Frequency Standards in the Optical Spectrum*

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1. Introduction

A long standing problem of spectroscopic measurements in the optical region of the spectrum has been the difficulty of obtaining accurate frequency reference standards to which spectral measurements can be compared. As will be discussed in this symposium on the hydrogen atom, this affects the value of the Rydberg constant as derived from optical measurements in hydrogen. As the experiments on hydrogen improve, the need for better reference points becomes more acute.

In this paper, we address two aspects of this general problem. First, we discuss the problem of frequency standards in the optical spectrum. (An analogue in the microwave region of the spectrum is the cesium beam frequency standard.) If one or a few of these reference frequencies can be accurately calibrated (perhaps by a frequency synthesis chain[1]) then it may be possible to compare optical spectra to these standards. As an example of the precision that might be achieved, we discuss only optical standards based on stored ions. Second, we discuss the problem of frequency comparison of unknown frequencies to the standards. Here we primarily restrict discussion to generation of wideband frequency "combs".

2. Optical Frequency Standards

One way an optical standard could be provided is by harmonic multiplication of a microwave frequency standard in a synthesis chain. By use of this technique, a laser at 88 THz (3.39 μm) has been made phase coherent with a microwave oscillator.[1] The best optical frequency standards may be made by locking a local oscillator (laser) to an atomic or molecular resonance line. State-of-the-art accuracies are characterized by measurements on methane stabilized He-Ne lasers in which reproducibilities in the 10^{-13} range have

* Supported by the U.S. Air Force Office of Scientific Research and the U.S. Office of Naval Research. Contribution of the U.S. National Bureau of Standards; not subject to U.S. Copyright.

** Supported by the DFG.

The Hydrogen Atom Editors: G.F. Bassani · M. Inguscio · T.W. Hänsch

been reported.[1] There are many other schemes for optical frequency standards.[1-3] Here, we will only discuss possibilities with single stored ions in order to give an idea of what stabilities and accuracies might be achieved.

3a. Single Ions

The use of single trapped ions as optical frequency standards was suggested by Dehmelt[4] in 1973. Since that time, demonstrations of laser cooling,[5] 100% detection efficiency,[6-8] and lasers with sub-Hz resolution,[1,9] have increased confidence that a single-ion frequency standard is viable. As an example, we summarize the current status[10] and discuss future prospects using single Hg^+ ions. Similar experiments using different kinds of ions have been reported at Seattle,[4,6] and Hamburg,[7,11] and other labs are initiating experiments.

In the NBS work, the transition of interest for a frequency standard is the Hg^+ $5d^{10}6s\ ^2S_{\frac{1}{2}} \rightarrow 5d^9 6s^2\ ^2D_{5/2}$ quadrupole transition at 281.5 nm shown in Fig. 1. The $^2D_{5/2}$ level has a lifetime of 86 ms, corresponding to a natural width of 1.8 Hz. Use of the single photon quadrupole transition has an advantage over two-photon Doppler free transitions[12] because ac Stark shifts are negligible.

A mercury atom that was ionized by a weak electron beam was captured in a miniature Paul (radio frequency) trap that has internal dimensions of $r_o \cong 466$ μm and $z_o \cong 330$ μm. The rf trapping frequency was 21.07 MHz with a peak voltage amplitude of about 730 V. The ion was laser cooled by a few microwatts of cw laser radiation that was frequency tuned below the $6s\ ^2S_{\frac{1}{2}} - 6p\ ^2P_{\frac{1}{2}}$ electric dipole transition near 194 nm. When the Hg^+ ion was cold and the 194 nm radiation had sufficient intensity to saturate the strongly allowed S-P transition, 2×10^8 photons/s were scattered. With our collection efficiency, this corresponded to an observed peak count rate of about $10^5\ s^{-1}$ against a background of less than $50\ s^{-1}$.

