Lifetime and oscillator-strength studies involving the $(6s6p)^3P_1$-$(6s7s)^1S_0$ transition of atomic Yb

Y. S. Bai and T. W. Mossberg
Department of Physics, Harvard University, Cambridge, Massachusetts 02138
(Received 15 August 1986)

The oscillator strength of the $(6s6p)^3P_1$-$(6s7s)^1S_0$ transition and the lifetime of the $(6s7s)^1S_0$ state in ^{174}Yb have been measured to be $(8.0\pm1.6)\times10^{-4}$ and 45.8 ± 1.0 nsec, respectively. The former value was obtained by use of coherent optical techniques (optical nutation and ac Stark splitting), while the latter was obtained by monitoring cascade fluorescence following pulsed excitation.

Simple level structure and transitions of convenient wavelength make nuclear-spin-free ^{174}Yb ideally suited for use in studies of basic light-matter interactions. The 555.6-nm $(6s^2)^1S_0$-$(6s6p)^3P_1$ intercombination line provides a natural true two-level system, and when combined with the coupled 611.1-nm $(6s6p)^3P_1$-$(6s7s)^1S_0$ transition an essentially perfect three-level system can be realized. All important parameters related to the former transition have previously been measured.[1,2] This is not true in the case of the latter transition. The purpose of this paper is to report measurements of the latter transition's oscillator strength and the $(6s7s)^1S_0$ state's lifetime. The oscillator strength was measured by two different coherent optical techniques (optical nutation[3-7] and ac Stark splitting[8-12]). These coherent optical techniques differ from conventional techniques for measuring oscillator strength in that information concerning level populations is not required. The lifetime of the $(6s7s)^1S_0$ level was measured by monitoring cascade fluorescence at 399 nm from the $(6s6p)^1P_1$ state following pulsed excitation of the $(6s7s)^1S_0$ state.

The relevant energy levels of ^{174}Yb are shown in Fig. 1. For simplicity, we refer to the levels as $|1\rangle$–$|4\rangle$ as shown. State $|1\rangle$ is the ground state, and the lifetimes of the excited levels $|2\rangle$ and $|4\rangle$ have been measured previously.[1,2] In all of our experiments, the 399-nm fluorescence, hence the population in state $|4\rangle$, was monitored. Since it is really the population in state $|3\rangle$ that is important in our measurements, we must investigate the relationship between the populations in states $|3\rangle$ and $|4\rangle$.

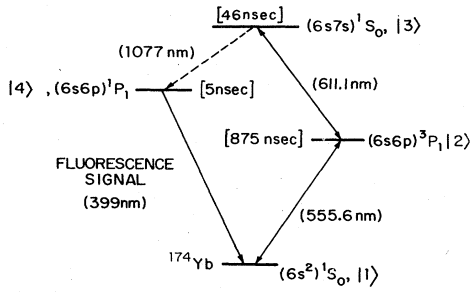

FIG. 1. ^{174}Yb energy levels. The numbers in brackets represent lifetimes.

Assuming that state $|4\rangle$ is populated only via decay of state $|3\rangle$, the population in state $|4\rangle$ obeys the following rate equation:

$$\frac{dN_4}{dt}=-\frac{N_4}{\tau_4}+W_{34}N_3\ , \tag{1}$$

where N_i and τ_i are, respectively, the population and lifetime of level $|i\rangle$, and W_{ij} is the rate of population decay from level $|i\rangle$ to level $|j\rangle$. Our ac Stark splitting measurements are made under steady-state conditions. It follows from Eq. (1) that N_4 and N_3 are strictly proportional, i.e., $N_4=W_{34}\tau_4N_3$. More generally, when N_3 varies with time, Eq. (1) can be formally integrated to give

$$N_4(t)=N_4(0)\exp(-t/\tau_4)+\int_0^t W_{34}N_3(t')\exp[(t'-t)/\tau_4]dt'\ . \tag{2}$$

