NEW POSSIBILITIES FOR FREQUENCY STANDARDS USING LASER COOLING AND DETECTION OF STORED IONS

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Abstract

Techniques for storing ~ 10^4 to 10^5 ions for periods of hours to days are described in detail. Ion dynamics and detection techniques are also covered. Experimental data is presented demonstrating that ions stored in a room temperature Penning style ion trap using dc electric and magnetic fields can be cooled to near ZERO Kelvin using a suitable laser. This cooling technique, which is appliciable to very many atomic and molecular ions, can reduce fractional frequency shifts due to the second order Doppler effect to smaller than 10^{-15}. This cooling, coupled with the other attractive features of ion storage, promises to make possible frequency standards with stabilities in the 10^{-16} to 10^{-18} range with accuracies of order 10^{-15} or better. One possible candidate for a microwave frequency standard is described. The projected fractional frequency stability is $\sigma_y(\tau) = 4\mathrm{x}10^{-15}\ \tau^{-1/2}$ for $16\mathrm{s} < \tau < 10^4\mathrm{s}$.

Introduction

All presently known techniques for achieving ultrahigh resolution and accuracy in atomic or molecular spectroscopy are limited by one or more of the following effects: 1) residual first order Doppler, 2) second order Doppler, 3) perturbations due to confinement - for example the wall shift in hydrogen, 4) transit time broading, and 5) signal to noise ratio. It has been realized for many years that the storage of ions in an electromagnetic trap reduces the uncertainty in the residual first order Doppler effect to a negiligible value, that the uncertainties in perturbations due to the confinement fields could be made fractionally smaller than 10^{-14}, and that the very long storage times permitted the realization of extremely narrow linewidths limited by the natural lifetimes of the states involved and/or by power broading. Until only very recently, however, the second order Doppler effect appeared to present a formidible obstacle to attaining fractional accuracies below 10^{-13}, moreover the signal to noise ratio in most experiments was rather poor, severely limiting attainable frequency stabilities.

Very recently we have succeeded in experimentally demonstrating that ions contained within a room temperature Penning style ion trap can be cooled close to zero Kelvin using laser induced refrigeration. This technique is very widely applicable and reduces the second order Doppler effect to below parts in 10^{-15} depending on the ion and the amount of cooling. Signal to noise problems have been greatly improved with the advent of stable tuneable dye lasers. As a result, ion storage techniques now present a very promising opportunity to achieve frequency stabilities of 10^{-16} to perhaps 10^{-18} and absolute frequency accuracies in excess of 1 part in 10^{15}.

In this paper we will describe the general techniques of ion storage, present the experimental verification of laser induced refrigeration and briefly describe one possible candidate for a microwave frequency standard including projected numbers for the obtainable frequency stability.

Ion Storage Techniques

Ion traps are available in two basic varieties, the Paul trap and the Penning Trap. Fig. 1 shows the cross section of the cylindrical symmetric electrodes generally used in both styles of traps. In the Paul trap an rf voltage is applied between the center ring electrode and the two end caps.[1] The resulting inhomogeneous rf electric fields produce a three dimensional well for ions of approximate depth $e/m\ (V_{rf}/\omega_{rf})^2\ (2Z_o)^2$ where e/m is the ion charge to mass ratio, $R_o = \sqrt{2}Z_o$, $2R_o$ is the diameter of the ring electrode, $2Z_o$ is the axial separation of the two end caps, V_{rf} is the amplitude and ω_{rf} the frequency of the applied rf electric field. The disadvantage of this type of trap is that the applied rf trapping fields impress a sympathetic micromotion on the ions thereby raising their kinetic temperature by an amount proportional to their average distance from the center to the trap. If many ions are simultaneously stored, the Coulomb potential will prevent them from all occupying the center of the trap, thereby preventing the ion cloud from being cooled to low temperatures.[1,2,3] If only a single ion is stored this problem is greatly reduced.[4]

In the Penning trap, three dimensional confinement is obtained by applying a dc potential, V_o, between the ring and end caps of appropriate polarity and applying a homogeneous magnetic field along the axis of trap symmetry (Z axis).[5] The electrostatic potential created is approximated by:

$$V = \frac{V_o (r^2 - 2 Z^2)}{R_o^2 + 2Z_o^2}$$

and is shown in Fig. 2.

