

Energy and Radiative Lifetime of the $5d^96s^2\ ^2D_{5/2}$ State in Hg II by Doppler-Free Two-Photon Laser Spectroscopy

J. C. Bergquist, D. J. Wineland, Wayne M. Itano, Hamid Hemmati,^(a) H.-U. Daniel,^(b)
and G. Leuchs^(c)

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

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The Doppler-free, two-photon $5d^{10}6s\ ^2S_{1/2} - 5d^96s^2\ ^2D_{5/2}$ transition in singly ionized Hg, attractive as an optical-frequency standard, has been observed for the first time on a small number of $^{198}\text{Hg}^+$ ions confined in a radio-frequency trap. The radiative lifetime of the $^2D_{5/2}$ state and the absolute wave number of the two-photon transition were measured to be 0.090(15) s and $17757.152(3)\text{ cm}^{-1}$, respectively. Optical amplitude-modulation sidebands, induced by the secular (thermal) motion of the harmonically bound ions, were observed also for the first time.

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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 $5d^{10}6s^2\ ^2S_{1/2} - 5d^96s^2\ ^2D_{5/2}\text{Hg}^+$ transition driven either by two photons with a wavelength near 563 nm⁴ or by one photon at half this wavelength.⁵ The lifetime of the $^2D_{5/2}$ state, which decays by the emission of electric quadrupole radiation at 281.5 nm, is calculated to be of order 0.1 s.⁶ This gives a potential optical-line Q of about 7×10^{14} . In this Letter we describe the first results of our investigation of the two-photon transition in $^{198}\text{Hg}^+$ (which is free of hyperfine structure) stored in a miniature rf trap.

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^+ and Ba^+ .⁸ A cross section of the trap electrodes is shown in Fig. 1. It is interesting to 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 partial pressure of the background gas, with the exception of

deliberately added mercury and helium, was $\leq 10^{-7}$ Pa (133 Pa \cong 1 Torr). After loading of 50–200 mercury ions the mercury vapor was frozen out in a liquid-nitrogen cold trap and the vacuum vessel was backfilled with He to the order of 10^{-3} – 10^{-2} Pa. This was sufficient to cool the trapped Hg^+ collisionally to near room temperature as verified by the Doppler width of the $^2S_{1/2} - ^2P_{1/2}$ resonance line near 194 nm.

Optical double resonance^{10,11} was used to detect the two-photon transition. About 5 μW of narrow-band cw sum-frequency-generated radiation near 194 nm¹² 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} . Our signal level depended on the ion number and temperature and, typically, was $(2-10) \times 10^3$ counts/s. The signal-to-background ratio was better than 10/1. When the ions

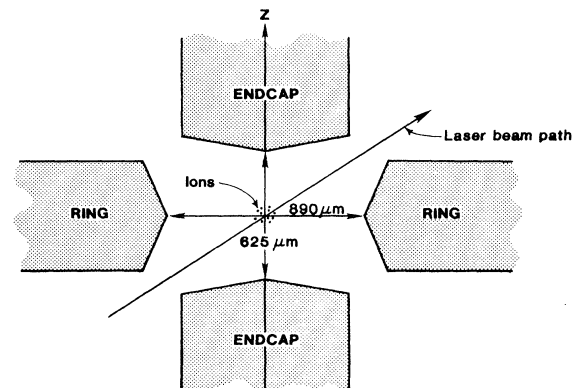


FIG. 1. Schematic showing a cross-section view of the trap electrodes. The electrodes are figures of revolution about the z axis and are made from molybdenum.

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 was also 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 better to ensure a nearly equal intensity of the counterpropagating beams. The cavity was positioned so that its waist ($w_0 \cong 25\text{--}30 \mu\text{m}$) 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 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 $^{127}\text{I}_2$. The linewidth of the swept laser was determined to be of the order of 300 kHz by spectrum analysis of the beat note between the two lasers.

A typical resonance curve and simplified energy-level diagram is shown in Fig. 2. The full scan width is 4 MHz. The electric field vector of the 563-nm laser radiation is nearly parallel to a small applied magnetic field of approximately $11.6 \times 10^{-4} \text{ T}$ (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 ($m_J = -\frac{1}{2} \leftrightarrow m_J = -\frac{1}{2}$)

but see sideband structure. This structure is due to amplitude modulation of the 563-nm laser intensity due to the secular motion of the ions in the rf trap. Any frequency modulation of the laser caused by the motion of the ions¹³ is strongly suppressed if the cavity is well aligned so that the k vectors of the counter-running beams are antiparallel. By a change in the well depth of the trap the harmonic frequency of the secular motion is changed, thereby shifting the sideband components of the two-photon signal. To our knowledge, this is the first observation of secular-motion sidebands at optical frequencies. In principle, the ion temperature could be derived from the relative amplitude of the sidebands.¹³ 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. For the data of Fig. 2, the 194-nm laser irradiates the ions continuously, and, since the Zeeman splitting is unresolved for the first resonance transition, it mixes the ground state, maintaining equal populations in the two electron spin states. The linewidth of the two-photon resonance is about 420 kHz, and is determined in nearly equal parts by the 563-nm laser linewidth of about 320 kHz and by the nearly 270-kHz excitation rate of the $^2S_{1/2}$ state by the 194-nm radiation. When the 194-nm radiation is chopped, the two-photon linewidth drops to approximately 320 kHz, consistent with the dye-laser linewidth. The pressure broadening of the resonance in Fig. 2 was estimated to be less than 50 kHz by observation of the resonance at 5 times higher He pressure.

