High Precision Linewidth Measurement of Laser-Cooled Atoms: Resolution of the Na $3p\,^2P_{3/2}$ Lifetime Discrepancy

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We report resolution of the 1% discrepancy between theory and experiment for the lifetime of the first excited state of Na. Determining the atomic lifetime by a novel method, we have measured the natural linewidth of the $3\,^2S_{1/2} \rightarrow 3\,^2P_{3/2}$ transition with <0.25% accuracy through precision spectroscopy on an optically prepared sample of ultracold, two-level atoms. The resulting $3\,^2P_{3/2}$ lifetime of 16.237(35) ns disagrees with previous time-domain measurements but is in good agreement with recent calculations and a new fast beam result.

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Advancing *ab initio* theory and quantifying its accuracy with experimental tests is important work and will be crucial in the interpretation of high-precision parity nonconservation measurements, particularly for Cs where the anticipated uncertainty will be limited by theory [1]. Precise lifetime measurements of low-lying excited states of alkali atoms provide useful benchmarks for testing atomic theory, especially now that improved theoretical and calculational techniques have produced *ab initio* lifetimes consistent to <0.5%, a level approaching experimental precision. Surprisingly, discrepancies of ≈1% arose for the first excited states of the two simplest alkali systems, lithium and sodium, although recent measurements in lithium are in agreement with theory [2]. For sodium, however, the discrepancy persists [3]. While improved relativistic many body perturbation theory [3] (MBPT) and multiconfigurational Hartree-Fock [4] (MCHF) calculations (two of the most successful *ab initio* techniques) agree at the 0.3% level, they still disagree with early fast beam measurements [5,6] (uncertainties <0.3%) by 1%. More recently, time-correlated photon-counting (TCPC) measurements have supported the fast beam results though their larger error bars (0.5%) make the disagreement with theory less compelling [7,8].

To help resolve such experimental puzzles, it is desirable to find alternative methods of measurement which offer comparable precision but different systematic limitations. We present here a technique in which an atomic lifetime is deduced from a precise measurement of the natural linewidth, and we have applied this method to the $3p\,^2P_{3/2}$ state of Na. The measurement strategy for obtaining the natural linewidth (≈10 MHz FWHM) required that all broadening mechanisms be either reduced or characterized below the desired uncertainty level. Suppression of two mechanisms in particular, Doppler broadening and probe-induced optical pumping, required optical preparation of the atomic sample before probing so we could derive line shapes from velocity-selected, two-level atoms.

The experimental schematic is shown in Fig. 1. A JILA-built ring dye laser, stabilized with an external Fabry-Pérot cavity and locked to an I_2 line, offered a linewidth and long-term frequency stability <25 kHz, and supplied trapping, probing, and optical pumping beams at 589 nm. About 10^7 Na atoms were collected and cooled to <50 μK using a magneto-optic vapor cell trap (MOT) [9] which is described in Ref. [10]. To enable precise control of the optical frequency, light twice diffracted in the first order by an acousto-optic modulator (AOM) provided the rf-tunable probe. Any uncompensated beam steering was suppressed by a polarization-holding single-mode fiber serving as a spatial filter, with the output intensity stabilized to 0.1%. This probe light was then split into two equal beams (<1% intensity imbalance), which passed through quarter-wave plates and were overlapped to produce a (σ^+) circularly polarized standing wave at the interaction region. The frequency of the probe was swept over the $F = 2$, $m = 2 \rightarrow F' = 3$, $m' = 3$ closed cycling transition, and the subsequent 589 nm fluorescence emitted in an $f/4$ cone around 45° to the laser axis was detected by a photomultiplier (PMT).