It can be seen from Fig. 2 that a low energy positive ion would be reflected from both end caps, however it would tend to fall out of the trap along the radial direction. The homogeneous magnetic field along the Z axis causes the ions to undergo cyclotron motion and therefore provides radial confinement. The net effect then is for the ions to undergo simple harmonic motion along the Z axis at frequency ω_z. In the radial direction they orbit on a circle with angular velocity $\omega_c^1 = \omega_c - \omega_m$ with the center of the circle precessing around the trap axis at angular frequency ω_m, where $\omega_c = eB/mc$, and $2\omega_m^2 - 2\omega_m\omega_c + \omega_z{}^2 = 0$. This is schematically shown in Fig. 3. The long range Coulomb collisions establish a Boltzmann distribution of kinetic energies in the ω_z and ω_c motions in a few ms.[1,2,3,5] The ω_m motion is essentially decoupled from the other motion.

The kinetic temperature of the ions can be measured by monitoring the first order Doppler width of one of their internal natural resonance lines or by monitoring the induced image current which flows from one end cap to the other in response to their thermally driven ω_z motion.[5,6,7] Such a detection scheme is shown in Fig. 4, while a typical signal for a single ion species is shown in Fig. 5. The area under the ion signal curve is porportional to the product of ion number, N, times absolute temperature, T.[3,6,7] Ion lifetimes are typically many hours to many days usually limited by chemical reactions with the background gas or radial diffusion, therefore variation in the integral over a few minutes is uniquely related to changes in ion termperature.

Laser Induced Cooling

The laser cooling is perhaps most easily described in terms of an energy arguement.[8,9] Assume that the stored ion species possesses an internal characteristic transition at frequency ν_o. Since the ions have finite kinetic energy this characteristic transition will be observed to be Doppler broadened when viewed in the laboratory frame.

If a laser of frequency ν is tuned to the low frequency side of the Doppler broadened line, then the ions will absorb the laser photons only when approaching the laser. However when they spontaneously re-radiate they will on the average radiate the proper, or unshifted resonance frequency, ν_o. The energy deficit must be accounted for by a reduction in kinetic energy, i.e. the kinetic energy decreases by $h(\nu_o - \nu)$ per scattering event. If $(\nu_o - \nu)$ is .5 GHz, a typical half width for an optical transition of a 300K absorber, then each scattering event removes ~ .02K on the average. Therefore only 15000 scatterings per ion are required in order to cool the entire ion cloud from 300K to almost zero Kelvin. Obtainable cooling rates can be very large for optically allowed transitions since one could scatter about 10^7 photons per second. This would provide a cooling rate of about 3 x 10^4 K/s assuming $\nu_c - \nu$ = .5 GHz. Laser frequency stability of even 100 MHz is sufficient for this cooling process.

Experimental verification of this laser induced cooling scheme has been obtained using approximately 10^4 Mg ions contained within a Penning style ion trap with $2R_o = 1.26$ cm, $2Z_o = .76$ cm, and B = 1T (10^4 Gauss). Mg^+ was chosen because of its simple level structure shown in Fig. 6, and the convenient overlap of available doubled dye lasers with the electric dipole allowed $2S_{1/2}$ to $2P_{3/2}$ transition at 279.6 nm. Fig. 7 shows the temperature of about 10^4 Mg ions as derived from the induced image current, during a laser induced cooling cycle. The ions were first heated to approximately 700K prior to turning on the 8 μW laser, which was tuned to the low side of the $2S_{1/2}$ to $2P_{3/2}$ transition. After about one minute the ion temperature is seen to approach zero Kelvin to within the measurement uncertainty of approximately 20K. The laser was turned off and the ions allowed to gradually rethermalize via collisions with the background gas. The background gas pressure was intentionally raised by turning off the ion pumps for two days so that the rethermalization time was fast enough to make a good demonstration figure. Normally the ion rethermalization time is at least 10 times longer than that seen in Fig. 7. We have also verified that the laser cooling does not eject ions from the trap.

