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Pulsed Field Recombination

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We study the production of neutral (anti-)matter via Pulsed Field Recombination of ions and electrons with three different experiments and with a theoretical description.

1. Introduction

In 1996 Baur et al.[1] observed nine statistically significant events of the formation of anti-hydrogen atoms, the bound state of a positron and an antiproton. However, the number of detected anti-hydrogen atoms was small and they were produced at relativistic velocities, making meaningful measurements essentially impossible. A breakthrough, paving the way to serious research into the properties of neutral antimatter, would be the production of cold anti-hydrogen atoms. To produce such cold anti-hydrogen atoms and to catch them before they annihilate with matter, is the primary goal of the ATRAP (Antihydrogen Trap) collaboration[2]. Once trapped, cold anti-hydrogen atoms can be used for experiments such as tests of CPT invariance or gravity measurements.

Making an (anti-) hydrogen atom by recombining an electron (positron) and a proton (anti-proton) is not simple. The recombination is only possible when a third object carries away the excess energy of the captured electron. This third particle can be a second electron (this process is known as three-body recombination), or a photon (radiative recombination).

Recently, we reported on the realization of a new,controllable and efficient recombination scheme using pulsed electric fields as slow as 1 ns. Pulsed field recombination (PFR) [3,4] can be regarded as intermediate between the radiative and the three-body process. One can say the effect of the third body, stimulating the recombination through its time dependent electric field, is now applied 'by hand' in the form of tailored pulses.

Here we report on new experiments where we use "ordinary" matter to study the recombination dynamics of the PFR scheme. There were three major motivations for performing these experiments. To test if the PFR scheme could indeed be used to recombine lighter ions with free electrons we have used lithium ions instead of rubidium ions that were used in previous experiments. Lithium being 12 times lighter than rubidium resembles much more a proton, although it is still 7 times heavier. The second motivation was to study PFR in an configuration which had a greater resemblance with the ATRAP experiment at CERN, where recombination of *trapped* positrons and *trapped* antiprotons will be studied in Penning traps [5]. The last motivation was to increase the recombination efficiency of the PFR scheme.

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We propose that PFR can be used in cation spectroscopy on molecular species where laser excitation of a Rydberg state fails. In Photo-induced InfraRed Ionization (PIRI) spectroscopy [6], the cation spectrum of a molecule is recorded with the aid of a Rydberg state in that specific molecule. Most commonly, such a Rydberg state is prepared by laser excitation. For complex molecules laser excitation of a well defined Rydberg state can fail, so that the PIRI technique can not be applied. Recently we have shown that the PFR method can be used to recombine a free electron with a large ionic carbon cluster such as C_{60}^+, inducing a highly excited bound state of such a molecule ($n \approx 180$) [4]. This was the first time the existence of C_{60} Rydberg states was observed convincingly.

In addition, a theoretical description is presented with semi-classical calculations. These semi-classical calculations give deeper insight on the states which are being formed in the recombination process and can be used to probe recombination dynamics not probed by the experiments.

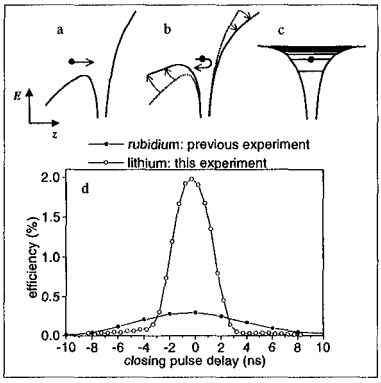

Fig. 1

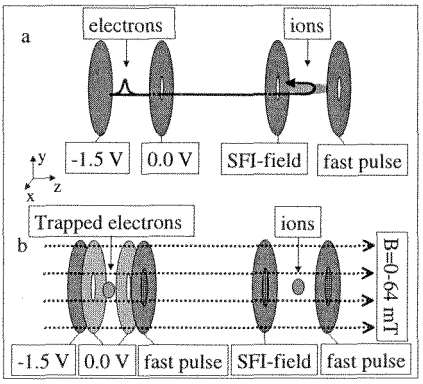

Fig. 2

Fig. 1, (a-b): Schematic representation of the Pulsed Field Recombination scheme (see text). (d): Experimentally observed efficiency as a function of the delay of the quick turnoff. The open circles represent recombination of free electrons and lithium ions with the use of an electron trap. The solid circles represent data from previous work[3] where we studied recombination of free electrons and rubidium ions in a trapless geometry.

