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13. ABSTRACT (Maximum 200 words)

Wave packet preparation and probing is investigated in molecules, with particular emphasis on the formation of Rydberg wave packets. A new apparatus has been developed to investigate Rydberg state excitation with ultrafast lasers. It consists of an ultrafast laser system, a cw dye laser for launch state preparation, and an electron time of flight spectrometer. Three photon plus one photon two-color photoionization of Kr has been achieved, with apparent resulting Rydberg wave packet signals. A wave packet has been prepared between the F and G electronic states of lithium dimers, which introduces the electronic degree of freedom into rovibrational wave packets in a simple diatomic molecule. This is directed towards quantum information manipulations.

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This project considers the new topic of preparing and investigating molecular Rydberg wave packets. Rydberg wave packets are of interest because of the electronic degree of freedom and the breakdown of the Born-Oppenheimer approximation due to interactions between the Rydberg wave packet and single vibrational and rotational states of the ionic core. The resulting wave packets are constructed from multiple degrees of freedom - i.e. nonstationary states in both the Rydberg and vibrational or rotational coordinates. The application of utilizing these wave packets for quantum information and quantum computing is a key goal. The grant period was shortened because of the principal investigator's change of institution. Therefore, this report from the University of Colorado covers the beginning of the project and a new grant has been issued to the University of California, Berkeley, for the continuation of the project.

A new experimental apparatus was constructed in the laboratory to prepare and probe Rydberg wave packets with ultrafast laser pulses (Fig. 1). This apparatus borrows features from the heat pipe cells that have been used in this laboratory for other studies of wave packets, but at lower densities and with differential pumping to extract electrons from the sample region and analyze for their kinetic energies. The experiments are based on a 1000 Hz, 100 fs, 1 mJ Ti:sapphire regenerative amplifier system, which is seeded by a 75 Mhz Ti:sapphire Kerr lens mode-locked oscillator. A cw ring dye laser is used for initial launch state preparation, and suitably time-delayed pulses are used to prepare and probe the Rydberg wave packets. A precision stepper time delay stage and a liquid crystal spatial light modulator are used to control the pulses. The molecules are generated in a heat pipe to form Li_2 or Na_2 dimers. When forming the dimers, high rotational states will be accessible, which will be central to the Born-Oppenheimer breakdown studies of the Rydberg wave packet timescales compared to the ion core rotational and vibrational motions. The apparatus has ion and electron counting electronics and provision for time-of-flight electron kinetic energy analysis and ion mass detection. The time-of-flight detection has recently been shown to give excellent signals from Kr Rydberg wave packets formed by a 3 photon excitation, as discussed further below. All of the laser and vacuum hardware, counting electronics, and optical diagnostics are dedicated to Rydberg wave packet studies and are being transferred to Berkeley.

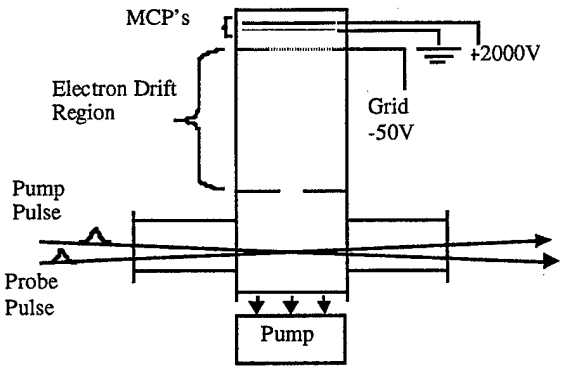

Fig. 1 Schematic of the Rydberg wave packet apparatus constructed for this work.

Rydberg wave packets, studied extensively in atomic systems, are notorious for producing weak signals by traditional optical-optical pump-probe ionization detection methods. This is because the high energy of the optical probe photon must be partitioned between the kinetic energies of the

outgoing Rydberg electron and the resulting ion, which is difficult to achieve because momentum must also be conserved. A classical picture is that the electron can only produce a sufficient "kick" to the core ion when it is at very close range. The weak signals from optical ionization of the wave packet are often obscured by multiphoton ionization "noise" by the individual laser pulses. A common way to detect Rydberg wave packets with improved signal-to-noise is the so-called optical Ramsey fringe method; two wave packets are launched in the atom with an intervening variable time delay, and the Rydberg packet is field ionized by voltages on plates. Interference between the two wave packets creates constructive or destructive amplitudes that enhance or diminish the ionization probability.

