

Accessing the $5S_{1/2} \rightarrow 5D_{5/2}$ Two-photon Transition in Rb Using a Diode Laser System

Abstract : We report observation of the $5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transition in Rb vapour at 778 nm, using an external cavity diode laser system and a heated vapour cell. The spectra in the two isotopes show well-resolved hyperfine transitions. The peaks are Doppler free, and have a Lorentzian lineshape with a typical linewidth of 2.2 MHz. This linewidth is larger than the natural linewidth of 300 kHz, but is still 5-10 times smaller than the linewidth for single-photon transitions in the D_2 line. Since the absolute frequency of this transition is measured with 8 kHz precision, it can form a better secondary reference in the optical regime compared to the D_2 line.

Precise measurement of optical frequencies (of order 10^{15} Hz) is complicated by the fact that the SI unit of time is defined in terms of a microwave hyperfine transition in ^{133}Cs — one at 9.19×10^9 Hz. Thus the frequency measurement requires a span of more than 5 orders of magnitude with no phase error, not an easy task. In recent times, the use of a femtosecond frequency comb has enabled such a precise span¹. An alternative technique, which is lower in cost and simpler to implement, is to use an optical transition as a secondary frequency standard, and compare an unknown frequency with the reference. In our laboratory, we have pioneered such a technique where we use a ring-cavity resonator as a transfer cavity between the unknown and reference frequencies²⁻⁵. The reference in these experiments was a laser locked to a hyperfine transition in the D_2 line of ^{87}Rb (at 780 nm), whose absolute frequency has been measured using a frequency chain to 10 kHz precision⁶.

The technique is particularly suited to the measurement of electric-dipole allowed (E1) transitions because the Q for such transitions is about 10^8 . Thus a precision of 10 kHz (which corresponds to a relative uncertainty of 10^{-11} in the measurement of an optical transition) requires splitting the spectral line by 1 in 1000, which is a considerable experimental challenge. In other words, the precision of measurements on such transitions

is limited by the linewidth of the transition itself and not the measurement technique, which makes the ring-cavity resonator a competitive alternative to the comb method.

Since the natural linewidth of the D_2 line used as the frequency reference is about 6 MHz, reaching a precision of 10 kHz in the lock point itself requires better than 1 in 500 reproducibility. Therefore, an attractive option is to use a two-photon transition, which are forbidden from E1 selection rules, and consequently have sub-MHz natural linewidth. One such transition is the $5S_{1/2} \rightarrow 5D_{5/2}$ transition in Rb with a natural linewidth of 300 kHz. The absolute frequency of this transition (at 778 nm) has been measured by Nez *et al.* with 8 kHz precision⁷, thus making it a useful secondary reference. In addition, a two-photon transition can be made Doppler free if the two photons come from counter-propagating beams, because only zero-velocity atoms are resonant with both beams.

In this work, we report observation of the $5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transition in Rb using a diode laser system. We are able to resolve the various hyperfine transitions in the two isotopes, ^{85}Rb and ^{87}Rb . We obtain a linewidth of about 2.2 MHz (limited by optical feedback into the diode laser system), which is about 5–10 times better than the linewidth obtained for the D_2 line used as a frequency reference in the ring-cavity resonator technique.

Experimental Details : The diode laser system is a home-built grating stabilized diode laser⁸. The laser has a free running wave length of 784 nm and a maximum power of 120 mW, which reduces to 70 mW after feedback. The linewidth after feedback is about 1 MHz. Using a combination of operating temperature and grating angle, the system is brought to operate near 778 nm required for accessing the two-photon transition. Coarse tuning is achieved using a commercial wavemeter (WS Ultimate from High Finesse Laser & Electronic Systems). The laser beam has elliptic cross-section with $1/e^2$ diameter of $3 \text{ mm} \times 5 \text{ mm}$. It is linearly polarized.

The schematic of the experiment is shown in the Fig. 1. The laser output first passes through a two-stage optical isolator (with 60 dB isolation), and then through a Rb vapour cell heated to 135 °C. The cell is cylindrical in

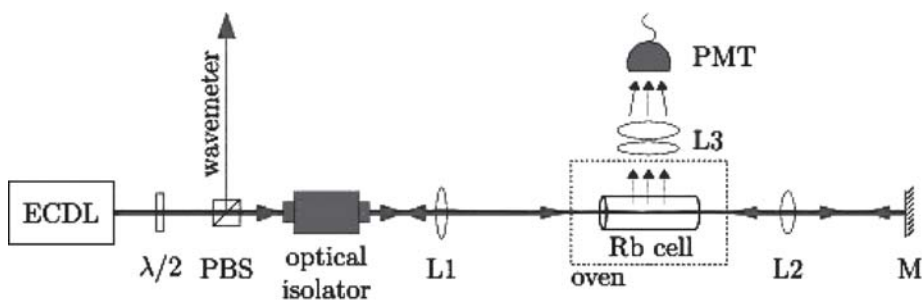


Fig. 1. Experimental schematic for two-photon spectroscopy in Rb. The vapour cell is heated to a temperature of 135 °C. Figure key — ECDL: external cavity diode laser; $\lambda/2$: half-wave plate; PBS: Polarization beam splitter cube; L1, L2, L3: lenses; M: mirror; PMT: photo multiplier tube.

shape with 25 mm diameter and 50 mm length. It is pure (no buffer gas) and contains the two isotopes of Rb in their natural abundances — 72% ^{85}Rb and 28% ^{87}Rb . The laser beam is focused into the cell using a biconvex lens (L1) of focal length 300 mm. After the cell, the beam passes through a second lens (L2) of the focal length 100 mm before being retro-reflected from a mirror.

