Two-Photon Spectroscopy of Trapped Atomic Hydrogen

Ronit Dutta

Final Year student of Integrated BS-MS in Physics

Indian Institute of Technology Kharagpur

Subject : LASER SPECTROSCOPY

Two-Photon Spectroscopy of Trapped Atomic Hydrogen

Claudio L. Cesar,* Dale G. Fried, Thomas C. Killian, Adam D. Polcyn, Jon C. Sandberg,† Ite A. Yu,‡ Thomas J. Greytak, and Daniel Kleppner

Department of Physics and Center for Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

John M. Doyle

Department of Physics, Harvard University, Cambridge, Massachusetts 02138

(Received 25 March 1996)

We report observation of the 1*S*-2*S* two-photon transition in hydrogen confined in a magnetic trap at submillikelvin temperatures. The excitation spectrum can display a sharp central feature arising from periodic motion of the trapped atoms. The metastable 2*S* 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. [S0031-9007(96)00604-7]

PACS numbers: 39.30.+w, 06.30.Ft, 32.30.Jc, 32.80.Pj

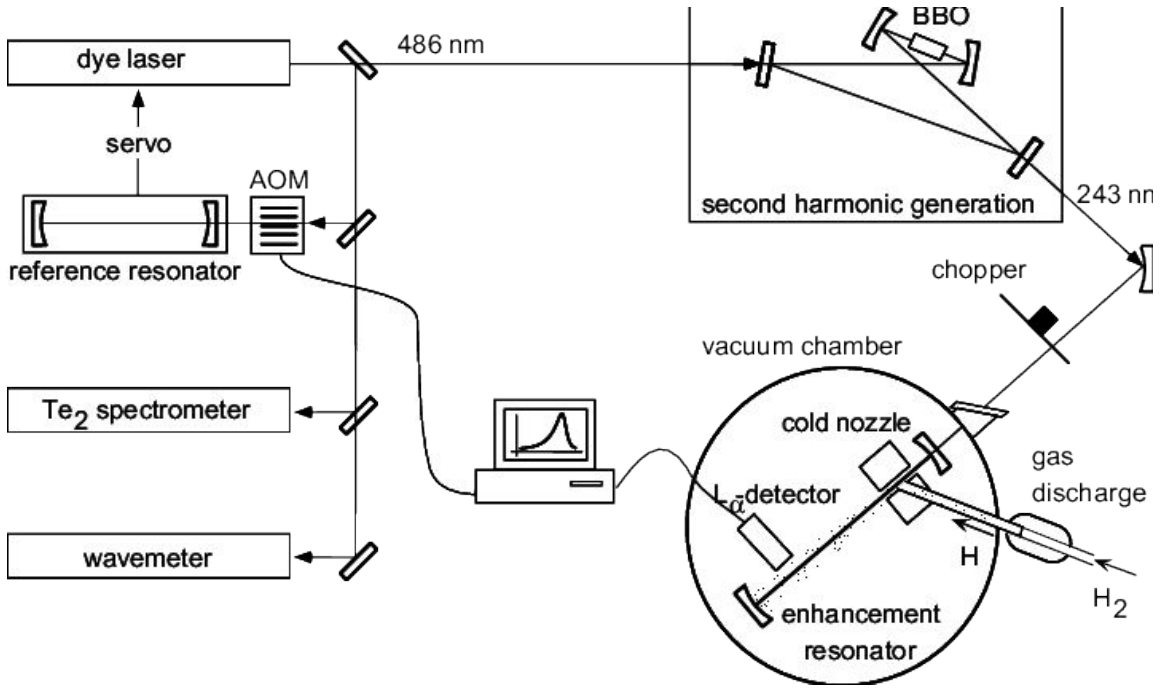
The 1*S*-2*S* two-photon transition in hydrogen plays a unique role in atomic studies because of its small natural linewidth (1.3 Hz), its sensitivity to the Lamb shift, and its potential for optical frequency metrology. First observed with the advent of two-photon Doppler-free laser spectroscopy two 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 combination of Lamb shift and proton size correction in hydrogen, 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 1*S*-2*S* 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 atomic hydrogen at submillikelvin temperatures confined in a magnetic trap. At these low temperatures the second order Doppler effect is negligible and periodic motion of the atoms produces 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 nonadiabatic 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 to 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 $B = \sqrt{B_\rho^2 + B_{z,0}^2}$. 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 T), and the axial bias field is $B_{z,0} \approx 2 \times 10^{-4}$ T. The magnetic potential energy of an atom is $V(\mathbf{r}) = \ldots |\mathbf{B}(\mathbf{r})|$. The density varies

Objectives



1s-2s two-photon spectroscopy setup of atomic Hydrogen at submillikelvin temperature confined in a magnetic trap

1. Observe 1S–2S two-photon transition in magnetically trapped hydrogen at sub millikelvin temperature.
2. Achieve high spectral resolution by reducing Doppler and Time-Of-Flight (Transit-Time) broadening.
3. Analyze natural line shape under ultracold, confined conditions.
4. Measure metastable 2S state lifetime in the trap.
5. Enable BEC studies and optical frequency standard development.

