LIMITATIONS ON LONG-TERM STABILITY AND
ACCURACY IN ATOMIC CLOCKS*

D. J. Wineland
National Bureau of Standards
Boulder, Colorado

ABSTRACT

The limits to accuracy and long-term stability in present atomic clocks are examined. In order to achieve a significant increase in performance, it appears that the limitations must be attacked on a fundamental level. For instance, the problem of residual first-order and second-order Doppler shifts has for many years been approached by asking how we can better measure these shifts. A more fundamental approach might be to ask how we can significantly lower the velocity of the atoms.

An attempt will be made to put recent proposals for new frequency standards into perspective. The advantages and disadvantages of frequency standards based on such ideas as laser transitions, single atoms, and atom cooling are examined. In addition, the applicability of some of these new techniques to existing standards is discussed.

INTRODUCTION

This paper attempts to answer the question: "What new ideas can lead to fundamental improvements in atomic frequency standards?" Since my answer can't be totally objective, what follows is vulnerable to criticism; nevertheless, it will be useful to examine some of these new ideas and speculate on what they might lead to.

This paper is not a survey of all new ideas for frequency standards; rather, some of these new ideas are used as examples to illustrate general areas in which fundamental problems might be attacked. Also, one will notice that the application of some of these ideas are impractical at the present time for a field usable standard, but most could be realized in a laboratory so that they may have more immediate application to a "primary" standard.

To make the problem somewhat more tractable, it will be assumed that the important question to ask is how to improve accuracy and that the improved long-term stability will naturally follow if the accuracy can be improved. I contend that this is generally true if not pushed too far; for example, if a way were found to drastically reduce wall shift, spin exchange shift, second-order Doppler shift, etc. in the H-maser then the fluctuations of these effects (which limit long-term stability) will also be reduced. We must also of course assume that we can improve the signal-to-noise so that the anticipated accuracy increase can be measured in a reasonable length of time.

In any case, the approach will be to ask not how we can better measure those effects in atomic clocks which limit their accuracy and long-term stability, but how we can get rid of them.

Later we will briefly ask what new ideas are likely to improve atomic clocks based on hydrogen, rubidium and cesium. More importantly, it will be interesting to look at other ideas for "atomic" clocks. First, however, it is useful to reexamine the ground rules—that is, what features do we really want in a frequency standard?

BASICS OF ATOMIC FREQUENCY STANDARDS

The requirements for making a good frequency standard are fairly simple[1]:

(1) it must be reproducible, and

(2) it must be "reasonably usable," in the sense that it should have an output frequency easily used in measurements.

The first requirement implies that bulk devices (such as quartz crystal resonators, macroscopic rigid rotors, or superconducting cavity oscillators) are undesirable, because the frequency depends on parameters, such as size, that are difficult to control. This shortcoming does not, however, rule out the use of these devices as calibrated "flywheels" (i.e., free running, stable clocks).

Atomic (or molecular) resonances provide the necessary reproducibility; one derives a "standard" frequency ν_o in terms of the energy difference between two states of the atom with energies E_1 and E_2 by the relation $h\nu_o = E_2 - E_1$ where h is Planck's constant. To ensure reproducibility between different observations, the measured frequency is usually referred to the value that would be obtained in free space; consequently, various corrections are necessary to take account of environmental factors, such as magnetic fields. The problem then reduces to correcting for the

various perturbations to the measured frequency. Our task in this paper is to ask how we can significantly reduce (or adequately resolve) these perturbations.

Of course, perturbations at some level will always exist so we will also ask how we can reduce their influence. In many cases, the perturbations to the measured resonance frequency are proportional to Q^{-1} where $Q = \nu_0/\Delta\nu$, and $\Delta\nu$ is the width (in frequency units) of the energy difference measured at the half power points; therefore a high Q is desirable. Also, all measurements are limited in precision by various sources of noise. The fractional frequency stability $\sigma_y(\tau)$ relates to Q and signal-to-noise by

$$\sigma_y(\tau) \doteq \left[Q \, \frac{S}{N} \, (\tau) \right]^{-1}$$

where $S/N(\tau)$ is the signal-to-noise ratio as a function of averaging time τ.

Satisfying the second requirement depends on technological limitations and may rule out some interesting frequency-standard possibilities. The output of the device (i.e., the operating frequency) must be convenient for general application. At the present time, this rules out for example, the use of certain transitions that can be observed at very high frequencies such as those in nuclei. Although the Q of these Mössbauer transitions can be as high as 10^{15},[2] they are not generally usable because neither frequency nor wavelengths can be accurately measured in the gamma-ray region. In general, we can say that if we have a frequency standard which operates at a frequency ν_0, we must be able to divide this frequency down (or multiply up a reference oscillator for comparison) in order to use the standard as a clock — that is, provide timing

signals. (We note however, that the use of the standard as a clock is not needed in some applications; for example, the gravitational red shift was first measured intercomparing the frequencies of two spatially separated samples using Mössbauer transitions.[2])

We can summarize our criteria for a good general frequency standard as:

(a) S/N large.

(b) Q large.

(c) Small perturbations to the frequency.

(d) Must be able to measure frequency.

REALIZATION OF THE CRITERIA

We, of course, quickly learn that it is not easy to satisfy all these criteria simultaneously. Some of the reasons for this are fundamental, some are practical.