The relevant energy levels in the two isotopes are shown in Fig. 2. The isotope ^{85}Rb has $I = 5/2$ and therefore 6 hyperfine levels in the $D_{5/2}$ state. The other isotope ^{87}Rb has $I = 3/2$ and therefore 4 hyperfine levels in the $D_{5/2}$ state. The signal from population in the $D_{5/2}$ state is the blue fluorescence at 420 nm due to radiative cascade through the intermediate $6P_{3/2}$ state, as shown in Fig. 2(a).

The fluorescence signal (which is also visible to the naked eye) is measured by a R928 Hamamatsu photo-multiplier tube (PMT) using a pair of lenses (L3 in the schematic figure). The solid angle subtended by the lens system is such that about 5% of the fluorescence light is collected. The PMT has a 420 nm interference filter in front with peak transmission of 72%.

Results and Discussion : The measured two-photon spectra in the two isotopes are shown in Fig. 3. There are four spectra corresponding to the two ground hyperfine levels in each isotope — $F = 2, 3$ in ^{85}Rb and $F = 1, 2$ in ^{87}Rb . The selection rules for a two-photon transition are $\Delta F \leq 2$, therefore each spectrum has several well-resolved peaks corresponding to this rule. Only the $F = 2 \rightarrow 1, 0$ peaks in ^{85}Rb are not resolved. As expected, the spectra appear on a flat Doppler-free background. The resolved peaks have a Lorentzian lineshape with a linewidth of about 2.2 MHz. This increase from the natural linewidth of 300 kHz is due to optical feedback into the laser from the system. Despite using an optical isolator in front of the laser, stable spectra of the kind shown are obtained only when the counter-propagating beam is slightly misaligned. For comparison, the linewidth seen in Ref.⁷ was 500 kHz,

but the experiment was done with a frequency stabilized Ti-sapphire laser with frequency uncertainty of only 2 kHz, and a laser that was much less sensitive to optical feedback.

We have studied the signal strength as function of laser power. For a two-photon transition, the rate varies as the square of the laser power. The results are shown in Fig. 4, along with a quadratic fit to the data. The error bars represent the standard deviation for each data set, which consists of 20 points. The quadratic curve fits the data well within the error bars.

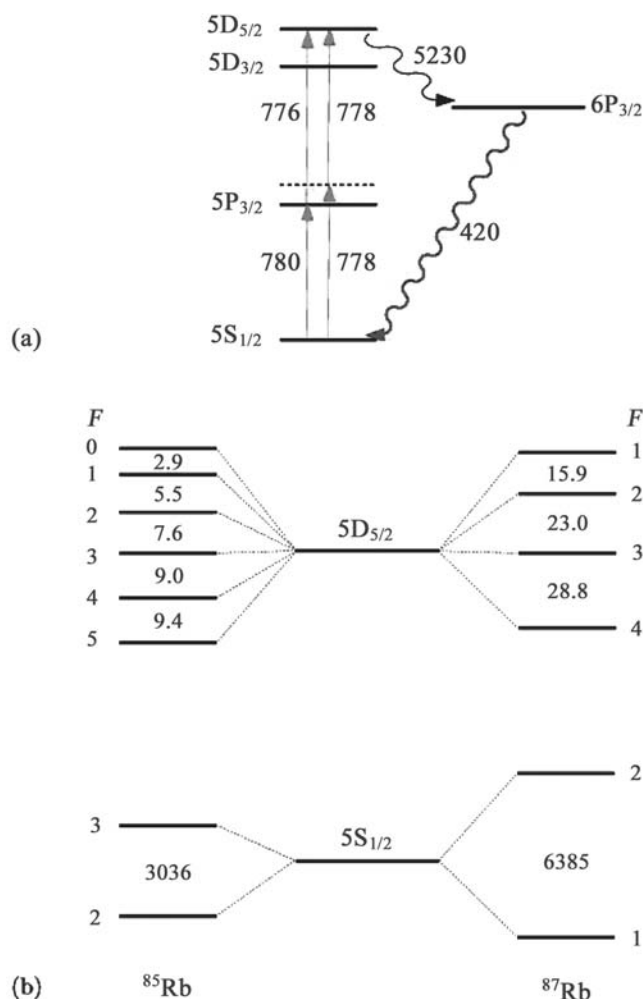


Fig. 2. (a) Energy levels of Rb. The laser at 778 nm drives the two-photon transition through an intermediate virtual state (dotted line). The two single-photon transitions at 780 nm (D_2 line) and at 776 nm are also shown. Population in the $5D$ state decays to the ground state by radiative cascade through the intermediate $6P$ state — the resulting blue fluorescence at 420 nm is the detected signal. (b) Hyperfine levels in the two isotopes in the $5S_{1/2}$ (ground) and $5D_{5/2}$ states. All intervals are in MHz. Note that the hyperfine structure in the two states are opposite.