In the case where $N_3(t)$ is allowed to decay freely, i.e., $N_3(t)=N_3(0)\exp(-t/\tau_3)$, as for example after pulsed laser excitation. The integral can be carried out and N_4 is found to consist of two terms. One term decays at the same rate as N_3, while the other dies away as $\exp(-t/\tau_4)$. Since $\tau_4\ll\tau_3$ in the present system, we can determine τ_3 directly from N_4 decay data provided that it is acquired at a suitable time after the excitation of level $|3\rangle$. The situation is more complicated when one studies an effect like optical nutation in which the population in state $|3\rangle$ displays a damped oscillatory behavior. However, under our experimental conditions and at our level of accuracy in the oscillator-strength measurement (20%), it turns out that the populations in the two states can be treated again as proportional.

The experiment was performed in a Yb atomic beam. Two single-mode cw dye lasers were tuned, respectively, near the 1-2 (556-nm) and 2-3 (611-nm) transition resonances of ^{174}Yb. The laser beams were perpendicular to the atomic beam and were counterpropagating so as to reduce residual Doppler effects resulting from the 2.6 m/sec transverse velocity spread of the atomic beam. A 5-G magnetic field oriented along the 556-nm laser beam propagation direction was applied in the interaction region to split the Zeeman levels in the $(6s6p)^3P_1$ state. The laser beams were circularly polarized (σ_-) and therefore only interacted with the $m=-1$ Zeeman level of the intermediate state. The 399-nm fluorescence was collect-

ed at right angles to both the laser and atomic beams and was detected by a photomultiplier tube. In translating measured powers into intensities, we assume that the beams have Gaussian profiles with $1/e$ intensity diameters given by the measured $1/e$ intensity widths of our beams. All intensities quoted below refer to peak values. In assuming Gaussian beam profiles, we ignore a slight ellipticity actually observed. In the oscillator-strength measurements, the relative beam diameters were adjusted to minimize the importance of knowing the precise transverse profiles. The pulsed excitation sequences, described in some cases below, were generated by acousto-optically gating the output of the cw dye lasers. Repetition rates of pulsed measurements were about 10^4/sec. The risetime of the modulators was 15—20 nsec. Time-dependent fluorescence signals were recorded using a 100-megasample/sec transient digitizer with high-speed averaging capacity.

$(6s\,7s)^1S_0$ LIFETIME MEASUREMENT

In the measurement of τ_3 both lasers were tuned exactly on resonance with their respective transitions. The 556-nm laser was actively locked to the 1-2 transition, while the 611-nm laser was locked to a reference cavity. The laser beams both had diameters of $\simeq$1.5 mm. The excitation sequence consisted of a 556-nm pulse (0.6 W/cm^2, 60-ns duration) which transferred most of the initially ground-state population to the $|2\rangle$ state, and a 611-nm pulse (0.2 W/cm^2, 100-ns duration) which transferred only a small fraction of the $|2\rangle$ state population to state $|3\rangle$. The two excitation pulses were temporally separated (rising edge to rising edge) by 100 ns. A typical decay curve, obtained by averaging about 6.4×10^5 single-shot decay curves, is shown in Fig. 2. In order to eliminate the effect of the finite τ_4 [see discussion after Eq. (2)] and the falling edge of the 611-nm excitation pulse, only those data points more than 50 ns after the maximum fluorescence point were used in deriving the $|3\rangle$ state lifetime. The measurement was repeated many times, yielding $\tau_3=45.8\pm1.0$ ns.

