Towards laser spectroscopy of antihydrogen

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The development of the first continuous coherent source at 121.56 nm is described. Radiation at this wavelength of Lyman-α can be used for laser-cooling of antihydrogen on the strong 1S–2P transition. It also opens up a possibility for precision spectroscopy that requires just a few antihydrogen atoms.

1. Introduction

The theory of the hydrogen atom and experiments keep challenging each other at ever increasing levels of precision [1]. That interplay stimulated the development of ultrahigh precision laser spectroscopy of the hydrogen atom which has recently been employed to measure fundamental constants, to establish stringent tests of quantum electrodynamics and even to investigate hadronic structure [2–4]. It would be fascinating to use these advanced tools for the investigation of antimatter. High resolution laser spectroscopy of antihydrogen could then open a new field for precise tests of the fundamental CPT symmetry [5]. Furthermore, laser cooling and laser spectroscopy techniques are essential for a possible measurement [6] of the gravitational force on antihydrogen [7]. Two collaborations have formed with the goal of precision measurements on magnetically trapped antihydrogen [8] using the new Antiproton Decelerator (AD) at CERN [9].

Recently, a few *fast* antihydrogen atoms have been produced [10,11]. This has given prospect to a new field of experiments with antihydrogen atoms. Simultaneous trapping of positrons and antiprotons for antihydrogen production has recently been demonstrated [12]. Nevertheless, the production of *cold* antihydrogen atoms still presents a considerable challenge.

The rate for spontaneous radiative recombination of antiprotons and positrons is rather low because the emission of photons is a slow process on the time scale of collisions. Laser-stimulated recombination can increase the antihydrogen formation rate by orders of magnitude [13]. The spectrum of the recombination rate vs. stimulating wavelength is expected to reflect the energy distribution of positrons and the population of high-lying levels. Therefore, the spectral resolution for the first step of laser-stimulated recombination will be rather limited. Laser-induced two-step recombination, first into a high-lying state with the subsequent stimulation of a bound–bound transition into a lower lying state, offers a first possibility for precise laser spectroscopy [14,15]. This has the advantage that it does not require magnetic trapping of antihydrogen.

Further experiments will most likely make use of magnetically trapped antihydrogen atoms [16]. The narrow 1S–2S transition is an especially intriguing candidate for ultimate precision experiments. The excitation rate for this two-photon transition, however, is typically rather small. Previous experiments on ordinary hydrogen compensated that by the use of many atoms (10^{15}–10^{17} atoms/s in a beam [17,18], 10^{10}–10^{13} atoms in a trap [19]). Given that the AD will deliver about 10^7 antiprotons/min at 100 MeV/c [9], it is clear that new techniques need to be developed for experiments with antihydrogen.

The strongest transition of the antihydrogen atom, $1S \rightarrow 2P$, is situated in the vacuum ultraviolet (VUV) spectral region at $\lambda = 121.6$ nm (Lyman-α). Narrowband continuous radiation at this wavelength is important for efficient laser cooling of antihydrogen atoms in a magnetic trap. Further, continuous radiation opens a possibility for ultrahigh resolution spectroscopy of the weak 1S–2S transition employing a "shelving" scheme that requires just a few antihydrogen atoms.

2. Generation of continuous wave coherent Lyman-α radiation

Producing coherent radiation at Lyman-α is a technological challenge as there are no tunable lasers and nonlinear crystals available for that spectral region. Sum-frequency generation of several incident laser beams utilizing the nonlinear susceptibility of atomic vapors and gases is commonly used to produce coherent radiation in the VUV. Four-wave mixing (FWM) can produce the sum-frequency of three fundamental colors. Many *pulsed* Lyman-α sources based on FWM with high-power lasers have been built over the years [20–24]. Several FWM schemes have been described in the literature which demonstrate the feasibility to produce *continuous* radiation in the VUV (down to 133 nm, where 11 pW has been obtained) [25–28].

The yield of near-resonant four-wave mixing scales as

$$P_1 \cdot P_2 \cdot P_3 \cdot \left[\text{density} \cdot \frac{\text{dipole-moments}}{\text{resonances}} \cdot \text{phasematch-integral}\right],$$

where P_1, P_2, and P_3 denote the powers of the fundamental beams [29]. For FWM with continuous wave beams the incident power levels are rather low. Tight focusing is, therefore, used to obtain high intensities. Especially important for continuous FWM is the resonant enhancement of the nonlinear susceptibility. An exact two-photon resonance is essential. Near resonances at one and three photon heights are chosen to give maximum enhancement without too much absorption. It is necessary to select a medium with strong dipole moments on transitions with convenient wavelengths so that powerful fundamental laser beams are possible. The requirements for efficient continuous wave FWM are rather different from pulsed FWM. Tight focusing and resonances are generally avoided for pulsed FWM with high-power lasers as they would lead to rapid saturation due to multi-photon ionization.