Significance of Precision Spectroscopy

1. **Ultra-narrow natural linewidth**: The 1s-2s transition of the hydrogen atom is 1.3 Hz enabling tests of quantum electrodynamics (QED), proton size, and an ultra-accurate estimation of the Rydberg constant.
2. **Doppler-free nature**: This is achieved via counter-propagating photons in a standing wave, which effectively cancels first-order Doppler shifts.
3. **Sensitivity to fundamental constants**: Shifts in the transition frequency reveal QED corrections (e.g., Lamb shift) and proton charge radius.

Challenges in Achieving Natural Linewidth

1. **Second-Order Doppler Broadening**: High precision spectroscopy measurements, and light atoms second order doppler effect is significant. It scales with $(v/c)^2$; significant at thermal velocities in non-cooled samples.
2. **Time-of-Flight (TOF) Broadening (aka Transit-time broadening)**: Limited interaction time with the radiation field as atoms quickly traverse the laser beam. For plain cosine oscillator amplitude we get a simple sinc squared profile while for a gaussian beam one would get a gaussian linewidth.
3. **Laser Instability**: Reference cavity fluctuations and mechanical vibrations add linewidths ≥3 kHz

1s-2s two-photon transition

Two-photon transitions require absorption of two photons with total angular momentum change $\Delta L = 0$. This however is not a part of Lyman series. For hydrogen's 1S (n = 1) to 2S (n = 2) transition, the energy is given by :

$$\text{Energy} = \frac{3}{4} R_H (1 + \frac{m_e}{m_p})^{-1} + \text{Lamb shift corrections}$$

Here R_H = Rydberg constant.

In Doppler-free spectroscopy, using counter-propagating laser beams to form a standing wave cancels the first-order Doppler shift:

$$\Delta v_{\text{Doppler}} = \frac{\vec{k_1}\vec{v} - \vec{k_2}\vec{v}}{2\pi} = 0 \ (\text{if } \vec{k_1} = -\vec{k_2})$$

Residual broadening arises from second-order Doppler effects $(\propto v^2/c^2)$ and finite interaction time with the radiation field.

The **transition rate** is proportional to the intensity squared, enabling narrow resonances.

Confinement in a Magnetic Trap

Atoms in the F = 1, $m_F = 1$hyperfine state experience a trapping potential given by

$$V(r) = \mu_B |B(r)|$$

where μ_B is the Bohr magneton, and B(r) is the magnetic field.

In thermal equilibrium, the density follows a Boltzmann distribution which is given by

$$n(r) \propto \exp\left(-\frac{V(r)}{k_B T}\right)$$

This defines a thermal radius which is given by :

$$\rho_{thermal} = \frac{2ak_B T}{\mu_B B_w}$$

where a = 22mm is the trap cell radius, and B_w is the quadrupole field strength.

Line Broadening Mechanisms

Coming back to the several broadening mechanisms we have :

1. **Second-order Doppler shift :**

$$\frac{\Delta v}{v_0} = -\frac{\langle v^2 \rangle}{2c^2}$$

where $\langle v^2 \rangle = \frac{3k_B T}{M}$. At T = 100K, $\Delta v \approx 10^{-18} v_0$, which is negligible

2. **Time-of-flight (TOF) broadening:**

Atoms moving at most probable velocity $u = \sqrt{\frac{2k_b T}{M}}$ cross the laser beam waist w_0 limiting the interaction time $t_{int} \propto \frac{w_0}{u}$. This introduces a Lorentzian linewidth.

$$\text{Linewidth parameter } (\delta_0) = \frac{u}{4\pi w_0} \propto \sqrt{T}$$

3. **Trap-induced motional narrowing**:

Atoms oscillate harmonially inside the trap with a frequency of $\nu_v = \frac{1}{2\pi a}\sqrt{\frac{B_w^2 \mu_B}{M B_{z,0}}}$

The reference paper notes that the trapped atoms, while oscillating, produce interference (motional narrowing) effects. These features have linewidths that are determined not by the typical transit-time but by the intrinsic properties of the trap and the periodic sampling of the excitation field. This results in a central feature that is significantly narrower than if TOF effects were dominant. Thus, coherent interactions over multiple oscillations reduce effective linewidth.

