ABSTRACT
A 445-THz (674nm), $^{88}$Sr$^+$ trapped and laser cooled single ion reference transition has been used at NRC to extend precision frequency measurements to other points in the electromagnetic (E.M.) spectrum. We are currently refining the single ion experiment to approach the uncertainty limited spectral resolution of $1 \times 10^{-15}$. Connected with these developments is the use of frequency grids based on mode-locked femtosecond lasers. A band of reference modes extending from 520 nm to beyond 1060 nm has been recently obtained at NRC. With such devices, the possibility of accurate, stable and compact sources at any wavelength is coming into being.

INTRODUCTION
The basis of modern precision measurement of time and length is founded on electromagnetic radiation stabilized on transitions in atoms and molecules. It has been a goal for many years to use a laser in conjunction with a nearly unperturbed quantum absorber system to create a reference that approximates as close as is physically possible a fundamental unit of measurement. Resolving spectral features below the Hz level at $10^{14}$-$10^{15}$ Hz allows one an unprecedented level of precision which can presumably lead to new frontiers of accuracy to probe the physical world.

SINGLE ION STANDARD
Following the first proposals by Dehmelt in the 1970’s [1], the concept of storing a single ion using electro-dynamic traps followed by probing it on a ultra-narrow forbidden transition has shown great promise. Subsequent advances in laser cooling of ions and atoms gave added strength to this approach. A number of single ion systems have since been examined and substantial progress has been made towards their ultimate implementation [2]. At NRC, activities have been focussed on the single ion of $^{88}$Sr$^+$ [3]. This system has also been examined by the group at the National Physical laboratory of the United Kingdom (NPL) and by others [4]. One of the advantages of the $^{88}$Sr$^+$ system is the ion’s resonance S-P transition lying within the visible region of the spectrum. This holds advantages in that the excitation used for laser cooling at 422 nm can be directly supplied by frequency doubled (and now direct) diode laser sources, thus making the system compact and relatively simple to implement. The ion fluorescence from the 22 MHz wide line is sufficiently strong at saturation that detection using photon counting optics can register count rates at the $10^4$-$10^5$ s$^{-1}$ level making unambiguous discrimination of the single ion. Moreover, the transition allows direct laser cooling to the Doppler-cooling limit to a level of 0.5 mK. This provides substantial reduction in Doppler and other perturbation effects on the reference transition to below the $10^{-15}$ level. The long-lived reference transition at 674 nm or 445 THz also possesses a number of attractive features. Diode laser sources are readily available at 674 nm and the lifetime of the $5S_{1/2} - 4D_{5/2}$ transition is 0.3 s making a resolution of 0.4 Hz attainable. An overview of the experimental arrangement is given in Fig.1. The excitation at 674 nm into the metastable $D_{5/2}$ level will cause a halt in the detected fluorescence at 422 nm. Thus even if the ion is excited on the reference transition at rates on the order of $10^4$ s$^{-1}$, every transition is effectively observed via the "quantum jump" in corresponding 422 nm fluorescence. By counting these jumps as a function of laser detuning, one obtains a spectral profile of the reference transition. Within our group, observed transition linewidths below 250 Hz have been obtained and widths below 150 Hz have been reported by the NPL team. The current primary limitation in resolution is the construction of laser probe systems of sufficient stability to match the relative linewidth of $10^{-15}$. It should be noted that the $^{88}$Sr$^+$ S-D system also possesses a first order Zeeman shift with applied magnetic field. Appropriate shielding is necessary, however the movement of the ion within the RF trap potential well is well below that of 0.1 $\mu$m$^3$. Sensitivities to field gradients are substantially reduced compared with other spectroscopic methods and modest levels of shielding are sufficient that other physical effects will be the major contributor to the uncertainty budget.

ABSOLUTE FREQUENCY MEASUREMENTS USING SINGLE IONS
By using several laser oscillators and nonlinear mixing stages spanning 28 THz (10 $\mu$m) to 445 THz (674nm), a coherent frequency chain was developed at NRC linking a Cs referenced RF signal and that of the optical source at
445 THz stabilized on the single ion [5]. The results of this 1998 measurement were the establishment of the absolute single ion frequency with a transition centre frequency of \( f_{SD} = 444.779 \pm 0.2 \text{kHz} \) (1sigma) and led to its formal adoption as a recommended realization of optical frequency and vacuum wavelength. This was the first direct frequency measurement of a trapped ion transition at visible frequency and the second direct frequency measurement of an optical visible frequency. Subsequent works by other groups using femtosecond laser frequency combs have recently yielded higher levels of accuracy [6] using other ions. With the current NRC \(^{88}\text{Sr}^+\) S-D reference system, the estimated systematic shifts are currently at the \(10^{-15}\) level, and the knowledge of the centre reference frequency is limited by the accuracy of the existing laser frequency chain and the instabilities of the probe laser source used. The obtained relative accuracy of \(5 \times 10^{-13}\) of the standard meant that it could be used as an effective reference point for absolute optical frequency for the measurement of other points in the electromagnetic spectrum.