We proposed to produce coherent continuous radiation at Lyman-α in magnesium vapor using the four-wave mixing scheme shown in figure 1(a) [30]. Radiation

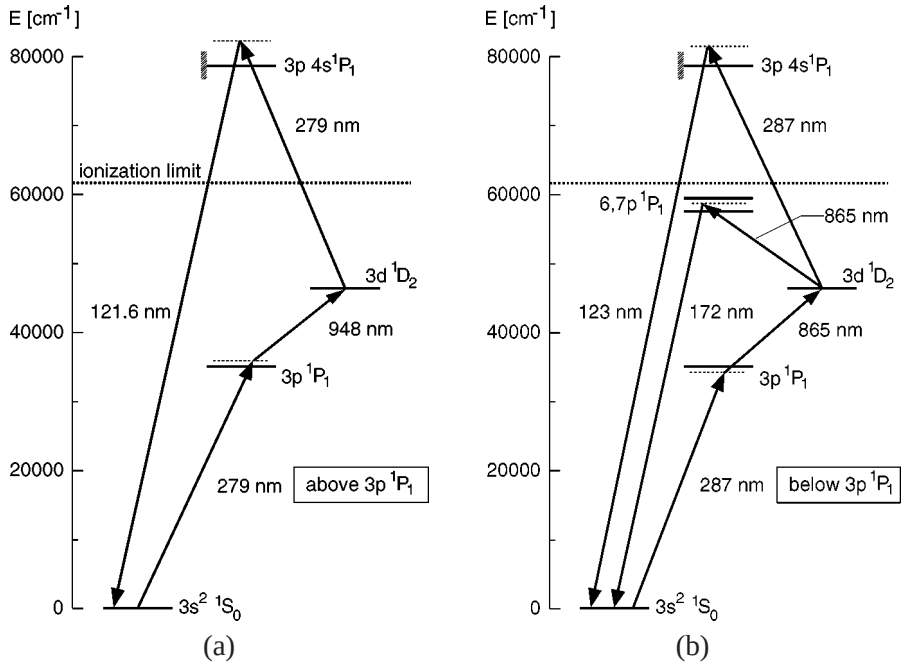

Figure 1. Four-wave mixing in magnesium vapor. (a) the original plan for generation of continuous wave Lyman-α radiation; (b) realized schemes to produce continuous radiation at 123 nm and at 172 nm.

at 279 nm, close to the first resonance transition at 285.2 nm, is obtained from a frequency-doubled dye laser. Together with the radiation at 948 nm from a titanium–sapphire laser, the two-photon resonance with the $3d^1D_2$ level is established. The third incoming light field is identical to the first one (279 nm). The energy of the resulting Lyman-α radiation corresponds to the short wavelength side of the $3p4s^1P_1$ autoionizing state which gives some additional enhancement as well. Since the fundamental ultraviolet beam at 279 nm is tuned to the *short* wavelength side of the $3s^2\,{}^1S_0$–$3s3p^1P_1$ transition, the phase mismatch is positive. This phase mismatch can in principle be compensated by mixing the magnesium vapor with negative disperse krypton gas. This technique is commonly used to establish phase matching for FWM at Lyman-α with pulsed lasers.

It turns out that this does not work in practice for continuous FWM. The additional krypton gas causes pressure broadening of the two-photon resonance with the $3d^1D_2$ level. The continuous FWM scheme depends heavily on the resonant enhancement of the nonlinear susceptibility. Additional pressure broadening spoils the resonant two-photon enhancement and thus caused a very low yield which we were unable to detect.

A successful scheme for continuous FWM in magnesium close to Lyman-α is shown in figure 1(b). It differs from the original plan mainly in that the fundamental ultraviolet wavelength is now at 287 nm which is on the *long* wavelength side of the $3s^2\,{}^1S_0$–$3s3p^1P_1$ transition. The wavelength of the titanium–sapphire laser is now at 865 nm in order to establish the exact two-photon resonance with the $3d^1D_2$ level.