The resolution and Q is fundamentally limited by the Heisenberg uncertainty relation on time and energy:

$$\Delta E \Delta t \gtrsim \hbar/2.$$

Thus, for a single atom, if we have a time Δt to measure the energy-level difference $E_2 - E_1$, the measurement will be uncertain by at least an amount ΔE. Specifically, Δt may arise from the time of flight (transit time) of an atom through the apparatus, or from the lifetime of the atom in one or both of its energy states.

The uncertainty relation yields a fractional uncertainty in energy of $\Delta E/(E_2 - E_1) = Q^{-1}$. We can, of course, make ΔE small by making Δt large; this may be accomplished by slowing down the atoms as much as possible or by confining them. Unfortunately, the process of confinement causes perturbations such as the wall shift in the H-maser and buffer gas shifts as in the Rb frequency standard.

Also, there is often a trade-off between signal-to-noise and Q. Extremely high Q does not guarantee a good frequency standard because, if the signal-to-noise ratio is small, it may take an impractical length of time to realize the accuracy. Conversely, we can increase S/N at the expense of Q, as in gas cell standards, where S/N can be increased by increasing number, but we also increase perturbations due to atom-atom collisions.

An important category of perturbations which exists in all frequency standards to varying degrees is that of Doppler shifts. Doppler effects are related to the particular method of confinement. They represent perhaps the most important problem limiting the accuracy and resolution of existing or proposed frequency standards. In the usual way we can say that if an absorber of radiation moves relative to the source, the observed resonance frequency is shifted to the value

$$\omega_{abs} \cong \omega_o + \vec{k}\cdot\vec{v} - \frac{1}{2}\,\omega_o\left(\frac{v}{c}\right)^2 + \frac{\hbar\omega_o^2}{2Mc^2}$$

where the velocity $\vec{v}$ and wave vector $\vec{k}$ are measured relative to the source and M is the atomic mass. The first-order Doppler shift ($\vec{k}\cdot\vec{v}$), the "second-order" Doppler shift, ($(v/c)^2$) and the recoil shift (the last term) can be understood in terms of conservation of energy and momentum in the absorption process. The

so-called "second-order" Doppler shift is merely the relativistic time-dilation factor resulting from the movement of the atom relative to the apparatus. Its effect is small but important, particularly if we are talking about improving the state of the art. We can describe the first-order shift in terms of time dependence of a plane electromagnetic wave as seen by an atom. We have for the electric field

$$\vec{E}(x,t) = \vec{E}_0 \sin (\vec{k} \cdot \vec{x} - \omega t + \phi)$$

where $\vec{x}$ is the atom position, $\vec{k}$ is the field wave vector ($\vec{k} \cdot \vec{E}_0 = 0$), and ϕ is an arbitrary phase factor. If $\vec{x} = \vec{v}_x t$ then

$$\vec{E}(t) = \vec{E}_0 \sin [(\vec{k} \cdot \vec{v}_x - \omega)t + \phi]$$

and the particle sees a sinusoidally varying field of frequency $\omega' = \omega - \vec{k} \cdot \vec{v}_x$. If uncompensated, the result is the familiar Doppler broadening because, typically, the atoms in a sample have a Maxwellian velocity distribution leading to a distribution of ω' values. If one observes the full Doppler width, then the Q of the transition is limited to about 10^6 for room-temperature samples. If the particle is confined within dimensions $|\vec{x}| < k^{-1}$ (the so-called "Dicke regime")[3] the resonance spectrum has a sharp central feature with natural width (Fig. 1).

This technique is of course used in the H-maser and Rb gas cell frequency standards and accounts for the negligible first-order Doppler effects. However, the price we have paid to avoid the first-order Doppler effect is the frequency shifts due to confinement (collision shifts).

To avoid the perturbing effects of confinement, a common approach is to use atomic beams. The most successful approach to avoid the first-order Doppler effect in this case is to make the atoms interact with the radiation in two, phase-coherent, spatially separated interaction regions. In each interaction region, the condition $\vec{k} \cdot \vec{v}_x \Delta t \lesssim 1$ is satisfied—where Wt is now the transit time through one of the interaction regions. However, the Q is now determined by the much longer transit time between interaction regions. This principle is the basis of Ramsey's separated oscillatory field technique[4] which is used in all commercial and laboratory cesium clocks. Because it uses an atomic beam, the cesium device is free of the confining shifts, but suffers from residual first-order Doppler shifts due to the presence of running-wave components in the interaction cavities. This inability to obtain pure standing waves generally affects all of the "sub-Doppler" techniques where there is a net velocity associated with the atoms. Thus, we have a tradeoff between the confinement techniques, which "eliminate" first-order Doppler shifts but introduce perturbations due to the confinement, and the "free"-atom techniques, which have no confinement perturbation but introduce Doppler shifts. Moreover, all of the techniques, including those employing Dicke narrowing, suffer from the "second-order" Doppler shift because the atoms have a non-zero motion. Thus, if we hope to fundamentally improve the performance of frequency standards, we must address the question of Doppler shifts.

