High Resolution Two-Photon Spectroscopy of Rb Rydberg Levels

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In this paper we report on our investigation of the interesting features, and problems, associated with ultrahigh resolution two-photon spectroscopy, using a frequency controlled dye laser to study the 52P-52S transitions in rubidium. The optical Ramsey technique of separated interactions is used to reduce transit broadening. Narrow resonances of 17 kHz full width half maximum are observed.

The use of counterpropagating laser beams to eliminate Doppler broadening in two-photon transitions by now is a well-known and standard method in laser spectroscopy. In principle, transitions between long-lived levels, such as the ground and metastable states, should lead to extremely narrow resonances, and various authors have proposed using these two-photon resonances as possible frequency standards in the optical region [1-5]. In practice, however, the resolution obtained in two-photon spectroscopy has thus far been limited either by the laser line width, or by the short lifetime of the states being investigated. Important aspects of high resolution two-photon spectroscopy, such as the transit-limited line shape, or small level shifts, have remained almost unexplored. The reasons may be summarized as a lack of suitable laser sources, and the lack of experimentally attractive candidate transitions.

The major disadvantage of two-photon transitions between long-lived states is the extremely low transition strength. Large laser intensity is needed and even so, two-photon signals usually require long periods of signal averaging (e.g., B1) [6,7]. This puts rather stringent requirements on the laser source: reasonable power output, narrow spectral width, and good long term frequency stability.

If the laser width may be neglected, transit broadening becomes the main limiting factor in two-photon line widths. The line shape of two-photon absorption, including the effects of the finite transit time of the atoms through a Gaussian laser field and the natural broadening of the levels, has been studied in detail by ROMNE [8], and by RAKLANOV and DUBETSKII [9]. Their treatment has recently been extended [7] to the

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experimental situation in an atomic beam, where the excited atoms are detected at a distance downstream from the interaction region. Due to the decay of the excited atoms in flight, the detected signal will show not only a reduction in strength, but also a broadening in width, since the faster atoms have a higher probability of reaching the detector before decaying. Taking this decay into account, the Doppler-free two-photon transit-limited line shape may be obtained by the following integral over all possible velocities \( v \) in the atomic beam [7]:

\[
I_{\text{det}} \sim \int_0^\infty dv \cdot v \exp \left( -\frac{v^2}{V} - \frac{G}{V} - \frac{r^2}{4V^2} \right).
\]

Here, \( V = v/u \), where \( u = (2\pi T/M)^{1/2} \); \( G = \gamma v/u \) is a decay parameter, where \( \gamma \) is the population decay rate of the upper state, and \( r \) is the detector to interaction distance; \( \xi = (\omega - \omega_0)V_0/u \) is the detuning parameter, where \( \omega \) and \( \omega_0 \) are the laser and atomic transition frequencies, respectively, and \( V_0 \) is the laser mode radius. Figure 1 shows the resulting line shape for different values of \( G \). For \( G = 0 \), i.e., detecting the atoms at the interaction region, the transit width \( \Delta \omega_{\text{FWHM}} \) (FWHM at the laser frequency scale) is \( \frac{1.25 \, \omega_0}{V_0} \) in agreement with Ref. 9.

The line shape calculated with \( G = 0.29 \) is found to be in good agreement with our recent experiment on the Bi \( 6p^3 \, ^4S_{3/2} \) to the metastable \( 6p^2 \, ^2P_{3/2} \) two-photon transition [7].

Fig. 1. Transit-limited two-photon line shape, calculated from Eq. (1). The full profile is symmetric around \( \xi = 0 \). In-flight decay between excitation and detection is parametrized by \( G \). Inset shows line width increasing effect of inflight decay.
To reduce transit broadening, BAKLANOV et al. [10] proposed using two spatially separated laser beams. The resulting fringe structure, arising from the interference between two separate interactions of an atom with the radiation field, will have a width determined by the atomic transit time between the two light beams. This is the optical analog of the well-known Ramsey fringes in microwave and rf spectroscopy. The first radiation zone creates a coherence between the atomic lower and upper states. In the region between the laser beams, the atomic polarization precesses freely, and at a rate which is, in general, different from the radiation field. The effect of the second radiation zone depends on the phase of the radiation relative to the phase of the atom, thereby producing interference dependent on the interzone transit time T. The fringe pattern is of the form \( \cos(2\omega - \omega_\text{in})T \). When the fringes are integrated over the velocity distribution, the side fringes tend to be averaged out, leaving only the central fringe.

