LIGHT INDUCED KINETIC EFFECTS ON ATOMS, IONS AND MOLECULES
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SINGLE ION OPTICAL SPECTROSCOPY*

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ABSTRACT

A single $^{199}Hg^+$ ion is confined in a miniature rf trap and is laser-cooled to nearly 0.001 K. We have studied the spectrum of the narrow $5d^{10}6s\ ^2S_{1/2}$ - $5d^96s^2\ ^2D_{5/2}$ electric-quadrupole-allowed transition near 282 nm and obtain a linewidth below 80 Hz (FWHM). The measured lifetime of the metastable $^2D_{5/2}$ state gives a natural linewidth limit for this transition of 1.8 Hz. The narrow linewidth, the ability to detect transitions with unit probability, and the small perturbations of a single laser-cooled ion make it an attractive candidate for an optical frequency standard. Our present resolution is limited by the spectral purity of the frequency-doubled dye laser at 563 nm. Optical heterodyne measurements between two laser beams locked to independent, high-finesse cavities give a beat note of less than 40 Hz (FWHM at 563 nm) that is dominated by noise in the frequency range from 0 to 10 Hz. This is caused by the insufficient isolation of the cavities from mechanical vibrations in this frequency range. Better isolation methods intended to improve the laser linewidth to about 1 Hz or less are being investigated. A linear Paul trap, in which it would be possible to trap and cool many ions unperturbed by rf micromotion, is being tested.

EXPERIMENT

The ion trapping and laser-cooling have been described elsewhere[1,2]. A ^{199}Hg atom is ionized and trapped in the harmonic pseudopotential well created by an rf potential applied between the electrodes of a miniature Paul trap. The separation between the endcap electrodes ($2z_0$) is about 650 μm. The frequency of the rf potential is about 21 MHz. Its amplitude can be varied up to 1.2 kV resulting in a secular frequency of up to 4 MHz. The ion is laser cooled to a few millikelvins by a few microwatts of radiation from two 194 nm sources. One source induces transitions from the $5d^{10}6s\ ^2S_{1/2}(F=1)$ to the $5d^{10}6p\ ^2P_{1/2}(F=0)$ level. This is essentially a two level system suitable for laser-cooling, except for weak off-resonance pumping into the $^2S_{\frac{1}{2}}(F=0)$ state. The second 194 nm source, tuned to the $^2S_{\frac{1}{2}}(F=0)$ to $^2P_{\frac{1}{2}}(F=1)$ transition, returns the ion to the ground state F=1 hyperfine level. The frequency separation between the two radiation sources is equal to the sum of the ground and excited state hyperfine splittings (about 47 GHz). The complication of two-laser cooling is dictated by the hyperfine structure of $^{199}Hg^+$. An isotope with hyperfine structure is required in order to have a first-order field-independent clock transition near $B \simeq 0$. The two 194 nm beams are made collinear and irradiate the ion at an angle of 55° with respect to the symmetry (z) axis of the trap. In this way, all motional degrees of freedom are cooled to near the Doppler cooling limit of 1.7 mK. 194 nm fluorescence from the ion, collected in a solid angle of about $5 \times 10^{-3} \times 4\pi$ sr, is detected with an efficiency of 10% to give a peak count rate on resonance of about 25 000 s^{-1}.

