAE)[7] has been shown to be a good approximation for the rare gases at the intensities and wavelengths that we will consider.[8] The SAE potential we use has an explicit angular momentum dependence which allows us to reproduce all of the excited state energies for xenon quite well. Xenon has two possible spin-orbit configurations when a single electron is excited, corresponding to a total angular momentum of J=3/2 or J=1/2. We will concentrate on the J=3/2 channel in what follows since its ionization potential, 12.15 eV, is 1.3 eV lower than that of the J=1/2 channel.

The pulses that we use have an intensity envelope given by $\cos^2[\pi t/2\tau_p]$, τ_p being the full width at half maximum (FWHM). This pulse is very close to a Gaussian pulse of the same FWHM, differing only for intensities far from the peak. It has the advantage of being finite in duration ($-\tau_p < t < \tau_p$), which allows for a precise determination of the final state populations and ionization probabilities. We have compared it to the analogous Gaussian pulses for several cases and have found it to be an excellent approximation.

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2. Rydberg Trapping

We first examine several pulses at a wavelength of 660 nm (1.88 eV). At this wavelength the ponderomotive shift ($I/4\omega^2$ in atomic units) of the ionization limit brings the top of the Rydberg series into resonance at $I_{res} = 2.50\ 10^{13}$ W/cm^2. This seven photon "channel closing" can bring many closely spaced Rydberg states into resonance each for a short time during a pulse that has a peak intensity that exceeds I_{res}. At the end of a pulse we project the final state wave function onto bound states of our xenon potential to determine the amount of probability "trapped" in excited states. This population, which is transferred resonantly during the pulse, assumedly does not have sufficient time to ionize out of the excited states. This is quite plausible given the long lifetimes of the high-lying, high ℓ states that can be accessed by seven photons from the $5p$ ground state. We can also think of this, loosely speaking, as the creation of a "Rydberg wave packet" which spends much of the time during the pulse at distances from the nucleus that are too great to allow for efficient ionization.[9-11] The ionized population can be defined as one minus the sum of the ground state population and the trapped population.

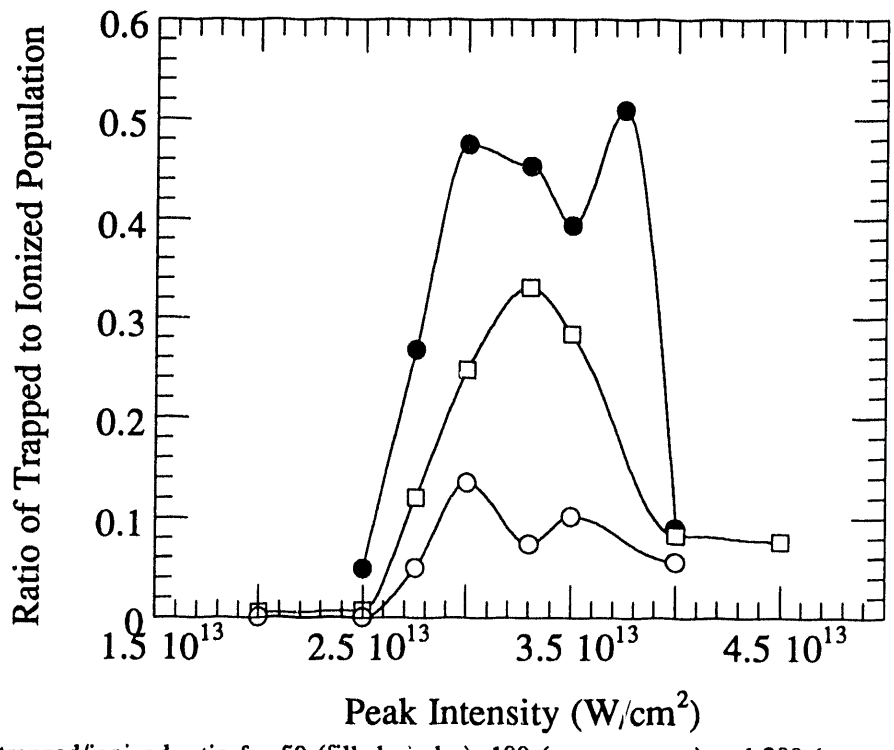

Fig. 1 The trapped/ionized ratio for 50 (filled circles), 100 (open squares) and 200 (open circles) fs FWHM pulses at a wavelength of 620 nm.