Line Broadening Mechanisms

At submillikelvin temperatures ($T \sim 100\,\mu\text{K}$), the thermal velocity v of hydrogen atoms is extremely small. For $T = 100\,\mu\text{K}$:

$$v = \sqrt{\frac{2k_B T}{M}} \approx \sqrt{\frac{2 \times 1.38 \times 10^{-23}\,\text{J/K} \times 100 \times 10^{-6}\,\text{K}}{1.67 \times 10^{-27}\,\text{kg}}} \approx 3.1\,\text{m/s}.$$

Substituting into the second-order Doppler term:

$$\frac{v^2}{c^2} \approx \frac{(3.1)^2}{(3 \times 10^8)^2} \approx 1.1 \times 10^{-16}.$$

For the 1S-2S transition frequency ($\nu_0 \approx 1.23 \times 10^{15}\,\text{Hz}$):

$$\delta\nu_{\text{2nd-order}} \approx \nu_0 \times \frac{v^2}{c^2} \approx 1.23 \times 10^{15}\,\text{Hz} \times 1.1 \times 10^{-16} \approx 0.14\,\text{Hz}.$$

This is **orders of magnitude smaller** than the natural linewidth ($1.3\,\text{Hz}$) and the experimental resolution ($3\,\text{kHz}$).

Justification for Neglecting second order Doppler and TOF effect.

1. **Low Thermal Velocities:**
   At $T \sim 100\,\mu\text{K}$, the velocity $v \approx 3.1\,\text{m/s}$. For a beam radius $w_b = 37\,\mu\text{m}$:

   $$\tau = \frac{w_b}{v} \approx \frac{37 \times 10^{-6}\,\text{m}}{3.1\,\text{m/s}} \approx 12\,\mu\text{s}.$$

   The corresponding TOF broadening would be:

   $$\delta\nu_{\text{TOF}} \approx \frac{1}{2\pi\tau} \approx 13\,\text{kHz}.$$

   However, this calculation assumes **single-pass interaction**, which does not apply here.
2. **Periodic Motion in the Trap:**
   Atoms are confined in a harmonic magnetic trap with vibrational frequency $\nu_v \approx 3\,\text{kHz}$. They oscillate radially, passing through the laser beam repeatedly. The interaction time becomes the coherence time of the oscillatory motion, which is much longer than τ.
   - **Coherence Time:** The atoms undergo many oscillations during the $2\,\text{ms}$ laser pulse (~ 6 cycles at $\nu_v = 3\,\text{kHz}$), leading to **motional narrowing**.
   - **Ramsey Interference Analogy:** Repeated interactions create interference fringes, narrowing the central peak (Fig. 4 in the paper).
3. **Laser Stability Dominates:**
   The observed linewidth ($3\,\text{kHz}$) is set by laser instability, not TOF effects. At higher temperatures ($T > 1\,\text{mK}$), TOF broadening dominates (e.g., $\delta\nu_{\text{TOF}} \approx 15\,\text{kHz}$ at $T = 1.7\,\text{mK}$), but at $T < 100\,\mu\text{K}$, motional narrowing suppresses TOF contributions.

Spectral Features in the trap

1. **Central narrowing**: Cold atoms near the trap center experience harmonic motion, leading to interference fringes (Ramsey-like).
2. **Sidebands**: are produced due to vibrational energy. Spaced by $2\nu_v$ due to quantized vibrational energy changes.
3. **Anharmonicity effects**: At larger radii, trap potential deviates from harmonic, broadening outer sidebands.

Comparison with Theory

- **Harmonic Oscillator Model:** Predicts equally spaced sidebands. Deviations in experiments confirm anharmonicity.
- **Ramsey Interference Analogy:** Explains narrowing via phase coherence over multiple interactions.
- **Parity Selection Rule:** Justifies even Δn transitions, consistent with two-photon selection rules.

This interplay of motion, symmetry, and potential shape underpins the experiment's success in approaching the 1S-2S transition's natural linewidth.

Spectral Features in the trap

Central narrowing :

Atoms in the harmonic magnetic trap oscillate radially with frequency $\nu_v \approx 3\,\text{kHz}$. During the $2\,\text{ms}$ laser pulse, they complete ~6 oscillations. This periodic motion allows them to interact with the laser field **multiple times**, analogous to the Ramsey separated oscillatory fields method.