The total phase of the Ramsey signal also includes a contribution from the relative phase difference between the laser fields themselves in the two zones [11]. This "cavity phase" introduces a displacement of the fringe system. Thus it is important to hold this "cavity phase" fixed in an experiment, otherwise the random fluctuations will wash out the fringe pattern. We note that in a folded standing wave configuration the phase factors cancel exactly, and if the laser beams are matched, the fringe pattern will be symmetric and unshifted. Ultimately, at very high resolution the second-order Doppler effect gives an interesting distortion -- as well as shift -- of the Ramsey fringe pattern. (See below.)

In the experimentally appropriate limit that the interzone separation \( d \) is much greater than the size of each zone, the Ramsey fringes have the form [10,11]

\[
I_{\text{det}} \sim \int_0^\infty dV \cos \left( \frac{2\pi}{V} + 2\pi \frac{yV}{u} \right) \left[ \frac{1}{V^2} \right] \left[ \exp \left( - \frac{G}{V^2} \right) \right] \left[ \exp \left( - \frac{V^3}{y^2} \right) \right]
\]

where as before \( V = \nu/u \) and the decay parameter \( G = (\gamma/u)[(\omega - d)/2] \). The cosine term arises from the Ramsey interference between excited state amplitudes produced in the two separate interaction regions. The detuning parameter is \( \nu = (\omega_c - \omega_\text{in})d/u \), where \( d \) is the interzone separation. The second-order Doppler shift is \( \omega_\text{D} = \nu/V \), normalized to the Ramsey period, \( \omega_\text{D} = \nu/V \). The \( [1/V^2] \) term arises from integration of the interaction through the laser mode, as may be shown with the diagrammatic method of BORDE [11]. The next factor represents exponential decay in the distance between excitation and detection regions. Finally the contributions of each velocity group need to be weighted with the appropriate beam velocity distribution. Figure 2A shows a Ramsey profile computed from Eq. (2) using conditions appropriate to our high resolution experiment (\( \omega_\text{D}/2\pi \) = 1.0 \( \times \) 10\(^{15} \) Hz, \( d = 0.5 \) cm, \( u = 3.13 \times 10^4 \) cm/sec, \( G = 2.0 \), \( y = 0.00087 \)). Figure 2B shows the effect of increasing the interzone spacing by a factor 10 to \( d = 5.0 \) cm (\( y = 0.087 \)) at which point the nominal Ramsey fringe spacing just equals the expected lifetime-limited line width (\( \Delta \) kHz at the laser frequency). Some fringe distortion is evident due to the influence of the second-order Doppler shift.

In the future we may expect to observe Ramsey fringes of even narrower spectral widths in atomic systems with longer lifetimes. For example, the Bi \(^{2}P_{3/2} \) metastable level mentioned earlier has a 4 msec...
Fig. 2. Calculated Ramsey fringe profiles in limit of negligible interaction time in laser beams. All curves correspond to detection downstream at 2 decay lengths (G=2), and differ in the ratio, $y$, of second-order Doppler redshift to the Ramsey spacing. A) Corresponds to experimental conditions of Fig. 1B, $y = 0.0087$; B) $y = 0.087$; C) $y = 0.2$; D) $y = 0.5$. Note that the fringe-like structure can appear on the blue side of the rest frequency.

estimated lifetime, leading potentially to 20 Hz wide two-photon resonances. However from a usual atomic beam source, the thermal velocity $\nu \sim 3.5 \times 10^4$ cm/sec leads to a second-order Doppler shift of 1.3 kHz. Such domination of the Ramsey line width by the quadratic Doppler shift gives rise to interesting spectral profiles. We may appreciate this effect physically from the following kind of arguments [22].