Since the third light field is again identical to the first one, the sum frequency is no longer at Lyman-α (121.6 nm) but at 123 nm. A crossed heat-pipe [31] was used to obtain a magnesium vapor length of 6 cm at pressures of 0.5–1 torr. A buffer gas, 60 torr of argon, prevented coating of the windows with magnesium. The laser beams of 8 mm diameter were focused with a 20 cm lens. Using fundamental powers of 100 mW at 287 nm and 900 mW at 865 nm we observed a signal of 10 counts/s for 123 nm at a background of 5 counts/s. The detection system consisted of a dispersion monochromator (20% efficiency), three narrow-band Lyman-α interference filters (17% each), and a solar-blind photomultiplier (15%). Taking the estimated detection efficiency into account, we conclude that $7 \cdot 10^4$ photons/s (0.1 pW) were produced at 123 nm. Using yet another frequency-doubled dye-laser it would thus be possible to produce continuous coherent radiation at exactly Lyman-α by FWM (287, 865 and 279 nm) in magnesium vapor.

A second FWM scheme in magnesium is also shown at the right side of figure 1. Using the same fundamental beams it produces radiation at 172 nm (287 nm and two times 865 nm). Radiation at 172 nm was detected employing a grating VUV monochromator and a photomultiplier. That second FWM process proved to be far more efficient than the FWM process giving 123 nm. More than 10^9 photons/s (1 nW) were produced at 172 nm. Bound states ($3s6p\,^1P_1$ and $3s7p\,^1P_1$) enhance the 172 nm FWM scheme at the three-photon height. This is much more efficient than the enhancement of 123 nm FWM by the broad $3p4s\,^1P_1$ autoionizing resonance.

Encouraged by the high yield at 172 nm we searched for another continuous FWM scheme to produce radiation at Lyman-α. Enhancement by bound states at the three-photon height requires that the candidate atom has a rather high ionization limit. A promising FWM scheme employing mercury vapor is shown in figure 2. Radiation at 257 nm, obtained from a frequency-doubled single-mode Ar^+-laser, is on the long wavelength side of the first resonance transition $6s\,^1S_0 \rightarrow 6p\,^3P_1$ in mercury at 253.7 nm. Radiation at 399 nm, obtained from a frequency-doubled titanium–sapphire laser, establishes an exact two-photon resonance with the $7\,^1S_0$ state. The wavelength of a third fundamental light field (545 nm from a dye-laser) is chosen such that the sum-frequency is at Lyman-α. Bound resonances such as the $11p\,^1P_1$ and $12p\,^1P_1$ contribute significantly to the non-linear susceptibility [32]. (As a point of historic interest we note that the lines $6\,^1S_0 \rightarrow 11\,^3P_1$, $11\,^1P_1$, $12\,^3P_1$, and $12\,^1P_1$ of ^{198}Hg have been used as wavelength standards in an early optical measurement of the Lamb shift in deuterium [33].)

The FWM scheme of figure 2 has been realized and indeed gives a very encouraging yield at Lyman-α. Results and a detailed description of the setup have been described elsewhere [34]. Using 180 mW at 257 nm, 300 mW at 399 nm, and 1.1 W at 545 nm, we now obtain a VUV yield of up to 1 nW ($6 \cdot 10^8$ photons/s) at Lyman-α.

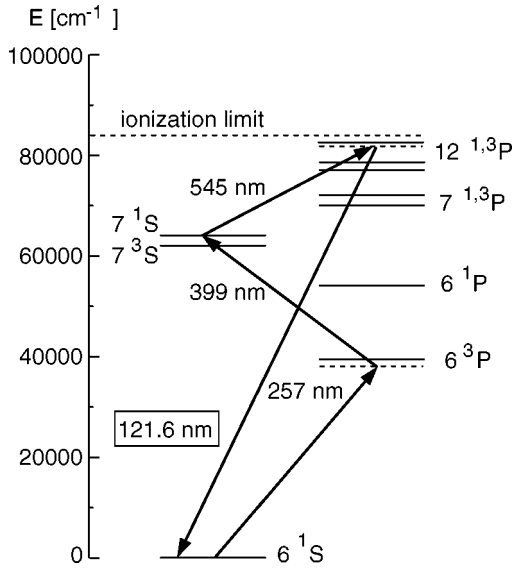

Figure 2. Four-wave mixing scheme to produce radiation at Lyman-α in mercury vapor.

3. Laser cooling and shelving spectroscopy

Let us now turn to possible applications of the continuous source for radiation at Lyman-α. Laser cooling of antihydrogen with pulsed Lyman-α radiation has been discussed by some authors [23,35,36]. A continuous source has clearly significant advantages over pulsed sources. Typical pulsed sources for radiation at Lyman-α have pulse lengths of nanoseconds. The lifetime of the $2\,\mathrm{P}$ states is 1.6 ns. Hence, sources with nanosecond pulses cause at most a few excitations per pulse. Laser cooling is effectively limited by the pulse repetition rate. Therefore, a continuous source can provide a larger rate for laser cooling. Furthermore, the spectral bandwidth of a continuous source can be much lower. This provides higher selectivity for magnetic substates of atoms in a trap thereby reducing losses due to spurious optical pumping to untrapped magnetic sublevels.