PRESENT LIMITS ON COMMON FREQUENCY STANDARDS

The limitations on accuracy and stability for present day frequency standards will be discussed more completely in another paper for this meeting by F. L. Walls. However, we briefly list the limitations and some possible cures.

Beam Devices (Cesium)

The dominant limitation appears to be due to cavity phase shift, which is a form of residual first-order Doppler effect. To first-order this effect can be measured by reversing the beam direction. It can be nulled [5] if measured periodically; however, exact beam retrace is required. This difficulty, along with the problem that the phase shift has a spatial dependence across the cross-section of the beam,[6] makes it difficult to deal with. The retrace problem and the problem of a spatially distributed phase shift are, in principle, eliminated if superconducting cavities are used.[7] These problems are substantially suppressed if an axially symmetric beam of small cross-section is used,[5] if a two-frequency interrogation method is used,[8] or if optical pumping state selection and detection is used.[8] Although the two-frequency method results in a degradation of Q by a factor of about 3, it has the advantage in possible commercial application in that beam reversal is not required to null the phase shift.[9] It would seem that more work needs to be invested to study these types of problems; a testament to this is the as yet unexplained frequency shift sometimes observed in cesium beam standards when the C field is reversed.[5,6,10,11]

Gas Cell Storage Devices (Hydrogen and Rubidium)

The fundamental limitations on accuracy appears to be due to our inability to measure the confinement shifts in the devices. For hydrogen, one might argue that the limit on long-term stability is due to cavity mistuning, but as this problem becomes solved by spin-exchange tuning[12] or cavity interrogating methods,[13] then the problem of the wall shift becomes more important. The varia-

ble volume[14,15,16] or "large box"[17] techniques are appealing, but have a problem in that the surfaces for both bulb configurations are not exactly the same.

The problems with the rubidium buffer gas standard are generically the same, although it suffers from incomplete spatial averaging.[18] If coated cells are used to combat this problem,[18] wall shifts become important. The problems associated with light shifts can be attacked using pulsing[19] or perhaps diode laser sources.

NEW IDEAS FOR FUNDAMENTAL IMPROVEMENTS

This section attempts to highlight new ideas which may bring about fundamental improvements in atomic frequency standards. The selection of topics in this section is, of course, somewhat arbitrary, but hopefully is representative of those methods which may be applicable in the not-too-distant future. With some of these ideas it may be difficult to envisage a practical device, let alone a commercial device; however, if one has faith in the advancement of the general technology, they may in the future become quite practical.

A. Cold Atoms

The advantage of using "cold" or low velocity atoms has been realized for quite some time. Not only is the interaction time of atoms increased—thereby increasing Q in a fundamental way, but the problem of second-order and residual first-order Doppler shifts are attacked in a fundamental way. The possibility of using very slow atomic beams was investigated as early as 1953 by Zacharias in his "Fountain" experiment.[20] In principle, only

the very slow atoms from an effusive source were selected by gravity. Unfortunately, the number of slow atoms available was too small to be useful. Since that time, various attempts have been made to produce slow atoms, but with very limited success. Some of the more recent experiments are mentioned here.

1. Cold Hydrogen: Very recently, hydrogen storage devices have been operated at cryogenic temperatures.[21,22,23,24] In the experiments of Crampton et.al.,[21] the storage bulb was coated with solid H_2 at 4.2K and although their experiments showed a rather large phase shift per collision ($\cong$ - 0.3 rad), this work may prove to be very useful in studies of the general problem. Moreover, a similar device might be used to generate a cold beam of polarized atomic hydrogen in other frequency standard schemes. In the work of Vessot et.al.,[22] maser oscillation was achieved down to 25K and Q's of ~2 x 10^9 were observed at 50K using a surface of tetrafluoromethane. Aside from the reduction in second-order Doppler shifts, low temperature H-storage devices have the following possible advantages:

a. Thermal noise is substantially reduced.[23,25] This affects the intrinsic maser stability and can also reduce the additive white phase noise, which is external to the maser.

b. Spin exchange collision rates are reduced by about two orders of magnitude.[23,25] Hence, the maser could operate with higher line Q at increased flux. At higher flux, the power could be increased[25] or the maser operated with lower cavity Q,[23] thus reducing cavity pulling.

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c. Mechanical rigidity should be more easily maintained at low temperatures[23,25] resulting in more stable cavity pulling, etc.

Investigations will continue to search for wall coatings (perhaps frozen inert gases) which will give stable surfaces with small wall shift. Such investigations may hopefully give very good results in the future.

2. Laser Cooling: Aside from the interesting results obtained with hydrogen, there have been many attempts to make cryogenic beam sources for other atoms, but temperatures below about 50K have been difficult to achieve. In 1975, independent proposals were made to cool down a gas of neutral atoms[26] or ions bound in an electromagnetic trap[27] using radiation pressure. Since then, substantial cooling (<0.5K) has been achieved for bound ions[28] and although only very limited atomic beam cooling has yet been obtained,[29] the potential for substantial cooling exists.

