Two-photon spectroscopy of the $6S_{1/2} \rightarrow 6D_{5/2}$ transition of trapped atomic cesium

N. Ph. Georgiades, E. S. Polzik,* and H. J. Kimble

Norman Bridge Laboratory of Physics, California Institute of Technology, Pasadena, California 91125

Received March 9, 1994

Two-photon spectroscopy of atomic cesium confined and cooled in a magneto-optical trap is reported. The hyperfine structure of the $6D_{5/2}$ state is determined with 1% accuracy. New capabilities for studying ac Stark shifts and kinetic transport for cold atoms are suggested.

Cold atoms confined in a magneto-optical trap[1] (MOT) are becoming a powerful tool for high-resolution spectroscopy.[2–5] The almost-Doppler-free environment and the high atomic density in a MOT are compelling advantages for using trapped atoms as a sample for spectroscopy. For example, by use of this method high-precision Doppler-less measurements of hyperfine structure in sodium were performed in the research reported in Ref. 4, and two-step excitation in trapped rubidium and cesium was studied in Ref. 5. Following this theme, in this Letter we report the results of two-photon spectroscopy for Cs atoms trapped in a MOT. The transition we have studied is the $6S_{1/2} \rightarrow 6D_{5/2}$ transition excited by two-photon absorption at $\lambda_0 = 883$ nm. Regarding usual Doppler-free two-photon spectroscopy in an atomic vapor,[6] note that the use of atoms in a MOT mitigates the residual Doppler broadening associated with the strong focusing required for most two-photon processes, which usually have relatively small cross sections and hence require rather high intensities.

The atomic energy levels for Cs relevant to our investigation are shown in Fig. 1. Together with the trapping beams for the MOT that are quasi-resonant to the $6S_{1/2} \rightarrow 6P_{3/2}$ transition at $\lambda_T = 852$ nm, the scheme for two-photon excitation effectively forms a V system including the ground state and the two excited states $6P_{3/2}$ and $6D_{5/2}$. By monitoring the fluorescence from the decay of the excited $6D_{5/2}$ state to the intermediate $6P_{3/2}$ level at $\lambda_F = 917$ nm as a function of excitation frequency near λ_0, we determine the excited-state structure of the $6D_{5/2}$ level and hence the magnetic dipole (a) hyperfine structure parameter with an accuracy of ~1%. With respect to earlier research we recall that Tai *et al.*[7] measured the magnetic dipole coefficient (a) by means of magnetic-level crossing in a vapor cell with an accuracy of 30%.[8] In addition to the determination of the excited-state hyperfine structure, we have also used the two-photon transition as a probe to observe ac Stark shifts and power broadening of the ground state caused by the presence of the trapping beams and to record some dynamical effects of thermal motion.

In our experiment we employ a homemade Ti:sapphire laser to produce the radiation P_0 at $\lambda_0 = 883$ nm for two-photon excitation of the $6D_{5/2}$ state. The laser is frequency stabilized relative to a reference cavity[9] (Fig. 2). The reference cavity itself is locked to the fluorescent signal ($6D_{5/2} \rightarrow 6P_{3/2}$) generated by Doppler-free two-photon absorption ($6S_{1/2} \rightarrow 6D_{5/2}$) obtained from an auxiliary Cs vapor cell. Before reaching the Cs cell the locking beam is double passed through two acousto-optic modulators (AOM's). By changing the rf drive frequency to one acousto-optic modulator with an HP8644A synthesized signal generator we can scan the laser frequency by ±40 MHz near λ_0, corresponding to $\Delta\Omega = \pm 80$ MHz detuning for the two-photon transition.

To trap the Cs atoms, we use a MOT in a vapor cell, as first implemented by Monroe *et al.*[1] The trap is sustained by five intersecting laser beams of circular polarization with three in the horizontal plane at 120° separation and two counterpropagating in the vertical (z) direction. A pair of anti-Helmholtz coils aligned along the z axis generates the necessary magnetic-field gradient for the MOT (Fig. 2). The trapping laser P_T is an external-cavity semiconductor diode laser[10] that is frequency locked at ~10 MHz below the $6S_{1/2}$, $F = 4 \rightarrow 6P_{3/2}$, $F = 5$ transition at $\lambda_T = 852$ nm through saturated absorption in an

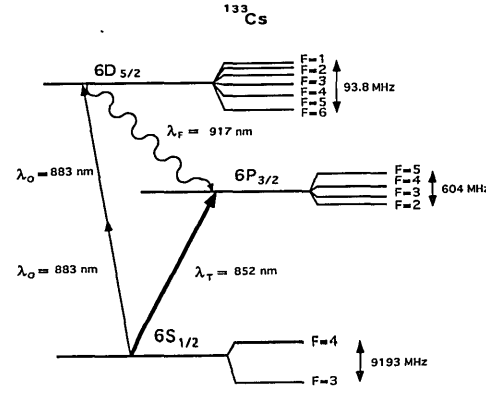

Fig. 1. Relevant energy levels of ^{133}Cs for trapping and for two-photon excitation.