Figure 1 shows the ratio of "trapped" to ionized population for several peak intensities and three pulse widths. This ratio is close to zero for peak intensities (I_{peak}) less than I_{res}, as expected. For $I_{peak} > I_{res}$ the ratio grows, and can be as much as one half. The trapped population is found mostly in states with $\ell > 4$ and quantum numbers $n > 7$.

Longer pulse widths result in more excitation and ionization, but a lower overall trapped/ionized ratio. Eventually, as the peak intensity is raised and the channel closing occurs earlier in the pulse, the ratio decreases since most of the ionization then occurs non-resonantly. The detailed shape of the curve for each pulse width is quite complicated, though the maximum in the ratio scales roughly as τ_p^{-2}. This is quite plausible since both the amount of excitation during the pulse as well as the ionization from the resonantly populated Rydberg states scale as ~ τ_p (i.e., the trapping probability is proportional to τ_p^{-1}). The structure in the curves may be indicative of interference between resonant excitation on the rising and falling branches of the pulse, a subject we will return to in the next section.

3. Single State Trapping

Population can also be transferred resonantly to a single low-lying, relatively isolated excited state during an intense pulse.[1] We have studied this process for 620 nm (2 eV) photons in detail using 40 fs pulses, a pulse width somewhat shorter than the experiments of de Boor[1] but within current experimental capabilities. A total of 40 pulses with peak intensities between 0.5 and 6.0 10^{13} W/cm^2 were calculated.

At a wavelength of 620 nm, xenon has several six photon resonances in the range of ~ 1-3 10^{13} W/cm^2. These include the 6f, 5f and 4f states at I_{res} = 0.8, 1.1 and 2.0 10^{13} W/cm^2 respectively and the 8p at 1.4 10^{13} W/cm^2, assuming that these states shift

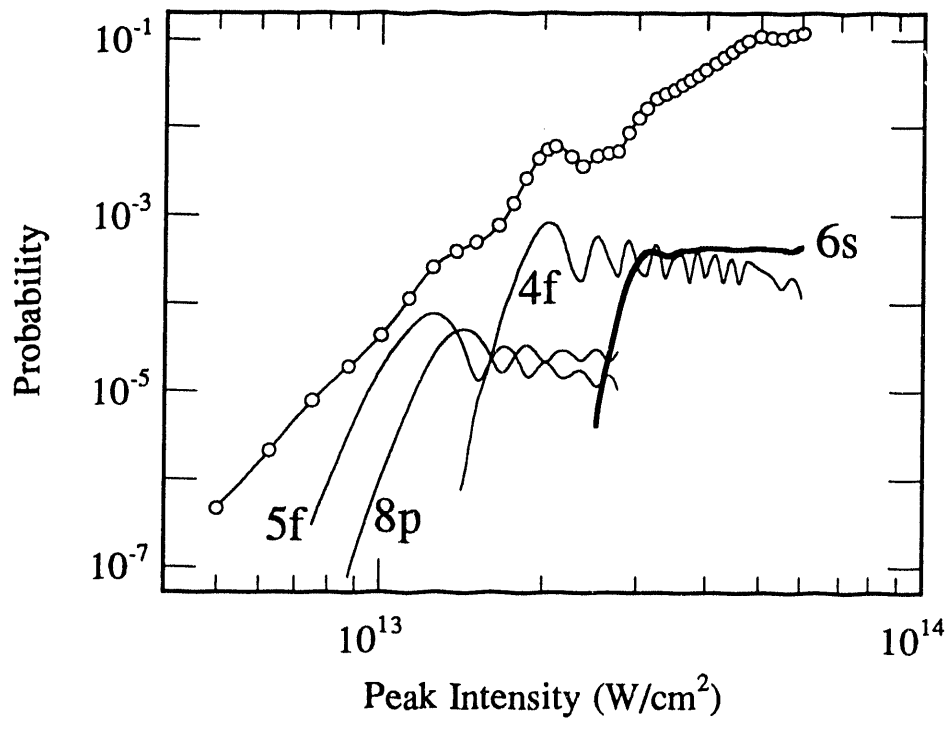

Fig. 2 The population remaining in several bound states at the end of a 40 fs pulse for 620 nm. Also shown is the total amount of ionization (open circles).