For free atoms or weakly bound ions (motional vibration frequency << optical transition linewidth), the cooling process is explained[30] by simply considering conservation of momentum and energy in a photon scattering event. If we average over the possible directions of reemission, we can find the average kinetic energy change per scattering event to be[30]

$$\Delta K.E. (\text{atom}) = \hbar \vec{k} \cdot \vec{v} + 2R \qquad (1)$$

where R is the "recoil" energy $R = (\hbar k)^2 / 2M$, $\vec{k}$ is the photon

wave vector and $\vec{v}$ is the atomic velocity before the scattering event. Since the scattering process is resonant, we can tune our light source (laser) below the atomic rest frequency, so that the atoms absorb only when they move toward the laser. Thus, we can make $\hbar\vec{k} \cdot \vec{v} < -2R$ and the atoms lose kinetic energy. Qualitatively, the atom's motion is retarded when it moves toward the laser because it receives a momentum kick in a direction opposite to $\vec{v}$ for each scattering event.

This cooling process makes the possibility of stored ion frequency standards more attractive (see below), but a practical scheme for producing a slow atomic beam of adequate flux has not yet been demonstrated. Nevertheless, the possibility[30] of very low temperatures ($< 10^{-3}$ to 10^{-4} K for strongly allowed transitions, less for weakly allowed transitions) makes this an attractive area of investigation.

B. Optical Traps

In the last few years, a fair number of papers have been written about the possibility of trapping neutral atoms in near-resonant light fields.[31] More recently, the dipole forces necessary for optical trapping have been demonstrated by a group at Bell Labs.[32] Such trapping is very attractive because atoms could be trapped in "cells" of a standing wave light field of dimensions $\lambda/2$. Hence, the potential confinement is extremely tight, i.e., the atoms would be well localized. The main disadvantage of this method for spectroscopy appears to be that in order to provide trapping, an optical transition must be driven at near saturation. Hence, any transition that might be interesting enough to provide a frequency standard would be broadened by the laser and also

subject to substantial light shifts. It would seem that the cure for this problem would be to turn off the "trapping" laser while the frequency standard transition is investigated. However, even if the theoretical limit[30,31] on laser radiation pressure cooling could be obtained, the velocity is still rather high (for Na atoms $v_{min} \simeq 30$ cm/sec) and the atoms would diffuse away from the trapping region while the trapping fields are off.

Even if a way around these difficulties is not found, the optical traps might be incorporated with laser cooling to provide a cold beam source. For example, the optical trap might be in the form of a tube (focused Gaussian laser beam) in which the atom could be laser cooled and then allowed to escape from one end. As yet, a practical scheme for this has not been suggested.

C. Stored Ions

The possibility of obtaining very high resolution spectroscopy with ions stored in electromagnetic traps had been realized very early by Dehmelt and was developed in the early stages by him and his co-workers.[33] Since that time, the radio-frequency (r.f.) trap has been developed to give rather encouraging numbers for an optically pumped ion standard. Noteworthy are the experiments of Major and Werth on mercury, which have been extended by others.[34] This device uses optical pumping—double resonance (pumping from a lamp) to detect the ground state hyperfine resonance in $^{199}Hg^+$ (~40 GHz) and gives estimated stabilities near $\sigma_y(\tau) \simeq 10^{-12} \tau^{-\frac{1}{2}}$. This success has prompted at least one commercial company to investigate the feasibility of such a standard.

The development of ion-storage frequency standards has been slowed somewhat because:

(1) The number of ions that can be stored is very small (typically a maximum of about 10^6 for a trap with ~1cm dimensions). This, coupled with the problem of the relatively low intensity of light from lamp sources for optical pumping and detection, has made resonance signals fairly weak.

(2) The presence of significant second-order Doppler shifts in experiments with traps (particularly r.f. traps) has been recognized for some time. Although one has various ways of measuring the velocity distribution,[35] this problem poses a serious limitation.

It may now be possible to overcome these limitations by using tunable lasers. By using a laser for optical pumping and detection, rather remarkable signals can be obtained. This is evidenced in two experiments[28].

(1) In the NBS experiments on Mg^+, the scattered photons from only about 500 Mg^+ ions stored in a Penning trap were observed with a signal to background of about 100. In this same experiment, the count rate was about 25,000/sec, while the net detection efficiency was only about 3×10^{-5}. This could be increased by 2 orders of magnitude in future designs.

(2) More dramatically, in the experiments at Heidelberg, a single Ba^+ ion contained in a miniture r.f. trap was photographed with good contrast.

In both of these experiments, the ions were laser cooled to substantially less than 1 K, hence the second-order Doppler shift was greatly suppressed.

Other advantages occur if one uses a laser in an ion storage experiment:

(1) Extremely weak optical pumping processes can be realized. This was demonstrated in the experiments at NBS where $^{25}Mg^+$ was pumped into the $(M_J = -\frac{1}{2}, M_I = -^5/_2)$ ground state. Although many absorption-reemission cycles are required for this pumping to occur, this is acceptable since the ions remain in the trap essentially indefinitely, and laser intensities can nearly saturate the optical transition.

