DOPPLER-FREE TWO-PHOTON LASER SPECTROSCOPY OF HgII*

J. C. Bergquist, D. J. Wineland, Wayne M. Itano, Hamid Hemmati**,
H.-U. Daniel†, and G. Leuchs††

Time and Frequency Division
National Bureau of Standards
Boulder, Colorado 80303

Abstract

The Doppler-free, two-photon $5d^{10}\ 6s\ ^2S_{1/2}-5d^9\ 6s^2\ ^2D_{5/2}$ transition in singly ionized Hg, attractive as an optical frequency standard, has been observed for the first time. A few $^{198}Hg^+$ ions were confined in a radio-frequency (rf) trap and the two-photon transition was detected by monitoring the change in the fluorescence light scattered by the ions from a laser beam tuned to the first resonance transition at 194 nm. The radiative lifetime of the $^2D_{5/2}$ state and the absolute wavenumber of the two-photon transition were measured to be 0.090(15) s and 17 757.152(3) cm^{-1} respectively.

Introduction

Microwave or optical transitions of laser cooled ions that are confined in electromagnetic traps offer the basis for frequency standards of high stability and accuracy.[1-5] The advantages of such devices are numerous: very long interrogation times and, therefore, high transition line Q's can be achieved; fractional frequency perturbations that are due to the trapping fields can be held below 10^{-15}; collisions with background gas and cell walls can be largely avoided; Doppler shifts are directly reduced by trapping and cooling and, finally, nearly unit detection efficiency of transitions to metastable states is possible so that the signal to noise ratio need be limited only by the statistical fluctuations in the number of ions that make the transition.[5] Details of ion traps and laser cooling have been published elsewhere.[1-3] A particularly attractive candidate for an optical frequency standard is the two-photon allowed (or single photon, electric quadrupole allowed) $5d^{10}\ 6s\ ^2S_{1/2}-5d^9\ 6s^2\ ^2D_{5/2}$ Hg^+ transition near 563 nm.[4,5] The lifetime of the $^2D_{5/2}$ state, expected to decay by emission of electric quadrupole radiation, is calculated to be of order 0.1 s.[6] This gives a potential optical line Q of about 7×10^{14}. Here we describe the first results of our investigation of the two-photon transition in $^{198}Hg^+$ (which is free of hyperfine structure) stored in a miniature rf trap.

Experiment

Our trap is similar to the small radio frequency traps used in the ion cooling experiments that were conducted at Heidelberg University on Ba^+,[7] and at the University of Washington on Mg^+.[8] A cross section of the trap electrodes is shown in Fig. 1. We note that, although the inner surfaces of our trap electrodes were machined with simple conical cuts, the trap dimensions were chosen to make the fourth and sixth order anharmonic contributions to the potential vanish.[9] The rf drive frequency was 21 MHz with a voltage amplitude, $V_0 \leq 1$ kV. The background pressure, excluding deliberately added mercury and helium, was $\leq 10^{-7}$ Pa (133 Pa ≡ 1 torr). After loading 50-200 mercury ions, the mercury vapor was frozen out in a liquid nitrogen cold trap and the vacuum vessel was back-filled with He to the order of $10^{-3}-10^{-2}$ Pa. This was sufficient to collisionally cool the trapped Hg^+ to near room temperature as verified by the Doppler width of the $^2S_{1/2}-^2P_{1/2}$ resonance line near 194 nm.

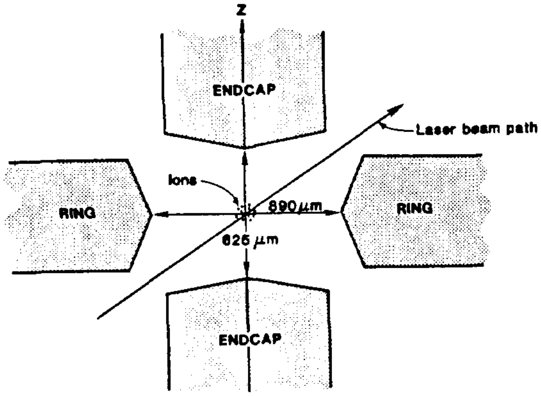

Fig. 1. Schematic showing cross section view of trap electrodes. The electrodes are figures of revolution about the z-axis and are made from molybdenum.