The 282 nm radiation that drives the narrow $5d^{10}6s\ ^2S_{\frac{1}{2}}$ - $5d^9 6s^2\ ^2D_{5/2}$ transition is obtained by frequency-doubling the radiation from a cw dye laser that is stabilized to a Fabry-Perot cavity with finesse[3] $F \simeq 60\,000$. Prior to being frequency doubled, the 563 nm radiation is passed through an acousto-optic modulator so that its frequency can be tuned through the S-D resonance. Since the Zerodur[4] reference cavity contracts at a nearly constant rate of 3.3 Hz/s[5], the acousto-optic modulator is driven by a frequency obtained by summing the rf output of two synthesizers. The frequency of one synthesizer sweeps opposite to the cavity drift; the frequency of the second synthesizer is stepped back and forth through the S-D resonance. The 282 nm laser and the two-frequency 194 nm source are turned on and off sequentially using shutters and the acousto-optic modulator. This prevents any broadening of the narrow S-D transition due to the 194 nm radiation. Optical-optical double resonance[1,6] (electron shelving)[7] is used to detect each transition made to the metastable D state as a function of the frequency of the 282 nm laser. At the beginning of each cycle, both 194 nm lasers irradiate the ion. The fluorescence counts in a 10 ms period must exceed a minimum threshold (typically 20 counts) before the interrogation sequence can continue. The 194 nm beams irradiate the ion for sequential 10 ms periods until the threshold is met. Then the 194 nm radiation tuned to the $^2S_{\frac{1}{2}}(F=0)$ - $^2P_{\frac{1}{2}}(F=1)$ transition is chopped off for 5 ms. During this time the 194 nm radiation tuned to the $^2S_{\frac{1}{2}}(F=1)$ - $^2P_{\frac{1}{2}}(F=0)$ transition optically pumps the ion into the $^2S_{½}$(F=0) ground state. Then this 194 nm source is turned off. One millisecond later, the 282 nm radiation, tuned to a frequency resonant or nearly resonant with the $^2S_{½}$(F=0, m_F=0) - $^2D_{5/2}$(F=2, m_F=0) transition irradiates the ion for an interrogation period that was varied up to as long as 15 ms. At the end of this period, the 282 nm radiation was turned off and both 194 nm sources were turned on. Another 10 ms detection period was used to determine whether a transition to the D state had been made (fluorescence counts > threshold, no; fluorescence counts < threshold, yes). The result was recorded as a 1 or 0 (no or yes) and averaged with the previous results at this frequency. Then the frequency of the 282 nm radiation was stepped and the measurement cycle repeated.

Since the frequency drift of the 282 nm laser depended not only on the reference cavity contraction rate, but also on small pressure and temperature changes, on laser power variations, etc.[8,9], we locked the frequency of the laser to the narrow S-D transition to remove any long term frequency drifts. To do this, we modified the measurement cycle to include a lock cycle. We began each measurement cycle by stepping the frequency of the 282 nm radiation to near the half maximum on each side of the resonance N times (N varied from 8 to 32). At each step, we probed for 5 ms, and then looked for any transition with the electron-shelving technique. We averaged the N results from each side of the resonance line, took the difference and corrected the frequency of the synthesizer drifting against the cavity. The gain of this lock needed to be properly adjusted to avoid adding frequency noise to the laser. In this way, variations in the frequency of the 282 nm laser for time periods exceeding a few seconds were reduced.

In Fig. 1, we show the spectrum obtained by scanning in this drift-free way through the Doppler-free resonance of the $^2S_{½}$(F=0, m_F=0) - $^2D_{5/2}$(F=2, m_F=0) transition. The lineshape shown is the result of 138 consecutive scans. The probe period was 15 ms and the step size was 15 Hz at 563 nm (30 Hz at 282 nm). The resonance shows a clearly resolved triplet with the linewidth of each component less than 40 Hz (< 80 Hz at 282 nm). We first thought that the triplet structure might be due to 60 Hz modulation of the frequency of the 563 nm laser either due to grounding problems, line pickup or inadequate servo gain. However, when the radiation from two independently stabilized laser beams were heterodyned together (see next section), the 60 Hz modulation index was far too small to account for the sideband structure observed on the S-D resonance. In addition, the frequency separation of the peaks is nearer to 50 Hz, not 60 Hz. We now think that, most likely, the triplet structure is caused by Rabi power broadening. The 282 nm radiation is focussed to a spot size of about 25 μm; therefore, on resonance, fewer than 10^6 photons/s (< 1 picowatt) will saturate the transition. Below the data is a theoretical lineshape calculated for an ion at rest, for no broadening due to collisions or laser bandwidth, for a pulse length of 15 ms and for sufficient power at resonance to give a 3.5 π-pulse. Qualitatively the figures compare well. The fluctuations from measurement cycle to measurement cycle in the quantum-occupation-number of the ion in the harmonic well of the trap cause variations in the transition probability of the ion. This, and the finite laser linewidth, likely cause the general broadening and weakening of the signal. We will investigate the lineshape and the effects of power broadening in more detail in future experiments.