Since the atoms must be in a perturbation-free environment during probing, a cycling strategy was employed in which the atoms were repeatedly trapped, prepared, and

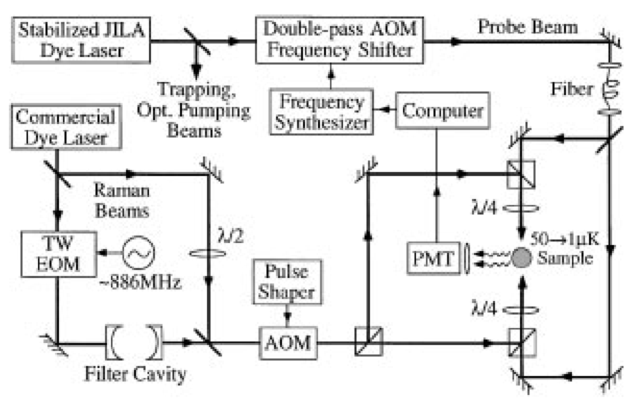

FIG. 1. Block diagram of experiment. Omitted for clarity are trapping and optical pumping beams, intensity-stabilization optics and electronics, and magnetic-field coils.

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probed (see Ref. [10]). A typical measurement cycle began with the MOT turned on for ≈ 1.5 ms, followed by a ≈ 130 μs optical molasses "postcooling" period [11] during which the gradient coils were turned off and the trap beam intensities were lowered for optimal cooling. The trap and molasses beams were then turned off and a 0.5 G magnetic bias field (aligned along the probe axis) turned on for the rest of the measurement cycle. A narrow velocity distribution of atoms was prepared in the $F = 2$, $m = 2$ stretched state by three periods of optical pumping to be described shortly. Next, this prepared atomic sample (5% of total number) was probed for 50 μs during which the PMT current was sampled by a gated integrator. Using a calibrated intensity input, the entire signal collection system was measured to be linear at the $<0.1\%$ level. In one set of linewidth measurements taken at lower collection efficiency, the probe cycle was preceded by a 30 μs normalization period (measured with resonant light), which was used to normalize the probe signal to the number of atoms prepared. A single line shape data point was typically an accumulation of ≈ 1000 identical measurement cycles made at a fixed optical frequency.

Though our atomic sample was cooled to 50 μK by optical molasses, first-order Doppler broadening still contributed $\approx 3\%$ to the linewidth, so we prepared a velocity-selected sample to reduce this contribution. This was achieved by first optically pumping the molasses-cooled atomic sample into the $F = 1$ hyperfine ground state, and then using a one-dimensional velocity-selective Raman transition [12] to transfer near-zero velocity atoms back to the $F = 2$ ground state to be used for probing. A Coherent 699-21 dye laser [13] produced the two Raman beams which were detuned -10 GHz from the D_2 line to prevent single-photon excitation to $F = 2$. To match the optical difference frequency to the Na ground-state hyperfine splitting (1.772 GHz), a traveling-wave electro-optic modulator (TWEOM) generated ≈ 886 MHz sidebands on one of the beams, after which a Fabry-Pérot filter cavity transmitted the $+2$ order. After passing through an AOM, the Raman beams (intensity stabilized to $<0.3\%$ at 200 mW/cm^2) were overlapped with the counterpropagating probe beams via polarizing beam splitters. The Raman transition probability spectrum was determined by the AOM's time envelope, which consisted of a 7 μs square-wave pulse that was filtered to produce a nearly Gaussian Raman line shape with an interaction-time limited FWHM of 140 kHz. This width corresponds to a $v_{\rm rms}$ of <2 cm/s (temperature <1 μK), a factor of ≈ 10 reduction from the original molasses velocity distribution, and its Gaussian shape was chosen to minimize its contribution to our measured Lorentzian linewidths.