0146-9592/94/181474-03$6.00/0

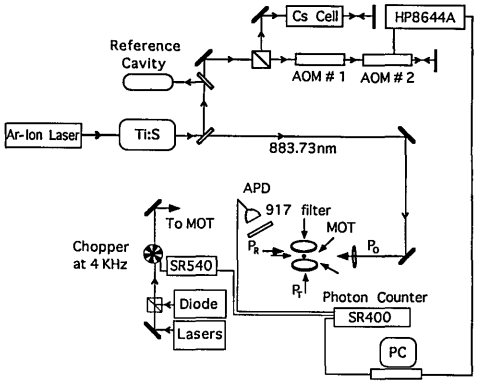

Fig. 2. Diagram of the experimental setup.

auxiliary Cs cell (not shown in Fig. 2). The waist for each beam is fixed at $\omega_T \simeq 4$ mm. A second diode laser, P_R, is operated without an external cavity and is manually tuned into resonance with the $6S_{1/2}$, $F = 3 \rightarrow 6P_{3/2}$, $F = 3$ or 4 transition to prevent depletion of the $F = 4$ ground state. The trap diameter is of the order of 0.5 mm.

We obtain spectroscopic information about the structure of the $6D_{5/2}$ level by observing the fluorescence from the $6D_{5/2} \rightarrow 6P_{3/2}$ transition at $\lambda_F = 917$ nm as a function of excitation frequency near $\lambda_0 = 883$ nm. To optimize the signal without saturating the two-photon transition, we must make sure in the excitation and imaging systems to match the Rayleigh length for the excitation beam to the size of the volume that is imaged. More explicitly, recall that the total fluorescence signal I_F for unsaturated excitation with Gaussian beam is given by $I_F = (2\Gamma_2 P_0^2 n_a \xi/\lambda_0)\arctan(L/2z_o)$, where n_a is the atomic density, P_0 is the excitation power at λ_0, ξ is the overall collection and detection efficiency, L is the length of the imaged volume, and $z_o = \pi\omega_o^2/\lambda_0$ is the Rayleigh length. Here Γ_2 is the two-photon cross section, defined by $R_2 = \Gamma_2 I_0^2$, with R_2 the rate of two-photon excitation for an intensity I_0.

In our experiment the waist size of the excitation laser is chosen to be ~ 7 μm and $L \cong 150$ μm so that $\arctan(L/2z_o) \simeq 0.4$. The saturation power with this geometry for the two-photon transition is measured to be ~ 10 mW, which permits $P_0 \simeq 2$–4 mW. Fluorescence at λ_F is collected by a telescope system consisting of two lenses with N.A. = 0.25 (opening angle 30°) and is focused with 1:1 imaging onto the surface of an avalanche photodiode (APD) with an active area of 150 μm × 150 μm. Two identical interference filters centered at 920 nm protect the avalanche photodiode from the stray light that is mainly at 852 nm. The electrical pulses from the avalanche photodiode are counted by an SR400 photon counter that is interfaced with a personal computer (PC).

To observe the spectra unperturbed by the trapping beams (while also recording the effect of this perturbation), we mechanically chop the trapping and repumping beams at 4 kHz and observe two-photon excitation spectra with the trapping beams both on and off. By gating the photon counting system accordingly, we are then able to obtain in a single-scan excitation spectra with the trapping beams both on and off. Excitation spectra are acquired by variation of the drive frequency to the acousto-optic modulator (and hence the laser frequency) to scan across the entire hyperfine structure of the $6D_{5/2}$ level at a rate of typically 0.05 Hz. An example of such a spectrum is shown in Fig. 3, in which two-photon excitation proceeds from the $F = 4$ component of the ground $6S_{1/2}$ level to five hyperfine components with $F = 6, 5, 4, 3, 2$ of the excited $6D_{5/2}$ level. Note that the two-photon excitation spectra taken in the on state permit observation of the effect of the strong trapping beams on the Cs ground state, complementing nicely other types of trapped-atom spectroscopy.[2–5]