Fig. 2: Sketch of the two experimental setups used to study PFR. In Fig. 2a the setup is depicted where we studied recombination of free electrons with Rb^+ and with C_{60}^+. In Fig. 2b the setup is depicted where we studied recombination of free electrons, originated from an electron trap, with Li^+.

2. Scheme & efficiency

In Fig. 1 the PFR scheme is sketched. An ion is situated in a static electric field that modifies the Coulomb potential such that a saddle point is created (Fig. 1a). If an electron passes over the saddle point in the modified Coulomb potential, it will take a small, but not negligible, amount of time to return to the saddle point and escape from the ion (Fig. 1a). If the static field is turned off (Fig. 1b) before the electron returns to the saddle point, it will remain bound in a highly excited state (Fig. 1c). The time

required for the electron to travel from the saddle point, to the nucleus, and back to the saddle point is about 1 ns for the fields and energies used in these experiments. This time is comparable to the turnoff time of the static electric field (90% → 10% in 0.8 ns). In Fig. 1d the experimentally determined efficiency (number of recombined rubidium atoms divided by the number of free ions) of this scheme is depicted as a function of the delay of the fast field turnoff. This delay is with respect to the time when the free electron has its turning point in the electric field. Clearly, a maximum number of recombination events is recorded at zero delay. A comparison with the previous experiments with rubidium ions shows that the efficiency ($3.0\text{x}10^{-3}$[3]) has gone up by almost an order of magnitude ($2.0\text{x}10^{-2}$) and that the electron pulses are much shorter. The increase in efficiency is mainly due to the fact that the electron pulses are shorter in time (from 11 ns[3] to 3.8 ns) and that we had better control over the overlap volume of the ion cloud and the volume of the electron pulses. We achieved the highest efficiency (6%) so far when we used the PFR scheme to recombine free electrons with free ionic carbon clusters in a trapless environment[4]. Note that we have shown that PFR can be used in the geometry of *trapped* electrons and *light* ions.

3. Experimental

We have used two different setups to study Pulsed Field Recombination. With the first setup we have studied recombination with rubidium ions and recombination with ionic carbon clusters. The second setup was designed to mimic as much as possible the conditions under which anti-protons and positrons should be recombined at CERN to produce cold anti-hydrogen.

3.1. Setup I

In the first setup a pulsed electron source was created by photoionizing lithium atoms in a static electric field of 1.50 V/cm, with a pulsed (9 ns duration) narrowband ($\Delta\lambda <$ 0.2 cm^{-1}) dye laser. Typically electron pulses of 11 ns duration, with about $(5\pm2)\text{x}10^4$ electrons in a volume of 0.02 mm^3 were produced. The electric field is created by two parallel capacitor plates (separation: 10.0 mm) over which a voltage is applied. After ionization, the free electrons are pushed towards the anode plate (connected to ground) through which a small hole is drilled, covered by a grid. After passing through the anode, the electrons enter a field-free region of 15.0 mm, after which another set of parallel capacitor plates (with small holes, covered by a grid) is situated, separated by 10.0 mm. In this region a cloud of ions is awaiting the electrons. To minimize the spread of the electrons during their travel towards the ions in directions perpendicular to the direction of propagation, a magnetic field was implemented, parallel to the direction of propagation ($B_z = 2.5$ mT). Typically $(5\pm2)\text{x}10^3$ Rb-ions were produced in a volume of 2.0 mm^3. The rubidium ions were produced by photoionizing rubidium atoms in a static electric field of 1.50 V/cm, 50 ns before the lithium atoms were ionized, in a two-photon process. Note that the heavy rubidium ions are virtually standing still during our experiment ($\Delta l = 1$ μm in 100 ns). The whole plate configuration is depicted in Fig. 2a.

The ionic carbon clusters were produced 500 ns before the photoelectrons, by photoionizing and fragmenting gas-phase C_{60} molecules (T$\sim$ 750 K) in a static electric field of 1.50

V/cm. We photoionized C_{60} molecules (IP = 7.6 eV) by the fourth harmonic (266 nm) of a Nd:YAG laser. The multiple ionization with 266 nm mainly produces the higher ionic carbon clusters. Detailed studies of the fragmentation and ionization dynamics of C_{60} are reported in Ref.[7]. By producing the ionic carbon clusters 500 ns prior to the arrival of the electron pulses assures that the major fraction of the excited C_{60} molecules have ionized at the moment of the recombination event. The ionic carbon cloud resembles a cylinder with a radius of 35 μm in the z-and y-direction and a length of 10 mm in the x-direction, with a density of $2.4(\pm 0.4)\text{x}10^7$ cm^{-3}.