(2) Transitions in double resonance experiments can be detected with nearly unit efficiency. In the experiments at NBS, the ions were caught in an optical trap (to be distinguished from spatial optical traps described earlier). That is, the ions were optically excited from a particular ground state level to a particular excited state level and (by selection rules) were forced to fall back into the original ground state level (this process can be repeated at very high rates). Thus, if we can arrange to drive a microwave transition (which will be, say, our frequency standard transition) that populates (or depopulates) the lower optical level, then we can produce (or exclude) many scattered optical photons for each ion that has made a microwave transition.[36] This process allows one to compensate for the

loss in collection efficiency due, for example, to small solid angle or small quantum efficiency in the photon detector so that we can effectively detect atoms with unit efficiency (The ability to achieve many scattered photons for one microwave photon was realized in ref. 36, but the S/N was incorrectly over-estimated). For example, if we can trap 10^6 ions, then the signal-to-noise in the detection process can be about 10^3, even though we may collect only about 1% of the total scattered photons.

It is useful to briefly compare the r.f. and Penning traps for possible frequency standard application. As is often pointed out, the r.f. trap has the potential advantage that magnetic fields are not required so that magnetic field induced frequency shifts do not pose a problem. However, this apparent difficulty can be overcome in the Penning trap by working at field extremum points.[36,37] A disadvantage of the r.f. trap not shared by the Penning trap is the effect of r.f. heating. Although not totally understood, it has prevented cold temperatures from being achieved except for very small numbers of ions. A possible disadvantage of the Penning trap is that the ions are in an unstable equilibrium in the trap; whereas, for the r.f. trap, the orbits are stable. This problem appears to have been overcome in recent experiments however, and indefinite confinement in a Penning trap should be possible.[38] At this point, it is unclear which type of trap will ultimately be better and more experiments are needed to decide this question. Perhaps a more important question to be addressed in the immediate future is how to get better optical sources for pumping and detection at the required frequencies. Simple schemes[39] are difficult to come by, but this difficulty

can be partially overcome by finding narrow band sources farther in the u.v.(< 210nm).

Nevertheless, ion trap derived standards are extremely attractive because they satisfy the confinement problem without causing significant perturbations (essentially indefinite confinement appears possible—implying no first-order Doppler shifts and confinement shifts are estimated to be below 10^{-15}). In this regard, they may be unique and deserve more attention in the future.

D. Laser Standards

With high probability, the frequency and time standards of the future will be based on optical transitions in atoms or molecules. This conjecture relies mainly on the idea that in a given system, if the lifetime of the transition remains reasonably fixed due to relaxation, transit time, etc., then the Q of the transition scales with frequency. However, before laser standards are realized, some crucial obstacles must be overcome. These problems are addressed below (see also a detailed review of precision, stable lasers by J. Hall[40]).

In contemplating laser standards, one first must realize that we are likely to be more susceptible to first-order Doppler shift since the wavelength of the radiation is so small; i.e., the Dicke criterion is harder to realize. However, the confinement criterion can be relaxed in important ways.

When we cannot meet the condition $|\vec{x}| \ll k^{-1}$, there is still the possibility of obtaining the same effect if we can satisfy the

more general condition $\vec{k} \cdot \vec{v}_x \Delta t \lesssim 1$, where Δt is the transit time of the atom through the apparatus. This is the general condition that must be met in a molecular beam apparatus. It allows for saturated absorption ("Lamb-dip") spectroscopy where atoms satisfying this condition are preferentially detected. Qualitatively, in this case, the detected atoms traverse the apparatus in a direction nearly normal to the traveling-wave propagation direction, and, therefore, the spatial phase change of the field experienced by the atoms is less than one radian.

The confinement problem has a rather unique solution, in the form of Doppler-free, two-photon spectroscopy. Here the atom interacts with counter-propajating plane waves of frequency $v_0/2$. The atom can resonantly absorb two photons simultaneously, one of frequency $^1/_2v_0(1 + \vec{v} \cdot \hat{k}/c)$ from one of the running waves and one with frequency $^1/_2v_0(1 - \vec{v} \cdot \hat{k}/c)$ from the counterrunning wave. The total energy from the two photons is hv_0, independent of the atomic velocity (to first order). Important applications exist in the optical region,[41] but the technique may be limited in accuracy by dynamic Stark shifts resulting from the required intense light field.

Therefore, the problem of first-order Doppler shifts can be solved, but as is true in the case of microwave frequency standards, residual first-order Doppler shifts can occur.[40] Moreover, the second-order Doppler shift remains unchanged.

As of now, rather impressive performance has been achieved with laser standards. For example,[40] methane stabilized He-Ne lasers have been used to probe external methane resonances using saturation spectroscopy with $Q > 10^{11}$ and stabilities of 10^{-14}. However,

the velocity distribution of the interrogated molecules is diffi-
cult to evaluate and accuracy capabilities better than 10^{-13} will
be difficult to achieve. In an experiment using a dye laser to
observe saturated absorption resonances in an atomic Ca beam,[42]
line Q's greater than 10^{11} were observed; however, a primary
limitation in accuracy in this experiment, as well as those on
CH_4, was the uncertainty in the second-order Doppler shift.
Therefore, we note that possible laser standards must be attacked
on a similar front as the microwave standards—that is, how can we
reduce the Doppler shifts? Certainly some of the same cooling
techniques as mentioned previously can be used; in addition, the
use of ions stored in a trap will have the advantage of long
confinement time with small perturbations.

