APPLICATION OF LASER-COOLED IONS TO FREQUENCY STANDARDS AND METROLOGY*

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ABSTRACT

With the first experiments on laser cooling, it was realized that a principal application of the technique would be to reduce Doppler shifts in frequency standards. Laser cooling is still thought to be essential to achieve uncertainty significantly smaller than 1 part in 10^{14}; however, a number of other problems must be solved to achieve this goal with stored atomic ions. For both RF/microwave and optical frequency standards based on trapped ions, some of these problems are discussed. Laser cooling of trapped ions also appears to be important for other reasons in metrology. For example, for frequency standards, cooling to the zero-point of motion should enable the creation of quantum mechanically correlated states for improved signal-to-noise ratio in spectroscopy, or provide accurate measurement of Stark shifts. Cooling to the zero-point of motion also enables the creation of nonclassical states of motion or correlated states which may be applicable to sensitive detection, quantum computation, or to test quantum measurement principles. Techniques to achieve laser cooling to the zero-point of motion are briefly described.

1. Introduction

The first laser cooling experiments[1,2] were accomplished on trapped ions in 1978. It was apparent then that one important application for laser cooling would be the reduction of the time-dilation (2nd-order-Doppler) shift in frequency standards. In fact by 1980, temperatures achieved from laser cooling on ions were low enough that the corresponding time-dilation shift was significantly below 1 part in 10^{16}. This is illustrated in Fig. 1, where we plot time-dilation shifts (shown as a fraction of the transition frequency) corresponding to temperatures reported in some laser cooling experiments. Since these shifts are significantly smaller than the smallest reported inaccuracies for atomic clocks, we can ask why the overall accuracies reported do not approach these numbers.

One source of the discrepancy is that, for trapped ions, laser cooling affects only the thermal or random modes of motion. However, ion motion in both Paul and Penning traps[3-5] in part consists of unavoidable coherent motion; this motion can result in large ion velocities and accompanying Doppler shifts. In a Penning trap, a cloud of ions must rotate about the symmetry axis of the trap. The velocity associated with this rotation gives rise to a $\vec{v} \times \vec{B}$ Lorentz force which provides trapping normal to the symmetry axis. For large clouds, the velocity in this rotation motion can be significantly larger than the thermal velocity in the axial and cyclotron motions (which can be laser-cooled), thereby limiting the degree to which the time-dilation shift can

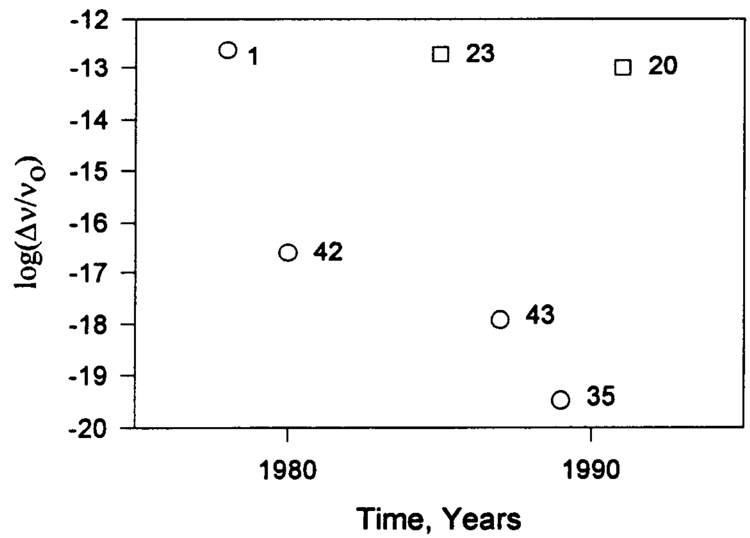

Fig. 1. The circles show magnitudes of calculated (fractional) time-dilation shifts $|\Delta\nu_{D2}/\nu_o|$ based on reported temperatures achieved in selected trapped-ion, laser cooling experiments. These calculations omit the contributions from the coherent modes of motion (see text). The squares show (fractional) uncertainties in trapped-ion, laser-cooled atomic frequency standard experiments in which overall errors were estimated. The numbers beside the symbols indicate the corresponding reference numbers.

be suppressed.[6] In a Paul trap, ions are forced to oscillate at the driving field frequency (the micromotion). Since the driving fields are spatially inhomogeneous, the force on an ion, averaged over one cycle of the driving field, gives rise to trapping (the ponderomotive force). For ions near the outside of a large ion cloud this force must be large to balance the Coulomb repulsion from the ions near the center of the trap. This implies that the velocity of this coherently driven motion is large; it is often much larger[6] than the velocity of the ions associated with their "secular" motion in the ponderomotive well (which can be laser-cooled).

