Measurement of the Quadrupole Moment in the Mercury Ion Optical Clock

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***Abstract*— We report a measurement of the ^{199}Hg^{+} $5d^9 6s^2\ ^2D_{5/2}$ electric quadrupole moment and discuss its implications for the accuracy of an optical clock based upon the $^2S_{1/2}(F=0) \longleftrightarrow {}^2D_{5/2}(F=2)$ transition at 282 nm.**

I. Introduction

A limiting systematic uncertainty for the accuracy of optical frequency standards based on several species of atomic ion has thus far been the quadrupole shift [1], a shift due to the interaction of an atomic quadrupole moment with static electric field gradients. Until recently, this shift has dominated the error budgets for the optical standards based on Hg$^+$ [2], Sr$^+$ [3], and Yb$^+$ [4]. The frequency of the mercury-ion optical clock is steered to resonance with the ^{199}Hg$^+$ $5d^{10}6s\ ^2S_{1/2}(F=0) \longleftrightarrow 5d^9 6s^2\ ^2D_{5/2}(F=2, m_F=0)$ electric-quadrupole "clock" transition at $\lambda = 282$ nm. The $^2S_{1/2}$ ground state is free of a quadrupole moment so the quadrupole shift in the clock transition frequency is entirely due to a shift in the $^2D_{5/2}$ excited state in the presence of external electric field gradients. Here we report a measurement of the electric quadrupole moment of the $^2D_{5/2}$ state [8], obtained by applying a calibrated electric field gradient and observing the resulting shift to the frequency of the clock transition.

II. The mercury ion optical clock

This mercury ion optical clock system has been described in detail previously [5]–[7]. Briefly, a single ^{199}Hg$^+$ ion is stored in a cryogenic spherical rf (Paul) trap and is laser cooled using the $^2S_{1/2} \longleftrightarrow {}^2P_{1/2}$ transition at 194 nm. The 282 nm clock transition is probed with the frequency-doubled output of a highly stabilized laser at 563 nm. The stability of the laser source derives from a thermally and mechanically isolated high-finesse ($\mathcal{F} \sim 200\,000$) Fabry-Perot cavity. The laser locked to the cavity has fractional frequency instability of $3-4\times10^{-16}$ between 1 s and 10 s, corresponding to a 640 mHz linewidth at 563 nm [5]. After stabilization to the cavity, the light is frequency shifted by two acousto-optic modulators (AO1, AO2) in series that shift the frequency away from that of the cavity resonance and, after frequency doubling, into resonance with the clock transition. For the measurement of the quadrupole moment, the clock transition was probed for a period of 40 ms, which gave a Fourier-transform-limited linewidth of about 20 Hz. Using AO2, the laser frequency is alternately stepped to probe the clock transition on either side of the resonance. A digital servo loop then uses AO1 to steer the average laser frequency to the atomic resonance.

III. Measuring the quadrupole moment

Our spherical rf trap consists of a ring electrode, driven with rf, and two endcap electrodes that are ac grounded. Normally, the endcaps are held near ground potential. By applying a static voltage $V_{\rm ec}$ to the endcaps relative to the ring electrode, we produce a electric quadrupole potential $\Phi = (V_{\rm ec}/d_0^2)(x^2+y^2-2z^2)$ centered at the location of the ion. The magnitude of the gradient along the symmetry axis $\hat{z}$ of the trap was calibrated to be $A_{\rm Q} \equiv V_{\rm ec}/d_0^2 = V_{\rm ec}/(1.08(1)\ {\rm mm}^2)$ by measuring the secular sideband frequencies [9] of the trapped mercury ion. The quadrupole shift $\Delta\nu_{\rm Q}$ in the frequency of the clock transition is given by

$$h\Delta\nu_{\rm Q} = \frac{4}{5}A_{\rm Q}\Theta(D, 5/2)(3\cos^2\beta - 1), \qquad (1)$$

where h is the Planck constant, $\Theta(D,\ 5/2)$ is the $^2D_{5/2}$ quadrupole moment, and β is the angle between the quantization axis (magnetic field orientation) and $\hat{z}$ [1].