An alternative experimental setup configured the Raman beams in a copropagating geometry in which *velocity-insensitive* Raman spectra were generated by sweeping the Raman detuning while keeping the optical probe frequency fixed on resonance. Since in this configuration the Zeeman structure could be easily resolved [extending the Raman interaction time led to narrow (<10 kHz) Raman components], analysis of the Raman spectra enabled the local magnetic field at the trapping region to be precisely set using three orthogonal pairs of Helmholtz coils. Note that in the presence of our magnetic bias field, selection rules for our σ^--σ^- Raman polarizations (in either geometry) allow only $\Delta m = 0$ Raman transitions. The resulting copropagating Raman spectrum thus contained only three peaks whose splittings (0.7 MHz) were proportional to the bias field. Comparison of the linewidth of the $m = 1$ peak with that of the magnetically insensitive $m = 0$ peak enabled a precise characterization of the magnetic field inhomogeneity across our sample (<13 mG—a fit by a Voigt profile yielded values of 7 kHz Lorentzian and 17 kHz Gaussian for the magnetic contribution to our $F = 2 \rightarrow F' = 3$ line shape).

Initially, when probing the $F = 2 \rightarrow F' = 3$ transition, optical pumping redistributed the m-level populations of the lower level in favor of transitions with larger Clebsch-Gordan coefficients. Since optical pumping was more pronounced on top of the peak, it produced non-Lorentzian line shapes with subnatural linewidths. We suppressed these effects by optically pumping our narrow velocity distribution to $F = 2$, $m = 2$ and probing the σ^+ transition to $F' = 3$, $m' = 3$, thereby effectively creating a two-level atom. As an additional benefit, the $F = 2 \rightarrow F' = 2$ transition (which lies -59 MHz away) was no longer accessed, thus simplifying the analysis. Optical preparation of the ultracold atomic sample in the $F = 2$ stretched state occurred in three stages. In the first, a 250 μs period of π light (intensity $I = 40$ mW/cm^2 connecting $F = 2 \rightarrow F' = 2$) and σ^+ light ($I = 30$ mW/ cm^2 connecting $F = 2 \rightarrow F' = 2$ and $I = 1$ mW/cm^2 connecting $F = 1 \rightarrow F' = 1$) pumped $>95\%$ of the atoms into $F = 1$, $m = 1$. We evaluated the efficiency of this first stage of optical pumping by comparing peak heights in the copropagating Raman spectrum. The 7 μs second stage used the Raman beams to transfer a narrow velocity slice from $F = 1$, $m = 1$ to $F = 2$, $m = 1$, with the Raman difference frequency centered on the Zeeman-shifted $m = 1$ resonance so as to excite zero velocity atoms. In the 3 μs final preparation stage, σ^+ light ($I = 30$ mW/ cm^2 connecting $F = 2 \rightarrow F' = 2$) transferred 42% of the velocity-selected atoms to the desired $F = 2$, $m = 2$ state (with minimal photon-recoil heating because it is dark for this excitation), while the remaining 58% returned to the $F = 1$ ground state. Our optically pumped, velocity-selected atomic sample now contained 10^5 atoms in the $F = 2$, $m = 2$ state with a $v_{\rm rms} < 4$ cm/s, ready to be probed.

When we scanned the probe frequency across the $F = 2$, $m = 2 \rightarrow F' = 3$, $m' = 3$ resonance, we observed line shapes (see Fig. 2) which were virtually pure Lorentzians with (1–10)% total measurement-induced

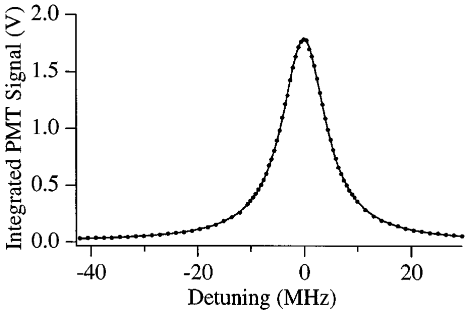

FIG. 2. Experimental line shape for $3s\,^2S_{1/2}(F = 2) \rightarrow 3p\,^2P_{3/2}(F' = 3)$ taken with probe intensity of 50 μW/cm^2. Circles represent experimental data, and solid curve is fit of a Voigt profile (FWHM Lorentzian parameter is 9.94 MHz) to the data. Note that fit is indistinguishable from pure Lorentzian.