- **Coherent Accumulation:** Repeated interactions lead to constructive/destructive interference, creating a narrow central peak.
- **Effective Interaction Time:** The coherence time is extended to the **oscillation period** ($\tau_{\text{coh}} \sim 1/\nu_v$), reducing the linewidth to:

$$\Delta\nu_{\text{central}} \approx \frac{1}{2\pi\tau_{\text{coh}}} \approx \nu_v \approx 3\,\text{kHz}.$$

- **Laser Stability Limit:** The observed $3\,\text{kHz}$ linewidth matches the laser instability, indicating central narrowing surpasses TOF broadening.

Sideband Formation :

The 1S-2S transition can involve changes in the atom's vibrational quantum number n in the trap. Due to spatial parity symmetry in the harmonic potential:

$$\Delta n = \text{even integer} \quad (\text{e.g., } \Delta n = 0, \pm 2, \pm 4, \dots).$$

This arises because the two-photon transition operator $\propto \mathbf{E}^2$ preserves parity.

- **Energy Splitting:**
  Each sideband corresponds to a vibrational energy change $\Delta E = 2n \cdot h\nu_v$, creating sidebands spaced by $2\nu_v \approx 6\,\text{kHz}$.
- **Observed Spectrum:**
  Sidebands appear as peaks flanking the central line (Fig. 4 in the paper). Their spacing provides a direct measure of ν_v, useful for characterizing trap properties.

Anharmonicity effects :

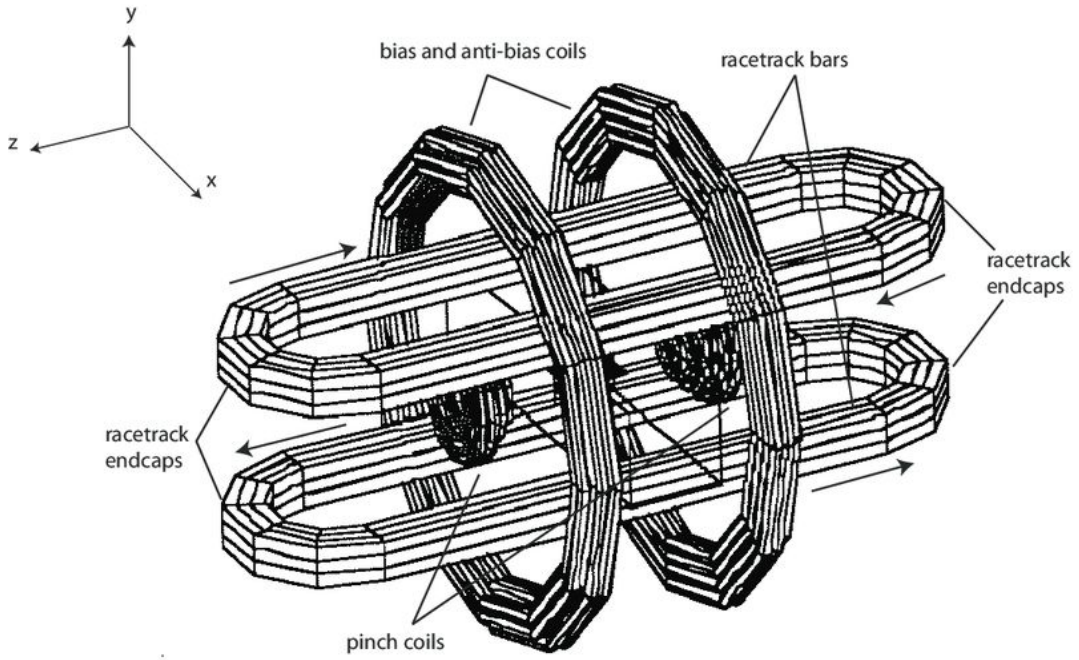
- **Trap Potential Realism:**
  The magnetic trap is only approximately harmonic. At larger amplitudes, the potential deviates as:

$$V(\rho) \propto \sqrt{B_\rho^2 + B_{z,0}^2} \quad (\text{non-quadratic in } \rho).$$

  This introduces **anharmonicity**, causing oscillation frequency ν_v to depend on amplitude.
- **Impact on Sidebands:**
  - **Frequency Spread:** Atoms with higher energy (larger ρ) oscillate at slightly lower frequencies, broadening the sidebands.
  - **Central Peak Immunity:** Atoms contributing to the central peak ($\Delta n = 0$) are localized near the trap center (small ρ), where the potential is nearly harmonic. Thus, the central line remains narrow.
- **Observed Spectrum:**
  Outer sidebands merge into a continuum (Fig. 4), but the central peak retains sub-kHz resolution. Anharmonicity calculations were included in the line-shape model to fit experimental data.

