Two-photon spectroscopy of trapped atomic hydrogen

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Abstract

We report observation of the 1S–2S two-photon transition in hydrogen confined in a magnetic trap at sub-millikelvin temperatures. The excitation spectrum can display a sharp central feature arising from periodic motion of the trapped atoms. The metastable 2S atoms remain trapped, and have a lifetime close to the natural lifetime of 122 ms. These developments open the way to achieving the transition's natural linewidth. We have also demonstrated that the temperature of the gas can be determined from the line shape, providing an important tool for the study of cold trapped hydrogen. The resolution in these experiments appears to be limited by laser stability, currently 3 kHz.

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The 1S–2S two-photon transition in hydrogen plays a unique role in atomic studies because of its small natural linewidth (1.3 Hz), its sensitivity to the Lamb shift, and its potential for optical frequency metrology. First observed with the advent of two-photon Doppler-free laser spectroscopy two decades ago [1], the experimental linewidth has been reduced over the years from a few hundred MHz to 3 kHz [2–4]. This 10^5 increase in resolution, accompanied by other advances in hydrogen spectroscopy, has yielded the most precise determination yet of the Lamb shift in hydrogen, new information on the proton radius, and a new value for the Rydberg constant [5,6]. Nevertheless, another factor of 10^3 stands between today's state of the art and the intrinsic resolution of the transition.

Current limits to resolution of the 1S–2S transition are the second order Doppler shift and the finite interaction time of the atoms moving through the radiation field (called time of flight broadening). We have performed two-photon spectroscopy of hydrogen confined in a magnetic trap at sub-millikelvin temperatures. At these low temperatures the second order Doppler effect is negligible and periodic motion of the atoms produces features which are narrower than the conventional time of flight profile. This opens a new regime for hydrogen spectroscopy in which the resolution should approach the natural linewidth. Currently our measurements are limited by the stability of our laser system (about 3 kHz on a 20 second time scale), but we believe that the intrinsic linewidth in our trap is much smaller.

Our method employs techniques we previously developed for confining a gas of atomic hydrogen in a magnetic trap and cooling it by evaporation to temperatures of 100 μK or below [7]. In that work we measured the energy distribution by monitoring the flux of atoms escaping from the trap as the confining field was rapidly lowered to zero [8]. However, by employing optical excitation we are now able to study the gas *in situ*.

Atoms are confined in the "low-field seeking" $F = 1$, $m_F = 1$ hyperfine state for which the energy $V(\mathbf{r})$ increases with magnetic field $|\mathbf{B}(\mathbf{r})|$. The trap is in the Ioffe-Pritchard configuration [9] as adapted by Hess [10]. Four elongated coils produce a quadrupole field in the plane perpendicular to the z-axis, and a pair of short solenoids creates a magnetic barrier along the z-axis at either end of the trap, thus forming a magnetic minimum at the center. In order to inhibit non-adiabatic spin flips, another coil provides a uniform bias field along the z-axis. Using evaporative cooling [11], we produce samples of 10^{13} – 10^{10} atoms at temperatures from 25 mK – 100 μK with atomic densities up to 6×10^{13} cm^{-3}.

The magnetic field strength at a small distance ρ from the axis of the trap is $B = \sqrt{B_\rho^2 + B_{z,o}^2}$. The quadrupole field strength is given by $B_\rho = \rho B_w / a$, where the cell radius is $a = 22$ mm, the field at the cell wall is B_w (maximum 0.91 tesla), and the axial bias field is $B_{z,o} \approx 2 \times 10^{-4}$ tesla. The magnetic potential energy of an atom is $V(\mathbf{r}) \approx \mu_B |\mathbf{B}(\mathbf{r})|$. The density varies as $\exp(-V(\mathbf{r})/k_B T)$. This establishes a characteristic thermal radius given by $\rho_{th} = 2ak_B T/\mu_B B_w$ for the cylindrically shaped sample. Depending on the temperature and conditions of confinement, ρ_{th} varies between 40 and 1100 μm.

The 243 nm laser radiation needed to drive the two-photon transition is generated in a system based on a design by Hänsch [3]. A 486 nm dye laser is locked to a reference cavity and its output is frequency doubled in an external ring [12]. In the trap, the 243 nm beam has a waist radius $w_o = 37$ μm and a divergence length of 18 mm. The typical power is 4 mW. The beam passes along the trap's axis and is retro-reflected by a spherical mirror to provide the standing wave required for two-photon Doppler-free excitation (see Fig. 1). The UV radiation is typically applied in a 2 ms pulse. Following each pulse, the excited atoms are quenched by applying an electric field of about 8 V/cm that Stark mixes the 2S and 2P states, causing prompt radiative decay by emission of a L_α photon (122 nm). The photons are detected by a microchannel plate detector mounted at the end of the trap, behind the retro-reflecting mirror. Due to the low collection solid angle (4×10^{-4} ster) and other losses, the detection efficiency is only 4×10^{-6}. Nevertheless, count rates up to 1000 s^{-1} have been observed.