Under these conditions of low laser power, the ions cool until a balance is reached with the heating due to collisions with the room temperature background gas. Based on the ratio of cooling time constant to the rethermalization time constant we would estimate an ion temperature of about 40K, for Fig. 7 which is somewhat higher than the direct measurement of T. Under normal vacuum conditions where the rethermalization time is much longer we estimate an ion temperature of about 1K to 2K. Further cooling could be obtained by increasing the laser power. The ultimate cooling limit is predicted to be $kT = 1/8h\Delta\nu_o$, where $\Delta\nu_o$ is the natural width of the resonance used in the cooling. For the $2s_{1/2}$ to $2P_{3/2}$ transition in Mg this limit is .5 mK.[9]

The above data and discussion clearly demonstrate that the second order Doppler shift can be reduced for below a part in 10^{15} for ions contained within a Penning style ion trap.

It should also be noted that the long range Coulomb interaction between ions can be used to cool other ion species contained within the trap. Since the cooling mechanism is separate from the trapping fields, the cooling can be periodically turned off to permit low power interogation of a narrow resonance feature. These features make this particular type of laser induced cooling a very widely applicable technique for realizing previously undreamed of precision in atomic and molecular spectroscopy.

Possible Microwave Frequency Standard Based on Laser Cooling

One possible candidate for a microwave frequency standard based on ions contained within a Penning style ion trap and laser cooled is Ba$^+$. The level diagram for Ba$^+$ is shown in Fig. 8. The $6s_{1/2}$ to $6p_{1/2}$ transition at 493 nm is within the range of available tunable dye lasers. Unfortunately the ions decay approximately one third of the time to the $5d_{3/2}$ level which has a lifetime of about 1 s. In order to obtain sufficient cooling rates, the $5d_{3/2}$ level must be pumped with another laser back to the $6p_{1/2}$ level.[10]

Ba$^+_{137}$ has a nuclear spin of 3/2 and a magnetic hyperfine splitting of about 8 GHz. The hyperfine separation vs magnetic field is shown in Fig. 9. Note the existence of extrema in the separation of several hyperfine levels. This feature is not unique to BA$^+_{137}$ but is generally found whenever one has transitions between levels having a multiplicity of 3 or more.

The existence of extrema in the hyperfine structure allows one to store the ions at a large magnetic field and still obtain relatively small shifts due to changes in magnetic field. By stabilizing the magnetic field to a part in 10^7 the uncertainty in the level separation could be reduced to a part in 10^{15}. Magnetic field stabilization to this order should be easily achieved by measuring a magnetic field dependent transition, e.g. the (2,-1) to (2,-2) transition frequency of the stored Ba$^+$ ions. Since the ion cloud is symmetrically distributed within the trap in a volume of ~ $1 mm^3$, and all ions oscillate back and forth along the Z axis while rotating about the Z axis, the magnetic field dependent line width and the magnetic field averaging should both be excellent.

Penning ion traps have the disadvantage that the magnetic field splits the various states, however, this fact allows one the possibility to cycle the ions many times between two well defined levels. In an optical-microwave double resonance scheme such as proposed here, each ion making a microwave transition can be made to emit many optical photons. A possible (although complicated) cycle for Ba$^+$ would be 1) prepare the ions in the (1,-1) level via optical pumping and selective microwave relaxation in the ground state, 2) drive the (1,-1) to (2,-1) transition at a low level with the laser off, 3) irradiate all the ions with a laser tuned about 100 MHz below the $6s_{1/2}$ (2,1) to $6p_{1/2}$ (2,2) transition simultaneously mixing the (2-1), (2,0), (2,1) and the (2,2) levels with microwaves and driving the $5d_{3/2}$ (eg (2,1)) level to the $6p_{1/2}$(2,2) level with another laser and appropriate microwave frequencies to mix the $5d_{3/2}$ levels. Scattered photons at 493.4 nm are collected and used as an indicator of how many ions made the (1,-1) to (2,-1) transition, 4) repeat step 1 using both the laser and appropriate microwave frequencies to pump all the ions into the (1,-1) level of the ground state. The magnetic field can also be measured during this cycle. Note that step 3 provides the ion cooling as well as serving as a monitor of the microwave frequency transition. In principle the ions which have made the (1,-1 to 2,-1) transition can be made to radiate approximately 10^7 photons per second without degrading the microwave line width because ions decaying from the $6p_{1/2}$(2,+2) level cannot reach the F=1 levels of the $6s_{1/2}$ ground state.

