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Precision determination of the ground-state hyperfine separation in $^{199}Hg^{+}$ using the ion-storage technique

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By an optical double-resonance experiment on $^{199}Hg^{+}$, suspended in a rf quadrupole trap, we determined the ground-state hyperfine separation. Linewidths as small as 4 Hz have been observed. The final result of $\Delta\nu_{\text{hfs}} = 40\,507\,347\,997.8 \pm 1.0$ Hz includes corrections to zero magnetic field and zero kinetic energy. The accuracy of the result and possible improvements lead to the feasibility of a new frequency standard in the microwave region.

I. INTRODUCTION

Since 1966 several experiments[1-3] on light ions have demonstrated that very precise spectroscopic data on charged particles may be obtained by taking advantage of some characteristic properties of the ion-storage technique. Since the ions are confined in a small volume by electromagnetic fields in ultrahigh vacuum for very long times, the energy levels remain unperturbed to a very high order. Furthermore, the long coherence times in the absence of collisions or strong resonant fields lead to very narrow spectral lines in microwave transitions.

Recently Major and Werth[4,5] applied the optical pumping technique to an alkalilike heavy ion $^{199}Hg^{+}$. While their experiment lacked a good signal-to-noise ratio, they achieved a spectral resolution of 10^{10} in the 40.5-GHz ground-state hyperfine transition. This result represents a first step in the realization of earlier proposals by Major[6] and Schuessler[7] for a frequency standard based on the ion-storage technique.

Stimulated by the results of Refs. 4 and 5 we started a new experiment on the same ion. Using essentially the same technique we introduced several improvements, which finally allowed the investigation of the line shape of the field-independent $\Delta F = 1$, $\Delta M = 0$ hyperfine transition. A line shift was observed, varying with the operating point of the trap, which could be ascribed to the second-order Doppler effect.

The main features of our experiment are identical with the one described in Ref. 5. Consequently we refer the reader to this publication and restrict ourselves to a brief outline of the principles of the experiment and the main experimental problems before we discuss our results.

II. PRINCIPLES OF THE EXPERIMENT

A. Ion storage

The theory of ion storage has been treated by several authors,[8] so we restrict ourselves to a brief outline. The ions are confined in a rf quadrupole trap. A dc voltage U_0 and an ac voltage $V_0 \cos\Omega t$ are applied between a ring electrode (radius r_0) and two endcaps (half distance $z = \sqrt{\frac{1}{2}}\, r_0$) of hyperbolic shape to create the potential

$$\Phi = (U_0 + V_0 \cos\Omega t) r_0^{-2} (x^2 + y^2 - 2z^2) .$$

Under the action of the inhomogeneous force, the ions follow trajectories given by the solution of a differential equation of the Mathieu type. A class of solution, depending on two parameters a and q, leads to finite amplitude of the ion motion, i.e., to confinement. Following the notation of Ref. 8, we can describe this motion in first order by a harmonic oscillation at the frequency whose amplitude ω is modulated at the frequency of the trapping field:

$$x_i = A_i (1 + \tfrac{1}{2} q_i \cos\Omega t) \cos\omega_i t ,$$

$$i = r, z,$$

$$\omega_i = \tfrac{1}{2}\beta_i \Omega ,$$

$$\beta_i^2 = a_i^2 + \tfrac{1}{2} q_i^2 ,$$

$$a_r = -\tfrac{1}{2} a_z = 4eU_0 / m\Omega^2 r_0^2 ,$$

$$q_r = \tfrac{1}{2} q_z = 2eV_0 / m\Omega^2 r_0^2 .$$

Assuming an equal distribution of kinetic energy between the r and z directions, the maximum allowed energy will occur at the point where the ion touches the electrodes. In the approximation where the ion motion can be described by a harmonic oscillator, this point is given by

$$W_{\max} = \tfrac{1}{8} m\Omega^2 (\beta_r^2 r_0^2 + \beta_z^2 z_0^2) .$$

The ions are created by electroionization of the neutral background atoms inside the trap. Several times in the course of the experiment the apparatus was exposed to a sample of ^{199}Hg for some hours. This was enough to establish an equilibrium pressure between 10^{-8} and 10^{-10} Torr, using an 8-liter/sec ion pump. The number of trapped ions was estimated to be 10^5–10^6 and was monitored by an absorption signal across a resonant tank circuit.[9]

TABLE I. Trap data.