1. **Central Narrowing:**
   - Result of coherent, repeated interactions in a harmonic trap.
   - Linewidth approaches vibrational frequency ν_v, limited by laser stability.
2. **Sidebands:**
   - Arise from parity-conserving vibrational state changes ($\Delta n = \text{even}$).
   - Spacing $2\nu_v$ serves as a diagnostic for trap parameters.
3. **Anharmonicity:**
   - Broadens sidebands but spares the central peak.
   - Highlights the importance of cold atoms (low ρ) for precision spectroscopy.

Confinement in a Magnetic Trap



a = 22 mm, B_w <= 0.91 T.
At T = 100 microKelvin, thermal radius is approx 40 micrometer.

- The experiment utilizes an **Ioffe-Pritchard trap configuration** that combines a quadrupole field for radial confinement and axial solenoids to create a longitudinal barrier, forming a three-dimensional magnetic minimum. The quadrupole field is described by :

$$B_\rho = \frac{\rho B_w}{a}$$

where ρ is the radial distance from the axis.
and the axial bias field is $B_{z,0} \approx 200\ \mu\mathrm{T}$

For small oscillations, the vibrational frequency is given by:

$$\nu_v = \frac{1}{2\pi}\sqrt{\frac{\mu_B B_w^2}{M B_{z,0} a^2}} \approx 3\ \mathrm{kHz}$$

Confinement in a Magnetic Trap

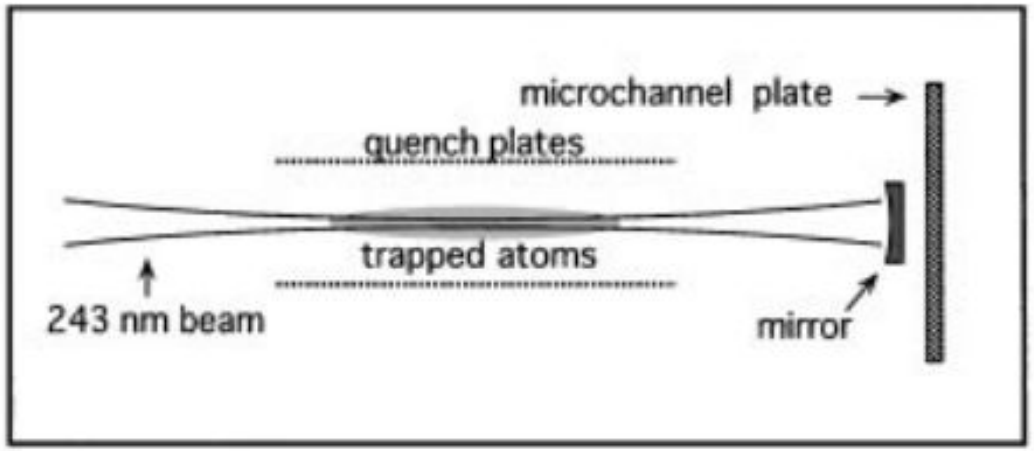


FIG. 1. Schematic diagram of optical excitation and detection apparatus. The UV beam is aligned along the axis of the magnetic trap. The beam traverses the cloud of trapped atoms (typically 10 cm long, 500 μm diameter), and is retroreflected. After a UV excitation pulse, the (metastable) $2S$ atoms are 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.

Confinement in a Magnetic Trap

- **Evaporative Cooling** : Forced evaporation removes high-energy atoms, cooling the sample to approximately *100 μK* by lowering the trap depth. The number of atoms varies from **10^13** to **10^10** and the density reaches up to **6 × 10^13 per cm** which is adjustable via trap depth.

- **Laser System :** The laser system starts with a **486 nm** dye laser, which is locked to a reference cavity, and then frequency-doubled to **243 nm**. The beam parameters include a waist radius of **w0 = 37 μm** stabilized to a reference cavity (**3 kHz linewidth**) with divergence length 18 mm. and Power = 4 mW, pulsed for 2 ms.

- **Detection scheme :** The detection scheme starts with the 2 ms UV pulses which are used to drive the 1S-2S transition; then, an electric field (8 V/cm) Stark-mixes the 2S and 2P states, inducing Lyman-alpha (λ = 122 nm) decay; finally, microchannel plates collect the photons with an efficiency of 4 × 10−6 .