Optical-optical double-resonance utilizing quantum amplification[4,6-8] was used to detect transitions of the ion driven by the 281.5 nm laser to the metastable $^2D_{5/2}$ state. This method makes use of the fact that the 194 nm fluorescence intensity level is bistable; high when the ion is cycling between the S and P states (the "on" state) and nearly zero when it is in the metastable D state (the "off" state). The fluorescence intensity in the on state is high enough that the state of the atom can be determined in a few milliseconds with nearly 100% efficiency. The full measurement cycle was as follows: A series of measurements of the 194 nm fluorescence was made, using

a counter with a 10-ms integration period. As soon as the counter reading per measurement period was high enough to indicate that the ion was in the on

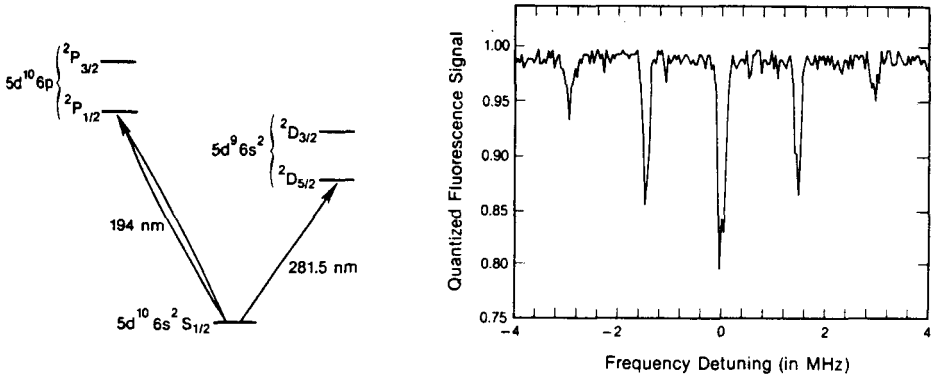

Fig. 1. On the left is a simplified energy-level diagram for $^{198}Hg^+$. The 281.5 nm quadrupole "clock" transition can be observed by monitoring the 194 nm fluorescence. If the ion has made a transition from the $^2S_{\frac{1}{2}}$ to the $^2D_{5/2}$ level the 194 nm flourescence disappears. For the figure on the right, on the horizontal axis is plotted the relative detuning from line center in frequency units at 281.5 nm. On the vertical axis is plotted the probability that the fluorescence from the 6s $^2S_{\frac{1}{2}}$ - 6p $^2P_{\frac{1}{2}}$ first resonance transition, excited by laser radiation at 194 nm, is on immediately after the 281.5 nm pulse. The electric-quadrupole-allowed S-D transition and the first-resonance S-P transition are probed sequentially in order to avoid light shifts and broadening of the narrow S-D transition. The recoilless absorption resonance or carrier (central feature) can provide a reference for an optical frequency standard. (From ref. 11)

state, the 194-nm radiation was shut off and the 281.5 nm radiation was pulsed on for 20 ms. Then, the 194 nm radiation was turned on again, and the counter was read. If the reading was low enough to indicate that the ion had made a transition to the $^2D_{5/2}$ state (the off state), the signal was defined to be 0. Otherwise, it was defined to be 1. The 281.5 nm laser frequency was then stepped, and the cycle was repeated. As the 281.5 nm laser frequency was swept back and forth through the resonance, the quantized measurement of the fluorescence signal at each frequency step was averaged with the previous measurements made at that same frequency. Since we could

detect the state of the ion with nearly 100% efficiency, there was essentially no instrumental noise in the measurement process. Occasionally, while the 194 nm radiation was on, the ion decayed from the $^2P_{\frac{1}{2}}$ state to the metastable $^2D_{3/2}$ state rather than directly to the ground state.[13] This process led to a background rate of false transitions which was minimized by the quantized data-collecting method described above and by decreasing the 194 nm fluorescence level thereby decreasing the $^2P_{\frac{1}{2}}$-$^2D_{3/2}$ decay rate. Neglecting decay to the $^2D_{3/2}$ level, the quantized measurement scheme removes any contribution to the signal base line due to intensity variations in the 194-nm source. The 281.5 and 194 nm radiation were chopped so that they were never on at the same time. This eliminated shifts and broadening of the narrow 281.5-nm resonance due to the 194 nm radiation.