The magnetic dipole a and electric quadruple b coefficients of the $6D_{5/2}$ state are determined from spectra similar to those in Fig. 3 recorded over a range of experimental conditions for the trapping beams, the atomic density, and the excitation power P_0 at λ_0. Spectra with the trapping and repumping lasers chopped off are fitted to multi-Lorenzian functions to extract line positions, amplitudes, and widths for each of the observed hyperfine components. From the fitted splittings of the hyperfine peaks [see, e.g., Eq. (1) of Ref. 7] we then calculate with a least-squares procedure the coefficients $a = -4.69 \pm 0.04$ MHz and $b = 0.18 \pm 0.73$MHz, which are to be compared with those of Ref. 7, namely, $a = -3.6 \pm 1.0$ MHz and no evidence for a nonzero value of b. Within our quoted uncertainties no observable dependence of the splittings on the intensity of the trapping beams has been found for single-beam intensities between 0.8 and 3 mW/cm^2. To address the issue of possible Zeeman shifts of the line positions, we have compared splittings of the various $6D_{5/2}$ hyperfine components and have also measured hyperfine splittings at two different magnetic-field gradients (6.2 and 4.8 G/cm in the z direction) and again have found no changes within our stated uncertainty.

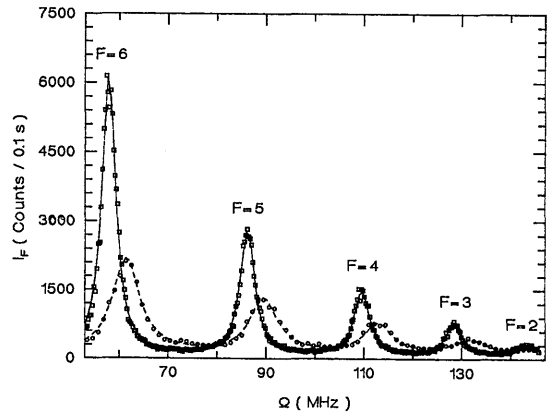

Fig. 3. Two-photon excitation spectra for $P_0 = 3.3$ mW showing the fluorescence I_F at λ_F versus excitation frequency Ω (cycles/s) near $\lambda_0 = 883$ nm with the trapping and repumping beams chopped on (circles) and off (squares). Stark shifts and power broadening of the ground state resulting from the presence of the trapping beams are clearly evident, here with a total trapping power of 3.1 mW. Note that Ω refers to the two-photon detuning (i.e., twice the actual frequency change near λ_0).

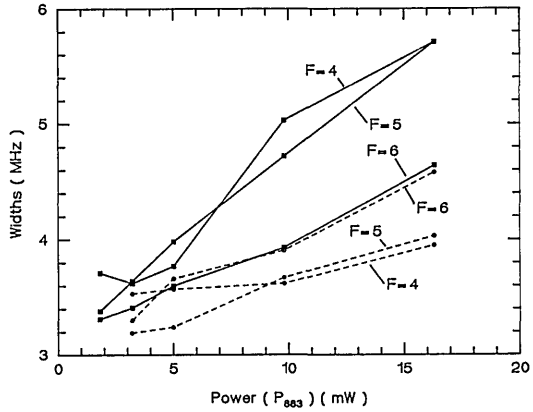

Fig. 4. Linewidths (FWHM) of the $F = 6, 5, 4$ hyperfine components of the $6D_{5/2}$ level versus the excitation power P_0 with the trapping beams off and the repumping beam either off (solid curves) or on (dashed curves).

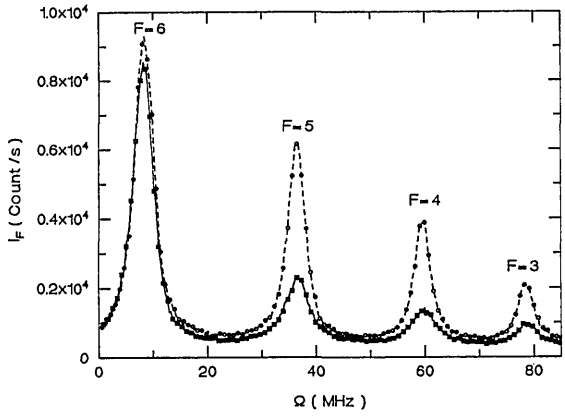

Fig. 5. Two-photon excitation spectra for $P_0 = 16$ mW showing fluorescence I_F versus detuning Ω obtained with the repumping and trapping beams off (squares) and with the repumping beam on and the trapping beam off (circles).