Optical double-resonance[10,11] was used to detect the two-photon transition. About 5 μW of narrowband cw sum-frequency-generated radiation near 194 nm[12] was tuned to the 6s $^2S_{1/2}$ to 6p $^2P_{1/2}$ first resonance transition and was directed diagonally through the trap (between the ring electrode and the end caps). The fluorescence light scattered by the ions was detected at right angles to the 194 nm beam with an overall detection efficiency of about 10^{-4}. Typically, our signal level was $2-10 \times 10^3$ counts/s and the signal to

---

*Work of the U.S. Government; not subject to U.S. copyright.

**Present address: Allied Bendix Aerospace Corporation, Columbia, MD

†Present address: Springer Verlag, Heidelberg, West Germany

††Heisenberg Fellow of the Deutsche Forschungsgemeinschaft.
Present address: Max Planck Institute für Quantenoptik, Garching, West Germany

background ratio was better than 10/1. When the ions were driven by the radiation from a 563 nm cw ring dye laser out of the $^2S_{1/2}$ ground state into the $^2D_{5/2}$ metastable state, there was a decrease in the 194 nm fluorescence corresponding to the number of ions in the D state.

The dye laser beam also was directed diagonally through the trap; the axes of the dye laser beam, the 194 nm beam and the collection optics were mutually perpendicular. A near-concentric standing wave cavity was placed around the trap in order to enhance the power of the 563 nm radiation and to better ensure nearly equal intensity counter-propagating beams. The cavity was positioned so that its waist ($w_0 \cong$ 25-30 μm) was located near the center of the cloud of trapped ions. The power buildup factor was approximately 50, giving nearly 5 W of circulating power for typical input power levels of 100 mW. The ring dye laser linewidth in these preliminary experiments was of the order of 300 kHz. The frequency of the laser was offset locked and precisely scanned with respect to a second dye laser locked to a hyperfine component in the Doppler-free, saturated absorption spectrum of $^{127}I_2$.

Results

A typical resonance curve and simplified energy level diagram is shown in Fig. 2. The full scanwidth is 4 MHz. The electric field vector of the 563 nm laser radiation is nearly parallel to a small applied magnetic field of approximately 11.6 x 10^{-4} T (11.6 G) which differentially Zeeman splits the ground and excited states. The selection rule for the two-photon transition for this polarization is $\Delta m_J = 0$, and, thus, only two components are observed, separated by approximately 13 MHz (approximately 6.5 MHz at the dye laser frequency). In Fig. 2, we scan over only one of these Zeeman components ($m_J = -1/2 \leftrightarrow m_J = -1/2$) but see sideband structure. This structure is due to amplitude modulation (AM) of the 563 nm laser intensity due to the secular motion of the ions in the rf trap.[13] To our knowledge, this is the first observation of secular motion sidebands at optical frequencies. The depth of the central feature in Fig. 2 is nearly 25% of full scale implying that we have nearly saturated the two-photon transition. The linewidth is about 420 kHz, and is determined in nearly equal parts by the laser linewidth of about 320 kHz and the power broadening by the 194 nm radiation of nearly 270 kHz. When the 194 nm radiation is chopped, the two-photon linewidth drops to approximately 320 kHz.