At moderate temperatures we observe the characteristic shape for time of flight broadened two-photon absorption in a Gaussian beam [13], proportional to $\exp(-|\delta|/\delta_o)$. Here, $\delta = \nu - \nu_o$ is the frequency detuning from resonance of the 243 nm radiation. The linewidth parameter is $\delta_o = u/4\pi w_b$, where $u = \sqrt{2k_BT/M}$ is the most probable speed for an atom of mass M and w_b is the radius of the light beam. This behavior can be seen in Fig. 2.

We determine the temperature of the gas by fitting an exponential curve to the observed spectrum (including suitable corrections for variation of the 243 nm beam radius w_b over the finite length of the trap). We have verified this technique by two other methods. The first consists of quickly dumping the trap and measuring the flux of escaping atoms. Comparing the observed energy distribution with theory yields the temperature [8]. The second method is based on a model of the balance between evaporative cooling and dipolar relaxation that was verified in earlier experiments [14]. As shown in Fig. 3, the three methods are in reasonable agreement. At the lowest temperatures, however, the sensitivity is limited by laser linewidth. The linewidth appears to be determined by reference cavity fluctuations and Doppler shifts arising from vibrations, both of which vary with the ambient conditions. Under favorable conditions the linewidth is 3 kHz.

An important feature of two-photon spectroscopy in the trap is that while the atoms interact with the light, they oscillate radially without losing coherence. This is possible since the 243 nm UV pulse (2 ms) is short compared to the mean collision time, but long compared to the vibrational period of the trap. Cold atoms, localized near the axis, sample a harmonic potential with energy level spacing $h\nu_v = h\sqrt{B_w^2\mu_B/MB_{z,0}}/2\pi a$. Typically, $\nu_v \approx 3$ kHz [15,16].

The laser field can induce the 1S–2S electronic transition accompanied by a change in the vibrational state of the atom—essentially a Raman transition. The change of the atom's trap state arises from its motion through the radial intensity profile of the laser beam. If the laser beam is aligned on the trap axis, spatial parity considerations require that the energy changes only by integer multiples of $2h\nu_v$. These changes in the vibrational state generate sidebands in the spectrum. One can also explain the lineshape as arising from a variant of the Ramsey separated oscillatory field method. In our case, the atoms periodically sample the field many times before losing coherence. Thus, one expects the 1S–2S excitation probability to display interference fringes with a separation of twice the vibrational frequency. The overall width of the spectrum is determined by the time of flight linewidth, while the fringe width is determined by the shortest relevant coherence time.

As an atom's total energy increases, it can sample the anharmonic region of the trap. Successive energy levels become closer and the oscillation period increases. The resulting distribution of energy level spacings broadens the outer fringes of the spectrum. However, the anharmonicity does not affect the central component. Figure 4 is a spectrum showing a motionally narrowed central peak. The calculated spectrum accounts for trap anharmonicity, small photoionization effects, and finite laser exposure time; the laser linewidth and sample temperature were adjustable parameters. The best fit to the data was found with a Gaussian linewidth of 3.0 kHz full width at half maximum, and a sample temperature of 150 μK. Trap anharmonicity and laser linewidth obscure the sidebands, but the spacing of the sideband features is reproducible.

To carry out 1S–2S spectroscopy at its natural resolution, it is essential that the 2S atoms live for their theoretically predicted lifetime. The experimental lifetime could be shortened by Stark quenching in stray electric fields, by collisions, or by some other artifact. To investigate the lifetime, we monitored the 2S decay by exciting the atoms and delaying the application of the quenching electric field for varying times. Measurements such as those displayed in Fig. 5 indicate lifetimes between 50 and 110 ms depending on trap conditions. Because the variations were not systematically studied, however, this should not be interpreted as a measurement of the natural lifetime (122 ms [17,18]). Nevertheless, these results give assurance that there is no major extraneous damping process.