In 1973, Dehmelt suggested storing a single ion in a Paul trap.[7] This system has the advantage that the kinetic energy in the ion micromotion is approximately equal to the kinetic energy in the secular motion. Therefore, if the secular motion is laser-cooled, the time-dilation shift from secular motion and micromotion can be very small. On the other hand, a single-ion frequency standard may not give sufficient short-term frequency stability. For N ions, the frequency stability (two-sample Allan variance) is limited to[6,8]

$$\sigma_y(\tau) = \frac{1}{\omega_o\sqrt{NT\tau}}, \tag{1}$$

where ω_o is the clock transition frequency, T is the transition interrogation time (the Ramsey method is assumed, where the time of the two Ramsey pulses is much less than the free-precession period T), τ is the averaging time, and we assume 100% detection efficiency. For a single-ion clock using RF or microwave transitions, this may imply a long averaging time to reach a measurement precision better than one part in 10^{14}. For example, for an ion with $\nu_o = \omega_o/2\pi = 10$ GHz, and T = 10 s, Eq. (1) implies $\sigma_y(\tau) \simeq 5.0\times10^{-12}\ \tau^{-1/2}$. Therefore, in general, for both Penning and Paul trap RF/microwave frequency standards, where we desire large N to make $\sigma_y(\tau)$ sufficiently small, we require methods to minimize the velocity in the coherent modes of motion. Some strategies are discussed below.

If the velocity in the coherent motion can be suppressed sufficiently, Doppler cooling[9] will suffice (at least for the next generation of frequency standards) to provide small enough time-dilation shifts for frequency standards. If we neglect the velocity in the coherent modes of motion, minimum temperatures achieved from Doppler cooling give rise to a time-dilation shift of $\Delta\nu_{D2}/\nu_o = -3.3\times10^{-18}(\gamma/2\pi)/M$ where $\gamma/2\pi$ is the laser-cooling transition linewidth in megahertz and M is the ion mass in relative atomic mass units. For example, for $^{199}Hg^+$ ions ($\gamma/2\pi \simeq 70$ MHz, M = 199 u), we could expect $\Delta\nu_{D2}/\nu_o = -1.2\times10^{-18}$.

However, there now appear to be other reasons for achieving better laser cooling - in particular for reaching the zero-point energy state of motion. New applications are emerging for this extreme form of laser cooling, both for frequency standards and other forms of metrology. Some of these applications and methods to reach the zero-point energy through laser cooling are discussed below. Reference 10 provides a more comprehensive review of cooling methods in ion traps.

2. Reduction of the coherent motion (micromotion) in a Paul trap

One way to suppress the velocity in the micromotion and still achieve a large number of ions for good signal-to-noise ratio is to use an elongated "linear" trap geometry. In this type of trap, shown schematically in Fig. 2, the RF electric fields vanish along the axis of the trap. Therefore, if ions are confined near the trap axis in an elongated ion cloud geometry, the micromotion can be suppressed. This idea was first developed and used by Prestage et al.[11] In part, because of the relatively small time-dilation shift, excellent stabilities have been achieved in clocks where the ions' secular motion in a linear trap is cooled by buffer gas (see the contributions at this conference by Tjoelker et al. and Fisk et al.).

Dehmelt suggested applying this idea to frequency standards in its limiting form,[12] that is, where a single string of ions is confined to the trap axis like "beads on a string." Here, as for a single ion in a spherical quadrupole trap, the velocity of micromotion can be approximately the same as for the secular motion. With laser cooling, the time-dilation shifts can therefore be very small. At NIST, this idea is being pursued for application to a frequency standard of high accuracy. Strings of laser cooled $^{199}Hg^+$ ions have been obtained in a room temperature apparatus[13] and in a cryogenic (4 K) apparatus[14] whose good vacuum suppresses collisional frequency shifts and ion loss from chemical reactions. The bottom part of Fig. 2 shows an image of a string of laser cooled $^{199}Hg^+$ ions taken in this cryogenic apparatus. The Munich group has also achieved strings of laser-cooled (Mg^+) ions in a race-track-type trap[15]; this type of apparatus could also be used for frequency standard applications.