Before such laser standards can be realized, we must solve two
other basic problems:

(1) Stable laser sources must be found. As in the microwave
case, the local oscillators used in optical frequency
standards must have the required short-term stability or
the desired accuracy will not be realized in a reason-
able length of time. At present, some gas-discharge
lasers meet this requirement[40]; however, these lasers
are very limited in tuning and therefore only in rare
instances have a frequency which coincides with a trans-
ition in a molecule or atom that might give a frequency
standard. Dye, diode, and color center lasers give the
desired tunability; however, this wide tunability and
fluctuations in the dye medium, for example, make them
far less stable. Nevertheless, superior stabilization
schemes and perhaps new lasers will undoubtedly be found

and the problem of (local-oscillator) short-term stability will be solved.

(2) If one desires to use a frequency standard as a time standard, one must in effect be able to count cycles of the oscillation. Phase-coherent measurements are at present very difficult to carry out at frequencies above about 100 GHz. However, by using harmonic mixing techniques in a boot strap fashion,[43] laser frequencies have now been compared to the cesium hyperfine frequency. This has so far only been done in a non phase-coherent way in a frequency synthesis chain such as that of ref. 43 shown in Fig 2. Accuracies of intercomparison are near the 10^{-10} level. In order for an optical frequency standard to provide time, phase-coherence would have to be included at each step in this chain. This seems to be a fairly complicated (although achievable) proposition even if somewhat more simplified chains [43] are realized. In dealing with this problem, one should of course recall that there are other uses of frequency standards than providing time. For example, some very interesting tests of gravitational effects can be examined[40,44] using optical frequency standards, if all that is required is to intercompare two optical frequencies—a task which is trivial compared to providing time. However, the time problem is very important and a solution to the frequency synthesis problem should be sought. A conceptually simple but unproven scheme[45] might be able to accomplish one-step frequency division from optical frequencies to microwave frequencies.

E. Single Atom (Ion) Frequency Standards

The idea of using a single atom is, of course, very appealing since, if suitably confined, it can be isolated from the perturbing influence of other atoms. Dehmelt was the first to suggest such an idea.[46] He proposed to use an optical transition in $T\ell^+$ ($^1S_0 \leftrightarrow {}^3P_0$ transition @ 202 nm $Q \cong 10^{14}$) in an r.f. trap. The additonal advantage of using a single ion confined to the center of an electromagnetic trap is that combined with laser cooling, it should be possible to closely satisfy the Dicke criterion in the optical region on an essentially unperturbed atom. Other possibilities exist, such as the B^+ ion, whose structure is shown in Fig. 3. This ion is also interesting because the fine structure transitions with $Q \gtrsim 10^{11}$ could provide a possible standard where the frequencies are fairly easily measured with state of the art precision. (This experiment could also of course be performed on a cloud of ions in a trap).

The primary drawback to using a single ion (or perhaps a single neutral atom in an optical trap) is that the S/N is rather poor. Therefore, single ion standards would seem to be more viable at very high frequencies where the Q can be quite high. Since the perturbations between many cold ions in an electromagnetic trap can be very small anyway, the use of a single ion may only be a philosophical advantage if one uses longer wavelengths. Of course, only the future will tell!

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(A)

gaussian

$\Delta \nu_0 \cong \frac{v}{c} \nu_0$

ν_0 $\nu \longrightarrow$

(B)

