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High Accuracy Spectroscopy of Stored Ions*

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ABSTRACT: Physical limitations to the achievement of accurate, high resolution spectroscopy on stored ions are briefly discussed. For experiments on ion clouds, a compromise between frequency stability and second order Doppler shift uncertainty must be made. Expected performance for specific examples using single ions and ion clouds is discussed.

1. Introduction

In this paper we discuss some of the basic physical limitations to the achievement of accurate, high resolution spectroscopy of electromagnetically confined ions. For brevity, we do not address many practical problems, such as local oscillator spectral purity. These problems, although important, will probably not cause the ultimate limit in accuracy and resolution.

This topic naturally divides between experiments on many ions and single trapped ions. We anticipate that for experiments using more than one stored ion, the uncertainty in the second order Doppler (time dilation) shift will be the accuracy-limiting systematic effect[1-3]. This has been the case in the past. For single ions, the uncertainty in the second order Doppler shift can be made extremely small. The limiting uncertainty for single ions may depend on the experiment[4-6], but inaccuracies should eventually be less than 1 part in 10^{18}. Use of single ions however might be precluded if the signal-to-noise ratio (S/N) is not high enough to achieve the desired measurement precision in a reasonable length of time. Therefore, we must consider a trade-off between second order Doppler shift and S/N, both of which increase as the ion number increases.

As in any spectroscopic experiment, we must account for the perturbations due to static and time varying multipole interactions for electric, magnetic, and gravitational fields. These include atom-trap field interactions, collisions, shifts due to uncontrolled electric and magnetic fields, gravitational red shifts, etc. (see refs 1-9 and other contributions in these proceedings).

2. Optical-Pumping, Double-Resonance Experiments on Stored Ions

We will assume ion spectra are observed by optical-pumping, double-resonance methods in which optical fluorescence changes are monitored. In Fig. 1, we

assume that level 3 is at an optical energy above the ground state 1. The transition of interest for high resolution spectroscopy is between levels 1 and 2 (frequency ω_0) which we designate the "clock" transition. A_2 and A_3 are radiative decay rates and R is the excitation rate between levels 1 and 3. R' and R" denote pumping rates between the clock levels due to optical pumping sources (independent of the clock radiation) applied simultaneously with R.

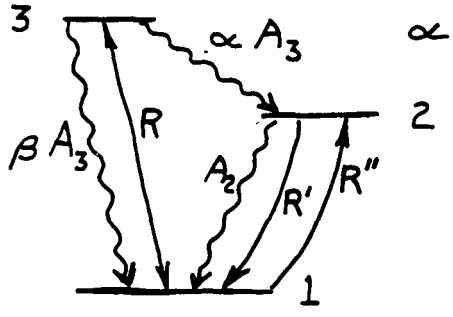

Fig. 1 Schematic diagram for optical pumping, double resonance experiments.

2.1. "Depletion" pumping. Assume A_2, R', R" « R < βA_3, and $\alpha \cong \beta$. When R is applied, atoms are pumped from level 1 to level 2 through level 3 (level 1 is depleted). If radiation (not shown in Fig. 1) is applied near the clock frequency $\omega_0 \equiv (E_2-E_1)/\hbar$, some of the atoms will be transferred back to level 1. This causes an increase in the detected fluorescence.[7,8]

2.2. "Electron Shelving."[4] We assume that the clock radiation and pumping radiation are applied sequentially to avoid light shifts on the clock transition. Assume $\alpha = 0$, R' » R", and A_3 » (A_2+R'-R"). In steady state with R, R' and R" applied, the ions spend most of the time in level 1. If ions are in level 2 after the clock radiation is applied and if $R \cong A_3$, we can observe the absence of about $N_d \cong \eta A_3(A_2+R'-R")^{-1}$ detected (3→1) photons for each photon absorbed on the clock transition (η is the net photon detection efficiency). When N_d » 1 and sources of technical noise are absent, the fluctuations in detected counts are caused not by the photon shot noise but by the fluctuations in the number of ions which have made the transition from level 1 to level 2 (Ref. 1). This is the fundamental noise limit. In Ref. 5, $\alpha \cong 10^{-7}$, R', R" = 0, $A_3 \cong 4.3\times10^8$ s^{-1}, $A_2 \cong 12$ s^{-1}, $\eta \cong 5\times10^{-4}$, $N_d \cong 20000$. In Ref. 9, $\alpha = 0$, $A_2 = 0$, $N_d \cong 1$, and R » R' » R" (R, R', and R" result from the same radiation source).