Spectroscopy and Line Shape Analysis

- **Time-of-Flight Broadened Spectrum:** At temperatures T > 1 mK, the observed line shape follows

$$\exp(-|\delta|/\delta_0) \text{ where } \delta_0 = \frac{u}{4\pi w_b}$$

  For example, for T = 1.7 mK, δ0 = 11 kHz yields a full width at half maximum (FWHM) of approximately 14 kHz.
- **Motional Narrowing and sidebands:** Due to the periodic radial motion of the atoms, they interact repeatedly with the laser in a manner akin to Ramsey interferometry. This periodic motion results in a sideband structure where energy changes of

$$\Delta E = 2nh\nu_v$$

  produce sidebands spaced by **2ν** ≈ 6 kHz. The central peak is narrowed to a width limited by laser stability (approximately 3 kHz), which demonstrates the motional narrowing effect.

Spectroscopy and Line Shape Analysis

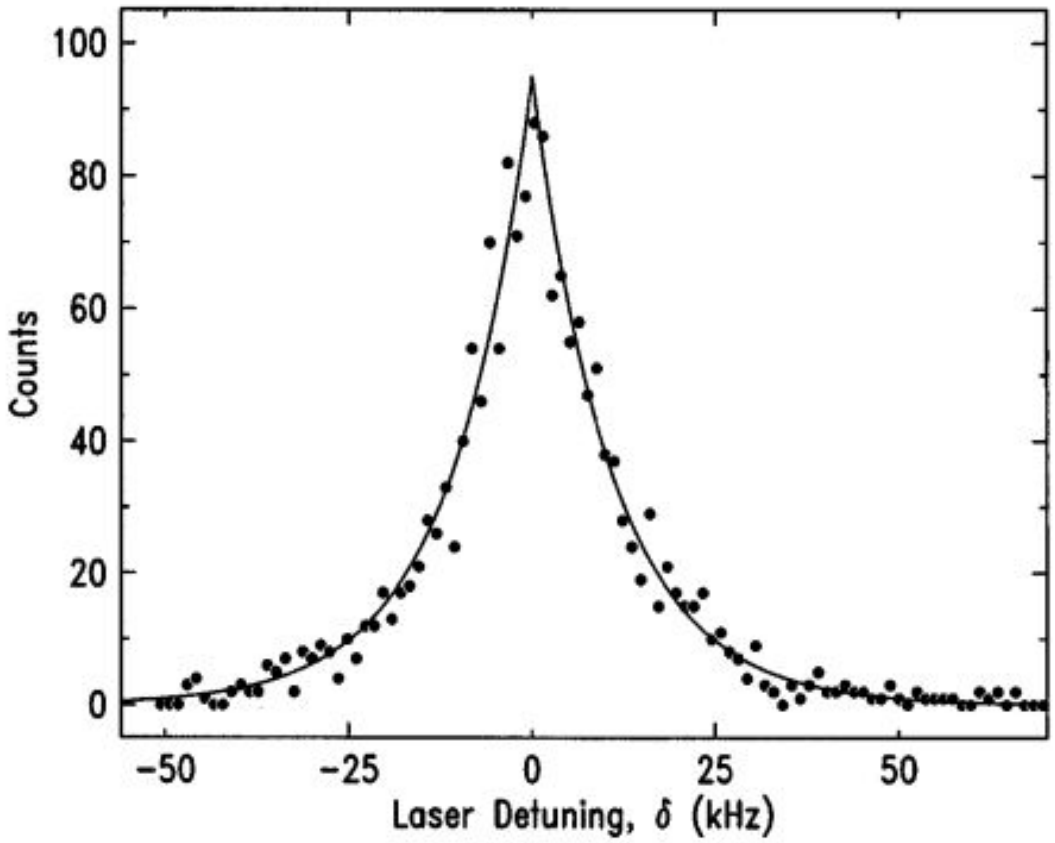


FIG. 2. $1S$-$2S$ excitation spectrum displaying a time of flight profile. The UV detuning (at 243 nm) is δ. The density is 3×10^{12} cm^{-3}, the temperature is 1.7 mK, and the UV power is $\approx$1.5 mW. The total UV exposure time at each point is 2.7 s. Here the dominant source of broadening is the 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.