There is some uncertainty in the rate at which the ions can be cycled from the $6s_{1/2}$ to the $6p_{1/2}$ state due to the decay to the metastable $5d_{3/2}$ state. The hyperfine seperation of the $5d_{3/2}$ levels, purity, and the overlap of the transitions from $6p_{1/2}$ to $6s_{1/2}$ and $5d_{3/2}$ are not well known. Such complications are specific to Ba$^+$ and need not be present in other possible ions. Therefore in the following discussion we assume that efficient pumping of ions from the $5d_{3/2}$ state to the $6p_{1/2}$(2, 2) level can be achieved.[10]

The possible attainable fractional frequency stability can be estimated from the following

$$\sigma_y(\tau) = \frac{1}{Q\ S/N} \qquad Q = \pi\tau_o\nu$$

Where Q is the microwave line quality factor, ν_o is the microwave frequency (8 GHz), τ_o the microwave observation time, and S/N, the signal to noise ratio, is given by:

$$S/N = \left[KN_sN_i\tau_\ell \frac{t}{2(\tau_\ell + \tau_o)}\right]^{\gamma_a}$$

where K is the photon collection efficiency, N_s is the number of scattered photons per second, N_i is the number of ions which make the microwave transition, and τ_ℓ is the laser cooling time.

For $\tau_\ell = 4s$, $\tau = 4S$, $N_s = 2.5 \times 10^{5}/S$, $N_i = 10^4$

$$S/N = \left[(.01)\ 10^6\ 10^4 \frac{1}{16}\ t\right]^{-1/2} = 1/4 \times 10^4\ t^{1/2}, Q = 10^{11}$$

$$\sigma_y(\tau) = \frac{1}{Q\ S/N} = 4 \times 10^{-15}\ t^{-1/2}$$

The achievement of such good short-term frequency stability would make it possible to reach frequency stabilities of order 10^{-17} and evaluate systematic shifts of less than 1×10^{-15}.

The numbers generated for Ba$^+_{137}$ are only to illustrate the great potential for improved microwave frequency standards. Similar techniques could be applied to achieve even better frequency stability at infrared, visible, or ultraviolet frequencies. For example several optical transitions have possible line Q's of order 10^{14}.[11] One would expect the following general possibilities for optical frequency standards based on ion storage:

$Q \sim 10^{12}$ to 10^{14}

$S/N \sim 10$ to 10^4

$\sigma_y(\tau) \sim 10^{-13}$ to $10^{-18} t^{-1/2}$

General estimates of systematic effects which would limit the accuracy of a frequency standard based on cooled ions are listed below under the assumption that the ions temperature is below 1 K and that magnetic field independent or independendent or nearly independent transitions such as the (2,-1) to (1,-1) hyperfine transition in Ba^+_{137} described above are used.

1. 1st order Doppler $<10^{-16}$
2. 2nd order Doppler 10^{-15} to 10^{-17}
3. Perturbations due to confinement
    a. Magnetic field $\leq 10^{-15}$
    b. Electric fields $<10^{-15}$
    c. Collisions with background gas $<10^{-16}$

Conclusion

The general concepts of ion storage in Penning style ion traps have been described. Experimental evidence demonstrating the effectiveness of laser induced refrigeration which will make it possible to reduce fractional frequency shifts due to the second order Doppler effect to $\leq 10^{-15}$ was presented. It was also shown that the ion storage techniques and the laser induced cooling were applicable to a large number of ions and should therefore lead to previously unattainable precision and accuracy in atomic and molecular ion spectroscopy. Specifically, a sample calculation showed that fractional frequency stabilities of order 10^{-17} and absolute accuracies of order 10^{-15} are likely possible.