To measure the quadrupole shift for a given value of $A_{\rm Q}$ and β, we first locked the laser to the atomic resonance as described in § II. We then drove $V_{\rm ec}$ as a modified square-wave with period 120 s at values $V_{\rm ec} = 0$ V and $V_{\rm ec} = V_{\rm A}$, for applied voltage $V_{\rm A}$. During this procedure, an electronic counter recorded the frequency applied to AO1 to keep the laser in resonance with the clock transition. The frequency record contains not only the quadrupole shift due to the applied field, but also the unmodeled drift of the optical cavity. The value of $\Delta\nu_{\rm Q}$ was extracted from this record as the component with a 120 s period. This procedure was repeated for four values of $A_{\rm Q}$, each with $\beta = 0°$ (magnetic field $\mathbf{B} \parallel \hat{z}$) and $\beta = 90°$ ($\mathbf{B} \perp \hat{z}$).

IV. Results and Conclusions

The results of the eight measurements of $\Delta\nu_{\rm Q}$ are shown in Fig. 1. As we would expect from Eqn. 1, the data with $\beta = 0°$ have a quadrupole shift twice the magnitude and

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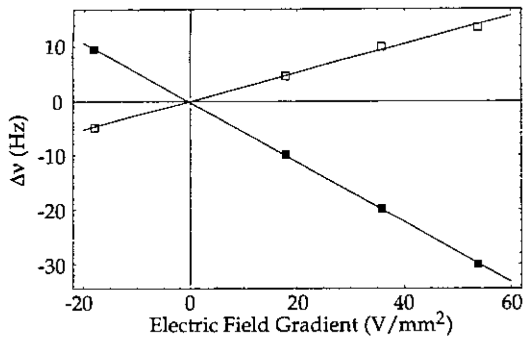

Fig. 1. Quadrupole shift as a function of the electric field gradient applied to the trap. Data are shown with the magnetic field $\mathbf{B} \parallel \hat{z}$ (filled boxes) and with $\mathbf{B} \perp \hat{z}$ (open boxes). The two curve fits shown are linear with slopes -0.550 Hz/(V/mm^2) and $+0.261$ Hz/(V/mm^2) respectively. Error bars representing the combined servo error uncertainty and statistical uncertainty would be smaller than the symbol at each data point.

opposite in sign of those with $\beta = 90°$. Using the weighted mean slope and Eqn. 1 gives the value for the quadrupole moment, $\Theta(D,\ 5/2) = -(5h/8) \cdot 0.552(19)$ Hz mm^2/V $= -2.29(8) \times 10^{-40}$ C $\cdot$ m^2 $= -0.510(18)$ ea_0^2, where e is the elementary charge and a_0 is the Bohr radius. The uncertainty of this measurement is limited by the $\pm$ 5° uncertainty in the alignment of the magnetic fields (uncertainty in β).

Knowing the quadrupole moment alone does not constrain the quadrupole shift due to stray electric field gradients, since the field configuration is generally unknown. One way to remove the quadrupole shift from the optical clock frequency is to average the measurement of the clock frequency over three orthogonal magnetic fields [1]. A second approach to removing the quadrupole shift is to average over magnetic sublevels rather than field orientation [10]. It is not yet clear which approach is more advantageous for Hg^+ since the $^2D_{5/2}(F = 2)$ magnetic sublevels have different second-order Zeeman shifts. We have measured the quadrupole shift in our trap several times, and each time obtained a value below ± 1 Hz. By averaging over orthogonal fields with our present accuracy of field alignment, we expect that the quadrupole shift can be controlled to the level of ± 10 mHz, a fractional frequency error of less than 10^{-17}.

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