For a given atomic velocity $\nu$ the Ramsey pattern contains, in addition to the central fringe near $\nu = \omega_0/2$, a number of side fringes of laser frequency spacing $\Delta \omega_{\text{Ramsey}} = (\nu/d)m$ where $m$ is the side-fringe order number. On the red side of line center this linear side-fringe frequency shift and the second-order Doppler shift are additive, both increasing with atomic velocity $\nu$. On the blue side of line center, however, the two contributions to the fringe offset have opposite signs. Thus with low but increasing velocities the side fringe resonance position moves first linearly to higher frequency. With further velocity increase the resonance position becomes stationary as the linear fringe offset and second-order Doppler shift approximately cancel. Ultimately at higher
velocities, the second-order shift dominates and the side fringe components again move to the red. Computer simulation under very high resolution conditions leads to "fringe-like" wiggles on the blue side of true line center superimposed on a nearly structureless background. Figure 2(C,D) shows two representative curves for $G = 2$ and $y = 0.2$ and 0.5. As the shape and position of these wiggles are highly sensitive to the assumed conditions, it is amply clear that some type of velocity-selection technique will be essential in this ultrahigh-resolution domain. Alternatively, the observed profiles may be used to infer the actual velocity distribution.

Turning now to experimental aspects, two-photon Ramsey fringes have been observed in Na 35-4D transition, in the time domain by SALOUR and COHEN-TANNOURDI [12] using two coherent pulses, and by CHEBOTAYEV and coworkers [13] using two spatially separated laser beams. As the short lifetime of the 4D state ($\tau \sim 50$ nsec) is really the main limiting factor in these experiments, they could not bring out the high resolution aspect of the Ramsey technique. We report now on an investigation of ultrahigh resolution two-photon spectroscopy using the Ramsey technique and a frequency-controlled dye laser to study the 5S-4S transitions in rubidium.

The choice of rubidium has the following rationale: The Rydberg levels of Rb can be reached by two photons of a Rhodamine 6G dye laser; and more importantly, they have reasonably long lifetimes and are easy to detect. For Rydberg levels, the lifetime varies $\sim (n^*)^3$, where $n^*$ is the effective principal quantum number. Thus for $n^* \sim 30$, the lifetime is $\sim 25$ usec, giving a two-photon line width of 3 kHz (in the laser frequency scale), which is adequate for our purpose. In addition, the Rydberg levels are easily ionized by an electric field, and so the two-photon transitions may be detected with high efficiency and sensitivity by field ionization and positive ion counting.

In our experiment a beam of Rb atoms is produced from a resistively heated oven ($T \sim 200^\circ$C), and is collimated to approximately 1 mm in diameter at the interaction region. Figure 3 shows a schematic of the atomic beam apparatus. To increase the circulating laser flux, we use a resonant cavity inside the vacuum chamber. This slightly folded cavity (see Fig. 3) produces a power buildup $\sim 30$ and has two separate waists, of 155 $\mu$m.

![Diagram of Rb atomic beam apparatus for two-photon Ramsey fringes.](image)

Fig. 3. Rb atomic beam apparatus for two-photon Ramsey fringes.
radius, displaced along the atomic beam axis. The use of careful input
mode matching and a high-finesse, non-degenerate cavity insures that its
internal fields accurately represent a spatially matched standing wave
laser field and so have the proper amplitude and phase conditions for a
clear study of the optical Ramsey effect. The atom beam crosses the laser
beams at right angles, and the excited atoms are field ionized at a dis-
tance of 1.5 cm downstream from the interaction region. The positive ions
are accelerated into the cone of a channeltron electron multiplier. The
multiplier's output pulses are stored in a signal averager, which is swept
synchronously with the laser frequency. The detection region is enclosed
in a metal box to shield its electric field from the interaction region.
A pair of parallel plate electrodes in the interaction region allow us to
apply a small, uniform electric field to study the dc Stark effect. We
note that for S levels, the Stark effect introduces a red shift in the
transition frequency but does not produce splitting. By observing the
Stark shift as the polarity of the applied electric field is reversed,
we conclude that stray electric field in the interaction region is less
than 0.15 V/cm, giving a frequency shift of less than 10 kHz. A small
helmholtz coil pair is used to generate -20 G of magnetic field for
Zeeman studies.