Acknowledgement

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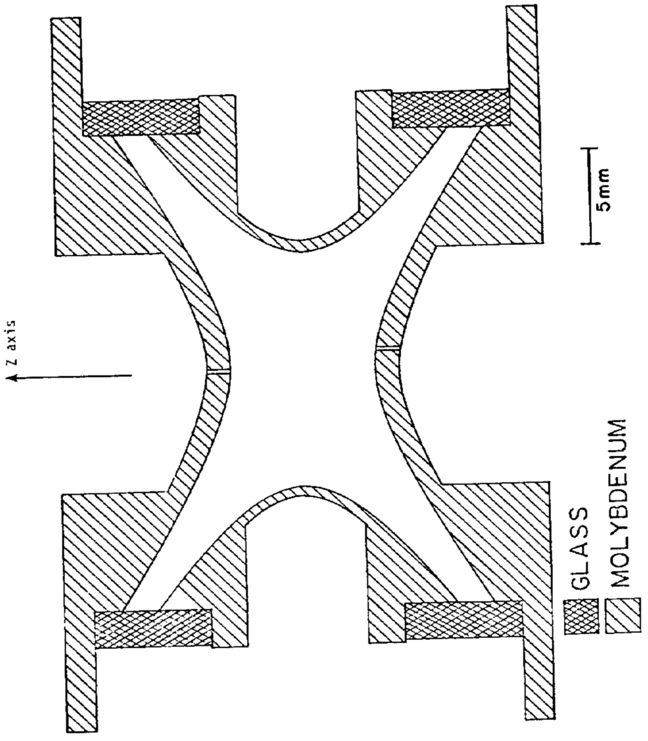


Fig. 1. Cross section of cylindrically symmetric electrodes generally used in Paul and Penning quadrupole ion traps.

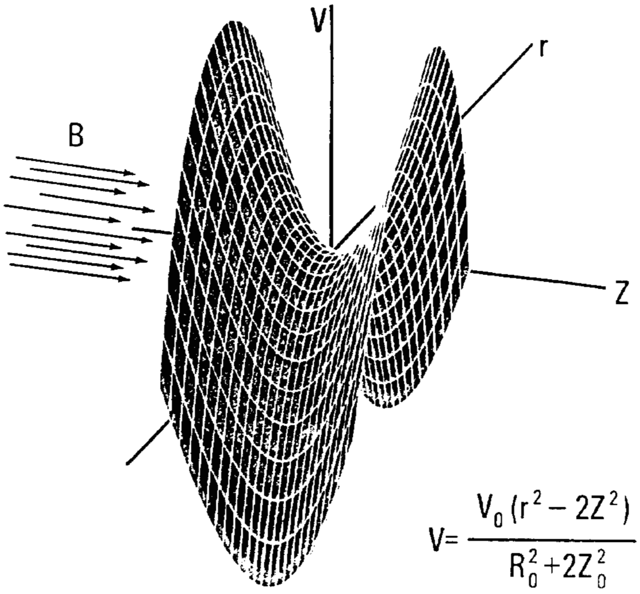


Fig. 2. Graphical plot of the electrostatic potential distribution resulting from biasing the trap end caps positive with respect to the ring.