One of the first applications of the single ion standard was the precision measurement of a 1.55 micron laser source (developed by a team at the Université Laval) which was frequency doubled and stabilized on a \(^{87}\text{Rb}^+\) S-D two photon transition. By sum mixing radiation of a 148 THz = (445 THz)/3 laser referenced to the single ion with a CO laser operating at 44 THz in a non-linear crystal of AgGaSe\(_2\), sufficient power (400nW) at 193 THz was synthesized for the heterodyne frequency measurement of the source [7]. Besides giving the most precise measurement of a reference in the 1.5 micron region, the measurement yielded an improved value of the Rb S-D two-photon transition. This transition has been used as a reference in work determining the Rydberg constant in hydrogen and may continue to play a significant role in future precision measurement.

A second application of single ion based measurement has been the precise measurement of the widely used 633-nm HeNe laser stabilized on iodine. The 633-nm standard forms the basis for the metre in most standard labs worldwide and its precise measurement served to consolidate its value. By difference frequency mixing a stabilized laser radiation at 28 THz (measured with the NRC infrared frequency chain) with 474 THz (633-nm) light referenced to the I\(_2\)/HeNe standard, radiation at 445 THz was generated which was heterodyned with the source probing the single ion [8,9]. A series of absolute measurements were conducted using reference lasers from the NRC, BIPM and NIST/JILA in the traceable determination of the accepted value for the standard. Based on this work, calibration of the 633nm standard at an accuracy of \(\pm 720 \text{Hz} \) (1sigma) [9] was achieved and a traceable determination of the international working value for transition frequency was obtained at a level of 1.4 kHz [8]. In addition, the work led to a significant comparison of frequency measurement methods; the laser frequency chain measurement combined with the NRC single ion, and the new femtosecond laser comb systems.

**IMPROVEMENT OF SINGLE ION STANDARD AND FUTURE MEASUREMENTS**

Work is underway to improve the accuracy of the standard to its full potential yielding resolutions of \(10^{-15}\) relative linewidth and an ultimate accuracy in the \(10^{-16}\) region and beyond. The work involves the upgrade of the probe laser system and ion trap containment apparatus. A new probe laser using a commercial extended cavity diode laser system as the source is currently in development. The laser is stabilized successively to two Fabry-Perot high finesse resonators in cascade. A 100-kHz linewidth evacuated resonator provides the initial line narrowing of the diode source. This
spectrally narrowed radiation, taken from the transmitted beam and passed through an acousto-optic modulator (AOM) is then further stabilized with an ultra-stable, temperature controlled and vibration isolated, 4-kHz linewidth resonator constructed from ULE glass with mirrors optically contacted to the spacer. Initial results of the first stage of line narrowing have now yielded operating linewidths on the kilohertz level. Heterodyne frequency comparisons of the new laser system using the first stage of narrowing are now being performed relative to the previous probe laser setup. Intercomparison of the two systems now allows a more rapid evaluation of such factors as servo optimization and the determination of the point of zero linear thermal expansion versus temperature for the new ULE ultra-high finesse cavity.

In concert with the probe laser developments, a new single ion trap is being developed which should allow a greater ability to evaluate and control the single ion in its environment. A chamber allowing probing and laser cooling along 3 orthogonal directions has been designed using the principle of an end-cap trap [4,10]. This will allow independent monitoring of the ion kinetic temperature along all directions and the possibility for further laser cooling using two-stage methods [11]. This should be able to bring the single ion kinetic energy to the fundamental ground state of the trapping field’s potential well. A double magnetic shield design should reduce the background AC magnetic field such that it will cause negligible broadening of the 0.4 Hz wide S-D reference line. By controlling the ion position and kinetic temperature at these ultimate levels, the main sources of frequency shift will be the thermal ac Stark shift of the transition and the electric quadrupole shift of the upper 3D_3 level due to the gradient of the trapping field. These shifts are on the 10^-16 level and it should be possible to evaluate them so that the uncertainties in these shifts may be known to the percent level or better.

OPTICAL FREQUENCY REFERENCES FOR TELECOM WAVELENGTHS

Several groups have proposed the use of acetylene as a series of useful reference transitions that coincide with the optical telecom region of the spectrum. With both isotopic varieties, there are a large number of well-spaced resonances from 1510 to 1550 nm. In addition, saturated absorption resonances can be obtained when an optical cavity is used to enhance the circulating optical power and effective path length [12]. Recently, a transition in ^13C_2H_2 has been selected by the Comité International des Poids et Mesures (CIPM) as a reference for this wavelength region. Given the potential importance to technology and science, work has been directed in our group toward its refinement. An extended cavity laser system using the first stage of narrowing are now being performed relative to the previous probe laser setup. Narrowing have now yielded operating linewidths on the kilohertz level. Heterodyne frequency comparisons of the new laser system using the first stage of narrowing are now being performed relative to the previous probe laser setup. Intercomparison of the two systems now allows a more rapid evaluation of such factors as servo optimization and the determination of the point of zero linear thermal expansion versus temperature for the new ULE ultra-high finesse cavity.

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Fig. 2 Optical setup of the self-referencing femtosecond optical frequency comb.

Fig. 3 Observed heterodyne beat of comb offset via 2nd harmonic light generated from low frequency side of comb vs. high frequency side at 540 nm.

REFERENCES