$(6s\,6p)^3P_1$-$(6s\,7s)^1S_0$ TRANSITION OSCILLATOR-STRENGTH MEASUREMENT

ac Stark effect. In this experiment the 2-3 transition was resonantly excited with a strong laser,[12] and the intensity of the 399-nm fluorescence was measured as a function of the frequency of a weak probe laser which was swept through the resonance of the 1-2 transition. As described elsewhere,[9,12] the 399-nm fluorescence should peak at two-probe frequencies separated by χ_{32}, where χ_{32}(rad/sec) is the Rabi frequency at which the 2-3 transition is being driven. Since

$$\chi_{32}(\text{rad/sec})=|\mu_{32}E_2|/\hbar\,,$$

where μ_{32} is the electric dipole moment of the transition excited and E_2 is the electric field amplitude of the 611-nm laser, one can determine μ_{32} by simply measuring E_2 and the splitting in the 399-nm fluorescence spectrum. E_2 is determined from the intensity I_2 of the 611-nm laser according to

$$E_2=(8\pi I_2/c)^{1/2}\,.$$

To achieve measurable splittings in the 399-nm fluorescence spectrum, it was necessary to focus the 611-nm laser to a diameter of 715 ± 25 μm. With the maximum laser power available, this provided a 611-nm intensity of 5.0 W/cm^2. A region of relatively homogeneous 611-nm intensity was selected by focusing the 556-nm laser to a spot size of 500 ± 25 μm. The intensity of the 556-nm laser was set at 0.5 mw/cm^2. The measured 399-nm fluorescence intensity versus probe laser frequency is shown in Fig. 3. The solid line in the figure is a numerically calculated spectrum appropriate to laser beams of Gaussian profile with the $1/e$ diameters mentioned above. The calculation also accounts for residual Doppler effects. The beam-center Rabi frequency χ_{32} used in the calculation was adjusted to obtain the best agreement with the measured spectrum. After analyzing several data sets like that shown in Fig. 3, we find that $\chi_{32}/2\pi=9.8\pm0.2$ MHz. Combining this measurement with our laser power measurement, we deduce that $|\mu_{32}|=(3.1\pm0.3)\times10^{-19}$ statcoulomb-cm. This corresponds to an oscillator

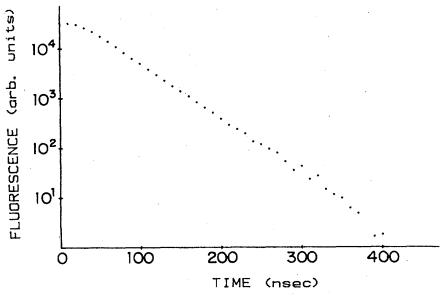

FIG. 2. Lifetime measurement. Vertical scale, 399-nm fluorescence intensity in arbitrary units; horizontal scale, time following pulsed excitation of the $(6s\,7s)^1S_0$ state. The data points between 50 and 300 ns after the fluorescence peak were used to derive the $(6s\,7s)^1S_0$ level lifetime.

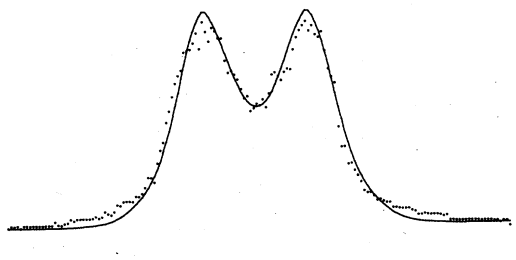

FIG. 3. ac Stark splitting. Vertical scale, 399-nm fluorescence intensity in arbitrary units; horizontal scale, detuning of the 556-nm probe laser about the 1-2 transition resonance frequency, the full scale is 38.4 MHz. The dots represent the data, and the solid line represents a best-fit computer simulation.

strength[13] for the $(6s\,6p)^3P_1$-$(6s\,7s)^1S_0$ transition of $f=(8.0\pm1.6)\times10^{-4}$. We note the uncertainty in our oscillator strength measurement arises primarily from the uncertainties in the power of the 611-nm laser and its transverse beam profile.