Material	Stainless steel
Ring diameter $2r_0$	40 mm
Distance between endcaps $2z_0$	28.6 mm
Light entrance hole diameter	12 mm
Light exit hole	12×18 mm^2
Transmission of endcap meshes	80%
Typical operating conditions	
Background pressure	10^{-8}–10^{-10} Torr
dc voltage U_0	25 V
ac voltage V_0	550 V
Driving frequency $\Omega/2\pi$	350 kHz
Ion oscillation frequency $\omega_z/2\pi$	49 kHz

Under ideal conditions the time of confinement will approach infinity. Collisions with background atoms reduce the storage time. In our experiment the lifetime of the ions inside the trap always exceeded several seconds and was typically several minutes.

Departures from the ideal trap geometry caused by the finite size of the electrodes, by holes for entrance and exit of the pumping light beam, and by a mesh (80% transmission) in both endcaps for fluorescent-light detection did not change the performance of the trap in a detectable manner. Table I gives the relevant data of the trap and the usual operating points.

B. Hyperfine transitions

The ground state of ^{199}Hg$^+$ is split by magnetic hyperfine interaction into two levels $F=0$ and $F=1$ of approximately 40.5 GHz separation. The precise measurement of this separation is performed by the optical double-resonance (ODR) method: A deviation from equilibrium population is introduced by optical pumping and a microwave transition is monitored by the change in fluorescence intensity. The 5-GHz Doppler width of the optical transition–due to the ion energy of several eV–is small compared to the level splitting and allows hyperfine pumping with unpolarized light. An easy way to pump is provided by a fortuitous matching of the unsplit 1942-Å line of ^{202}Hg$^+$ with one of the hyperfine components in the same transition of ^{199}Hg$^+$. Figure 1 illustrates the pumping scheme. Three microwave transitions between the $F=0$ and $F=1$ hyperfine levels are possible. The $\Delta F=1$, $\Delta M=\pm 1$ transitions serve to determine the magnetic field. The $\Delta F=1$, $\Delta M=0$ transition shows a quadratic field dependence according to the Breit-Rabi formula. A narrow linewidth limited by relaxation processes can be expected at this transition.

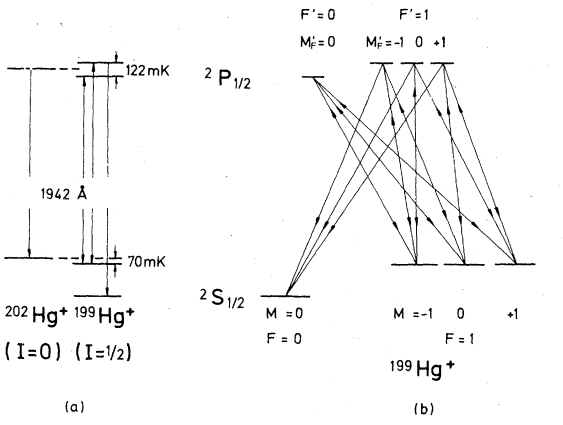

FIG. 1. Optical pumping of ^{199}Hg$^+$ by ^{202}Hg$^+$ radiation at 1942 Å.

The shape of a magnetic dipole transition between the hyperfine levels is a Lorentzian in the absence of collisions and under the action of a weak resonant microwave field. The oscillatory ion motion leads to a Doppler modulation of the field and results in an unshifted central line and sidebands at the distance of the oscillation frequencies. Major and Duchene[10] have shown that the amplitude of the sideband contains information about the mean energy of the ions and thus might be used to determine the second-order Doppler correction. In our experiment, however, the amplitude of the sidebands was too small to give useful information. Since the separation of (nominal) 50 kHz is large versus the linewidth of the $\Delta M=0$ transition, their influence on the line shape can be totally neglected.

C. Detection of microwave transitions

Figure 2 shows the experimental setup for optical pumping and detection of fluorescent radia-

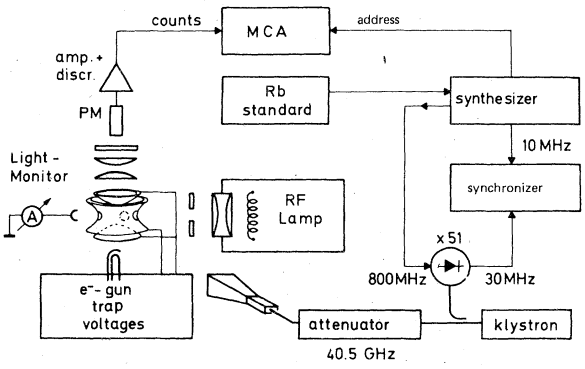

FIG. 2. Block diagram of the experiment.

