Ultrafast Quantum Control in Atoms

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This paper discusses recent experiments to control quantum dynamics in atoms and simple molecules, using shaped ultrafast radiation. The shaped pulses excite shaped quantum wave packets, which can perform various tasks. One application is quantum information science, where shaped wave packets interact with shaped laser fields to load, store, manipulate, and retrieve data from the quantum system. A second application is active control of molecular dynamics. Here the optical pulse shaper receives feedback signals from the quantum system. Efficient search strategies such as genetic evolutionary algorithms make it possible to find and optimize excitation pathways, or enhance weaker reaction channels over stronger ones.

Programmable ultrafast pulse shapers were developed as modulators for optical communication. In high field laser physics these modulators are now used widely to compensate for dispersion in the laser system, in order to produce the shortest possible high intensity radiation pulses. Nonlinear feedback from a second-harmonic generator is often used to provide an indication of the shortest pulse. This even has been shown to work for very high order nonlinearities, such as high harmonic generation in atoms [1].

The shortest optical pulse may not always be the aim of the pulse shaping experiment. More sophisticated pulse shapes were programmed to control nonlinear atomic absorption [2], molecular fluorescence [3], and photodissociation. Control experiments that rely on only two known interfering pathways allow for calculations of appropriate pulse shapes to effect control [2]. Strongly coupled systems, especially large molecules in condensed phase are so complicated that it is difficult and often impossible to calculate optimal pulse shapes in advance. As a result, recent efforts have used experimental feedback as suggested by Rabitz and Judson [4] to determine the optimal optical pulse shape in order to achieve a particular control goal [5, 3]

We have studied new methods to discover specific pulse shapes that produce specific photoionization reaction channels in molecular sodium, and the excitation of specific bonds in methanol. We have constructed an automated learning apparatus capable of using feedback from the molecular. The feedback was used as input for an evolutionary learning strategy based on a genetic algorithm (GA) [6]. This algorithm ran on a computer, which controlled an acousto-optic modulator (AOM) pulse shaper [7]. The algorithm generated new pulse shapes based on the success of previous ones in achieving a predetermined goal. The GA is a useful tool for learning about complicated physical systems, but its use in quantum dynamics has been relatively limited. We have discovered new ways to use the GA-found solutions to yield new information about the quantum system under study.

In systems where the Schrdinger Hamiltonian is understood well, such at atomic Rydberg states, feedback can still be used to compensate for imperfections in the pulse shaping apparatus. Rydberg states often contain hundreds or thousands of different states within the bandwidth of the optical pulse shaper. Programmable can be produced readily, and viewed using simple holographic techniques [8]. One application of such states is a quantum data register. The storage of information as quantum phase has been proposed in connection with new computational algorithms based on the rules of quantum mechanics.

We have investigated the storage and retrieval of information in the quantum phase of a coherent superposition state of energy levels in a Rydberg atom [9]. We prepared a data base in which one of the items was marked, and demonstrated Grover's contention that the marked item can be retrieved by a quantum computation in a single query of the data base [10]. We analyzed the influence of ensemble averaging as well as errors introduced by technical imperfections in our physical system. We also showed that the same quantum system and measurement techniques can be extended in order to store and retrieve large numbers. We have demonstrated storage of numbers up to 2^{N-1} for $N = 8$. A straightforward extrapolation of our results suggests that numbers as large as 2^{100} may be stored in a single N-level atom, where $N = 20$.

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