Optical nutation. In the nutation measurement, the lasers were tuned and locked precisely as described above for the lifetime measurement. However, beam diameters identical to those of the ac Stark effect experiment were employed. The 611-nm laser was constantly on and had an intensity of 5.0 W/cm^2. The 556-nm laser, on the other hand, was pulsed on (0.3 W/cm^2 intensity, and 20-ns duration) to initiate each optical nutation event. The 399-nm fluorescence was recorded as a function of time after the 556-nm laser pulse, and averaged over typically 6.4×10^5 experimental cycles (see Fig. 4). In general, the relationship between the nutation period $T_{\rm nu}$ and the Rabi period χ_{32} can be complicated.[6,7] However, at the level of accuracy set by our laser power and transverse beam profile measurements, we can simply write $T_{\rm nu}=2\pi/\chi_{32}$. The value of χ_{32} deduced from our optical nutation measurements (Fig. 4) is consistent with the result from the ac Stark effect measurement.

In conclusion, we have measured the $(6s\,6p)^3P_1$-$(6s\,7s)^1S_0$ transition oscillator strength and the $(6s\,7s)^1S_0$ level lifetime of the Yb atom. The coherent optical technique employed to measure the oscillator strength do not require knowledge of level populations, and the optical nutation technique should be applicable[6] in strongly Doppler-broadened systems as well. Potentially high accuracy is possible. In our case, however, a means of accurate absolute laser power measurement was not available.

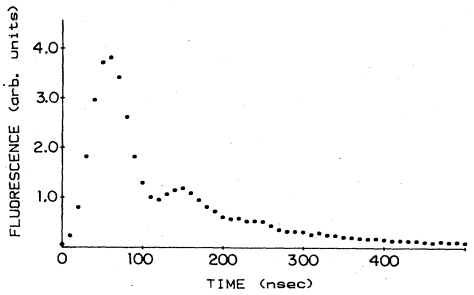

FIG. 4. Optical nutation. Vertical scale, 399-nm fluorescence intensity in arbitrary units; horizontal scale, time in nsec.

ACKNOWLEDGMENTS

We acknowledge financial support from the National Science Foundation under Grant No. PHY85-04260 and from the office of Naval Research under Contract No. N000 14-85-K-0724.

[1]M. Gustavsson, H. Lundberg, L. Nilsson, and S. Svanberg, J. Opt. Soc. Am. **69**, 984 (1979).
[2]F. H. K. Rambow and L. D. Schearer, Phys. Rev. A **14**, 738 (1976).
[3]R. G. Brewer and R. L. Shoemaker, Phys. Rev. Lett. **27**, 631 (1971).
[4]R. L. Shoemaker and E. W. Van Stryland, J. Chem. Phys. **64**, 1733 (1976).
[5]R. L. Shoemaker, in *Laser and Coherence Spectroscopy,* edited by J. I. Steinfeld (Plenum, New York, 1978).
[6]R. G. Brewer, in *Frontiers in Laser Spectroscopy, Les Houches, Session XXVII, 1975,* edited by R. Balian, S. Haroche, and S. Liberman (North-Holland, Amsterdam, 1977).
[7]L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975), Chap. 3.
[8]S. H. Autler and C. H. Townes, Phys. Rev. **100**, 703 (1955).
[9]R. M. Whitley and C. R. Stroud, Jr., Phys. Rev. A **14**, 1498 (1976); H. R. Gray and C. R. Stroud, Jr., Opt. Commun. **25**, 359 (1978).
[10]J. E. Bjorkholm and P. F. Liao, Opt. Commun. **21**, 132 (1977).
[11]P. R. Hemmer, B. W. Peuse, F. Y. Wu, J. E. Thomas, and S. Ezekiel, Opt. Lett. **6**, 531 (1981).
[12]P. T. H. Fisk, H. A. Bachor, and R. J. Sandeman, Phys. Rev. A **33**, 2418 (1986).
[13]W. Demtröder, *Laser Spectroscopy* (Springer-Verlag, Berlin, 1982), p. 60.