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Optical Heterodyne Measurement of Xenon Isotope Shifts

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Isotope-shift measurements with a precision of two parts in 10^9 of the optical transition frequency were made for the 2.02-μ line of the three even isotopes of xenon: Xe^{132}, Xe^{134}, and Xe^{136}. This shift is extremely small due to the large xenon mass, the proximity of the xenon isotopes to the neutron magic number $N=82$, and the weak interaction of the nucleus with the $5d$ and $6p$ electron states. The measurements were 500 ± 300 kHz ($0.17 \pm 0.01 \times 10^{-3}$ cm^{-1}) for $Xe^{132}-Xe^{134}$ and 1500 ± 300 kHz ($0.050 \pm 0.01 \times 10^{-3}$ cm^{-1}) for $Xe^{134}-Xe^{136}$, where the lighter isotope had the larger transition frequency in each case. These two shifts are sufficiently different to suggest that nuclear field effects contribute to the results.

I. INTRODUCTION

Since optical isotope shifts are very small, better measuring techniques are constantly sought. Recently, lasers have been employed by several groups for such measurements.[1-3] Vetter *et al.*[4-7] have made a systematic study of argon and xenon isotope shifts using a variety of methods involving lasers. The xenon isotopic series falls just before the neutron number 82, and Xe^{136} has this magic number of neutrons. King *et al.*[8] have shown that the total nuclear expansion due to the addition of a neutron decreases rapidly just before $N=82$, and it ought to be extremely small for xenon, so that xenon should exhibit a very small nuclear field effect. Measurements confirm this prediction.[7,9,10]

By far the largest field-effect shifts are due to interactions between the nuclear charge and s or $p_{1/2}$ electron states.[11] For xenon transitions in the infrared, which do not originate or terminate in these levels, the isotope shifts ought to be extremely minute. The transition

$$(^2P^0_{3/2})5d[3/2]^0_1 - (P^0_{3/2})6p[3/2]_1$$

at 2.0268 μ, involving no s levels and probably only a small admixture of $p_{1/2}$ state, was found by Vetter to have the smallest total shift of all those measured:

$$600 \pm 1500\,\text{kHz}\ (0.02 \pm 0.05 \times 10^{-3}\ \text{cm}^{-1})$$

between Xe^{132} and Xe^{134}. In the present investigation, an optical heterodyne technique was used in the measurement of this shift resulting in a fivefold increase in precision. This technique required only a few hundred micrograms of separated isotopes.

II. EXPERIMENT

The two lasers used were constructed identically except for the different isotope of xenon in each. Laser plasma tubes with Brewster's-angle windows were made from 3-mm-bore quartz tubing. Quartz side tubes, attached at right angles to the plasma tubes, were immersed in liquid nitrogen. This froze the xenon and supplied a stable reference temperature for the xenon vapor. The tubes also served as xenon reservoirs. The xenon vapor pressure could be varied over two orders of magnitude by heating these side tubes. Usually the pressure was maintained near 10 mTorr, and the laser power output was commonly about 10 μW. However, both the pressure and the power were varied extensively.

Lasers were prepared with xenon from two sources. Xe^{136} of 99% purity was obtained from the Monsanto Research Corporation in fairly large quantities. About 1% of the gas was Xe^{134} and perhaps 0.1% was nitrogen. Xe^{132} and Xe^{134} were obtained from the Argonne National Laboratory in about 1-mg quantities using electromagnetic separation methods. 2 or 3 mg of Xe^{136} were obtained in the same way. Before the laser plasma tubes were filled with xenon, the filling system was pumped down to 10^{-9} Torr, and a rf helium discharge was used to clean contaminants from the laser tube walls. Then the xenon gas was introduced and frozen on these walls by cooling the plasma tubes with liquid nitrogen. Unfrozen contaminants were pumped out while the xenon remained frozen. Finally the lasers were sealed.

Since the laser walls collected large quantities of xenon during operation, low-pressure discharges could not be maintained without large xenon reservoirs. Heating the laser tube drove contaminants as well as xenon from the walls, and getters removed xenon as well as these contaminants, so that the laser lifetime was severely limited by the amounts of separated isotopes available. However, rf laser excitation eliminated the need for internal metal electrodes which would absorb xenon, and the laser tube volume was kept as small as possible. With these precautions, a laser filled with 100 μg of xenon would operate almost 1 h while one filled with 500 μg would run more than 15 h.

A large source impedence for the laser plasma and a 200-MHz discharge excitation were used to stabilize the discharge, but only partial success was achieved. Xenon discharges are not known for their quiet operation. Plasma instabilities of all kinds were seen; laser frequency drift as well as power drift was observed. Nevertheless, the average beat frequencies measured as well as the frequency distributions appeared independent of these instabilities, so that it is believed their direct effect on the experimental precision was small. It might be noted that helium should quiet a xenon discharge. The noisiness of the discharges in this experiment is an indication that the lasers had been cleaned of helium which might have produced unknown pressure shifts.