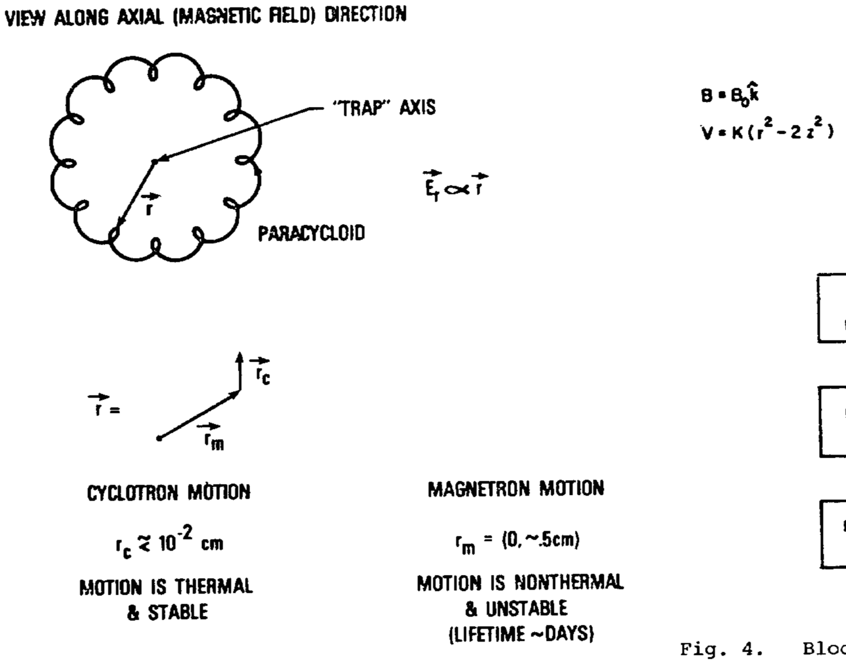


Fig. 3. View along the trap axis showing the slow precision of the ion cyclotron orbits about the Z axis due to the ω_m motion.

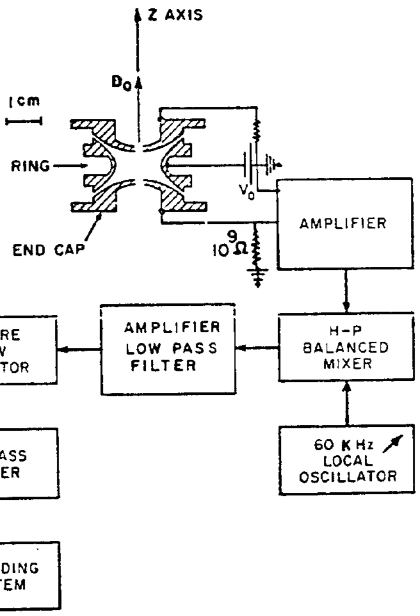
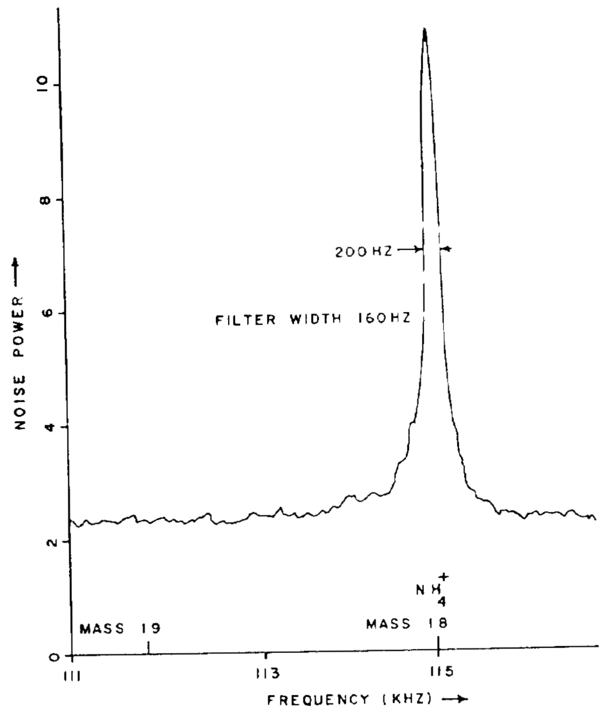


Fig. 4. Block diagram of the circuit used to detect induced ion image currents.