A schematic of the frequency-controlled dye laser spectrometer [14]
is shown in Fig. 4. The dye laser is basically a three-mirror astigma-
tically compensated cavity, with a birefringent tunable and two etalons for
single frequency operation. The dye cavity optical length may be varied by
applying voltages to an intracavity ADP phase modulator, and to a
piezo-driven mirror. For frequency control, a part of the dye laser output
is mode matched into a high-finesse, invar-spaced cavity. The laser fre-
quency is servo-controlled to the side of a transmission fringe of the

![Schematic of a frequency-controlled dye laser spectrometer](image)

*Fig. 4. Narrow-bandwidth frequency-offset-locked dye laser spectrometer.*

Lock of dye laser on side of "invar cavity" fringe narrows dye laser line
width. Long-term frequency control is via first-derivative lock to the
transfer cavity which is similarly controlled to the HeNe offset laser.
The offset frequency of this laser from the I2-stabilized laser is locked
to the programmable frequency synthesizer. A multichannel signal averager
and the frequency synthesizer are swept synchronously.

135
cavity, using the usual fast differencing technique. With an improved amplifier system and an intra-dye-laser-cavity AQP phase modulator to provide fast response, the dye laser spectral width is narrowed to 2 kHz RMS. The long term stability and frequency information is obtained by servo-controlling the invar cavity length so that the dye laser is locked on the center of the transmission fringe of a quartz-spaced transfer cavity. The length of the transfer cavity is servo-controlled to a 633 nm HeNe local oscillator laser, which is in turn frequency offset locked [15] to an $^{129}$I$_2$ stabilized HeNe laser. Digital scanning of the dye laser frequency is accomplished by the frequency synthesizer in the frequency-offset-locked loop. The system is capable of a continuous scan of 900 MHz, at 1 kHz step resolution. The limit on the scan range is due to the gain profile of the HeNe laser. The use of a separate transfer cavity greatly improves the long term stability and frequency reproducibility of the dye laser, particularly against spurious frequency shifts introduced by very small angular drifts in the direction of the dye laser output beam (which degrade the mode-matching into the invar cavity). Drift rate of the absolute optical frequency was $< 1$ kHz/min under favorable conditions. The useful dye laser output for the experiment is $\sim 35$ mW, at 2 kHz spectral width.

We note that strong realtime two-photon 5S-5S transitions in Rb may be obtained with a simple electrostatically shielded, space-charge limited ionization cell [16,17]. In Fig. 3 we show a spectrum of the 5S-3S transition in natural rubidium. As illustrated in the lower part of the figure, the two larger central components arise from the more abundant $^{85}$Rb and are separated in laser frequency by one half of the 3.03573 GHz ground state hyperfine splitting. The outer two peaks are from $^{87}$Rb, and are separated by one half of its 6.78468 GHz ground state hfs. The upper state hfs is negligible at this resolution level. Unfortunately, condi-
tions in a cell are rather complex. Stray electric fields induce Stark shifts, and pressure shifts and broadenings of ~1.6 GHz/Torr and (2.6±1.0) GHz/Torr are observed for the 5S-3S2 transition [17].

In an atomic beam the collision effects are negligible. When the spectrum was obtained with high resolution, each of the lines described above showed a complex structure. This was found to be caused by the presence of a small magnetic field (~1 G) in the apparatus, which caused Zeeman splitting of the transitions. To investigate the Zeeman effect more clearly, a magnetic field of about 20 G was applied. For 39Kr, I = 5/2, giving F = 3 or 2. The F = 3-3 transition consists of 11 components, whereas in the F = 2-2 transition the central component is missing. See Fig. 6. The effect may be explained by the following simple model. The 3S levels have a total angular momentum F = I+S, where I and S are the nuclear and electronic spin angular momentum, respectively. The spin Hamiltonian may be expressed as

\[ H = A(I,J) + g_u B_s I^z - g_I \mu_B I^z I^z, \]

where \( g_u, g_I \) are respectively the electronic and nuclear g-factors, and \( \mu_B, \mu_N \) are the electronic and nuclear magnetons. The selection rules for transitions between Zeeman levels for <S states were discussed by BLOEMBERGEN et al. [18]. For small magnetic fields such that \( g_u B_s \ll A \), the selection rules are \( \Delta F = 0 \), and \( \Delta m_F = 0 \), assuming the spin orbit coupling may be neglected in the intermediate F states. The Zeeman levels may be labeled by the quantum numbers F and m_F. We note that in a hydrogenic atom, the hyperfine constant A varies as \( \langle n^* \rangle^{-3} \). For 85Rb, \( A_{ss} = 1011.9 \text{ MHz} \), giving \( A_{ss} \approx 200 \text{ kHz} \). Thus the weak field criterion, \( B \ll A/g_u \) (~0.1 G) is not satisfied even in the earth's magnetic field. For a field of 20 G which we used, the upper level is in the Paschen-Back region.