TABLE II. Typical counting rates.

Stray-light background	165 000 s^{-1}
Ion background fluorescence by relaxation processes	2 700 s^{-1}
Microwave-induced fluorescence at resonance	2 050 s^{-1}

tion from the trapped ions. The pumping light came from a rf excited ^{202}Hg lamp. At input power of 100 W at 120 MHz the light flux in the 1942-Å ionic line was estimated to exceed 10^{13} photons per second. Because of the low ion density and consequently the small number of fluorescent quanta, it is crucial for the experiment to provide effective suppression of background light. A small part of it came from ion fluorescence caused by excitation by the ionizing electron beam, which was on all the time in order to increase the ion density by partial space-charge compensation. However the main source of background was stray light from the lamp. Careful adjustment was necessary to guide the pump light beam through a series of apertures and to achieve sufficiently low count rates. Table II give typical values.

A detailed treatment of the optical pumping process is given in Ref. 5. An important result is that the time dependence of the different magnetic substate populations is governed by the pumping time constant τ_p. To avoid line broadening by fast pumping, the pump light intensity has to be sufficiently low. Experimental results (Fig. 3) are of the order of 1 sec.

The hyperfine transitions are induced by microwaves which are simply blown into the apparatus. This has the advantage of simplicity, but does not provide any knowledge of the field distribution inside the trap. We used a klystron, thermally isolated in an oil bath, as a source, and phase locked the output frequency to the 51st harmonic of a (nominal) 800-MHz synthesizer. It in turn was controlled by a rubidium frequency standard. Long-term frequency stability was assured by comparison of our laboratory standard to the broadcast Cs-standard frequency of the Physikalisch-Technische Bunderanstalt.

The ion-fluorescence quanta were detected by a standard photon counting technique, using a solar-blind photomultiplier, a uv interference filter (full width at half maximum 200 Å, transmission 7%) and uv grade quartz lenses. The total solid angle was 5%. The output of the photomultiplier was passed through a photon counting amplifier discriminator chain and fed to the input of a multichannel analyzer, whose channel number varied with the frequency of the 800 MHz synthesizer.

III. MEASUREMENT

Sweeping the klystron output frequency across the resonance, we observed the expected increase in fluorescence intensity at the $\Delta M = 0$ resonance. The intensity of the microwaves was reduced to achieve minimum linewidth, which finally was limited by relaxation processes (Fig. 4). At 10^{-10} Torr we observed lines as narrow as 4 Hz, however we did not succeed in maintaining this pressure over the whole period of the experiment. So most of the measurements were performed at pressures between 10^{-8} and 10^{-9} Torr. Typical linewidths were 15 Hz (Fig. 5). To increase the signal-to-noise ratio we averaged up to 50 min across the resonance.

A least-squares Lorentzian fit, allowing a linear drift in background counting rate, did not show any significant deviation from the expected theoretical line shape. The normalized χ^2 values were about 1.2 at all measurements. The statistical error in the determination of the line center was 0.1 Hz.

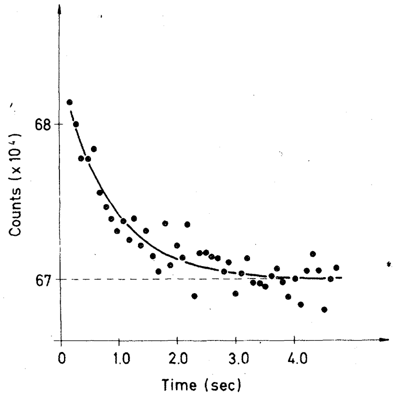

FIG. 3. Time constant for the optical-pumping process.

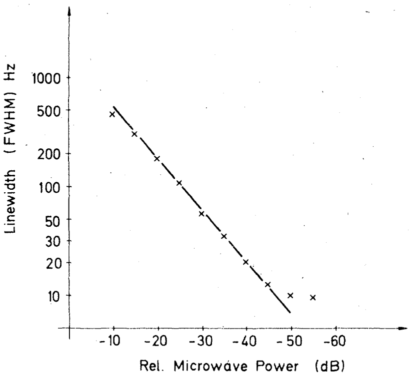

FIG. 4. Half width of $\Delta M = 0$ resonance as a function of microwave power.