In the ion source region the electrons are decelerated by the electric field of 1.50 V/cm, and have their turning point at the position of the ion cloud. At that time the field is turned off by dipping the voltage on the cathode plate from -1.50 V to -0.20 V. The electric field turnoff is realized by connecting an impedance matched, fast pulse generator to the cathode plate.

3.2. Setup II

The second setup was designed to mimic as much as possible the ATRAP-experiment at CERN, where one will study recombination of *trapped* positrons and *trapped* antiprotons in Penning traps[5]. In this setup we studied recombination starting out with trapped electrons and free ions. We have used SIMION[8] to calculate the shape of the potential well, created by a specific set of electrodes, in order to design a plate configuration which produces as much as possible a harmonic potential in order to mimic the conditions of the ATRAP setup at CERN.

In these experiments we filled an electron trap by photo-ionizing gas-phase lithium atoms. The lithium atoms were ionized by means of two synchronized dye lasers pumped by the second harmonic of a Nd:YAG laser, which produced laser pulses of about 7 ns duration with a repetition rate of 10 Hz. By connecting a fast pulse generator on one of the electrodes the voltage could be switched. This enabled us to empty the trap at any time and produce electron pulses of 3.8 ns duration. Care was taken for correct impedance matching of the pulsegenerator to the trap electrode. We could fill the trap with about $7.0(\pm 2)\text{x}10^5$ electrons and the recorded lifetime was about 1.5 ms depending on the background pressure (10^{-6} Torr).

At 20.0 mm from the Penning trap plate configuration two parallel capacitor plates where situated separated by 5.0 mm. A hole of 10.0 mm was drilled through these parallel capacitor plates covered by a grid to minimize distortions on the electric field (3.0 V/cm) inbetween the capacitor plates. A second atomic beam of lithium atoms was directed inbetween these capacitor plates, by means of a resistively heated oven. Lithium ions were produced by using ionizing laser light. Typically we would produce a cylindrically shaped ion cloud containing $\sim 5\text{x}10^2$ ions, with a diameter of 2 mm and a length of 10 mm. The last plate was connected to a fast pulse generator (SRS DG535) which could ramp the voltage down from -1.5 V to any voltage in between -1.5 V and + 1.0 V within 0.8 ns. Again care was taken to match the impedance of the parallel capacitor plate to the output of the fast pulse generator. The whole plate configuration was placed in the heart of 2 coils (outer diameter 12.5 cm) in the Helmholtz configuration producing a tunable magnetic field of 0.0-64.0 mT. The plate configuration is depicted in Fig. 2b.

In all the experiments a small static electric field is present in this region of 200 mV/cm

to ensure that any unrecombined free electron is accelerated out of this region and to prevent the effect of "trapping" the electrons in the attractive potential of the ion cloud. For the volumes and densities used in the experiments, the electrons feel an attractive field of <10 mV/cm at the edge of these volumes. Such a low density plasma is therefore not stable when the ion region is biased with an electric field of 200 mV/cm. The experiments were performed in a vacuum chamber with a background pressure of $5\text{x}10^{-7}$ Torr.

4. Discussion I: lithium and rubidium

Timing is essential in a PFR experiment. This can be seen in Fig. 1d: if we turnoff the static electric field before the electrons reach the ions halfway between the parallel capacitor plates, the electrons will hardly be slowed down and will pass the ions without being recombined. If we turnoff the static electric field too late the electrons will turn around at the ions but will gain kinetic energy again due to the static electric field and will leave the ion region in the opposite direction without being recombined. If we turnoff the static electric field at the moment the electrons turn and overlap with the ions, we record a maximum recombination efficiency of 2.0 %. From Fig. 1d we also retrieve the time profile from the electrons pulses since we record a Guassian profile with a width of 3.8 ns (solid line) which means that we have created electron pulses of 3.8 ns. In this case the electron pulses are produced by pulsing open the electron trap in 0.8 ns and therefore one can expect that electron pulses are created of 0.8 ns. The fact that the pulse duration is 6 times longer is mainly due to Coulomb repulsion which results in an enlargement of the electron cloud. The enlargement of the electron cloud is mainly restricted in the z-direction since the presence of the magnetic field prevents the cloud from blowing up in directions perpendicular to the z-axis. This, in turn, will result in different travelling times towards the ions and therefore a broadening of the time duration of the electron pulses. Note that this time duration of the electron pulses is much shorter than in the previous experiments with Rb where the (photo-)electron pulse duration was limited by the duration of the laser pulses ($\sim$ 10 ns) and that PFR works for both heavy (Rb) ions as for light (Li) ions.