lorentzian

$\Delta \nu$ = natural linewidth

gaussian

ν_0 $\nu \longrightarrow$
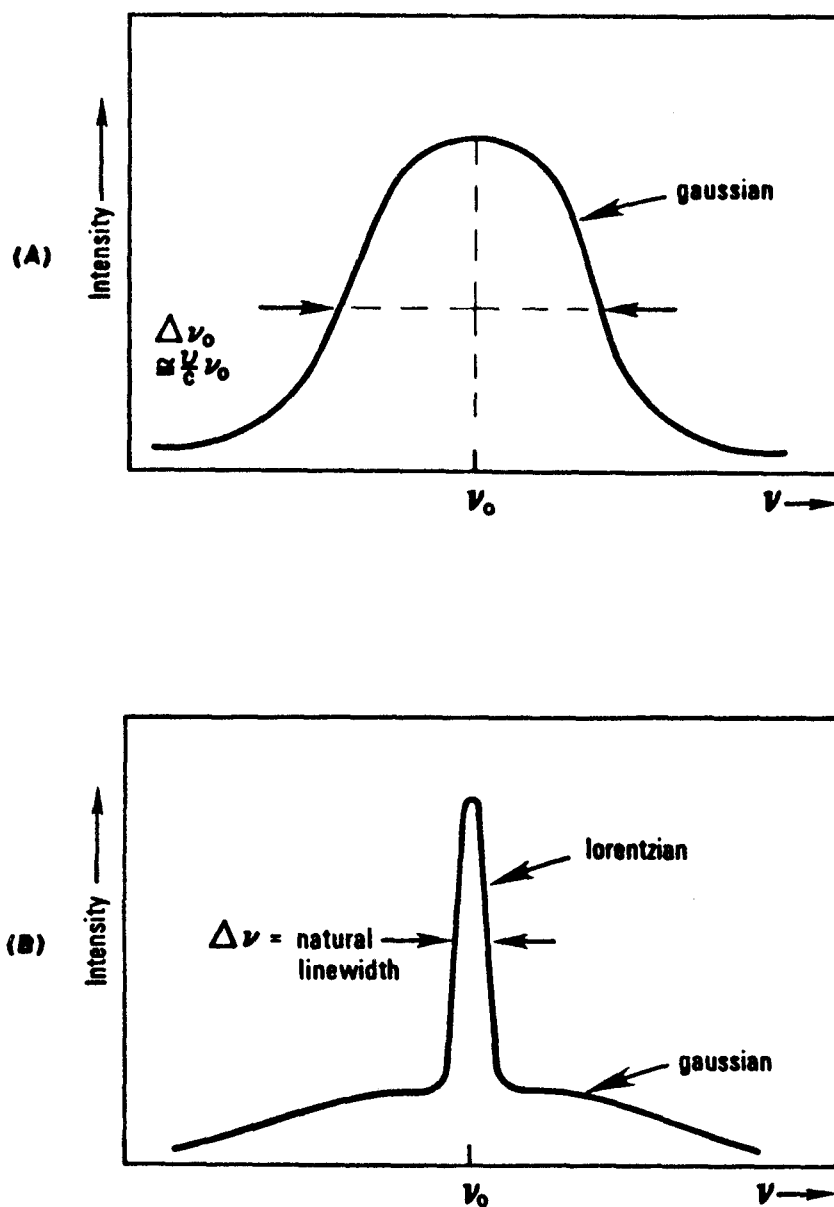
FIGURE 1. Part A shows the situation when the atoms are unbound and the resonance feature has the full Doppler width $\Delta \nu_D \cong (v/c)\nu_0$. When the atom is confined to dimensions less than the wavelength, the Doppler profile is suppressed and the central feature has the natural width $\Delta \nu$. This condition is most easily realized in the microwave region of the spectrum.
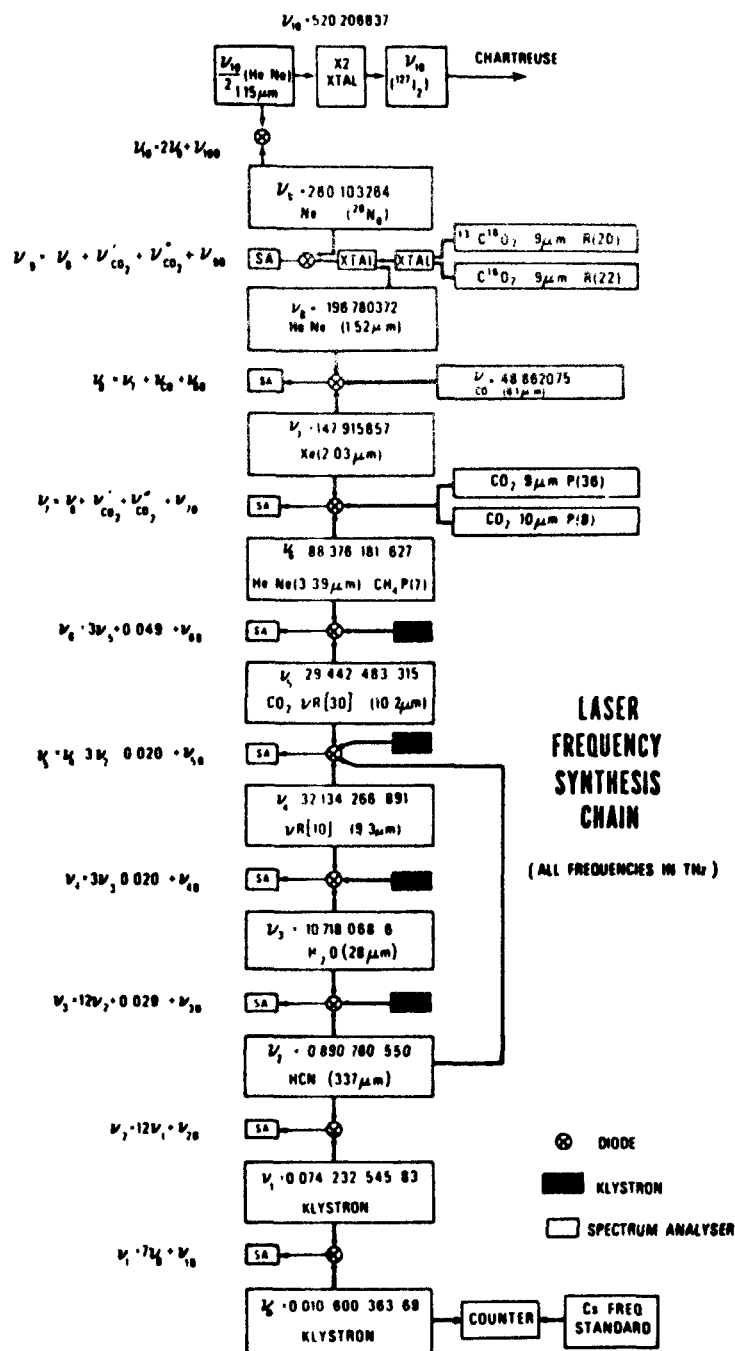
$\nu_{10} \cdot 520\ 206837$

| $\frac{\nu_{10}}{2}$ (He Ne) 1.15μm | X2 XTAL | ν_{10} ($^{127}I_2$) | → CHARTREUSE |