broadening. From these line shapes, we derived Lorentzian linewidths from nonlinear least squares fits by a Voigt profile with a fixed Gaussian parameter [$\delta\nu_{\mathrm{FWHM}} = 141$ kHz, associated with velocity-selection width (138 kHz) plus laser width (24 kHz) and magnetic field inhomogeneity (17 kHz)]. One of the dominant broadening mechanisms (beyond the natural width) was due to saturation (saturation intensity $I_0 = 6.3$ mW/cm^2) by our standing wave probe and was linear for this low saturation limit ($I/I_0 < 0.05$). Figure 3 shows the experimental Lorentzian FWHM (with 1σ error bars) as a function of probe power taken with and without a normalization period. Though saturation broadening is responsible for a majority of the slope, detailed Monte Carlo simulations revealed that 15% of this slope can be attributed to diffusive broadening of the atomic velocity

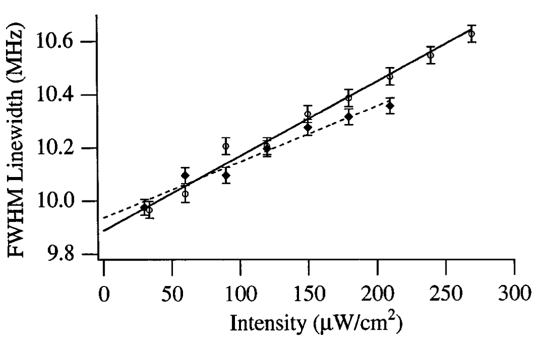

FIG. 3. Graph of FWHM linewidth vs probe intensity with and without normalization. Circles represent data where normalization (intensity equal to probe intensity) preceded probing, resulting in a larger slope due to additional heating. Linear fit to this data (solid curve) extrapolates to a zero-intensity FWHM linewidth of 9.887(21) MHz, which when corrected for 3.0% opacity corresponds to 9.812(27) MHz. Diamonds depict data taken without normalization. Dashed line is a linear fit to this data yielding a zero-intensity FWHM linewidth of 9.935(25) MHz, or 9.845(31) MHz after correction for 3.6% opacity. Adopted experimental result is average, 9.827(21) MHz.

distribution due to photon-recoil heating from the probe. Furthermore, Monte Carlo simulations correctly predict the 30% added to this slope when a normalization pulse precedes the probing due to additional heating of the prepared atomic sample. In both cases the broadening due to the combination of probe heating and saturation linearly extrapolates to the appropriate zero-power linewidth.

The high density offered by our ultracold atomic sample presented a significant optical thickness ($\approx$3% after sample preparation) which differentially attenuated the probe beam versus detuning, leading to broadened line shapes. To determine the effect of the optical opacity, we measured the linewidth as a function of the number of prepared atoms, which was changed by altering the Raman excitation efficiency. An extrapolation to zero optical opacity lead to an $\approx$0.8% correction (the two lines in Fig. 3 were taken at opacities differing by 20%), in excellent agreement with computer simulations. For this measurement, taken at 10^{-7} torr background pressure, other density and number dependent effects such as background and cold atom collisions play a negligible role for our line shapes.

Two less important systematics resulted from incomplete optical pumping. First, atoms excited to $F = 2$ by velocity-insensitive processes broadened the lines by 0.13% due to the larger Doppler contribution of these 50 μK atoms, which contributed 5% of our total signal. Secondly, Zeeman splitting in the Raman spectrum led to excitation of a different velocity class ($v = 30$ cm/s) for atoms left in $m = 0$ after the first round of optical pumping, broadening the resonance by only 0.02%. Remaining systematics contributed at the <0.1% level—Table I summarizes our systematic corrections and their associated uncertainties for our measurement.

Our final result for the full linewidth of the Na $3p\,^2P_{3/2}$ excited state is 9.802(22) MHz (1σ uncertainty), which corresponds to a lifetime of 16.237(35) ns. Figure 4 shows a comparison of this result with recent measurements (with 1σ error bars) and theoretical calculations.