The resonant absorption cross section for radiation at Lyman-α can be as high as $3\lambda_\alpha^2/2\pi$ [37]. Consider a volume of 1 mm diameter being illuminated with 1 nW radiation at Lyman-α. The resonant excitation rate for an atom is then $5\ \mathrm{s}^{-1}$. Suppose that we would like to cool antihydrogen atoms in a magnetic trap starting with an initial temperature of 1 K which corresponds to an average velocity of 150 $\mathrm{m/s}$. The average velocity change per excitation is 3.3 $\mathrm{m/s}$. Cooling could thus be done in about 10 s with only 1 nW radiation at Lyman-α available from our source at its early stage.

A continuous coherent Lyman-α source opens also a possibility for high-resolution spectroscopy with only a few antihydrogen atoms [38]. Consider the excitation scheme shown in figure 3. Radiation at Lyman-α excites a single atom in a magnetic trap from the $1\,^2\mathrm{S}_{1/2}$ ground state to the $2\,^2\mathrm{P}_{3/2}$ excited state. The $2\,^2\mathrm{P}_{3/2}$ has a very short natural lifetime of 1.6 ns. Thus intense resonance fluorescence can

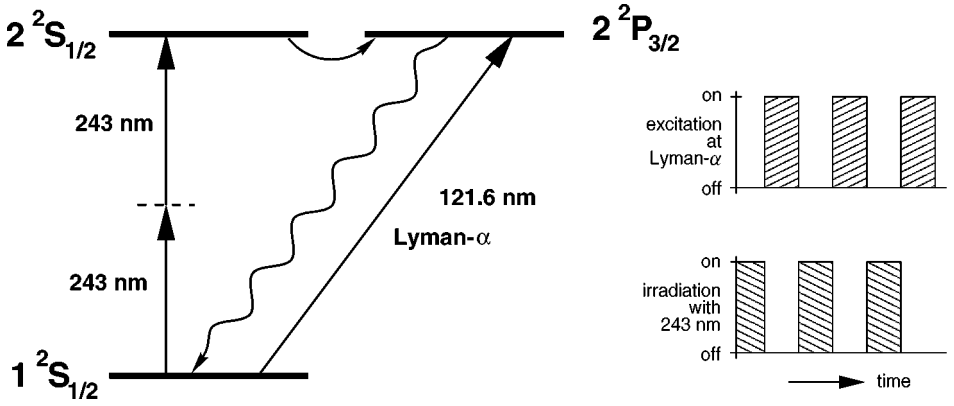

Figure 3. Excitation scheme for shelving spectroscopy of antihydrogen.

be emitted by the decay of the $2\,^2P_{3/2}$ state back into the ground state. Excitation on the strong Lyman-α transition is alternated with irradiation of the atom by ultraviolet light at 243 nm. Simultaneous Doppler-free absorption of two photons leaves the atom in the $2\,^2S_{1/2}$ state. This state is metastable and decays by two-photon emission to the ground state with a natural lifetime of 122 ms. Observation of resonance fluorescence at Lyman-α in the following cycle is used to determine whether the weak two-photon excitation into the metastable state was successful or not. The absence of resonance fluorescence indicates that the atom has been excited ("shelved") into the metastable state. In that case the atom can be "reset" into the ground state by applying a microwave field to couple the long-lived $2\,^2S_{1/2}$ state with the rapidly decaying $2\,^2P_{3/2}$ state. The fraction of cycles with no resonance scattering represents the probability of the two-photon excitation. An absorption spectrum is obtained by stepwise scanning the 243 nm laser frequency while measuring the probability of excitation.

Shelving spectroscopy thus involves many decisions whether the antihydrogen atom has been excited to the metastable $2\,^2S_{1/2}$ state or not. These decisions have to be made somewhat quicker than the natural lifetime of the metastable state and are based on the observation or the non-observation of fluorescent light at Lyman-α. The detection efficiency for fluorescent light from an antihydrogen sample in a magnetic trap with superconducting coils is probably rather low. Shelving spectroscopy requires thus far more power at Lyman-α than laser cooling.

To conclude, the photon flux from our source for continuous Lyman-α radiation at its early stage is promising for laser cooling of antihydrogen in a magnetic trap. It is expected that the yield can be increased by several orders of magnitude. It would also be very interesting to replace one of the continuous beams in the FWM scheme by a pulse-amplified beam with a long duration, say μs to ms. Combining the advantages of narrow bandwidth and high intensity, such a hybrid Lyman-α source could be ideal for laser cooling, Zeeman slowing, and shelving spectroscopy of antihydrogen.

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