One might expect a frequency shift due to the difference in atom–atom interaction energies of the 1S–1S system and the 2S–1S system—the low temperature equivalent of a pressure shift [19]. A recent calculation [20] indicates that this shift should not present a fundamental limit to the spectroscopic accuracy. At a density of 10^{10} cm^{-3} the predicted shift is comparable to the natural linewidth.

Another factor affecting the ultimate spectroscopic accuracy is a residual Zeeman shift. The electron g-factors of the 1S and 2S states differ because of a small relativistic effect [18], so that in a magnetic field B there is a frequency shift $\delta\nu_Z = \alpha^2\, \mu_B B/4h$, where α is the fine structure constant. In the trap, the atoms experience a characteristic magnetic field $B = 2k_BT/\mu_B$, so that at a temperature of 100 μK the shift is about 50 Hz. The shift can be reduced by operating at lower temperature and by restricting attention to the central part of the line (the coldest atoms make the dominant contribution to the center of the line, and they sample only small magnetic fields).

Our trap is operating close to the regime of Bose-Einstein condensation (BEC). The techniques reported here are well suited for studying BEC. In particular, the condensate is predicted to display a narrow line that is recoil shifted by about 7 MHz from the Doppler-free line [12]. Atoms excited from the condensate could be ejected from the trap due to photon recoil, forming a coherent beam. In addition, the spatial distribution of the atoms develops a sharp peak at the trap center. We can measure the density profile by displacing the laser beam relative to the atoms.

The developments described here may also be usefully applied to the creation of an optical frequency standard. At present the most promising such standards probe a forbidden transition in a small number of trapped ions. A trapped ion is close to an ideal system for high resolution spectroscopy. For an optical clock, however, the signal rate is also an important consideration, and the intrinsic signal rate from a trapped ion is one count per radiative lifetime. The spectral resolution of the 1S–2S transition can be comparable to that of such ions, but the signal rate can be many thousands of times higher.

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FIGURES

FIG. 1. Schematic diagram of optical excitation and detection apparatus. The UV beam is aligned along the axis of the magnetic trap. The beam traverses the cloud of trapped atoms (typically 10 cm long, 500 μm diameter), and is retro-reflected. After a UV excitation pulse, the (metastable) 2S atoms are detected by applying an electric field across the quench plates which mixes the 2S state with the 2P state (1.6 ns lifetime). The resulting L_α photons are detected with the microchannel plate.

FIG. 2. 1S–2S excitation spectrum displaying a time of flight profile. The UV detuning (at 243 nm) is δ. The density is 3×10^{12} cm^{-3}, the temperature is 1.7 mK, and the UV power is ≈ 1.5 mW. The total UV exposure time at each point is 2.7 s. Here the dominant source of broadening is the finite interaction time of an atom moving across the UV beam, which leads to an exponential spectrum: $\exp(-|\delta|/\delta_0)$. The solid line corresponds to $\delta_0 = 11$ kHz, which yields a full width at half maximum of 15 kHz.

FIG. 3. Comparison of the temperature inferred from the time of flight linewidth (dots) with predictions from a model of heating and cooling effects in the trap (solid line). The temperature was varied by changing the depth of the trap, and measured by recording a spectrum at each point. The dashed line indicates the temperature at which the typical laser linewidth (3 kHz) dominates the observed linewidth. Inset (same axes): Comparison of the temperature inferred from the linewidth (dots) with the temperature obtained by quickly dumping the trap and measuring the energy distribution (triangles). The scatter of the points is indicative of the error.

FIG. 4. 1S–2S excitation spectrum in the regime of motional narrowing. Each data point represents a total exposure of 350 ms. The upper solid line is the calculated spectrum including the effects of trap anharmonicity, photoionization, and an estimated laser linewidth. The best fit is for a laser linewidth of 3.0 kHz (full width, half maximum) at 243 nm. From the curve the sample temperature is found to be 150 μK. The lower curve is the line shape calculated for a monochromatic light source. Inset: schematic diagram of trap vibarational states on the 1S and 2S electronic manifolds, indicating allowed transitions from the lowest three 1S trap states.

FIG. 5. Determination of the lifetime of 2S atoms in the trap: $T \approx 250$ μK, $n \approx 4 \times 10^{12}$cm^{-3}. The atoms were excited by short UV pulses and quenched after a variable delay. This procedure was repeated 10^4 times. The straight line is the best fit to an exponential decay. The error bars represent statistical fluctuations only. In this particular trap configuration, the lifetime is 110 ms. Lifetimes between 50 and 100 ms have been observed for other trap configurations. A systematic study of the experimental factors that affect the lifetime measurements has not been carried out.