Laser mirror separation was 24 cm for single-frequency axial-mode operation within the 160-MHz Doppler linewidth. Apertures in the cavity eliminated nonaxial modes, and the lasers were usually run near the lowest power levels sufficient to produce measurable signal levels. An electronic feedback system, which used the first derivative of the Doppler line shape, stabilized each laser on line center. Initially it had been hoped that the 2.02-μ line would show a narrow Lamb dip so that the laser stability could be enhanced. However, no trace of a dip could be found under any conditions in which the lasers would operate. This presumably means that the natural linewidth for this line is large. A schematic diagram of the feedback system is shown in Fig. 1. A piezoelectric crystal modulated the laser cavity length at 4800 Hz to produce a laser signal modulated about the line center. The detected signal was amplified, and the second harmonic, which contained information only about the laser power, was removed using an active twin-tee notch filter. Phase-sensitive detection and low-pass filtering of the first harmonic produced a dc signal which was amplified and used to adjust the laser signal to line center. The low-pass filter attenuated electronic signals at a 10-dB/octave rate in order to achieve maximum feedback bandwidth without causing oscillations while introducing a minimum of noise of electronic origin to the laser frequency.

The two lasers were mounted side by side, as shown in Fig. 2, with a beam splitter used to bring the two beams to coincidence. Since undesirable laser output at 3.5 μ could not easily be eliminated, dielectric filters were used to remove this signal component before the laser beams reached the photodiodes in the servo-system. The two lasers faced in opposite directions, and the photomixer was placed at the point where the two beam diameters coincided. Wavefront-correcting mirrors were used to reduce phase differences between the two beams, and these phase differences were small enough to allow almost the whole wavefront to pro-

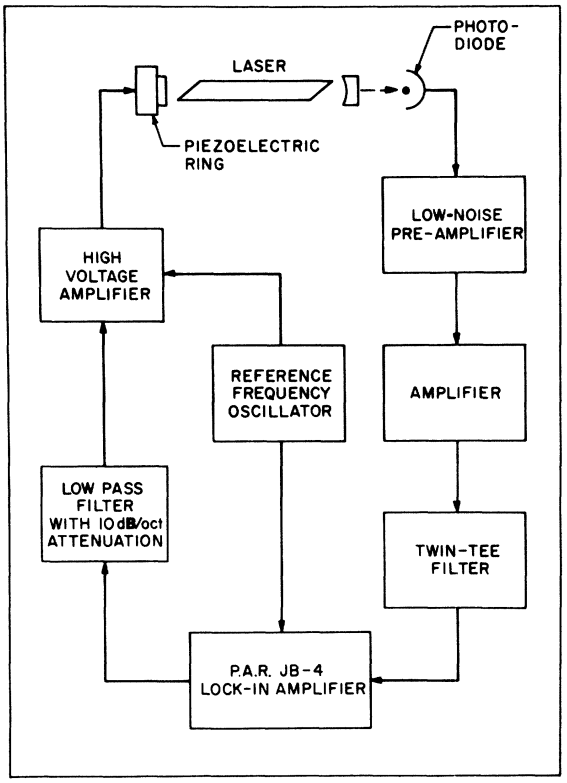

FIG. 1. Laser servo-system electronics.

duce useful beat frequency signal. In order to eliminate specular reflections of the beams back into the cavities, all reflecting surfaces were skewed until laser-frequency fluctuations disappeared. The back surfaces of the cavity mirrors were antireflection coated.

Photomixing was accomplished on the surface of a Philco-Ford L4530 indium-arsenide photodiode, and digital-frequency measuring techniques were used. Since the average beat frequency was usually smaller than the frequency fluctuations, and since the electronics could not distinguish between positive and negative frequencies, an external signal was introduced into the feedback loop to provide a laser frequency offset from the line center. The offset was chosen so that beat frequency excursions never reached zero frequency in order that linear measurements could be made. With one laser offset in this fashion, the measured frequencies no longer had any direct relation to the atomic line. If subsequently, both lasers were offset to increase the average beat frequency further, and then only the second laser was offset, the true frequency separation of the two lasers stabilized on their line centers could be retrieved. The measurements needed are shown in Fig. 3. The isotope separation has been much exaggerated. The true frequency separation of the lines is given by

$$\Delta\nu = \nu_1 + \nu_2 - \nu_{12} , \quad (1)$$

where the quantities are identified in Fig. 3. Automatic cycling of the offsets as well as synchronous control of the counting was used. One offset cycle was completed in 75 sec.