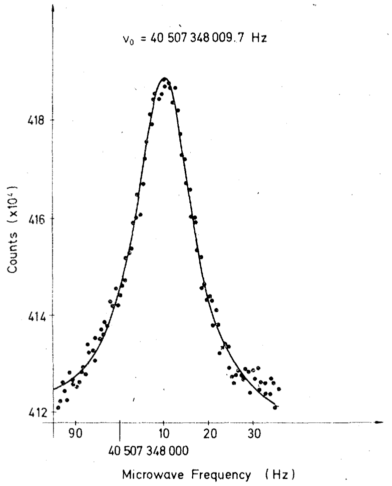

FIG. 5. $\Delta F=1$, $\Delta M=0$ microwave transition. Solid line: Lorentzian least-squares fit. Averaging time: 50 min, background pressure. $\sim 2\times 10^{-9}$ Torr. Linewidth: 16 Hz. Statistical error of center frequency: 0.09 Hz.

All measurements were made at the operating point $a_z=-0.03$, $q_z=0.5$ which is close to the point of highest ion density.[11]

A. Magnetic field dependence

The $\Delta M=\pm 1$ transitions served to determine the magnetic field inside the trap. Because of insufficient shielding, stray and 50-Hz fields in the lab caused a residual width of about 200 kHz. The line center was determined to 1% of the linewidth, which was sufficient to keep the error of the Zeeman shift below 0.5 Hz.

The $\Delta M=0$ transition was fitted to the Breit-Rabi formula. The values $g_J=2.004\,12$ (from isoelectronic gold) and $g_I=5\times 10^{-4}$ give

$$\delta\nu = 97.2H^2\,,$$

if $\delta\nu$ is given in hertz and H in oersted. The parabola fit is shown in Fig. 6 and gives a total error in the extrapolation to zero field of 0.2 Hz.

B. Doppler shift

Since the microwave field inside the trap certainly has a running component, a possibility for a shift in the central line of the Doppler-modulated spectrum exists if the ions have a residual linear motion during a time of the order of the inverse linewidth. Laser measurements on trapped Ba^+ ions indicate that the linear dimension of the ion cloud usually does not exceed 6 mm.[12] As a worst case for a shift, one may consider a movement of 3 mm in a time of 10 sec, which leads to a first-order Doppler shift of 0.4 Hz. Averaged over the total ion cloud this shift will be much smaller and we take it for negligible. Because of the ion energy of several eV the second-order Doppler shift is of importance at our level of accuracy.

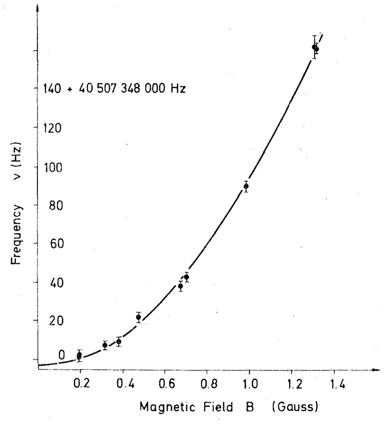

FIG. 6. Magnetic field dependence of the $\Delta M=0$ transition.

Iffländer and Werth[11] have shown, using laser spectroscopy on trapped Ba^+ ions, that the average ion energy varies linearly with the potential depth. This provides a means to vary the ion energy. We were able to vary our operating point to a certain extent, limited by the loss of ions near the boundary of the stable region. The span of potential depth was from 25 to 78 eV. The observed shift of the transition frequency (Fig. 7) can be fitted by a straight line for extrapolation to zero energy. The experimental value $\delta\nu/\nu=(5.3\pm 0.6)\times 10^{-12}$ per ion energy change of 1 eV is in accordance with the calculated value of 5×10^{-13} from the second-order Doppler effect, assuming that the average ion energy is $\frac{1}{10}$ of the potential depth as in Ref. 11. The total correction from the usual operating point to zero energy amounts to 1.1 ± 0.1 Hz.

