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Introduction

The observed imbalance between matter and antimatter in the universe remains one of the unsolved mysteries of modern physics. One test that might shed some light on this puzzle is a measurement of how antimatter acts in a gravitational field. One possible way to measure this directly is to produce a beam of neutral positronium (Ps, the bound state of an electron and its antiparticle, the positron) and observe its free-fall in the gravitational field of the Earth [1]. However, the triplet ground state of Ps has a lifetime against annihilation of 142 ns, allowing free-fall of only around 100 fm (if $\bar{g} = g$). Therefore, any such experiment must be done using longer lived excited states, and only Rydberg states have lifetimes that are experimentally feasible (which is a few ms). Atoms in these highly excited states can also possess extremely large electric dipole moments, allowing manipulation with inhomogeneous electric fields [2].

Typical methods of Ps generation produce very fast atoms $(v/c \sim 10^{-3})$, resulting in significant Doppler broadening (e.g., ~0.5 THz for 1s-2p). Efficient production of Rydberg Ps has been achieved with broad bandwidth (85 GHz) lasers that give reasonable overlap with these large Doppler profiles [3]. However, this technique limits the resolution and prevents accurate state selection at higher n. To overcome this we plan to implement non-resonant Doppler-free two-photon excitation with much narrower bandwidth (3 GHz) pulsed dye lasers. This scheme allows state-selectivity at high-n, opening up possibilities for transfer to higher angular momentum states. As a demonstration of this scheme we have used the Central Laser Facility's NSL2 laser system to drive non-resonant two-photon transitions to Rydberg states in a pulsed supersonic beam of helium. Using circularly polarized light we have driven transitions from the $1s2s 2^{3}S_{1}$ state to the 1snd Rydberg states in the range from n = 20 up to $n \sim 100$, clearly resolving states up to n = 76.

Experiment

A beam of helium atoms is made by supersonic expansion into a vacuum chamber followed by population of the 1s2s ${}^{3}S_{1}$ state in an electric discharge. The resulting beam of metastable atoms travels with a forward speed of 1940 m/s and a temperature of around 5 K. After passing through a skimmer the atoms can be excited to high Rydberg states by non-resonant two-photon excitation with light at wavelengths in the range 520 - 524 nm from a pulsed dye laser (up to 15 mJ in a 5 ns duration pulse). The laser polarization is set and manipulated using a Glan-Taylor polarizer (GT) and a quarter-wave plate (WP) before being focused into the atomic beam, with a waist of around 100 µm. Excitation is performed between two electrodes (labelled E1 and E2 in Figure 1). Applying a potential difference across these electrodes allows the laser excitation to take place in a uniform electric field. This field can be used to separate out the Stark states, allowing state-selective excitation, which will be



Figure 1: Experimental setup. The light from the pulsed dye laser is incident upon a supersonic beam of helium atoms in the metastable 1s2s ${}^{3}S_{1}$ state. By scanning the wavelength between 520 nm and 524 nm two-photon transitions are driven to Rydberg states in the range from n = 20 up to the ionization limit. The Rydberg atoms are detected by pulsed field ionization in a field of strength 800 V/cm between electrodes E3 and E4.

very important for future manipulation of these atoms using inhomogeneous electric fields (such as those in Rydberg-Stark decelerators [2], or electrostatic mirrors [4]). After traversing the excitation region the atoms travel for 118.5 mm before being ionized by a pulsed electric field applied between electrodes E3 and E4. The resulting ions are then detected by a micro-channel plate (MCP).

Excitation scheme

The two-photon scheme employed in our experiment is nonresonance enhanced, with the virtual intermediate state fardetuned from any bound states (Fig. 2). The excitation path depends on the laser polarization; excitation with circularly polarized light drives transitions in which $|\Delta m|$ = +1. Thus a twophoton transition drives the atoms from the 2s state to *n*d Rydberg states. Alternatively linearly polarized light can drive transitions in which $|\Delta m|$ = ±1, allowing the population of both *ns*- and *n*d-states. With the goal of populating long-lived higher angular momentum states, we use circularly polarized light for most of our measurements.



Figure 2: Energy level diagram of helium, showing the possible excitation channels for the two-photon transition, depending on the laser light polarization.



Figure 3: Recorded spectrum of |m|=2Rydberg states of helium, populated by nonresonant two-photon excitation using circularly polarized light. Rydberg states in the range $20 \le n < 100$ are efficiently populated, with states up to n = 76 clearly resolved.

Rydberg spectrum

Figure 3 shows a recorded spectrum of *n*d Rydberg states ranging from n = 20 up to the ionization limit, driven by circularly polarized light. The Rydberg states are clearly resolved up to n = 76. The linewidths are greater than the laser bandwidth, and are dominated by the ac Stark shift experienced by the 2s state, which has contributions from the off-resonant coupling to many higher *n*-states. The ac Stark shift reduces the energy of the 2s state, resulting in an asymmetric broadening. The overall shape of the spectrum is influenced by the radiative lifetimes and black-body interactions (which dominate for lower *n*-states) and the *n*-dependence of the transition strengths (which dominates the shape for higher *n*).

Stark states

An electric field causes the Stark states of a given n to shift in energy. Applying such a field in the excitation region allows us to selectively populate individual states. This is important for manipulation of the atoms with electric fields as the Stark shift experienced by an atom depends on which of these Stark states it is in. Thus, to be able to control the motion of an atom in a given electric field we need to be able to choose which Stark state it occupies.

Figure 4 shows the effect of various uniform electric fields applied in the excitation region. As this field is increased the Stark energy shifts broaden the measured lines. By around 150 V/cm we are able to resolve and selectively populate individual states. The effect of the ac Stark shift induced by the intense laser field can be clearly seen in the spectra in Fig. 4. The broad asymmetry limits the resolution somewhat and requires the application of quite strong fields before we achieve good Stark state resolution. The effect that this ac Stark shift has on the



two-photon transitions has been found to be significant, and we now know how important it will be to account for this in future experiments.

Conclusions

We have shown that non-resonant two-photon excitation of metastable He to Rydberg states can be efficiently achieved under experimental conditions that are not dissimilar to those expected in experiments with Ps atoms. Extraneous influences, such as ac stark shifts and effects of black body radiation have been observed and understood and will not adversely affect Ps atoms excited in the same way. By comparing the expected transition strengths of hydrogenic atoms [5] with an estimate of the He transition strengths [6], we anticipate that a laser with the same power and bandwidth of the NSL2 system (10 mJ, 5 GHz) would allow 1s-nd excitation of Ps atoms with an efficiency of 10 - 15%.

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