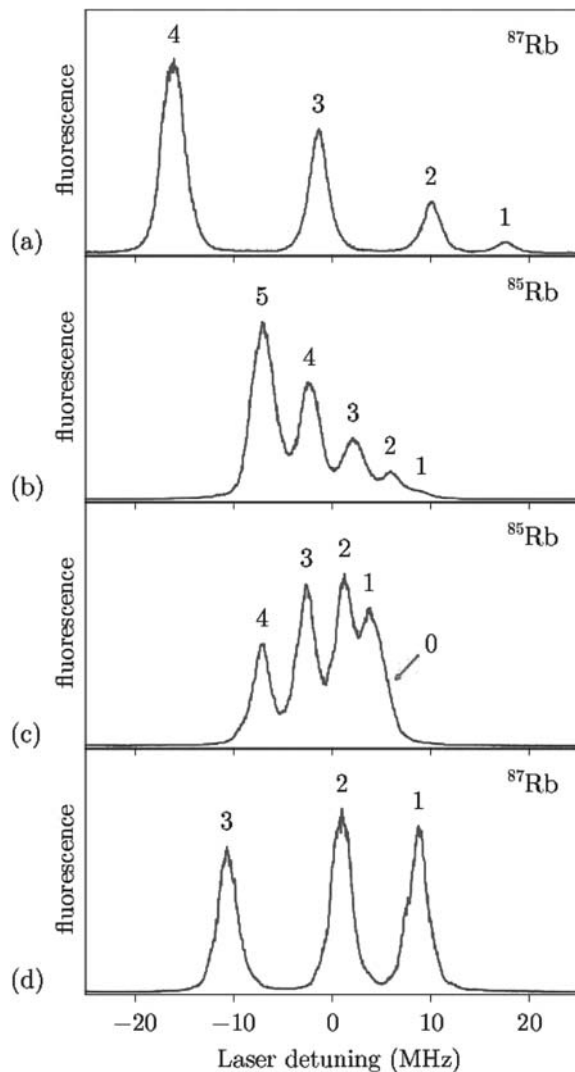


Fig. 3. The $5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transitions in the two isotopes of Rb. The selection rule is $\Delta F \leq 2$. (a) ^{87}Rb : $F = 2 \rightarrow F' = 4$ to 1. (b) ^{85}Rb : $F = 3 \rightarrow F' = 5$ to 1. (c) ^{85}Rb : $F = 2 \rightarrow F' = 4$ to 0; the $F' = 1$ and 0 levels are not resolved—the location of $F' = 0$ is indicated by an arrow. (d) ^{87}Rb : $F = 1 \rightarrow F' = 3$ to 1.

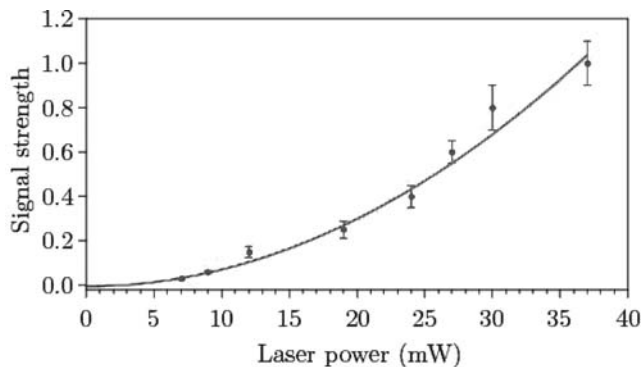


Fig. 4. Fluorescence amplitude for the two-photon transitions as a function of laser power. The error bars show the fluctuation observed. The fluctuations are larger at higher powers due to increased optical feedback.

Conclusion : In summary, we have observed the $5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transitions in a Rb vapour cell using a diode laser system. The various hyperfine transitions in the two isotopes are well resolved and have a linewidth of about 2.2 MHz. This is somewhat larger than the 300 kHz natural linewidth, and is due to laser jitter caused by optical feedback. But the linewidth is still about an order-of-magnitude smaller than the linewidth obtained for the D_2 line, which is used in our laboratory as a frequency reference in a ring-cavity resonator [5]. This makes the two-photon transition a useful frequency reference for improved precision in the ring-cavity technique.

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**K. D. RATHOD AND
VASANT NATARAJAN**

Department of Physics,
Indian Institute of Science,
Bangalore 560012, India
e-mail : profvasant@gmail.com

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1. See, for example, the Nobel Prize lectures – John L. Hall. Nobel lecture: Defining and measuring optical frequencies. *Rev. Mod. Phys.*, **78** :1279–1295, (2006); and Theodor W. Hansch. Nobel lecture: Passion for precision. *ibid.*, **78** :1297–1309, (2006); and the references therein.
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