A free electron is most likely to get recombined with the PFR scheme when it is close to the nucleus with zero kinetic energy, but the coordinates (for a certain fixed distance to the ion) of the electron relative to the ion essentially do not play a role. This means that it is not appropriate to characterize PFR with a cross section as it is with most other recombination processes. Therefore we introduced[3] an interaction volume (V_{int}) to describe the observed efficiency, which is the volume in space around the ion in which the electron has to be with a certain velocity to be recombined. A simple formula then predicts what this V_{int} should be:

$$N_{rec} = \rho_e \rho_{ion} V_{overl} V_{int} \tag{1}$$

where N_{rec} is the number of recorded atoms ($10\pm(3)$), ρ_e is the electron density in the electron pulses, ρ_{ion} is the ion density ($6\pm(2)\text{x}10^4$ cm^{-3}), and V_{overl} is the overlap volume of the electron cloud and the ion cloud. The size of V_{overl} is determined by the smallest cloud: in our case the electron cloud. We thus obtain a V_{int} on the order of 10^{-10} cm^{-3}. Such an interaction volume would correspond to a Rydberg state around $n\approx$ 180. We

therefore conclude that high Rydberg states are produced around $n\approx$ 180, by means of the PFR scheme.

To verify this conclusion we used the technique of Selective Field Ionization (SFI) [9], to probe the bound state distribution after the recombination event. Every bound state in an atomic or molecular system has a static electric field associated with it by which it will be ionized. A highly excited state will be ionized by relative small electric field compared to a more deeply bound state. The relation between the value of the electric field at which field ionization classically occurs and the principal quantum number n is (in atomic units):

$$F = (\frac{E}{2})^2 = \frac{1}{16n^4} \tag{2}$$

If one ramps the static electric field in time and monitors at which time ionization occurs one can deduce the bound state distribution of an atomic or molecular system. Typically we ramp the electric field with a slew rate of 1.70 V/cm per μs, from 0.20 V/cm to 5.0 V/cm. In Fig. 3 a SFI trace is depicted of the recombined states when during the recombination event the electric field in the ion region is switched from 3.0 V/cm to 0.20 V/cm in 0.8 ns (solid triangles). The SFI detection started 1.4 μs after the recombination event. Clearly it is seen that we mainly produce states which are ionized by the lowest SFI fields. If we use equation(2) to determine which bound states we have produced we indeed observe that we are making Rydberg states with a principal quantum number around $n\approx$ 180, in perfect agreement with the principal quantum number we estimated from our observed efficiency and the V_{int} deduced from it.

A numerical estimate of V_{int} is obtained by solving Newton's equations, with a force equal to: $\mathbf{F}(t) = q(\mathbf{E}(t) + \mathbf{v}(t) \times \mathbf{B}/c)$. The electric field ($\mathbf{E}$) is the superposition of the Coulomb potential of the ions and external field, $\mathbf{v}$ is the velocity of the electrons and $\mathbf{B}$ is the magnetic field. An interaction volume of $0.25\text{x}10^{-10}\text{cm}^3$ is found. Using the obtained state distribution obtained from solving Newton's equations ($< n >= 180$ with $\Delta n = \sqrt{< n^2 > - < n >^2} = 20$) the SFI trace is calculated (using the Landau-Zener approximation[10]) and a very good agreement between theory and experiment is observed (Fig 3, thin line).