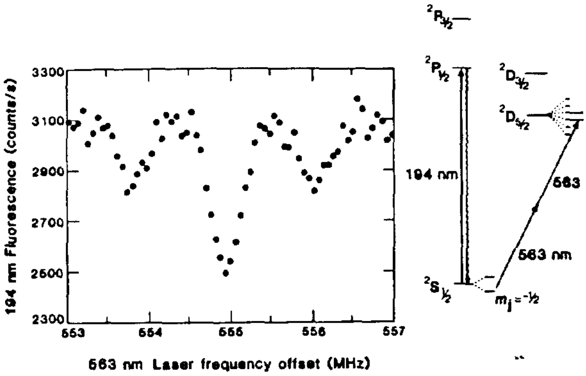

Fig. 2. Two-photon $^2S_{1/2}$-$^2D_{5/2}$ transition in $^{198}Hg^+$. AM sidebands caused by the harmonic secular motion of the ions are visible in this scan. The frequency scan is 4 MHz at the fundamental laser frequency (λ ≅ 563 nm). The depth of the central component is about 25% of full scale. The integration time is 2 s/point. In the inset is a simplified energy level diagram of $^{198}Hg^+$, depicting the levels of interest.

We have experimentally measured the radiative lifetime of the $5d^9\ 6s^2\ ^2D_{5/2}$ state to be 0.090(15) s in good agreement with the calculated lifetime of 0.105 s.[6] Again in this measurement, the ground state population was monitored by measuring the laser induced fluoreseence of the 194 nm transition. The radiation from the dye laser near 563 nm was tuned to resonance with the two-photon transition and chopped on and off. During the time that the laser radiation was on, it drove 10-20% of the ion population into the D state. The time constant for the atomic system to relax during the radiation-off period could be determined from the exponential return of the 194 nm fluorescence to steady state. An example is shown in Fig. 3. The relaxation rate was measured over a range of He pressures differing by a factor of four. The reported radiative lifetime is the result of an extrapolation to zero pressure of a linear least-squares fit to the data. The pressure-induced decay rate was determined poorly, but amounted to only about 25% of the radiative decay rate at the highest pressure (about 6 x 10^{-2} Pa).

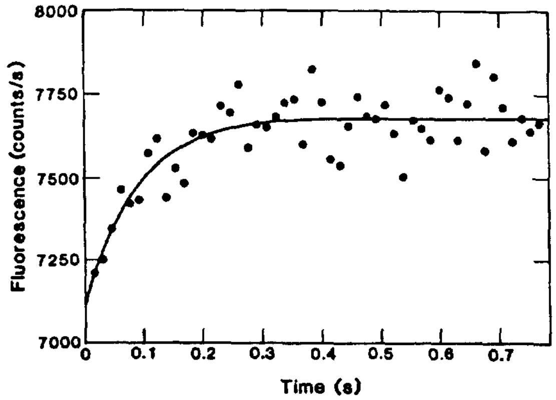

Fig. 3. Measurement of the radiative lifetime of the $5d^9\ 6s^2\ ^2D_{5/2}$ state in Hg^+. The dots are the experimental points indicating the return of the atomic system to steady state after chopping off the radiation from the 563 nm dye laser that drives the ions from the ground state to the metastable D state by two-photon absorption. The solid line is a least squares fit of an exponential to the experimental data. The vertical axis is the counts per second obtained by measuring the laser induced fluorescence from the continuously driven $^2P_{1/2}$-$^2S_{1/2}$ transition. The horizontal axis shows time after turning off the 563 nm laser radiation.

We have also measured the absolute wavenumber of the $^2S_{1/2}$-$^2D_{5/2}$ two-photon transition by measuring the frequency difference between the two photon resonance and the "t" hyperfine component of the nearby R(33) line of the 21-1 band in $^{127}I_2$ (line #1220 in the iodine atlas).[14,15] The two-photon transition in $^{198}Hg^+$ lies 551(2) MHz to the red of this component. From this we determine the wavenumber of the two-photon transition to be 17 757.152(3) cm^{-1}.

In the near future, we anticipate narrowing the 563 nm laser linewidth to the order of a few kHz and studying various systematic effects including pressure broadening and shifts, power broadening, and light shifts. Ultimately, we would like to narrow the laser linewidth to a value near that imposed by the natural lifetime of the D state, and to drive the two-photon (or single photon, electric quadrupole) transition on a single, laser-cooled ion.