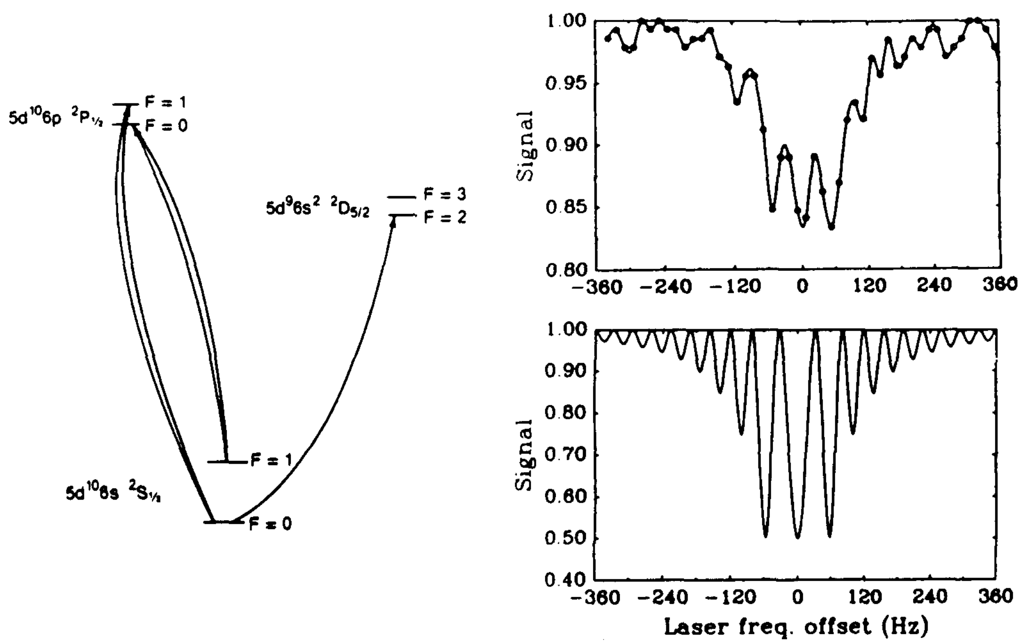

Figure 1. On the left is a simplified energy-level diagram for $^{199}Hg^+$ at zero field. Shown in the upper figure on the right is the power-broadened lineshape obtained by scanning through the Doppler-free resonance of the $^2S_{½}$(F=0, m_F=0) - $^2D_{5/2}$(F=2, m_F=0) transition in a single laser cooled $^{199}Hg^+$ ion. A 563 nm laser that is stabilized to a high finesse reference cavity, which in turn is long-term stabilized to the ion, is frequency doubled and stepped through the resonance for 138 consecutive sweeps. The

step size is 15 Hz at 563 nm (30 Hz at 282 nm). The lower-right figure shows the lineshape calculated for conditions similar to the experimental conditions for the upper figure, except that the ion is assumed to have zero temperature and the laser is assumed to have zero linewidth.

REFERENCE CAVITY STABILITY

To simplify the study of the performance of a laser locked to a reference cavity against external perturbations, we constructed a second Zerodur reference cavity. This study was carried out by heterodyning the light from laser beams locked to the two independent cavities and analyzing the fluctuations in the power spectrum of the beat frequency. Each laser beam was locked to its reference cavity using the reflection sideband technique discussed in detail by Drever et al.[8)] In attempting to narrow the 563 nm laser to the order of 1 Hz or less, great care must be taken in the electrical and optical set-up. Many of the techniques and subtle difficulties are treated in earlier papers by Hough et al.[9)] and by Salomon et al.[10)] The optical and electrical problems limit the laser linewidth in our work at or below the 1 Hz level; the dominant contribution to the laser linewidth is the length stability of the reference cavities.[10-12)] Some aspects of our stabilized laser systems and efforts made to isolate the cavities from external perturbations will be summarized here.[13)]