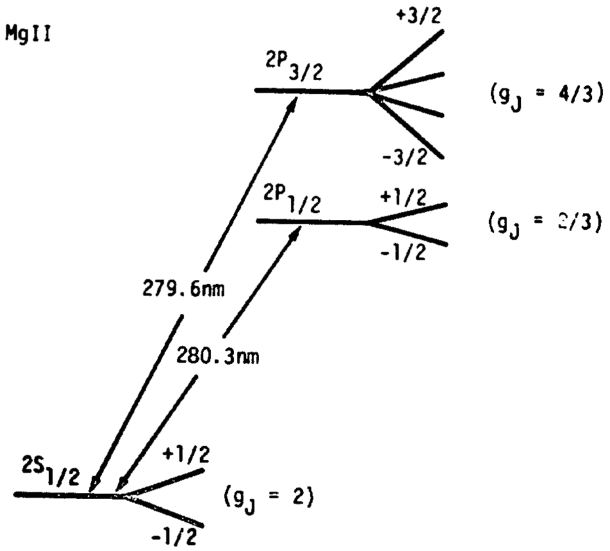


Fig. 6. Low lying energy levels of Mg^+.

Fig. 5. Plot of detected image power vs frequency. The large peak corresponds to the presence of NH_4^+ ions in the trap.

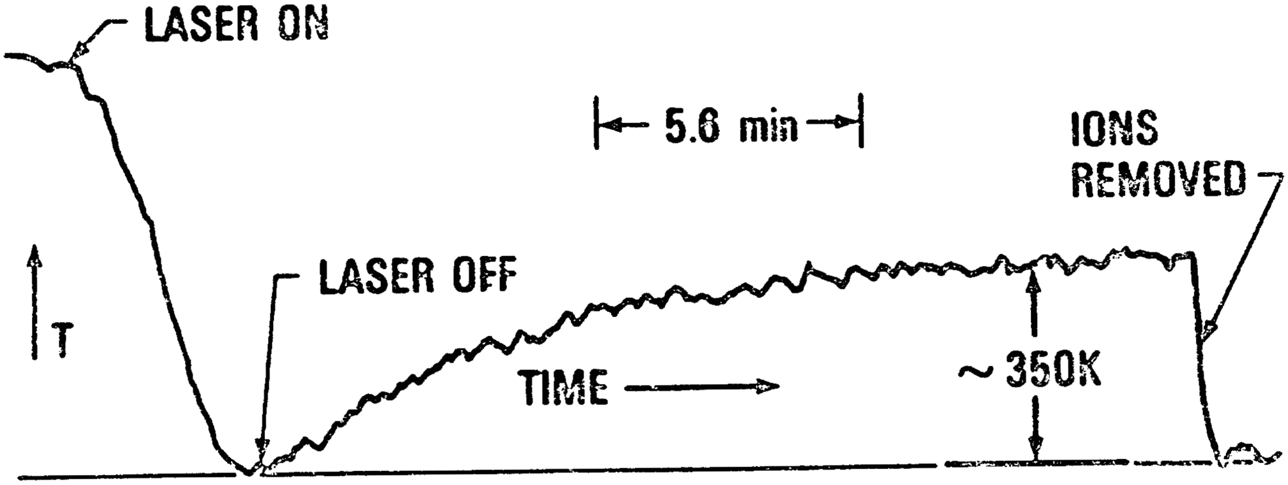


Fig. 7. Shows the cooling of Mg^+ due to laser induced refrigeration. Rethermalization when laser is off is due to collisions with the background gas at 350K.