TOF broadening

Spectroscopy and Line Shape Analysis

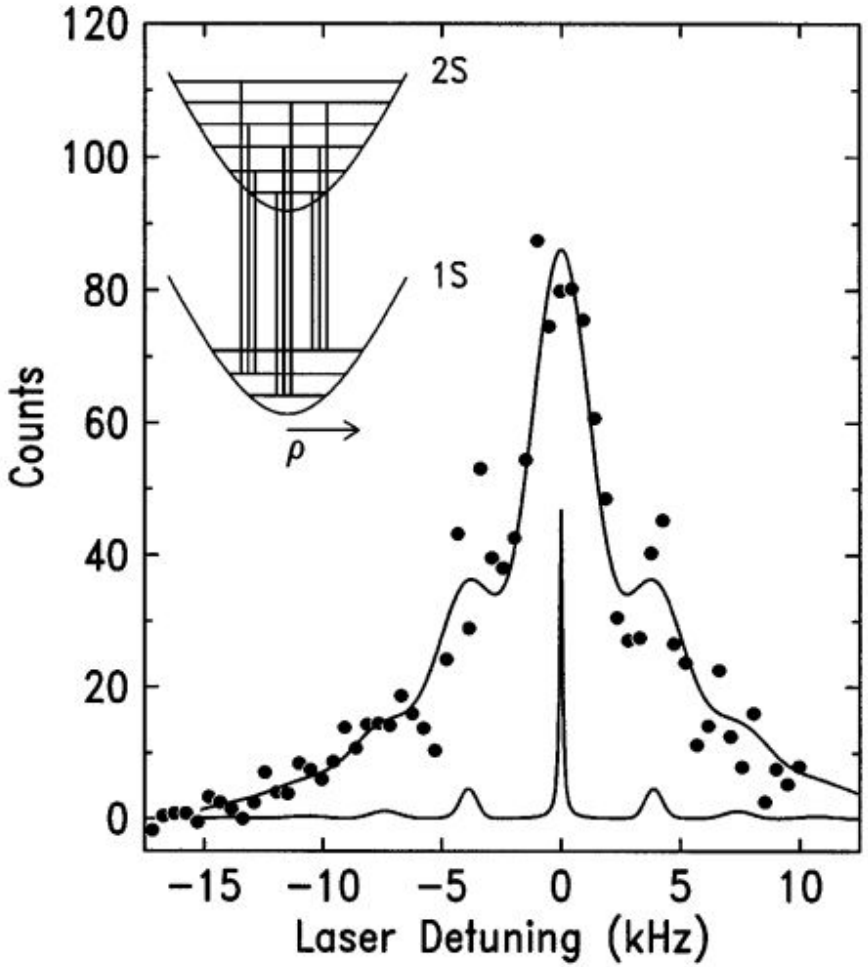


FIG. 4. 1*S*-2*S* excitation spectrum in the regime of motional narrowing. Each data point represents a total exposure of 350 ms. The upper solid line is the calculated spectrum including the effects of trap anharmonicity, photoionization, and an estimated laser linewidth. The best fit is for a laser linewidth of 3.0 kHz (full width at 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 vibrational states on the 1*S* and 2*S* electronic manifolds, indicating allowed transitions from the lowest three 1*S* trap states.

Motional Narrowing effect

Spectroscopy and Line Shape Analysis

- **Temperature Determination:**

  Temperature was determined by three methods: (a) linewidth fitting using

$$T = \frac{M\delta_0^2 w_b^2}{2k_B}$$

  (b) Trap dumping by measuring the energy distribution and velocity of escaping atoms, and (c) Evaporative cooling model that balances between heating and cooling rates.

  All three methods agreed within experimental error, validating spectroscopy as a reliable tool for thermometry.