In addition to line positions, the peak widths have been measured as a function of excitation power P_0, with the broadening exhibited in Fig. 4 due to saturation of the two-photon transition. At low power the widths of the resonances extrapolate to 3.2 ± 0.2 MHz. Considering residual Doppler broadening from the finite trap temperature, assumed to be the Doppler cooling limit $T_D = 120$ μK, we obtain a value for the natural linewidth of 3.1 ± 0.2 MHz. The discrepancy between our measurement and the theoretical prediction of 2.5 MHz (Ref. 11) may be attributable to fluctuations in laser frequency (linewidth ~500 kHz), to inhomogeneities in the trap environment (especially spatially dependent Zeeman splittings), to temperatures in excess of T_D, and/or to uncertainties of the theoretical value itself. One interesting feature of our measurements in Fig. 4 is the dependence of the line broadening of the $F = 4$ and $F = 5$ states on the presence or absence of the repumping beam, whereas the widths of the $F = 6$ state seem insensitive to this.

To investigate this point further, we show in Fig. 5 two spectra, one obtained with the repumping beam on and the other with the repumping beam off. The difference in relative heights of the $F = 5, 4, 3$ components between the two curves is due to the fact that, without the repumping beam, the $F = 4$ component of $6S_{1/2}$ is depleted as a result of the decay of the $F = 5, 4, 3, 2$ components of $6D_{5/2}$ to the $F = 3$ component of $6S_{1/2}$. Indeed, without the repumping beam the transitions with $F = 5, 4, 3, 2$ should normally have near-zero amplitudes since the time to deplete the $F = 4$ ground level under the conditions of Fig. 5 is only $\sim 10^{-6}$ s, compared with the 10^{-4}-s interval of the off cycle during which the spectra are acquired. That significant amplitudes are observed in Fig. 5 without the repumping beam is principally because of thermal motion, which brings fresh atoms from the trap into the small excitation volume. Further studies of spectra such as those in Fig. 5 with systematic variations in trap parameters as well as in the size of the excitation volume might be of interest in providing a relatively nonintrusive probe of the trap kinetics.

In conclusion, we have investigated two-photon spectroscopy of cold trapped Cs atoms and have determined the a coefficient for the $6D_{5/2}$ state with 1% accuracy, which represents a 30-fold improvement over the results of Ref. 7. We have observed ac Stark shifts for the ground state of cold atoms (Fig. 3) and have demonstrated a possible avenue for exploring transport processes in the trap (Figs. 4 and 5).

We gratefully acknowledge contributions of H. Tsuchida and correspondence from S. Svanberg. This study was funded in part by the U.S. Army Research Office and the National Science Foundation. E. S. Polzik acknowledges support from Aarhus University.

*Present address, Institute of Physics and Astronomy, Aarhus University, Aarhus C, DK-8000, Denmark.

References

1. E. L. Raab, M. Prentiss, A. Cable, S. Chu, and D. Pritchard, Phys. Rev. Lett. **59,** 2631 (1987); C. Monroe, W. Swann, H. Robinson, and C. Wieman, Phys. Rev. Lett. **65,** 1571 (1990).
2. J. W. R. Tobosa, G. Chen, Z. Hu, R. B. Lee, and H. J. Kimble, Phys. Rev. Lett. **66,** 3245 (1991); D. Grison, B. Lounis, C. Salomon, J. Y. Courtis, and G. Grynberg, Europhys. Lett. **15,** 149 (1991).
3. For a recent summary of spectroscopic measurements in optical lattices see Phys. Today **46**(6), 17 (1993).
4. M. Zhu, C. W. Oates, and J. L. Hall, Opt. Lett. **18,** 1186 (1993).
5. R. W. Fox, S. L. Gilbert, L. Hollberg, and J. H. Marquardt, Opt. Lett. **18,** 1456 (1993).
6. L. S. Vasilenko, V. P. Chebotaev, and A. V. Shishaev, Sov. Phys. JETP Lett. **12,** 13 (1970).
7. C. Tai, W. Happer, and R. Guta, Phys. Rev. A **12,** 723 (1975).
8. For work on other excited states in Cs see G. Belin, L. Holmgren, and S. Svanberg, Phys. Scr. **14,** 39 (1976).
9. R. W. P. Drever, J. L. Hall, F. V. Kowalski, J. K. Hough, G. M. Ford, A. J. Munley, and H. Ward, Appl. Phys. B **31,** 97 (1983).
10. K. Libbrecht and J. L. Hall, Rev. Sci. Instrum. **64,** 2133 (1993).
11. O. S. Heavens, J. Opt. Soc. Am. **51,** 1058 (1961).