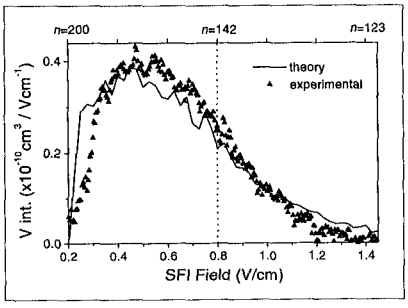

Fig. 3

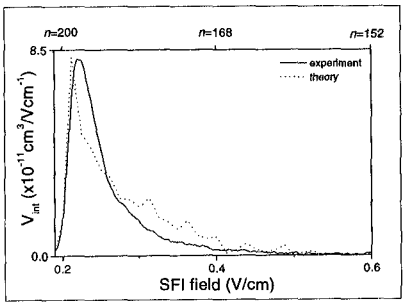

Fig. 4

Fig. 3: Calculated state distribution (thin line) of the recombined Rydberg atoms compared with experimental state distribution (solid triangles). The experimentally determined interaction

volume ($(1.0\pm0.7)\text{x}10^{-10}$ cm^3) is varied within its error to optimize the comparison with theory ($V_{int} = 0.3\text{x}10^{-10}$ cm^3).

Fig. 4: Calculated state distribution (dotted line) of the recombined Rydberg clusters, compared with the experimental state distribution (solid line). The experimentally determined interaction volume is varied within its uncertainty to optimize the comparison with theory.

5. Discussion II: carbon clusters

The efficiency we obtained when we recombined ionic carbon clusters with free electrons with the PFR scheme was as high as $6(\pm4)\text{x}10^{-2}$[4]. Typically we would detect about 15($\pm$5) recombined molecules. The fact that in this experiment the highest efficiency was recorded, is attributed to an optimal overlap volume of the electronic and ionic clouds, and the inertia of the large molecular ions (a C_{60}^+ ion is 720 times heavier than a proton). From the achieved efficiency we again conclude with the help of equation(1) that we are producing high Rydberg states around $n\approx 200$.

In Fig. 4, the Rydberg state distribution after recombination with PFR is depicted for recombination of the carbon clusters (C_{50} - C_{60}), showing that the state distribution peaks for small SFI fields. Since the 1 ns turnoff reduces the electric field to 0.2 V/cm, states with a principal quantum number beyond $n = 200$ are not stable. From Fig. 4, it is deduced that Rydberg states are produced around $n = 190$. The calculation depicted in Fig. 4 shows a direct comparison between theory and experiment, and clearly good agreement is obtained. To obtain this level of agreement, the polarizability of the ionic carbon clusters and autoionization of the recombined carbon clusters is included. The polarizability is accounted for through a local potential $-\alpha/2r^4$ at large distances; $\alpha \sim 540$ atomic units. Autoionization of the Rydberg states by interactions of the electron with the hot C_{60}^+, is included through a model that allows the electron energy to change: every time the recombined electron gets within 5 atomic units of the center, it gets a kick in a random direction so that the energy of the electron is just as likely to increase as to decrease. If the kick increases the energy, the electron leaves (autoionization). If the kick decreases the energy, the electron will be in a lower Rydberg state. For this deeper bound state the time to return to the C_{60}^+ decreases, so the electron will rapidly get kicked again. This is like a random walk where the time between steps decreases in one direction and the time between steps increases in the other direction. The resulting V_{int} from this calculation is $0.7\text{x}10^{-11}$ cm^3, in good agreement with the experimental observation: $(1.3\pm1.2)\text{x}10^{-11}$ cm^3. From the calculation it follows that $\sim$ 75% of the originally recombined electrons are ejected before the SFI detection.

6. Discussion III: implications for anti-hydrogen

In Ref.[11] a combined trap is described, where typically $>10^5$ protons and 10^6 electrons (corresponding density: 10^8 cm^{-3}) in the combined trap were detected. Implementing PFR in such a configuration and if the overlap volume is maximal (the anti-proton plasma takes up the same volume as the positron plasma), the positron density is $10^7 - 10^8$ cm^{-3}, and the number of trapped anti-protons is 10^5, our scheme can produce 10^2-10^3 recombined anti-hydrogen atoms in a single experiment (assuming the same V_{int} as is observed in our

measurements). The exact implementation of PFR for the experimental realization of atomic anti-hydrogen production is currently investigated.

7. Conclusion

In conclusion, a detailed study of the Pulsed Field Recombination scheme is presented in which we have shown that PFR is an universal method to recombine a free electron with any sort of ion. Therefore we propose that PFR can be used in the pursuit of the production of cold anti-hydrogen.

Moreover, we have shown that PFR can be used to create a Rydberg state in a large carbon cluster such as C_{60}. The recombination dynamics for these large molecules was shown to be the same, but the survival- and SFI detection dynamics differ. A simple model, including autoionization is presented to understand the survival dynamics. We propose that PFR can be used for cation spectroscopy.

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