ponderomotively. Other, lower lying states such as the 7p and 6s (five photons) come into resonance at higher intensities and are not expected to have an ac Stark shift equal to the ponderomotive shift. Fig. 2 shows the population remaining in each of these states at the end of the pulse as a function of the peak intensity. Also shown is the total ionization probability. The curves all show a similar shape, with the exception of the 6s which we will comment on below. When $I_{peak} < I_{res}$ little population can be excited to the resonant state. At $I_{peak} \sim I_{res}$ the probability to remain in the resonant state peaks, and for $I_{peak} > I_{res}$ the probability is smaller and shows oscillations with respect to I_{peak}. This is shown in more detail for the 4f resonance in Fig. 3.

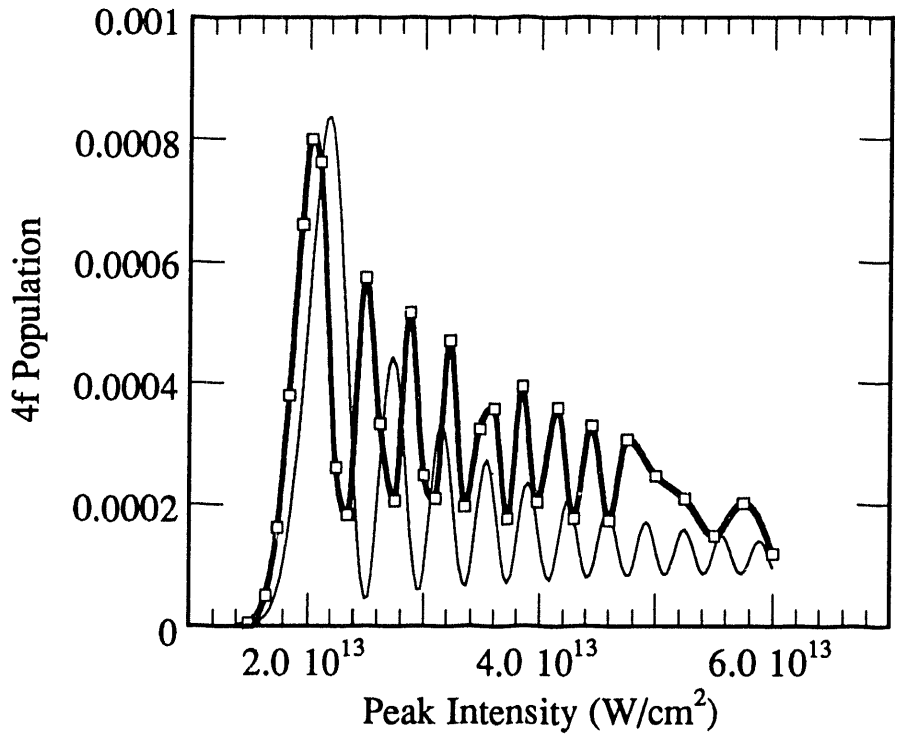

Fig. 3 Population in the 4f state at the end of a 40 fs pulse (the heavy line is a spline fit to the calculated points). The lighter curve is a two state RWA calculation.