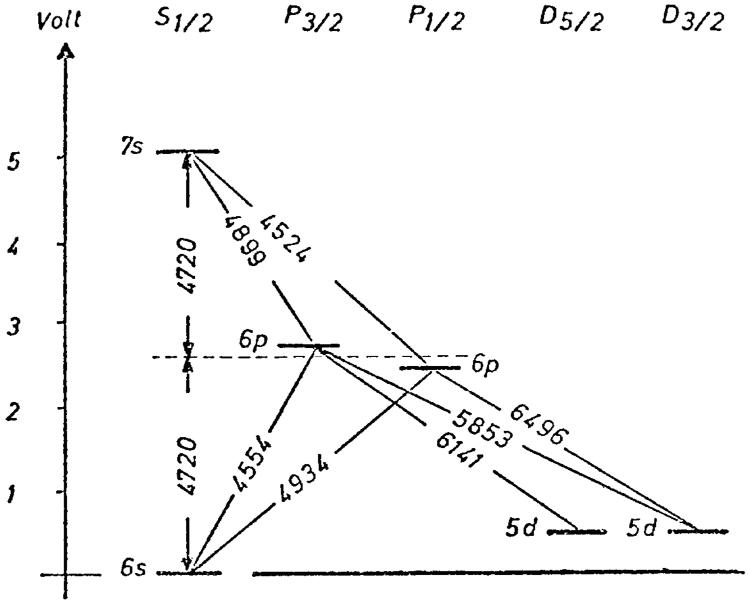


Fig. 8. Low lying energy levels of Ba^+.

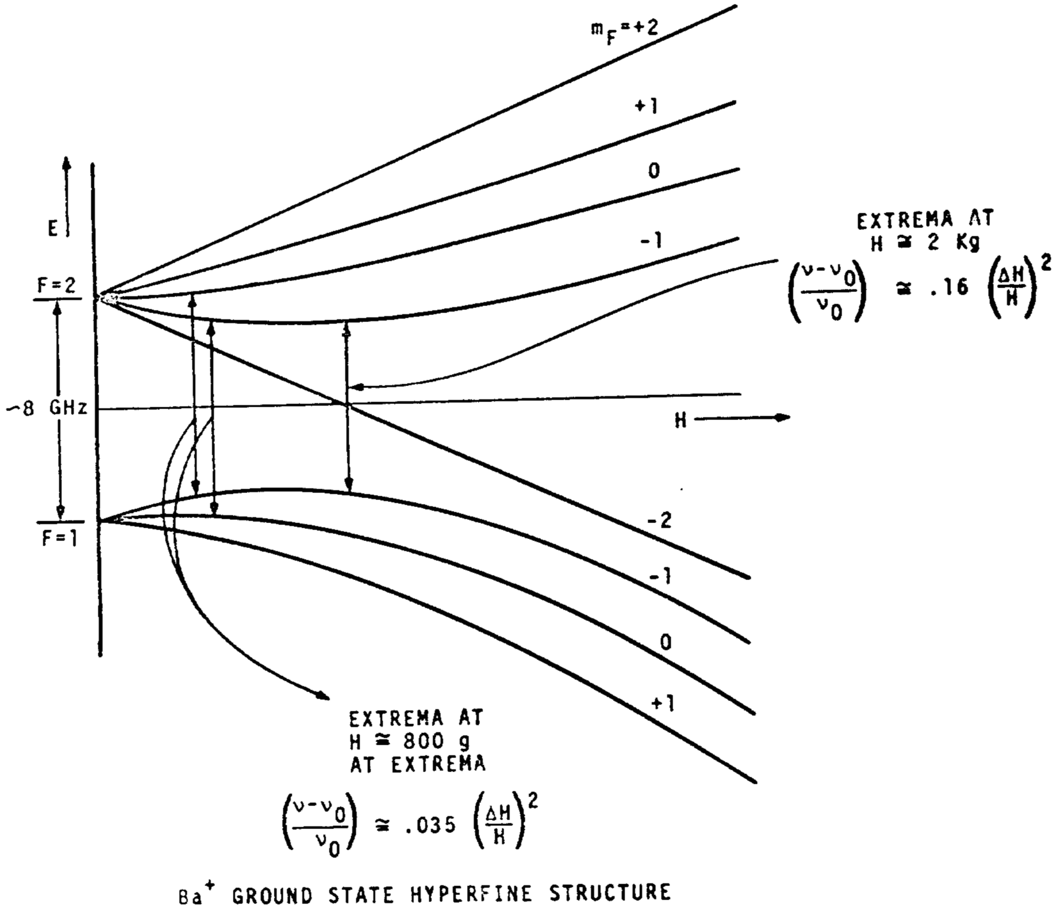


Fig. 9. Magnetic hyperfine splitting of the $6S_{1/2}$ ground state of Ba^+_{137}.