Acknowledgments

We gratefully acknowledge the support of the US Air Force Office of Scientific Research and the US Office of Naval Research. We wish to thank R. Blatt (University of Hamburg) for technical assistance in the construction of the rf trap and H. Layer (NBS) for supplying the ^{198}Hg. We take particular pleasure in acknowledging the help of W. Martin, J. Reader and C. Sansonetti (NBS) who provided the correct wavelength for the $^2D_{5/2}-^2S_{1/2}$ transition. We also wish to thank D. Huestis (SRI) and our colleague R. Drullinger for many useful conversations and insights.

References

[1] H. G. Dehmelt, Adv. At. Mol. Phys. 3, 53 (1967); H. G. Dehmelt, Adv. At. Mol. Phys. 5, 109 (1969); H. G. Dehmelt, IEEE Trans. Instrum. Meas. IM-31, 83 (1982).

[2] D. J. Wineland, W. M. Itano, and R. S. Van Dyck, Jr., Adv. At. Mol. Phys. 19, 135 (1983).

[3] D. J. Wineland, W. M. Itano, J. C. Bergquist, J. J. Bollinger, and J. D. Prestage, in "Atomic Physics 9", R. S. Van Dyck, Jr. and E. N. Fortson, eds. (World Scientific Publ. Co., Singapore, 1984) p. 3.

[4] P. L. Bender, J. L. Hall, R. H. Garstang, F. M. Pichanick, W. W. Smith, R. L. Barger, and J. B. West, Bull. Am. Phys. Soc. 21, 599 (1976).

[5] D. J. Wineland, J. C. Bergquist, R. E. Drullinger, H. Hemmati, W. M. Itano, and F. L. Walls, J. Phys. (Orsay, Fr.) 42 C8-307 (1981); D. J. Wineland, W. M. Itano, J. C. Bergquist, and F. L. Walls, Proc. of 35th Annu. Symp. on Freq. Control, (1981) p. 602 (copies available from Electronic Industries Assoc., 2001 Eye St., NY, Washington, DC 20006).

[6] R. H. Garstang, J. Research NBS 68A, 61 (1964).

[7] W. Neuhauser, M. Hohenstatt, P. Toschek, and H. G. Dehmelt, Phys. Rev. Lett. 41, 233 (1978).

[8] W. Nagourney, G. Janik, and H. G. Dehmelt, Proc. Natl. Acad. Sci. USA 80, 643 (1983).

[9] E. C. Beaty, to be published.

[10] D. J. Wineland, J. C. Bergquist, W. M. Itano, and R. E. Drullinger, Opt. Lett. 5, 245 (1980).

[11] H. G. Dehmelt, in "Advances in Laser Spectroscopy", F. T. Arecchi, F. Strumia, and H. Walther, eds., (Plenum, New York, 1983) p. 153.

[12] H. Hemmati, J. C. Bergquist, and W. M. Itano, Opt. Lett. 8, 73 (1983).

[13] H. A. Schuessler, Appl. Phys. Lett. 18, 117 (1971); F. G. Major and J. L. Duchene, J. Phys. (Orsay) 36, 953 (1975); H. S. Lakkaraju and H. A. Schuessler, J. Appl. Phys. 53, 3967 (1982); M. Jardino, F. Plumelle, and M. Desaintfuscien, in "Laser Spectroscopy VI", H. P. Weber and W. Lüthy, eds. (Springer-Verlag, NY, 1983) p. 173; L. S. Cutler, R. P. Giffard and M. D. McGuire, Appl. Phys. B36, 137 (1985).

[14] S. Gerstenkorn and P. Luc, Atlas du Spectre d'absorption de la Molecule de L'iode Entre 14,800-20,000 cm^{-1} (Editions du C.N.R.S., 15, quai Anatole-France, 75700 Paris, 1978).

[15] S. Gerstenkorn and P. Luc, Rev. Phys. Appl. 14, 791 (1979).