In the strong field domain, where the levels may be labeled by their magnetic quantum numbers \( m_F \) and \( m_J \), the Zeeman energy of the levels is simply \( \Delta m_F \mu_B H \approx g_u \mu_B B \Delta m_F \). The selection rules are \( \Delta m_F = 0 \), \( \Delta m_J = 0 \).

In our case we have \( A_{2S,ss} \ll g_u B_s \ll A_{ss} \). Consider the transitions from the F = 3 hyperfine level only. The lower and upper states have different spin wave functions, and the projection of the lower \( m_F \) levels
onto the upper \( m_F \) levels give two different transition frequencies for each \( m_F \) value, except for \( m_F = \pm 3 \) which are pure states. These latter two transitions contribute to a single line at the zero field \( F=3 \) position. Thus 11 transitions are possible, in agreement with experiment. Similarly, the \( F=2 \) transition can be shown to have 10 Zeeman components and to be missing the central component (which derives from \( m_F = \pm 3 \)), in agreement with Fig. 6.

These Zeeman patterns can be used to obtain the hyperfine constant of the upper level. The frequency interval of adjacent transitions belonging to the \( m_F = \pm 1/2 \) group is different from the interval of the \( m_F = \pm 1/2 \) group, the difference being \( A_{1/2} \). Here the factor of 2 is introduced so that the frequency scale is in terms of the laser frequency. We measured \( A_{1/2} = (0.23 \pm 0.07) \text{MHz} \). A precise knowledge of the magnitude of the magnetic field is not needed to deduce the excited state hyperfine structure constants but it would allow determination of the \( g \) factors.

We note that laser stepwise excitation in conjunction with rf resonance technique has been very successful in measuring the hyperfine structure of \( nS \) levels in alkali atoms, where \( n \leq 10 \) [19]. The extension of the double resonance technique to higher \( n \) values proves to be difficult, due to the rather poor signal-to-noise ratio in observing fluorescence. The present method has a comparable frequency resolution but with extremely good S/N. Typical time spent in obtaining a Zeeman pattern is 1 min. Thus our method should be very useful in measuring hyperfine parameters of high Rydberg levels.

Another interesting type of possible measurement with the \( m_F \)-state–resolved spectrum is the determination of the excited state static polarizabilities. For example when even small voltages were applied to the electric field plates mentioned earlier, rather dramatic frequency shifts were observed. For the \( 5S-3S \) (\( F = 3, m_F = -2 \)) transition a quadratic Stark frequency shift rate of \( -620 \text{kHz}/(V/cm)^2 \) was measured. Presumably more complicated behavior would occur in the \( nD \) levels where a tensor polarizability term can also be non-zero.

To investigate the optical Ramsey effect, the \(^{85}\text{Rb} 5S-3P \) \( F = 3 \) line was resolved into its 11 components with a 20 G magnetic field. The central Zeeman component (\( m_F = \pm 3 \)) was chosen for its strength, and also because this line is least perturbed by magnetic field inhomogeneity in the interaction regions. The velocity of the beam is \( u = 3 \times 10^8 \) cm/sec. With an upper state radiative lifetime of \( -25 \mu \text{sec} \), the population decay length is \( -0.75 \) cm.