$\nu_{10} \cdot 2\nu_9 + \nu_{100}$

$\nu_9 \cdot \nu_8 + \nu'_{CO_2} + \nu^*_{CO_2} + \nu_{90}$

| $\nu_9 \cdot 280\ 103264$ Ne (^{20}Ne) |

SA — ⊗ — XTAL — XTAL

| $^{13}C^{18}O_2$ 9μm R(20) |
| $C^{18}O_2$ 9μm R(22) |

| $\nu_8 \cdot 196\ 780372$ He Ne (1.52μm) |

$\nu_7 \cdot \nu_6 + \nu_{CO} + \nu_{80}$

SA — ⊗ — | $\nu \cdot 48\ 862075$ CO (61μm) |

| $\nu_7 \cdot 147\ 915857$ Xe(2.03μm) |

$\nu_7 \cdot \nu_6 + \nu'_{CO_2} + \nu''_{CO_2} + \nu_{70}$

SA — ⊗ — | CO_2 9μm P(36) | CO_2 10μm P(8) |

| $\nu_6 \cdot 88\ 376\ 181\ 627$ He Ne(3.39μm) CH_4 P(7) |

$\nu_6 \cdot 3\nu_5 + 0.049 + \nu_{65}$

SA — ⊗ — ■

| $\nu_5 \cdot 29\ 442\ 483\ 315$ CO_2 νR(30) (10.2μm) |

$\nu_5 \cdot \nu_4 + 3\nu_3 + 0.020 + \nu_{54}$

SA — ⊗ — ■

| $\nu_4 \cdot 32\ 134\ 266\ 891$ νR(10) (9.3μm) |

$\nu_4 \cdot 3\nu_3 + 0.020 + \nu_{43}$

SA — ⊗ — ■

| $\nu_3 \cdot 10\ 718\ 068\ 6$ H_2O(28μm) |

$\nu_3 \cdot 12\nu_2 + 0.029 + \nu_{32}$

SA — ⊗ — ■

| $\nu_2 \cdot 0.890\ 760\ 550$ HCN (337μm) |

$\nu_2 \cdot 12\nu_1 + \nu_{21}$

SA — ⊗

| $\nu_1 \cdot 0.074\ 232\ 545\ 83$ KLYSTRON |

$\nu_1 \cdot 7\nu_0 + \nu_{10}$

SA — ⊗

| $\nu_0 \cdot 0.010\ 600\ 363\ 69$ KLYSTRON | — | COUNTER | — | Cs FREQ STANDARD |

LASER FREQUENCY SYNTHESIS CHAIN

(ALL FREQUENCIES IN THz)

⊗ DIODE

■ KLYSTRON

▭ SPECTRUM ANALYSER

FIGURE 2. Synthesis "chain" used at NBS[43] to measure visible laser frequency.
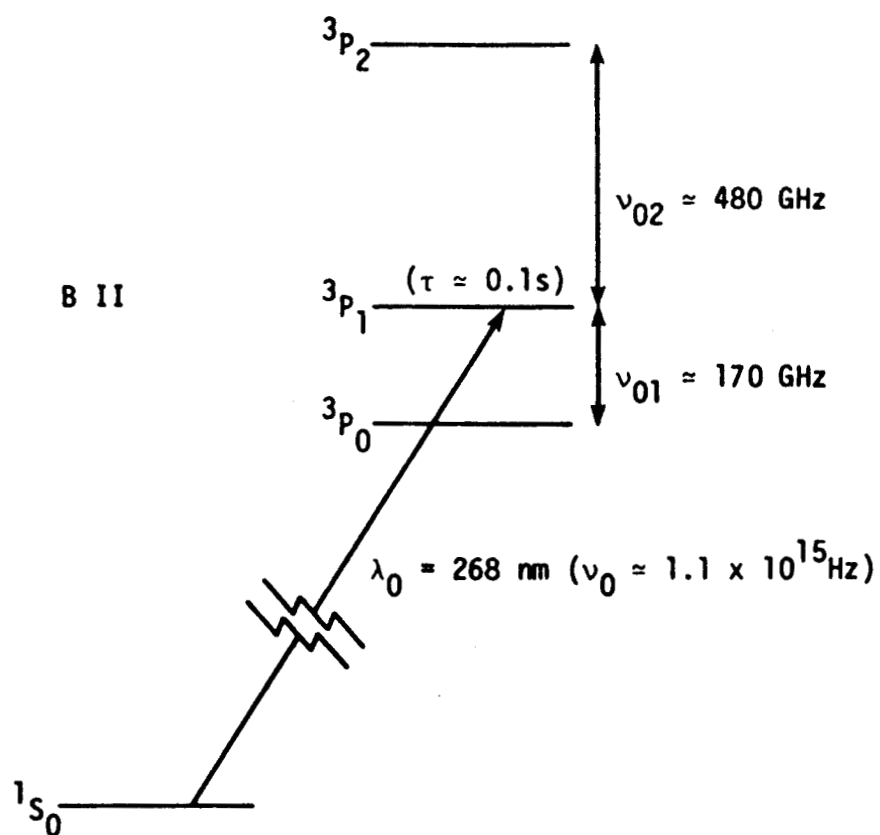
FIGURE 3. Energy level scheme of singly ionized Boron showing energy separations of interest.