Each cavity is constructed with a Zerodur[4)] cylindrical spacer that has a diameter of 10.2 cm with a 1 cm holed bored down the axis. The mirrors are highly polished Zerodur[4)] substrates that are coated to give high finesse and good efficiency and then optically contacted to the polished ends of the spacer. The cavities are suspended by two thin molybdenum wires inside an aluminum vacuum vessel that has an inner diameter of 26.7 cm. The wires were slung around the spacers at the nodal positions for the lowest order bending mode (about L/5 from each end where L is the cavity length) and attached to the walls of the vacuum vessel by small clamps. The two wires open into a symmetric "V" at each end; with opening angles of about 10° at one end and about 15° at the other end. This allowed free movement of the cavities along the direction of their axis. The damping of this motion was primarily into the aluminum housing and from there into the table and padding used for isolation. The aluminum vacuum vessels were thermally insulated and temperature controlled to the order of a few mK. The temperature coefficient of our Zerodur[4)] spacers was approximately $6 \times 10^{-9}/^{\circ}C$ near room temperature. The thermal time constant from the walls of the evacuated aluminum housing to the spacers was on the order of 1 day. Low pressure was maintained in each vacuum vessel by an ion pump that was directly attached to the vessel. The pressure in one vessel was about 8×10^{-9} Torr (1.1×10^{-6} Pa) but only 2×10^{-7} Torr (2.7×10^{-5} Pa) in the second system. This is marginally adequate, since 10% pressure fluctuations in the second system would cause frequency fluctuations of a few Hz to the laser stabilized to its cavity from changes in the index of refraction.

The length of one Zerodur[4)] rod results in a free spectral range of 562 MHz while the other gives 622 MHz. The finesse of the longer cavity is 60 000 and the transmission efficiency on resonance exceeds 23%. The shorter cavity has a finesse of 90 000 and an efficiency of about 27%. (Two Fabry-Perot cavities have been constructed from ULE[4)] spacers that are about 10 cm long by about 7.6 cm in diameter. Both cavities have a measured finesse that exceeds 130 000. The frequencies of the mechanical resonances of these shorter, stiffer bars will be about a factor of 3 higher than the Zerodur bars. In general, if the frequencies of the mechanical vibrations are high enough, the clock transition can be probed by the "unperturbed" carrier of the laser spectrum.) The aluminum vacuum vessels rested on Viton[4)] rubber strips attached to V-blocks made of aluminum. The V-blocks were secured to a rigid plexiglass[4)] plate. Each reference cavity system was mounted on separate optical tables that (initially) consisted of surface plates damped into sand. The sand box sat on soft rubber pads and cinder block legs in one case and on low-pressure inner tubes and cinder block legs in the second. Noise vibrations on the floor and on the table tops were monitored with sensitive seismometers. Some isolation from mechanical vibrations at frequencies above 5 Hz was achieved for both table tops.

For heterodyning purposes a small fraction of the frequency stabilized light from each cavity was combined on a beam splitter and detected with a fast diode. The heterodyned signal was amplified and analyzed with two spectrum analyzers used in tandem. This allowed us not only to look directly at the beat note but also to Fourier analyze the noise terms that contribute to its linewidth. The first analyzer could be used to observe the beat signal or as a frequency discriminator. As a discriminator, the scan was stopped and the analyzer was used as a tunable rf receiver with a variable bandwidth. The center frequency was shifted so that the heterodyne signal lay at the 3 dB point of the response curve. The bandwidth was adjusted so that the frequency excursions of the beat signal were much less than the bandwidth. This produced a one-to-one map of frequency excursions to voltage excursions whose Fourier power spectrum could be analyzed in the second, low-frequency (0-100 kHz) spectrum analyzer. This allowed us to study the nature and origin of the vibrational noise that contributes to the linewidth of the stabilized laser.