Another approach to suppress micromotion is to use arrays of conventional (spherical quadrupole) ion traps.[16,17] Here, single laser-cooled ions could be stored in each trap thereby ensuring small time-dilation shifts. Important technical problems appear to be the loading, addressing, and detecting of the individual ions.

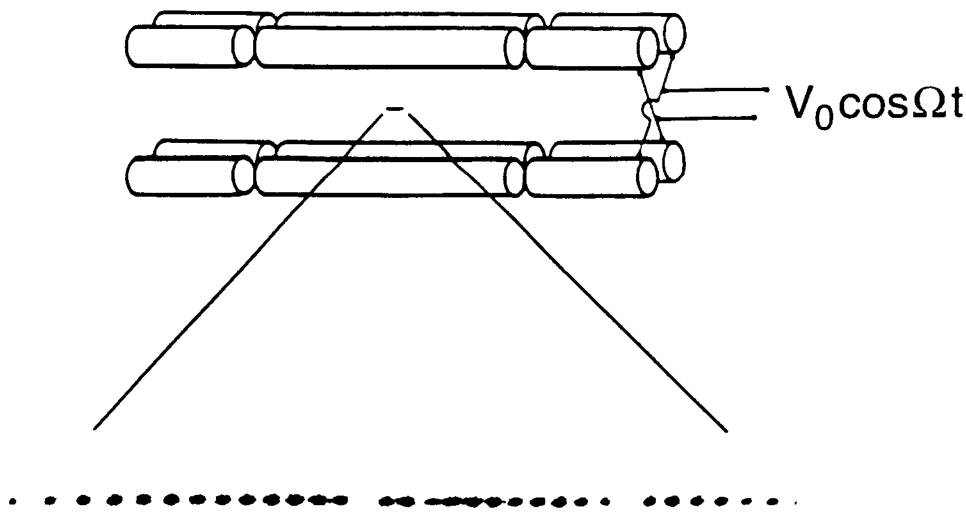

Fig. 2. The upper part of the figure shows a schematic diagram of the electrode configuration for a linear Paul-RF trap.[13] Typical parameters for trapping $^{199}Hg^+$ ions are: rod-electrode separation of approximately 1 mm, $V_0 \simeq 400$ V, and $\Omega/2\pi \simeq 10$ MHz. The lower part of the figure shows an image of a string of $^{199}Hg^+$ ions, illuminated with 194 nm radiation, taken with a UV-sensitive, photon counting imaging tube. The spacing between adjacent ions is approximately 10 μm. The "gaps" in the string are occupied by impurity ions, most likely other isotopes of Hg^+, which do not fluoresce because the frequencies of their resonant transitions do not coincide with the 194 nm $^2S_{1/2} \rightarrow {}^2P_{1/2}$ transition of $^{199}Hg^+$.

3. Reduction of the coherent (rotation) motion in a Penning trap

Following the example of the linear or racetrack Paul traps, the coherent motion in a Penning trap could be suppressed by storing ions near the axis of the trap - ideally as a single string along the axis. A more modest approach might be the following. For a cloud of laser-cooled ions in a Penning trap, as the radius of the cloud is varied, the time-dilation shift goes through a minimum (see Ref. 18 and Tan, et al., this conference). Therefore if the cloud of ions is stabilized at this extremum point, the time-dilation shift and its fluctuations can be made very small.

Any configuration of ions in a Penning trap is in an unstable equilibrium (as opposed to a sample of ions in a Paul trap). Therefore, once created, the configuration would not be preserved if, for example, collisions are present. Torques from laser scattering could restore the configuration but at the expense of possible perturbing effects from the cooling laser.

4. Sympathetic laser cooling

One solution to the problems associated with Penning trap cloud stability is sympathetic laser cooling.[19] As an example, in the laser-cooled frequency standard experiment reported in Ref. 20, a $^9Be^+$ ion sample was cooled sympathetically, through Coulomb coupling, by a simultaneously stored, surrounding cloud of Mg^+ ions which were directly laser-cooled. This method has the advantage that the laser cooling (and laser torques) can be applied continuously to the Mg^+ ions, thereby maintaining the geometry and temperature of the $^9Be^+$ ions. Because the frequency of the Mg^+ optical cooling transition is significantly different than the optical transitions in $^9Be^+$, the $^9Be^+$ ions are not appreciably perturbed by the Mg^+ cooling laser beam. It should be possible to apply this technique to sympathetically cool a string of ions in a Penning trap or a cloud that is configured to give the minimum time-dilation shift.[18]

Sympathetic cooling might also be employed to advantage in a (linear) Paul trap. When long Ramsey interrogation times T are desired, the ions may heat from, for example, RF heating. However, in a long string of ions, cooling could be applied to ions on one end which then cool the "clock" ions sympathetically. In principle, the "cooling ions" could be of the same species if the string is long enough or, if the string is bent around a (light-baffled) corner or, if the clock ions are shielded by a coupling electrode[21] to minimize the perturbing effects of scattered light from the cooling ions.