TABLE I. Summary of systematic corrections and uncertainties for our measurement of the lifetime of the $3P_{3/2}$ level of Na (all units in MHz).

Systematic	Linewidth	Correction	Uncertainty
Data run 1	9.887		0.021
For 3.0% opacity		−0.075	0.017
Data run 2	9.935		0.025
For 3.6% opacity		−0.090	0.018
Averaged linewidth	9.827		0.021
Residual 50 μK atoms		−0.013	0.003
B-field inhomogeneity		−0.007	0.001
Residual $m = 0$ atoms		−0.002	0.0003
Laser linewidth		−0.0015	0.003
Residual velocity		−0.0015	0.001
Result	9.802		0.022

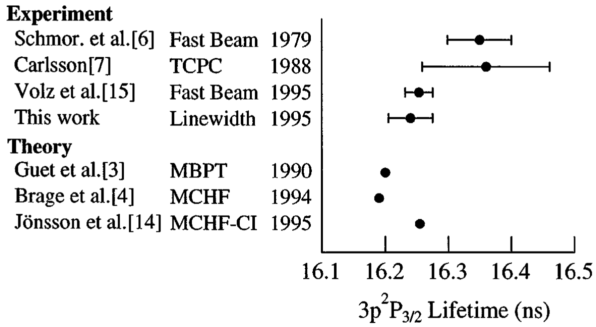

FIG. 4. Comparison of Na $3p\,^2P_{3/2}$ lifetime measurements and calculations. A $3p\,^2P_{3/2}$ lifetime has been derived from the calculation of the total $3s$-$3p$ line strength ($S = 37.26$ a.u.) by Jönssen *et al.* [14]. This derivation assumes an oscillator strength ratio $f(3p\,^2P_{3/2})/f(3p\,^2P_{1/2}) = 2$, consistent with Refs. [3,15].

Here we see that our result disagrees with previous (time-domain) measurements, but is in good agreement with state-of-the-art calculations, especially the recent configuration interaction calculation (MCHF-CI) by Jönsson *et al.* [14]. Furthermore, it agrees extremely well with a very recent fast-beam measurement by Volz *et al.* [15]. These results collectively bring an end to the long-standing Na $3p$ lifetime discrepancy between experiment and *ab initio* theory.

Our result encourages us to put this method forward as an alternative high precision method for measuring atomic lifetimes. This technique is complementary to time-domain methods in that it is ideally suited for broad transitions (>30 MHz) from short-lived levels, whose lifetimes (<5 ns) are difficult to measure precisely in the time domain. Moreover, Doppler broadening effects would be greatly reduced for heavier atoms, so the measurement could be performed (without Raman velocity selection) with a single frequency-stabilized laser. Thus frequency-domain lifetime measurements would be best performed on short-lived levels of reasonably heavy, cooled atoms or ions. Attractive candidates include the first excited state of heavy alkali atoms such as Rb, Cs, or even Fr, for which the lifetime has only been theoretically estimated [16]. Also, the strong singlet S-P transitions in the alkaline earths [such as the 34 MHz natural width $(5s^2)\,^1S_0$-$(5s5p)\,^1P_1$ transition at 461 nm in Sr] are certainly enticing, although second-stage cooling may be required [17]. Finally, laser-cooled trapped ions could provide a good environment for precision linewidth measurements since high-frequency ion traps can greatly suppress recoil effects. For example, the 24 MHz natural width $6\,^2S_{1/2}$-$6\,^2P_{3/2}$ transition at 455 nm in Ba^+ would be an interesting candidate, since the analogous transition in Cs is of particular importance [1]. Moreover, since Ba^+ is itself a parity-violation candidate, precise measurements which test the $6S$-$6P$ *ab initio* wave function would be essential [18].

In summary, we have measured the natural Lorentzian linewidth of an atomic transition at the 0.22% level and have thus demonstrated a new precision technique for determining atomic lifetimes. We have determined an improved value for the Na $3p\,^2P_{3/2}$ lifetime, thus resolving the 1% discrepancy which has existed for this atomic level.

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