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Title → Amplitude modulation of atomic wave functions

Research accomplished under the previous award

During our first three years of support by the Department of Energy, we have made advances in our theoretical understanding of the modulation process, and in our development of the equipment necessary to perform wave function modulation, and we have begun experiments using the two pulse method which will be crucial for creating modulations. Following that, we moved our laboratory from the University of Southern California to the College of William and Mary. In addition, we were given a fully functional high-power, short-pulse laser laboratory from AT&T Bell Laboratories. This laboratory, and our laboratory from USC, have now been completely set up at William and Mary, and we are in the final stages of coordinating the equipment from the two.

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Phase modulation

Our major theoretical advance has been to show that we can modulate Rydberg wave functions using either of two methods: (1) the amplitude modulation technique which depends on autoionization to deplete part of the wave function, or (2) a phase modulation method, which uses a change in the core potential to create a localized phase shift in the wave function. Essentially, these two methods can both be seen as using the core potential to change the Rydberg wave function, using the imaginary part of the potential to do amplitude modulation, or using the real part of the potential to do phase modulation. This work will be published as we acquire experimental results which show the differences between the two methods.

One of the results of this theoretical study is that our initial proposal to study Barium $6snd$ states had a significant flaw. Neither the autoionization time, nor the quantum defect shifts are very large in these cases. This means that the modulation is relatively small. This shows itself primarily in the difficulty of seeing significant population redistribution into different $6snd$ states. We intend to correct this in the next funding cycle either: (a) by using the more quickly decaying Ba $6pnf$ states to modulate $6snd$ states, or (b) by using Sr $5snd$ states, as outlined below.

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Laser development

Our equipment development is now almost complete. We have installed a Tsunami Ti:Sapphire laser system which can produce either 1-2 ps pulses, or 70 fs pulses. In our present experiments, we have been using the ps mode to generate pulse trains of 908 nm light, which we then frequency double to produce the required 454 nm light. Also, we have constructed a five-stage dye amplifier, pumped by a Nd:YAG laser which is synchronized to the Tsunami pulse train. This system produces single pulses of 454 nm light with pulse energies up to 100 μ J per pulse. We have made an extensive effort to characterize these pulses, using a variety of auto-correlation methods, and by examining the pulses' spectra. All of these diagnostics have been necessary to produce repeatable, well-characterized pulses.

Time dependent studies

Our first, low power experiments are complete. These experiments have used two pulses to do a temporal version of the Ramsey separated oscillatory fields excitation. The two pulses are generated by passing the single pulse through a Michelson-Morley interferometer, which is computer controlled to sweep one arm through 2.5 μ m in steps of 10 nm. The second pulse's excitation interferes with that of the first pulse, and so the total excitation has a sinusoidal variation (with a time period equal to the optical period) on top of a constant background. The amplitude of the total variation should decay at half of the rate decay rate of the autoionizing state, so this produces a time-resolved measurement of the very rapid autoionization decay. Although this does not yet show that the atom stores modulations in the bound coherent state, it does demonstrate that the atom can be excited to an autoionizing state with high efficiency, and then brought back to a bound state at a later time.

Figure 1 shows the type of oscillations we typically obtain after sweeping the interferometer arm. This figure was obtained using the 6s37d state as the initial Rydberg state, and a 4 ps overall offset between the two pulses. A series of these measurements at different overall pulse separations leads to a decay signal which illustrates the time

Figure 1: Ionization efficiency for excitation by two pulses separated by approximately 4ps.

evolution of the doubly excited wave packet.

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view by convoluting the two pulse laser spectrum with the atomic absorption line, the low power experiments do not show the unique power of the timed pulse method. High power results, on the other hand, are not perturbative, and thus cannot be obtained from a simple time independent spectral analysis.

Saturated line shapes

Figure 2 is an example of a saturated spectrum, which shows that the modulation process will be sensitive to laser tuning, even at high powers. Robiscoⁱ first showed that lineshapes, when the exciting laser has a sech^2 intensity profile, are insensitive to power broadening. Model calculations show that Gaussian profiles behave more like sech^2 pulses than like square pulses at high powers.

Figure 2: Saturated lineshape of the 6s48d→6p48d transition.

Consequently, we have used fluences greater than $10 \mu\text{J}/\text{mm}^2$ to measure the high power behavior of the core transitions. We have observed interesting effects using either one or two pulses. First, a single laser pulse saturates the core transition, instead of producing Rabi oscillations, as shown in Figure 2. This is consistent with a small frequency chirp in our laser pulse (which is also consistent with the small excess bandwidth in our pulse after amplification). The chirp causes adiabatic rapid passage to the doubly excited states, and transfers nearly 100% of the Rydberg atoms without significant return to the bound states. This is an effective way of eliminating the adverse effects of pulse-to-pulse power fluctuations. It is worth noting that the linewidth of this saturated transition *does not change with power*.

Chirped π pulses

We have observed that a second pulse (also chirped) returns nearly 60% of the atoms to the bound states. Figure 3 shows a two pulse, high power ionization signal obtained

starting from the $6s48d$ state, with a delay near 4.5 ps between the two pulses. Figure 3 also shows the interferometric signal when the relative delay between the two pulses is varied; however, the total transfer to the doubly excited state is always less with two pulses than with one. This is very clear evidence of transitions in the strong coupling regime. The residual interferometric signal is presumably due to insufficient laser power and we are in the process of eliminating it by increasing our laser power, and modifying our laser chirp.

Figure 3: Two high power pulses return most of the autoionizing population to the bound states.

ⁱ R.T. Robisco, Phys. Rev. A 17, 247 (1978).