We have experimentally measured the radiative lifetime of the $5d^96s^2D_{5/2}$ state to be 0.090(15) s (one standard deviation) in agreement with the calculated lifetime of 0.105 s.⁶ Again, in this measurement the ground-state population was monitored by measure-

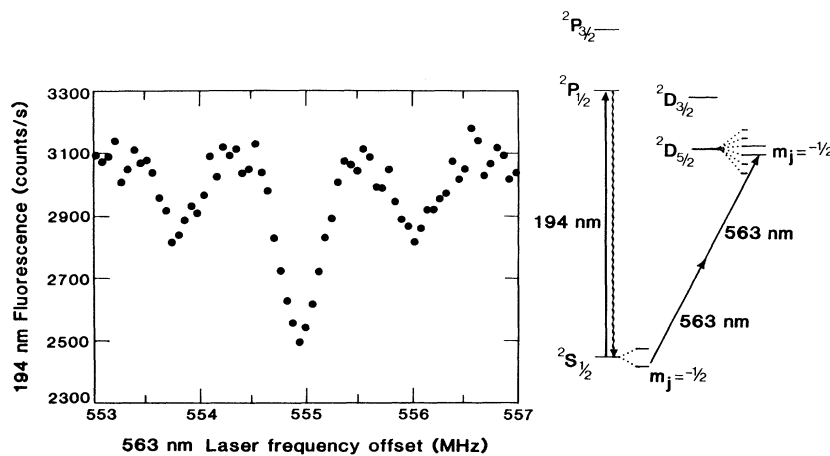


FIG. 2. Two-photon $^2S_{1/2}$ - $^2D_{5/2}$ transition in $^{198}\text{Hg}^+$. Amplitude modulation by the harmonic secular motion of the ions is visible in this scan. The frequency scan is 4 MHz at the fundamental laser frequency ($\lambda \cong 563 \text{ nm}$). The depth of the central component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

ment of the laser-induced fluorescence 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 4. 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 6×10^{-2} Pa).

We have also measured the absolute wave number 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 $^{127}\text{I}_2$ (line No. 1220 in the iodine atlas).¹⁴ At zero magnetic field, the two-photon transition in $^{198}\text{Hg}^+$ lies 551(2) MHz to the red of this component. To a fairly good approximation, the center of gravity for the $R(33)$ line is situated at a point $\frac{5}{9}$ the distance between the t and b hyperfine components.¹⁵ From this approximation, we find that the two-photon signal is 998(5) MHz to the red of the center of gravity of this iodine line. Thus, the meas-

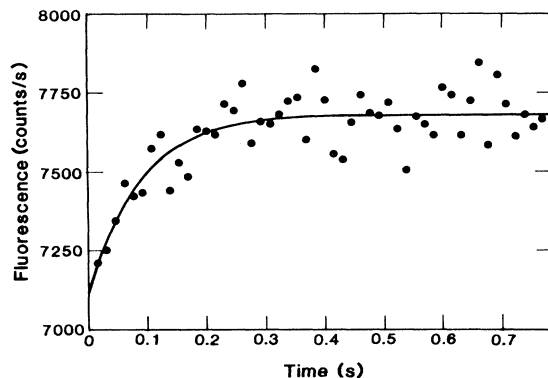


FIG. 3. Measurement of the radiative lifetime of the $5d^96s^2^2D_{5/2}$ state in Hg^+ . The dots are the experimental points indicating the return of the atomic system to steady state after chopping off of the radiation from the 563-nm dye laser that drives the ions from the ground state to the metastable 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 measurement of the laser-induced fluorescence from the continuously driven $^2P_{1/2} - ^2S_{1/2}$ transition. The horizontal axis shows time after the turning off of the 563-nm laser radiation.

ured wave number is $17757.1522(20) \text{ cm}^{-1}$ as determined relative to the Gerstenkorn and Luc value of $17757.1855(20) \text{ cm}^{-1}$ for the center of gravity of the $R(33)$ 21-1 line in iodine.^{14,15} Tellinghuisen finds a consistent discrepancy of the order of $0.002\text{--}0.003 \text{ cm}^{-1}$ between his predictions for the line center of gravity and those of Gerstenkorn and Luc for lines with odd J values in this spectral region (there is good agreement for lines with even J values).¹⁶ Tellinghuisen calculates a value for the line center of gravity of $17757.182(2) \text{ cm}^{-1}$. Using this value, we find the two-photon transition to be at $17757.149(2) \text{ cm}^{-1}$. These values are in good agreement with the direct wavelength measurement of the electric quadrupole emission of the $^2D_{5/2} - ^2S_{1/2}$ transition obtained from a mild electrodeless discharge in a ^{198}Hg cell backfilled with about 700 Pa of neon and from a standard ^{198}Hg cell backfilled with 33 Pa of argon.¹⁷ The wavelength measured in standard air is $2814.9389(5) \text{ \AA}$, which corresponds to a vacuum wave number of $17757.146(3) \text{ cm}^{-1}$ for the two-photon transition. In this Letter, we report the wave number of the two-photon transition to be $17757.152(3) \text{ cm}^{-1}$. The quoted uncertainty allows for the discrepancy in the determinations of the iodine-line center of gravity.

In the near future, we anticipate narrowing the 563-nm laser linewidth to the order of a few kilohertz 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 this two-photon, or single-photon, electric quadrupole transition on a single, laser-cooled ion. Single-photon excitation of the $^2D_{5/2}$ state is particularly attractive because light shifts are eliminated. Light shifts are expected to contribute the largest uncertainty to the resonance frequency for two-photon excitation.⁵

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(a) Present address: Allied Bendix Aerospace Corporation,

Columbia, Md.

^(b)Present address: Springer-Verlag, Heidelberg, West Germany.

^(c)Present address: Max-Planck-Institute für Quantenoptik, Garching, West Germany.

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