Considering the low signals expected with Rydberg wave packets, exploratory work was undertaken using a 1000 Hz Ti:sapphire laser system already available in our laboratory while the new 1000 Hz laser for this project was being procured and installed with DURIP equipment grant support. Using a three photon excitation at 267 nm (third harmonic of the Ti:sapphire laser), Rydberg wave packets were excited directly in Kr atoms in the $nd_{3/2}$ and $nd_{5/2}$ series. A third photon at 800 nm was used to ionize the Rydberg wave packets. With an electron time-of-flight spectrometer, the 3+1' multiphoton process that probes the wave packet was isolated by the precise kinetic energy of the electrons, with good signal-to-noise ratios (Fig. 2), after removal of a slow background variation by normalizing to the signal with that from a different final spin orbit state of the Kr^+. Quantum beat interferences among a small number of Rydberg wave packet states were observed.

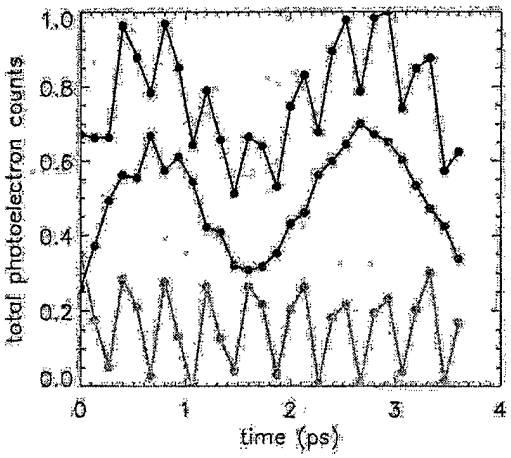

Fig. 2 Rydberg wave packets observed in Kr, showing the original signal (top), the slowly varying component (middle) and the fast oscillations (bottom) of the wave packet.

Using the kinetic energy resolution to isolate the two-color process of interest appears to be a good way to study Rydberg wave packets. These results were performed initially in a magnetic bottle time-of-flight spectrometer, and the magnetic field can have a strong influence on the Rydberg levels. Thus we have constructed a normal linear time-of-flight spectrometer to eliminate the magnetic field, shown in Figure 1 above.

Recently the new 1000 Hz Ti:sapphire laser amplifier for this project was installed and is operating. Preliminary signals were obtained on rovibrational Li_2 molecular wave packets, and

then again on Kr, as mentioned above. Investigations are still in progress to determine whether the oscillations are the desired Rydberg wave packet signals. With the new time-of-flight spectrometer, two directions will be pursued to obtain Rydberg wave packets in molecules. The first is to tune the total energies of the photons in the multistep excitations to higher electronic levels in Li_2 or Na_2 to excite pairs or multiple Rydberg states simultaneously with vibration/rotation states in the dimers. A second option is to explore Rydberg wave packets in molecules such as NO, using the direct 3+1' type of multiphoton ionization as discussed for Kr atoms above.

To begin on the project of tuning to higher total energies to access multiple electronic states using Li_2 wave packets, wave packets have recently been prepared between the F and G electronic states for the first time. This is a first example of an electronic wave packet. It can involve similar or different vibrational and rotational levels in each electronic state. The new aspect is that the electronic degree of freedom is being accesses. By tuning to higher states, this method of stepping up to higher levels in lithium dimer has a good chance of preparing desirable Rydberg wave packets.

Based on the signals achieved in the recent Li_2 studies in a heat pipe (presently we do not even require an electron counting system), and scaling for the lower densities needed for the Rydberg wave packet studies, we estimate that good signal-to-noise should be achieved for electron time-of-flight detection of Rydberg wave packets by using a combination of the time-of-flight and heat pipe methods. Once a system is working, with single state selection and phase control, we plan to pursue the conceptual experiments related to quantum information with that system.

Finally, a project was undertaken to study of the Deutsch-Josza algorithm by using rovibronic states in the E electronic manifold of Li_2 as a model system by addressing multiple wave packet states. In this case, 8 wave packet states are used to mimic 3 qubits. The balanced or constant functions were prepared and measured, demonstrating the determinations of balanced or constant functions with greater than 99% accuracy.

Personnel on project:

Elizabeth Mirowski - Ph.D. expected July 2002
Stefan Gilb - postdoctoral associate
Joshua Ballard - graduate student
Lutz Hüwel - visiting professor

Publications:

J. Vala, Z. Amitay, B. Zhang, S. R. Leone, R. Kosloff, "Experimental implementation of the Deutsch-Josza algorithm for three-qubit functions using rovibrational molecular wave packets representation," Phys. Rev. Lett. submitted.

Annual Technical Report submitted in December 2001

Invited talks:

Symposium on Molecular Spectroscopy, Columbus, Ohio, June 2002

Inventions: none