5. Laser-cooled, trapped-ion, optical frequency standards

Most researchers agree that, in the future, the most accurate and stable clocks will be made using optical or higher frequency transitions. From Eq. (1), if the clock transition frequency is high enough, then a single ion would give good short term stability. For example for the $^2S_{1/2} \rightarrow {}^2D_{5/2}$ 282 nm transition in $^{199}Hg^+$, assuming N = 1, T = 0.01 s (the radiative lifetime of the $^2D_{5/2}$ level is approximately 0.1 s), Eq. (1) implies $\sigma_y(\tau) \simeq 1.5\times10^{-15}\,\tau^{-1/2}$. As discussed above, the time-dilation shift could be very small. In addition, shorter interrogation times will mitigate the effects of various sources of ion heating, since clock cycles could be alternated with relatively frequent laser cooling. Work on optical frequency standards is represented by several groups at this conference; see, for example, the stored-ion contributions by Barwood, et al., Fermigier, et al., Gill, et al., Knoop, et al., Madej, et al., Nagourney, et al., Peik, et al., and Tamm and Engelke.

Currently, laser local oscillators are not as stable as desired. A representation of the state of the art for trapped-ion optical frequency standards is given in Ref. 22, where a laser with frequency bandwidth of less than 25 Hz (for 60 s averaging times) was locked to the 282 nm quadrupole transition in a single laser-cooled $^{199}Hg^+$ ion.

Finally, an additional technical difficulty for all optical frequency standards is that of comparing laser local oscillators to other oscillators (particularly oscillators at low frequency). Several approaches are discussed in these proceedings.

6. Present and future laser-cooled ion frequency standards

So far, there has been only one report of an ion-based, laser-cooled atomic frequency standard whose accuracy has been evaluated. In this experiment, laser-cooled $^9Be^+$ ions in Penning trap were used.[23] A second evaluation[20] of this system uncovered an unexpected background gas pressure shift. This shift was apparently caused, in part, by background methane gas, which has a strong perturbing effect on $^9Be^+$ ion hyperfine structure.[24] (Interestingly, methane does not have nearly as large a perturbing effect in the $^{171}Yb^+$ experiments reported by Bauch and Tamm at this conference.) At present, the inaccuracy of the laser-cooled $^9Be^+$ ion clock is limited to about one part in 10^{13} from this pressure shift.[20]

Further work is required to minimize possible pressure shifts and a number of other typical systematic effects (for example, magnetic field shifts) present in all frequency standards. However, we can be optimistic that frequency standards based on laser-cooled ions will eventually have inaccuracies considerably smaller than 1 part in 10^{14}, perhaps as small as 1 part in 10^{18} or less.[6,25,26]

7. Applications of trapped ions which are laser-cooled to the zero-point of motion

Although Doppler cooling should yield sufficiently small time-dilation shifts for frequency standards, there now appear to be other reasons for achieving better laser cooling. At very low kinetic energies, the quantum nature of the motion becomes apparent; this quantized motion, particularly the zero-point state, can be used for various purposes.

For example, when inaccuracies of optical clocks become smaller than one part in 10^{15}, Stark shifts may become significant.[27] As Dehmelt has pointed out[25], the intercombination transitions in group IIIA ions are superior in this regard, although for various technical reasons, optical quadrupole S $\rightarrow$ D transitions in other ions can be easier to implement. The quadratic Stark shifts on these latter transitions depend on the kinetic energy of the ions. Since the secular motion is quantized, the quadratic Stark shift from the secular motion becomes quantized. This could be used to identify the zero-point energy states of the ion[27] or, conversely, the Stark shift could be calibrated from its discrete spectrum caused by quantized motion.

Another possible frequency standard application of zero-point cooling is to improve the quantum-limited signal-to-noise ratio in the detection process (see Ref. 28 and Bollinger, et al., this conference). If spectroscopy is performed on atoms which are first prepared in particular quantum mechanically correlated, or entangled, states, the frequency stability would be given by $\sigma_y(\tau) = 1/(\omega_o N(T\tau)^{1/2})$ rather than that given by Eq. (1). In this case, the time to reach a desired measurement precision would be reduced in proportion to 1/N.