Figure 1 shows the signal from an 8 MHz scan of the 281.5 nm laser through a Zeeman component of the ion. The central feature or "carrier" can be the reference for an optical frequency standard. The laser could be servoed to this resonance by probing on both sides of the resonance to develop an error signal which forces the average frequency of the laser to the center of the resonance. Two primary requirements for good performance are: (1) immunity from and ability to characterize systematic frequency shifts (accuracy) and (2) sufficient signal-to-noise ratio and line Q to reach the accuracy level in a reasonable length of time (stability).

3b. Stability

If we make the simplifying assumption that the time-domain Ramsey method is used to interrogate the clock transition, then the frequency stability (two-sample Allan variance[14]) is given as[15]

$$\sigma_y(\tau) \equiv [\langle(\langle\omega_k\rangle_\tau - \langle\omega_{k+1}\rangle_\tau)^2\rangle_k/2\omega_0^2]^{\frac{1}{2}} = (2\omega_0^2 T_R \tau)^{-\frac{1}{2}} . \qquad (1)$$

In this expression, $\langle\omega_k\rangle_\tau$ is the kth measurement of the frequency of the locked oscillator averaged over time τ, $\langle\ \rangle_k$ denotes an average over many measurements, ω_0 is the nominal frequency of the clock transition, and T_R is the interrogation time between pulses in the Ramsey method. Thus $\sigma_y(\tau)$ is a measurement of the rms fluctuations of the average frequency between adjacent measurements of duration τ. Eq. 1 assumes that optical state preparation (cooling and optical pumping) and fluorescence detection takes a time much less than T_R. We have also assumed $T_R \ll \tau(D_{5/2})$ and that the frequency of the 281.5 nm laser changes by much less than $(2T_R)^{-1}$ in time T_R. For our Hg^+ example, assuming T_R = 25 ms, $\sigma_y(\tau) \cong 7 \times 10^{-16}\tau^{-\frac{1}{2}}$. Therefore, at averaging

times of 1 s, the measurement imprecision on a single ion should be on the order of 1 part in 10^{15} or 1 Hz. Even better performance is expected on ions with narrower linewidths. To realize such stabilities, very narrow, tunable lasers are required but this technology is now available.[9]

3c. Accuracy

The advantage of single ions over other experimental configurations is the relative immunity from systematic frequency shifts. Laser cooling gives the lowest second order Doppler shifts for single (as opposed to many) trapped ions.[16] In the Doppler cooling limit,[5] the magnitude of the second order Doppler shift for Hg^+ ions in an rf trap is fractionally 2.4×10^{-18}. Laser cooling to the zero point energy, which has recently been achieved,[17] gives even smaller shifts. One must also account for all perturbing influences on the ion's internal structure.[4,18] For example, one must consider the perturbations due to static and time varying multipole interactions for electric, magnetic, and gravitational fields. These include atom trapping field interactions, collisions with neutral atoms, ac Stark shifts due to laser beams, stray electric and magnetic fields, gravitational red shifts, etc.

For the $^2S_{\frac{1}{2}} \rightarrow {}^2D_{5/2}$ quadrupole transition in Hg^+, the limiting accuracy may be due to the uncertainty in the interaction of the $^2D_{5/2}$ atomic quadrupole moment with static electric fields of quadrupole symmetry.[4,18] The interaction with the quadrupolar electric fields of the trap can be calibrated since they affect the ion oscillation frequencies of the trap in a known way. More difficult to control are the effects due to patch fields or stray charge build up. Shifts may be on the order of 0.1 Hz. We can significantly reduce the uncertainty due to these shifts, however, by taking the mean quadrupole transition frequency for three mutually orthogonal orientations of a quantizing magnetic field. In this case, the mean quadrupole shift is zero. For the $^1S_0 \rightarrow {}^3P_0$ transitions of the group IIIA ions, such shifts are absent.[4]

Although experimental verification is still lacking, it would appear that single ion optical frequency standards will eventually yield extremely high performance. Accuracies and stabilities better than 1 part in 10^{15} seem quite feasible; eventually they could exceed 1 part in 10^{18}.

4. Frequency Comparison

To appreciate the problem of frequency comparison in the optical region of the spectrum one needs only to plot the electromagnetic spectrum on a linear

frequency scale. The number (and spectral density) of accurate optical frequency standards is very small to begin with; this is to be compared with the situation in the microwave spectrum (and below) where commercially available synthesizers provide state-of-the-art precision at any frequency.