We can use a simple two state model in the rotating wave approximation (RWA) to explain the main features of Fig. 3.[12] Consider a state that comes into resonance with the ground state during a pulse that peaks at I_{peak}. The two states are connected by an effective six photon Rabi frequency. The detuning from resonance is simply the zero field detuning minus the ac Stark shift, which is proportional to the intensity if the upper state is assumed to shift ponderomotively. For $I_{peak} < I_{res}$ the detuning is always positive and very little population is transferred and hence little remains after the pulse. For $I_{peak} = I_{res}$ the detuning is close to zero for a time proportional to τ_p^2 leading to a relatively large population transfer and, even allowing for ionization, population remains after the pulse. For $I_{peak} > I_{res}$ the detuning is close to zero twice during the pulse for a time proportional to $\sim \tau_p$. If the accumulated phase between the two resonant times is an odd multiple of pi,

for instance, then destructive interference occurs and the upper state population is zero, in the absence of ionization. Of course there is loss from the upper state in the form of one photon ionization, which is also proportional to the intensity in lowest order. This leads to less than perfect cancellation between the two resonant amplitudes and so the minima in Fig. 3 are not equal to zero. It is interesting to note that we observe oscillations in the upper state population as a function of I_{peak} even though the population in the upper state is always small compared to the lower state. This is because the oscillations come from the interference between the two small amplitudes excited at the two resonant times. Although the two state model gives a qualitative explanation of the oscillations, the RWA result shown in Fig. 3, calculated for a reasonable set of parameters, is deficient in several respects, indicating that the full calculation includes features not captured in the simple two state RWA.

Oscillations in the remaining population are only visible when the lifetime of the upper state is as long or longer than the pulse width. In Fig. 3 this is visible in the damping out of the oscillations at higher peak intensities. In Fig. 2, the 6s state, which comes into resonance at high intensity and which has a short lifetime owing to its low angular momentum, shows no oscillations. This effect can be illustrated directly by comparing 40 and 100 fs pulses. As shown in Fig. 4, the longer pulse leads to more ionization and *much* less trapping, as expected since the lifetime of the 4f state is expected to be shorter than 100 fs at a few times 10^{13} W/cm^2. The remaining population shows no oscillations for the longer pulse and continues to rise for $I_{peak} > I_{res}$. Again, this behavior is qualitatively reproduced by the two state RWA.

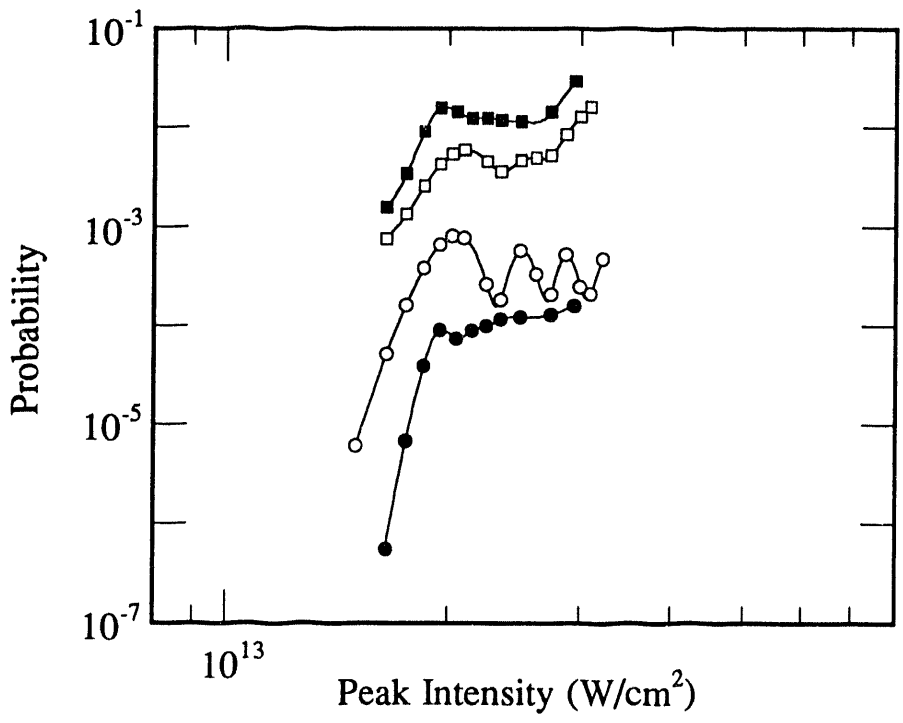

Fig. 4 Ionization (squares) and 4f population (circles) for 100 (filled) and 40 (open) fs pulses.

4. Acknowledgment

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