Outside the realm of frequency standards, there may be other metrological applications of ions which are laser-cooled to the zero-point of motion. Cirac and Zoller have recently suggested a very interesting scheme to perform quantum computation using an array of ions confined in a linear trap.[29] Quantum computers have received attention lately because of their ability to crack the most common form of public-key encryption. The first quantum logic gates using prepared input "qubits" have now been realized using trapped ions which are first cooled to the zero-point energy.[30] A second possible application might be to the generation of multiparticle "EPR"-type correlated states which would be interesting from the standpoint of quantum measurement theory.[31] Finally, if a single ion or single mode of oscillation is cooled to the zero-point state, it is possible to generate nonclassical states of motion such as squeezed states[21,28,32,33] (see also Blatt et al., this conference). Such states may be useful for sensitive detection, for example, mass spectroscopy at the quantum level[21]. In all of these applications, efficient cooling to the zero-point of motion is a crucial prerequisite.

8. Methods for zero-point laser cooling

With the use of Doppler cooling,[9] minimum energies for a bound atom in one dimension are approximately equal to $\hbar\gamma/2$, where $2\pi\hbar$ is Planck's constant and γ is the natural linewidth of the cooling transition. For an atom bound in a harmonic well with "vibrational" frequency ω_v, this energy can be expressed as $\hbar\omega_v(<n_v> + ½)$ where n_v is the harmonic oscillator quantum number. Since Doppler cooling is valid in the regime where $\gamma \gg \omega_v$, limiting kinetic energies for Doppler cooling necessarily imply $<n_v> \gg 1$.

When $\omega_v \gg \gamma$, cooling can be achieved in the "resolved sideband" regime.[34] Consider a two-level atom characterized by resonant transition frequency ω_0 and radiative linewidth γ. If a laser beam (frequency ω_L) is incident along the direction of the atomic motion, the absorption spectrum is composed of a "carrier" at frequency ω_0 and resolved frequency-modulation sidebands spaced by ω_v which are generated from the Doppler effect. Cooling occurs if the laser is tuned to a lower sideband, for example, at $\omega_L = \omega_0 - \omega_v$. In this case, photons of energy $\hbar(\omega_0 - \omega_v)$ are absorbed and spontaneously emitted photons of average energy $\hbar\omega_0$ return the atom to its ground internal state (assuming that $\hbar\omega_v$ is much greater than the photon recoil energy). Therefore, the atom's kinetic energy is reduced by $\hbar\omega_v$ per scattering event. Cooling proceeds until the atom's mean vibrational quantum number in the harmonic well is given by[2,9] $\langle n_v \rangle_{min} \simeq (\gamma/2\omega_v)^2 \ll 1$. Two experiments on single ions have reported laser cooling to the zero-point of motion.[35,36] In Ref. 36, resolved-sideband stimulated Raman transitions replaced single photon transitions; cooling to the zero-point of motion was achieved 98% of the time in 1-D and 92% of the time in 3-D. Before correlated states between ions can be obtained, zero-point cooling must be extended to certain modes (such as the center-of-mass oscillation) on collections of N ($\geq$ 2) ions.

Dramatic advances in laser cooling of neutral atoms were made with the introduction of polarization-gradient and Sisyphus cooling.[37] Because of this success, it was interesting to investigate whether or not these methods would yield $<n_v> \simeq 0$ for bound atoms. Theoretical investigations have shown that particular applications of Sisyphus cooling or polarization-gradient cooling[38,39,40] to trapped atoms appear to give cooling near $<n> \simeq 1$, but not $<n> \rightarrow 0$. More recently, it has been suggested that ions in the $n = 0$ state could be selected from a distribution by null detection.[41] Other discussions of laser cooling appear in the contributions to this conference by Knoop et al., Sugiyama and Yoda, and Alekseev and Krylova.

9. Summary

Laser cooling, which can suppress time-dilation shifts, appears to be essential to achieve high accuracy in atomic frequency standards. However, before the benefits of laser cooling can be realized with trapped ions, the velocity in the coherent or nonthermal modes of motion must be suppressed. We have discussed several approaches which address this problem. It also appears that laser cooling may have other benefits in metrology. We have pointed out the application of zero-point laser cooling to the generation of correlated states for improved signal-to-noise ratio in spectroscopy, fundamental quantum measurements and quantum computation.

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