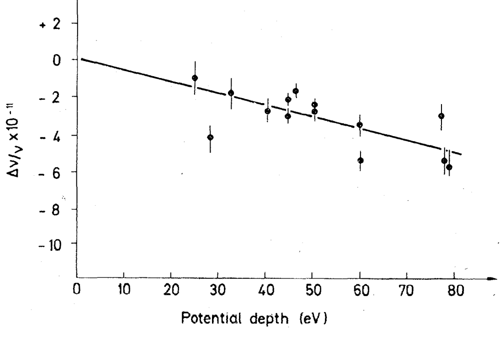

FIG. 7. Shift of $\Delta M=0$ resonance at different trap potential depth (second-order Doppler effect).

C. Other possible sources of line shifts

Stark-effect measurements on the hyperfine structure of neutral atoms indicate that a shift arising from the trapping field should be at most of the order of 10^{-3} Hz and we do not consider it here.

Light shifts are possible due to incomplete overlap of the spectral profiles of the emission by the ^{202}Hg$^+$ and absorption by the ^{199}Hg$^+$. Optical measurements[5] indicate that the overlap is rather good. No shift with varying light intensity has been observed. However systematic investigation is planned.

IV. RESULT

The final result of our measurements gives for the ground-state hyperfine splitting of ^{199}Hg$^+$, including corrections to zero magnetic field and energy, a value of

$$\delta\nu_{\mathrm{hfs}} = 40\,507\,347\,997.8 \pm 1.0\ \mathrm{Hz}\,.$$

While the errors are due to statistics, Zeeman and Doppler corrections are only 0.1 and 0.2 Hz, respectively the main part is a conservative estimate of the 1-h stability of our Rb frequency standard (Efratom FRK-H) in the thermally nonstabilized environment of our lab.

Improved equipment will obviously increase the accuracy of our measurements. In addition we observed very narrow lines, when the relaxation rate was reduced at lower background pressures. An example is given in Fig. 8, which shows a $\Delta M = 0$ transition at pressures of some 10^{-10} Torr. In spite of the poor statistics due to a short measuring time, there is no deviation from the Lorentzian line shape within the statistical error, and from the 4-Hz linewidth, we could determine the line center to 0.08 Hz.

V. CONCLUSIONS

The relative accuracy of 2.5×10^{-11} in the hyperfine transition demonstrates the high potential of the ion-storage technique for high-precision microwave spectroscopy. Since further improvements of our measurements are possible, the ^{199}Hg$^+$ ion may now be regarded as a serious candidate for a new frequency standard—provided that the intrinsic problem of the high second-order Doppler effect can be solved by more accurate determination of the ion energy distribution or by ion cooling. While radiative cooling to room temperature has been demonstrated on light ions,[13] proposals for laser cooling[14] are promising for the future.

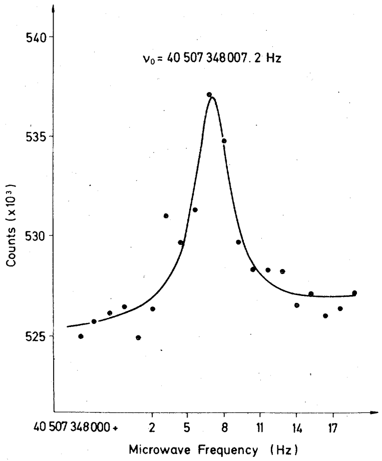

FIG. 8. $\Delta F = 1$, $\Delta M = 0$ microwave transition. Solid line: Lorentzian least-squares fit. Averaging time: 15 min, background pressure: $\sim 10^{-10}$ torr. Linewidth: 4 Hz. Statistical error of center frequency: 0.08 Hz.

The short-term frequency stability (Allan variance—$\delta f/f$) of this system operated in feedback mode as a frequency standard can be estimated. The optimum discriminator (feedback) signal is obtained when the frequency of the microwave excitation is chopped between the opposite steepest points on the Lorentzian curve—the points of maximum signal change. For this case the Allan variance is given by

$$\delta f/f = \frac{3}{4}\,\frac{1}{Q}\,\frac{1}{\Delta_{\mathrm{SNR}}}\,,$$

where Q is the "Q" of the resonance and Δ_{SNR} is the signal-to-noise ratio. This gives a result

$$\delta f/f = 1.5 \times 10^{-12}/\sqrt{t}\ .$$

This is in the same region as the short-term stability of commercially available cesium standards.

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