3. Frequency Stability

The signal is the response of the fluorescence detector to changes in frequency of the oscillator which drives the clock transition. As a specific example, we assume use of the Ramsey separated field method in time domain. That is, excitation of the clock transition is by two phase coherent pulses of radiation of duration ΔT_R separated by T_R.[9] For mathematical simplicity we assume ΔT_R « T_R « A_2^{-1} and R'-R" » A_2 (In Ref. 5, R', R" = 0). The electron shelving method of detection is assumed in the case where N_d » 1 in a time much less than (A_2 + R'-R")$^{-1}$ « T_R; that is, the detection time is less than the repumping time, which

is less than T_R. If we probe the clock transition on each side of the central Ramsey peak, we can develop an error signal which steers the average frequency to ω_0. Under the above assumptions, the measurement precision as characterized by the two-sample Allan variance[10] for the locked oscillator is[1,11]

$$\sigma_y(\tau) = (\tau N T_R \omega_0^2)^{-\frac{1}{2}} \qquad (1)$$

where τ is the measurement time and N is the ion number.

4. Second Order Doppler Shift in Ion Clouds

From Eq. 1, if ω_0 or T_R is too small, it may be necessary to use large N to obtain the desired measurement precision in a reasonable length of time. We anticipate that the largest systematic effect in an experiment on many stored ions will be the uncertainty in the second order Doppler shift, $\Delta\nu_{D2}$. Fractionally, this shift is equal to $\langle\Delta\nu_{D2}/\nu_0\rangle \cong -\langle v^2\rangle/2c^2$ where $\nu_0 = \omega_0/2\pi$, $\vec{v}$ is the ion velocity, c is the speed of light, and $\langle\ \rangle$ denotes an average over the N ions and over time. We would like to minimize the second order Doppler shift while simultaneously making the number of ions as large as possible to increase stability. These two requirements are contradictory and so a compromise between accuracy and stability must be made.

4.1. rf Trap

We assume that the pseudopotential approximation[12] is valid and the kinetic energy of the secular motion (the motion associated with the pseudopotential) is reduced to a value where the Debye length of the ion cloud is less than the cloud dimensions[13]. If the cloud is large enough, this can be accomplished by bringing the secular motion nearly into thermal equilibrium with a background gas (e.g., He) through ion-neutral collisions[7,14]. In principle it can also be accomplished by the method of laser cooling but so far this has been achieved with only a few ions in an rf trap because of rf heating.

Under the above conditions, the average kinetic energy per ion is dominated by the rf driven micromotion [3,14]. For a spheroidal cloud of radius r_{c1} and height $2z_{c1}$ we calculate $\langle\Delta\nu_{D2}/\nu_0\rangle = -2a(2z_{c1}^2 + r_{c1}^2)/5mc^2$ where $a \equiv q^2V_0^2/(m\Omega^2(r_0^2+2z_0^2)^2)$. Here, q and m are the ion mass and charge, V_0 and Ω are the applied rf potential and frequency and r_0 and z_0 are characteristic trap dimensions[12]. From this expression and the ion density[12,13], we have for a spherical cloud [3,14]

$$\langle\Delta\nu_{D2}/\nu_0\rangle = -3\omega_s^2 r_{c1}^2/10c^2 = -3q^2N/10r_{c1}mc^2 = -3(\omega_s N q^2/m)^{2/3}/10c^2 \qquad (2)$$

where ω_s is the secular frequency for a single ion in the trap. From Eq. 2, we derive a relation between N and $\langle\Delta\nu_{D2}/\nu_0\rangle$,

$$N = -2.16 \times 10^{16} r_{cl} M \langle \Delta\nu_{D2}/\nu_0 \rangle / Z^2 \qquad (3)$$

where Z is the ion charge (in units of the proton charge), M is the ion mass in atomic mass units, and r_{cl} is the radius of the cloud in centimeters. Equation 3 shows that for an assumed value of $\langle \Delta\nu_{D2}/\nu_0 \rangle$, we would like to work with as large a cloud (low density) and ion mass as possible.

4.2. Penning Trap

We will assume that the internal (cyclotron and axial) temperature of the ions is reduced to a value where the second order Doppler shift is dominated by the velocity of ions due to cloud rotation at angular frequency ω[3,13,15]. This rotation is an essential part of ion storage in the Penning trap since it provides the inward Lorentz $(q(\vec{v}\times\vec{B})/c)$ force which overcomes the radially outward force due to the trapping fields and space charge. We have $\langle \Delta\nu_{D2}/\nu_0 \rangle = -(\omega r_{cl})^2/5c^2$ where the cloud is a uniformly charged spheroid of height $2z_{cl}$ and radius r_{cl}[13]. From Ref. 13, the density of ions is given by $n_0 = m\omega(\Omega_c - \omega)/2\pi q^2$ where $\Omega_c = qB_0/mc$ is the ion cyclotron frequency. From these two expressions, the total number of ions in the cloud is [3]

$$N = 3.10 \times 10^{13} B(T) \langle -\Delta\nu_{D2}/\nu_0 \rangle^{\frac{1}{2}} z_{cl}(r_{cl} - r_c')/Z \qquad (4)$$

where B is in teslas, z_{cl} and r_{cl} in centimeters and $r_c' \equiv (5 \langle -\Delta\nu_{D2}/\nu_0 \rangle)^{\frac{1}{2}} c/\Omega_c$. For a given value of $\langle \Delta\nu_{D2}/\nu_0 \rangle$, large B, z_{cl} and r_{cl} (low density) are desirable. N is independent of M (for the typical case $r_c' \ll r_{cl}$). We may choose B to make the clock transition field independent to first order[1,9].