The width of the heterodyne signal between the two stabilized lasers was less than 40 Hz. The noise power spectrum revealed that low frequency fluctuations in the range from near 0 to 30 Hz dominated this linewidth. The vibrational noise spectrum measured by the seismometers on the table tops matched the largest noise components of the beat note. The frequencies of the pendulum motion of the suspended cavities were about 1.4 Hz and 1.48 Hz which gave FM at these frequencies. There were also bright features in both the laser and the seismometer power spectra that came from floor motion at 14.6 Hz, 18.9 Hz and at 29.2 Hz. These all had enough power to contribute to the beat note linewidth. When the pendulum motions of the bars were quiet, the integral of the nearly featureless noise power spectrum from ~ 0 Hz to 10 Hz most strongly contaminated the spectral purity of the lasers. Some, if not all of this noise was mechanical in origin, but it was not clear how it coupled to the suspended cavity. To help elucidate the connection, we drove the table tops in either the horizontal or vertical direction with a small loudspeaker connected to an audio signal generator. The motion of the speaker diaphragm was coupled to the table by a rod glued to the diaphragm and gently loaded against the table. The table could be driven at frequencies from a few Hz to about 500 Hz with enough power to be 40 dB above any background noise term. When the loudspeaker drove the table in the horizontal plane in a direction parallel to the axis of the cavity, the isolation of the suspended cavity was sufficiently good that the beat signal showed no evidence of the perturbation even at the high drive levels. However when the drive was applied vertically at a level barely perceptible above the vertical background noise, the heterodyne signal revealed added noise power at the drive frequency. The stiff support in the vertical direction strongly coupled vertical motion into effective cavity length changes. The sensitivity of the Fabry-Perot cavity to vertical motion was orders of magnitude higher than for horizontal motion parallel to the cavity axis.

For practical reasons, we were unable to drive the table in the horizontal plane in a direction perpendicular to the cavity axis.

In order to improve the vertical isolation, one table was suspended just above the floor with latex-rubber tubing attached to the ceiling. The resonance frequencies for both the vertical motion and the horizontal pendulum motion of the suspended table were near 0.33 Hz. These were damped to the floor with two small dabs of grease. The linewidth of the heterodyne signal between the laser radiation stabilized to the cavity supported on this table and the laser radiation stabilized to the cavity supported on the best sandbox table dropped from 40 Hz to less than 20 Hz. The Fourier noise power spectrum from 0-10 Hz is shown in Fig. 2. The beatnote linewidth obtained by integrating the power spectrum is about 15 Hz. We suspect that the laser stabilized to the cavity on the sandbox table is the dominant contributor to the width of the heterodyne signal since the vibrational noise measured on this table is greater. Current efforts are devoted to measuring the narrow S-D transition in $^{199}Hg^+$ using the laser stabilized to the cavity on the suspended table. A linear rf Paul trap [14-17] has been constructed and is also being tested. With this trap it is possible to store and laser-cool many ions which gives a better signal-to-noise ratio (thereby better stability), but it is still possible to have a small second-order Doppler shift.

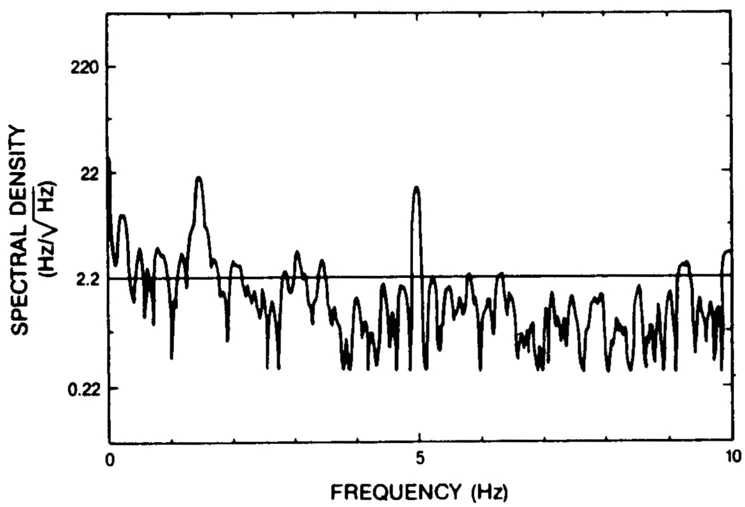

Figure 2. Fourier noise-power spectrum of laser heterodyne signal.

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