Spectroscopy and Line Shape Analysis

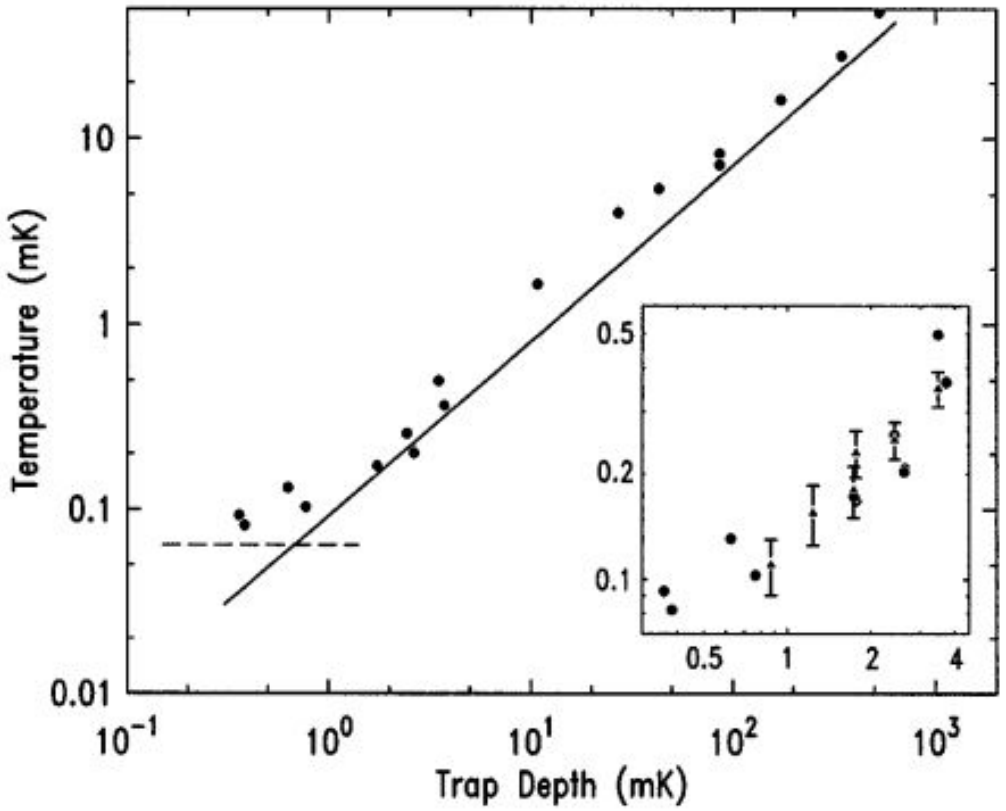


FIG. 3. Comparison of the temperature inferred from the time of flight linewidth (dots) with predictions from a model of heating and cooling effects in the trap (solid line). The temperature was varied by changing the depth of the trap, and measured by recording a spectrum at each point. The dashed line 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.

Temperature Determination

Lifetime of the metastable 2S State

Delayed quenching measurements revealed 2S lifetimes in the range of 50 – 110 ms, which are close to the natural lifetime of 122 ms. Although stray electric fields and collisions may reduce the lifetime, no systematic damping was observed.

Quenching mechanisms: (a) Stray electric fields: Dominant loss at E > 1 V/cm.

Collisions: Negligible due to low density (n ~ 10^13 cm^−3).

Sources of Line Shifts and Broadening

- **Zeeman Shift :** The Zeeman shift is calculated using the formula:

$$\delta\nu_Z = \frac{\alpha^2 \mu_B B}{4h} \text{ , } B \sim \frac{2k_B T}{\mu_B}$$

where α is the fine-structure constant. $\text{At T} = 100\ \mu\text{K}, \delta\nu_z \approx\ 50\ \text{Hz}$

- **Density-Dependent Shifts:** Atom-atom interactions result in pressure shifts that alter the transition frequency.

$$\text{Predicted shift } \Delta\nu \sim 1\ \text{Hz at } n = 10^{10} cm^{-3}\ \text{(negligible)}$$

- **Technical Limitations:** The current spectral resolution is limited by the laser's stability, which is about 3 kHz.

Discussion & Implications for Future Research

Achievements and Limitations :

- *Resolution* : Laser stability (3 kHz) now limits resolution, not TOF or Doppler effects.
- *Anharmonicity* : Broadens sidebands but spares the central peak.
- *Lifetime* : Confirms 2S metastability, enabling long interrogation times.

Implications for BEC and Optical Clocks Metrology :

- At T ~ 100 μK, hydrogen's phase-space density is close to the condensation threshold. Spectroscopy can be used to **detect BEC** via phenomena such as the **recoil shift** (approximately 7 MHz shift for condensed atoms) and the observation of spatial localization through a density peak at the trap center.
- Hydrogen spectroscopy offers advantages over ion clocks by providing higher signal rates (around ~ 103 counts/s) while achieving comparable resolution, thus promising improved optical frequency standards.

Conclusion

- This work demonstrates two-photon spectroscopy of trapped hydrogen with a **resolution** of **3 kHz**, **limited solely by laser stability**.
- Key achievements include the **observation of motional narrowing**, which reveals a **central peak** that is **100 times narrower** than those observed with previous methods; the **validation of in situ thermometry** against independent techniques; and **lifetime measurements** that **confirm minimal environmental perturbations**.
- Future **improvements** in **laser stability** and **further lowering of temperatures** (**T < 10 μK**) could allow the intrinsic **1.3 Hz natural linewidth** of the transition to be resolved, thereby **enabling more rigorous tests of QED** and the development of new optical frequency standards.