5. Single Ions

From Eq. 1, if ω_0 and T_R are large enough, stability can be high enough for practical use, even with N = 1. For single ions, many systematic effects become negligible[1-6]. For example, from our recent experiments on single Hg^+ ions[5], we estimate uncertainties in the second order Doppler shift correction to be less than 1 part in 10^{20}. What will actually be the limiting systematic effect will depend on practical considerations and which ion is used. For the sake of definiteness we consider two cases of interest at Boulder and list what we think are the limiting systematic effects.

5.1 $^{199}Hg^+$, $5d^{10}6s\ ^2S_{\frac{1}{2}}(F=0, m_F=0) \leftrightarrow 5d^9 6s^2\ ^2D_{5/2}(F=2, m_F=0)$ optical quadrupole transition ($\nu_0 = 1.07 \times 10^{15}$ Hz).

(1) Magnetic field sensitivity. From the Breit-Rabi formula, we obtain $(\nu_0(B) - \nu_0(B=0))/\nu_0 = -1.8 \times 10^{-5} B^2$ where B is expressed in teslas. It should be possible to obtain fractional precision of better than 1 part in 10^{18}.

(2) $^2D_{5/2}$ state quadrupole interaction. The quadrupole moment of the $^2D_{5/2}$ state can couple to static electric field inhomogeneities to cause a shift of this level. In a suitable (Cartesian) coordinate system, the atomic quadrupole moment

interacts with a potential of the form $\varphi = A(x^2+y^2-2z^2) + \delta A(x^2-y^2)$. In the specific case where $\delta A = 0$ and $\vec{B}$ (quantization axis) is parallel to z, then $\Delta\nu_Q \cong$ -1.8 Hz for 1 V applied to the ring electrode for the trap of Ref. 5. The presence of contact potentials on the trap electrodes makes the direction of z (in φ) uncertain but $\Delta\nu_Q$ averages to zero for three measurements of ν_0 where the directions of $\vec{B}$ are mutually orthogonal.[16]

5.2 $^{199}Hg^+$ ground state (F=0, m_F=0) $\leftrightarrow$ (F=1, m_F=0) hyperfine transition. (ν_0 = 40.5 GHz)

(1) Magnetic field sensitivity. $(\nu_0(B) - \nu_0(B=0))/\nu_0 \cong 0.24\ B^2$ where B is expressed in teslas. For single ions, magnetic field homogeneity requirements are not stringent.

Table I: Expected performance in particular systems. In rows 1 and 2, N is calculated from Eq. 4. Eq. 1 is used to determine $\sigma_y(1\ s)$. For N > 1, the ion cloud is assumed to be spherical (radius = r_{cl}). B is chosen to minimize field sensitivity. For T_R > 1 s, quoted stabilities are referred to 1 s.

SYSTEM	$\langle -\Delta\nu_{D2}/\nu_0 \rangle$	B(T)	r_{cl}(cm)	N	T_R(s)	$\sigma_y(1\ s)$
$^9Be^+$ hyperfine $\nu_0 \cong$ 303 MHz (Penning trap)[9]	10^{-15}	0.819	0.5	2.0×10^5	100	1.2×10^{-13}
$^{201}Hg^+$ hyperfine ν_0 = 25.9 GHz (Penning trap)[1]	10^{-16}	0.534	0.5	4.1×10^4	100	1.9×10^{-15}
$^{199}Hg^+$ hyperfine ν_0 = 40.5 GHz (rf trap)	$<10^{-15}$	$\cong$ 0	--	1	100	3.9×10^{-13}
$^{199}Hg^+$ optical quadrupole $\nu_0 = 1.07\times10^{15}$ Hz (rf trap)[5]	$<10^{-18}$	$\cong$ 0	--	1	0.025	9.4×10^{-16}

6. Examples

Table I lists some possibilities of interest at Boulder. Implicit in the Table is the use of laser cooling. Sympathetic cooling [9,17] may be necessary for N > 1. Reasonable short term stability is obtained for single $^{199}Hg^+$ ions using the ground state hyperfine transition. Systematic effects should be more easily

controlled using single ions.[1-6] Fluctuations in cloud density and size (and therefore detected fluorescence) could degrade stability over that shown in the first two rows. For example, laser induced instabilities similar to those cited in Refs. 15 and 18 have been observed in all Penning traps at NBS; these instabilities must be suppressed to achieve $\sigma_y(\tau)$ given by Eq. 1.

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