A laser whose frequency is unknown can be compared to a reference laser by heterodyne methods to high precision.[1] Beat frequency measurements up to 2.5 THz in the visible spectrum have already been made.[19] An alternative to simple heterodyne schemes is harmonic mixing by use of synthesis chains.[1] Here, the unknown laser frequency is compared (via heterodyne methods) to a harmonic of some well known reference line such as the methane stabilized He-Ne laser at 3.39 μm.[1]

Comparisons can also be made by wavelength methods. These have the advantage that two lasers of quite different frequency can be compared but are currently limited in accuracy to about the 10^{-10} to 10^{-11} range.[20]

For high accuracy comparison, heterodyne detection and synthesis chains are proven methods but have not been demonstrated in the visible yet. Moreover, since they are somewhat cumbersome and their coverage in the optical spectrum somewhat sparse, it is useful to pursue alternative methods.

5a. Wideband Comb Generation

Precise frequencies relative to a frequency standard (at frequency ω_0) can be provided by comb generation. If we amplitude or frequency modulate (at frequency Ω) a source at the standard frequency, then spectral components at frequencies $\omega_0 \pm n\Omega$ (n an integer) are generated. If n and Ω are chosen so that $\omega_0 + n\Omega \cong \omega$(unknown) then ω(unknown) can be precisely determined by heterodyne methods. The challenge is to make $(n\Omega)$ very large.

5b. Amplitude Modulation

As a specific example, let

$$A(t) = \sin(\omega_0 t + \varphi)\, f(t), \qquad (2)$$

where f(t) is periodic with period $2\pi/\Omega$. We will assume f(t) is an even function and near t = 0, has the form

$$f(t) \cong \exp[-(t/\tau)^2] \qquad \left(-\frac{\pi}{\Omega} < t < \frac{\pi}{\Omega}\right), \qquad (3)$$

where we also assume $\tau \ll 2\pi/\Omega$. (See Fig. 2) Expanding A(t) in a Fourier series, we find

$$A(t) \cong \frac{\Omega\tau}{2\sqrt{\pi}} \left\{ \sin(\omega_0 t+\varphi) + \sum_{n=1}^{\infty} \exp(-(n\Omega\tau/2)^2)\ (\sin[(\omega_0+n\Omega)t+\varphi_0] + \sin[(\omega_0-n\Omega)t + \varphi_0]) \right\}. \quad (4)$$

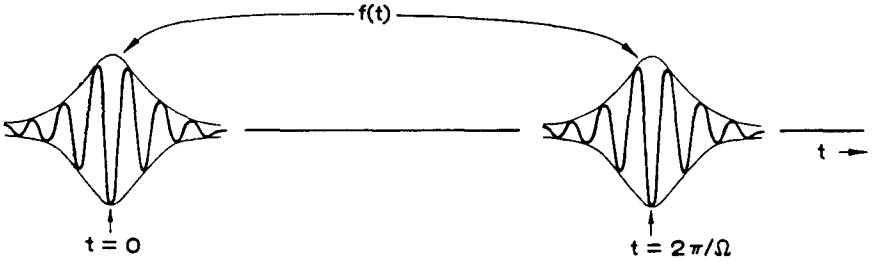

Fig. 2. A sine-wave carrier (frequency ω_0) is assumed to be periodically amplitude modulated at frequency Ω with a Gaussian envelope. Exact frequency division (ω_0/Ω = integer) is obtained if the phase of the carrier is fixed to the phase of the modulation envelope.

To make the spectral component at $\omega_0 \pm n\Omega$ as large as possible we want $n\Omega\tau/2$ close to unity. For harmonic generation, $\omega_0 = \Omega$. For either harmonic generation or comb generation around a high frequency ω_0, the comb bandwidth is approximately equal to $1/\tau$. High order multiplication to the visible or large bandwidth combs in the visible are difficult to achieve for two reasons: (1) it is difficult to make τ small enough and (2) the modulator must be sufficiently broadband to pass the fundamental and its harmonics or sidebands.