Figure 7A shows the Ramsey feature obtained with two spatially separated laser beams. The fringe contrast is \( -35\% \). The signal was obtained in one sweep, at 1 sec/channel, and 5 kHz step size (at 633 nm). The two cavity waist, of mode radius 155 \( \mu \text{m} \), were separated by approximately 1.5 cm. As expected, the side fringes rapidly wash out, due to the velocity distribution in the atomic beam. The estimated experimental fringe full width of \( \pm 40 \) kHz was somewhat larger than the expected 30 kHz width in this early experiment. (The dye laser width was \( -5 \) kHz when this spectrum was taken.) The large background in Fig. 7A is the transit broadened signal from those atoms which interact with only one radiation zone. The full width, according to calculations described above, is 390 kHz. But with a detection to interaction length of 1.5 cm, corresponding to a decay parameter \( G = 2 \), the width is broadened to \( -630 \) kHz. The ex-
Fig. 7. Two-Photon Ramsey Fringes. Frequency increases to the left.
A) Waist separation = 1.5 mm. Single zone background resonance width ~600 kHz, Ramsey fringe width ~40 kHz. Dwell time 0.25 sec per point. B) High resolution, waist separation = 4.2 mm. Single sweep at 1.06 kHz/channel. Dwell was 0.35 sec per point.

Experimentally observed width is ~600 kHz. On the peak the inA counting rate is ~10 kcps.

Figure 7B shows the narrowest Ramsey fringes that we have obtained, with an interzone separation of 0.42 cm. The fringe full width of 17 kHz is almost entirely due to the interzone transit time [20]. Further increase of the separation unfortunately led to a severe reduction in the Ramsey signal. (We note that as the separation is increased, fewer atoms will be intercepted by both zones. If the oven aperture is too large the reduction in fringe contrast will be thus quadratic in the separation.) Furthermore, at n = 32 the natural decay is beginning to take its toll.

In the spectra we obtained, the Ramsey fringe pattern always appears to be displaced relative to the background peak. (The central Ramsey fringe is to the high frequency side of the background peak.) See Fig. 7A for example. These data were obtained on the magnetic field-independent $|m| = 2$ transitions so that magnetic field inhomogeneities can play no role in producing the somewhat asymmetric shape. In principle asymmetric Ramsey fringes could be caused by a residual phase difference between the radiation zones. However, the high finesse and accurate mode-matching of our cavity makes this explanation unlikely. A more tantalizing cause may be the reduction in the ac Stark shift with the Ramsey method: the background peak is shifted the full amount, while the shift of the Ramsey peak is diluted by a factor proportional to the one-zone to interzone transit time ratio. At our wavelength, the 8h 5S level is pushed upwards in energy by the ac Stark effect by 40 Hz/cm², when the 5 to 10P levels contributions are taken into account. The 32S level shift is very difficult to calculate accurately. We note that the largest radial matrix elements occur for nearby 10P states, but their nearly symmetric locations relative to 32S give rise to strong cancellation via the $(E_{32S} - E_{10P})^{-1}$ denominators in the summation which appears in the ac Stark shift formula. Only very weak matrix elements connect to lower levels (10P) where the energy denominators are small. Thus we believe that the dominant effect is an upward displacement of the 5S level and a consequent redshift of the two-photon transition frequency.
In conclusion, we have explored the two-photon optical Ramsey effect at high resolution using transitions to rubidium Rydberg levels. A Ramsey fringe as narrow as 17 kHz was easily observed [20]. We are anxious to test the idea that the ac Stark shift is responsible for our displaced Ramsey resonances and that the shift is diluted by a factor $F - W_{cd}$ relative to the transit-limited background profile. Certainly higher laser power with still narrower line width would be helpful and presumably ring laser techniques will be attractive in future work. However, as Fig. 7A illustrates, reasonably large shifts are possible even with our modest laser power. It is clear that a quantitative understanding of the intensity shift and its reduction via the "Ramsey dilution effect" are prerequisites to the use of two-photon Ramsey spectroscopy for optical frequency standards and precision spectroscopy. A possibly more serious challenge is presented by the distortion of the Ramsey profiles due to the second-order Doppler effect. Perhaps radiative cooling or other techniques can be used to significantly reduce the kinetic temperature, or perhaps supersonic expansion can be used to narrow the velocity distribution. Another clear area of interest is the influence of thermal fields [21] in depopulating -- and more interestingly -- in shifting the Rydberg levels.

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References

20. Although the width of the observed Ramsey feature can be brought into agreement with the prediction of Eq. (2), an unrealistically small value of G is implied. After the apparatus is modified to reduce the uncertainty in G caused by fringing fields near the Rydberg atom quench region, it will be interesting to pursue a quantitative comparison with theory.
22. These ideas were developed by R.L. Barger in collaboration with one of us (J.L.H.).