We remark that Fig. 2 is characteristic of the output of a mode locked laser[21] or a free electron laser with pulsed input electron beam.[22] Mode-locked lasers have already been suggested as devices for frequency translation in the optical spectrum.[21]

5c. Frequency Modulation

Consider sine wave modulation of a carrier at frequency ω_0:

$$A(t) = A_0 \sin[\omega_0 t + \frac{\Delta\omega}{\Omega} \sin\Omega t + \varphi]$$
$$= A_0 \sum_{n=-\infty}^{\infty} J_n(\frac{\Delta\omega}{\Omega}) \sin[(\omega_0 + n\Omega)t + \varphi], \quad (5)$$

where $\Delta\omega$ is the frequency swing. Assume we are interested in the spectral component at a particular (large) value of n. We make the substitution $\Delta\omega/\Omega = n\beta$, and will limit ourselves to the case where $\beta < 1$. From Meissel's formula[23] for $J_n(n\beta)$, for large n and $\beta < 1$, we can write

$$J_n(n\beta) \cong \left[\frac{\beta e^{(1-\beta^2)^{\frac{1}{2}}}}{1+(1-\beta^2)^{\frac{1}{2}}}\right]^n [2\pi n(1-\beta^2)^{\frac{1}{2}}]^{-\frac{1}{2}} . \quad (6)$$

We can make the spectral component at $\omega_0 + n\Omega$ reasonably large by making β close to unity or the modulation index $\Delta\omega/\Omega$ close to n. This is the primary practical problem for n large.

6a. Frequency Division

Using amplitude or frequency modulation of a carrier at frequency ω_0, we can achieve exact frequency division if we make sidebands of the carrier to such low frequency that we can force the condition $\omega_0 - n\Omega = (n+2)\Omega - \omega_0 = \Omega$ so that $\omega_0/\Omega = n+1$. For example, if we examine Fig. 2, we can achieve exact frequency division by any means which locks the phase of the carrier to the phase of the amplitude modulation; that is, the undulations of the carrier do not "slip" under the envelope of the amplitude modulation. A divider based on these principles would be quite useful if Ω is in the microwave region (or below) where precise frequency synthesis is possible. Since Ω and n could be freely chosen, any value of ω_0 could be measured in a single device.

6b. Single Electron Frequency Divider

The required extreme amplitude or frequency modulation might be accomplished with the interaction of an electromagnetic wave (at frequency ω_0) with a single electron oscillating at frequency Ω. For example, Ω might be the cyclotron frequency of the electron in a magnetic field.

As originally proposed,[24] extreme amplitude modulation of the electromagnetic wave (at the site of the electron) could be accomplished by

focussing the wave with a fast lens or slow wave structure (miniature waveguide) onto a portion of the electron's cyclotron orbit. The energy coupled into the electron at frequency $\omega_0-n\Omega = (n+2)\Omega-\omega_0 = \Omega$ is balanced by synchrotron radiation and phase locking can occur. Lens focussing requires the electron to be relativistic, $\gamma \equiv (1-(v/c)^2)^{-\frac{1}{2}} \geq 2$ to achieve a significant coupling[24] (v = electron speed, c = speed of light). Use of a slow wave structure would allow use of lower energy electrons.

Driving the electron could also be accomplished using an inverse synchrotron radiation geometry.[25] Here, a collimated laser beam is made to coincide with a tangent to the electron's orbit. We require the electron to be relativistic to achieve a significant interaction. If the laser beam waist is made much larger than the electron orbit dimensions, then we have the case of extreme frequency modulation of a plane wave as first analyzed by Kaplan.[26] Here, the electric field at the site of the electron is given by Eqs. 5 and 6 with $\beta = v/c$. Again, we require the electron to be somewhat relativistic to make J_n and J_{n+2} sufficiently large ($\omega_0-n\Omega = \Omega$).

In all cases above, the center of the electron's orbit (in the plane of the orbit) must be held within $\lambda\!\!\!^{-}(= c/\omega_0)$. A specific scheme for accomplishing this is discussed in Ref. 24. At NBS we have initiated a project along these lines. At first, low order division ($n < 10$) where ω_0 and Ω are both microwave frequencies will be tried. Aside from the practical application discussed here, such an experiment is interesting in terms of the fundamental nonlinear interactions of simple elementary systems.

We thank L. Hollberg and D. Jennings for helpful comments on the manuscript.

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