III. BEAT FREQUENCY PROPERTIES

The stability, reproducibility, and average frequency offset were established using nominally identical lasers, each filled with 99% Xe^{136} or with mass-separated Xe^{136}. It was found exceedingly difficult to run the lasers at arbitrary power levels and vapor pressures. Laser power drift and pressure drift sometimes occurred simultaneously with laser frequency drift, but since this was not always the case, these were most likely not responsible for the frequency drifts. On the other hand, they are closely associated with the plasma conditions, whose behavior could not be well regulated. It is most likely that gas density gradients, electron temperature fluctuations, Stark effects, and other such phenomena were responsible for the frequency instabilities observed in the stabilized lasers, which amounted

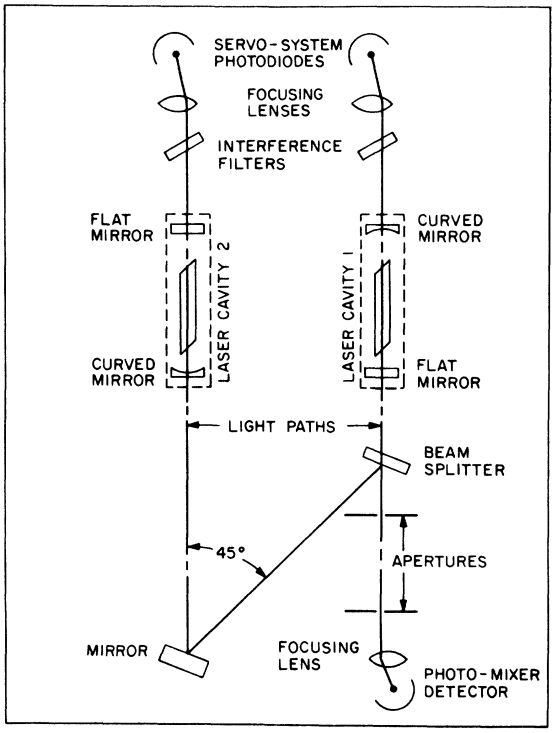

FIG. 2. Laser optics. Where the photomixer was placed, both beams had the same cross section. Surfaces were tilted to avoid specular reflections into the cavities.

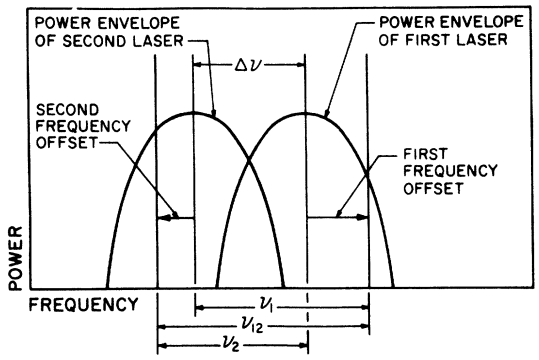

FIG. 3. Offset frequency measurements. The two laser gain curves have a greatly magnified separation. $\Delta\nu = \nu_1 + \nu_2 - \nu_{12}$ is the separation of the peaks.

to 1 to 3 parts in 10^9 of the transition frequency.

The histogram of Fig. 4 is a compilation of all data taken with both lasers containing Xe^{136}. Measurements are specified only within 50 kHz for convenience. The average frequency is -4.3 ± 50 kHz. However, frequencies measured on any given day were not necessarily compatible with this. For example, results of the operations on two different days were $+158 \pm 95$ and -327 ± 42 kHz, where the errors represent one standard deviation of the mean. The data from the histogram have a standard deviation of 485 kHz. It is unlikely that the reproducibility of a single measurement would be much better than this number, which represents about 3 parts in 10^9 of the transition frequency.

IV. ISOTOPE SHIFTS

Two groups of measurements were made for each of the shifts Xe^{132}-Xe^{134}, Xe^{132}-Xe^{136}, and Xe^{134}-Xe^{136}. For one of these groups, the laser plasma tubes were switched so that any undesirable frequency offsets between the lasers might be discovered. Each measurement consisted of ten or more offset cycles. Five of the six sets contained at least eight measurements; the sixth contained only two due to electronics problems. Its average value is labeled $\Delta\nu_1$ (132–134) below, where the notation indicates that the Xe^{132}-Xe^{134} shift was measured, that Xe^{132} had the higher frequency, and that the heavier isotope was in unit one cavity. Although the two measurements in $\Delta\nu_1$(132–134) were close together, there was no reason to assume this was not fortuitous, so that $\Delta\nu_1$(132–134) was assigned the same standard deviation that was found for $\Delta\nu_2$(132–134). The standard deviation of the mean was then taken to be $2^{-1/2}$ times this number.

The results of the measurements, including the standard deviation of the mean for each frequency, are given below. The labels *a*, *b*, *c*, *r*, *s*, and *t*

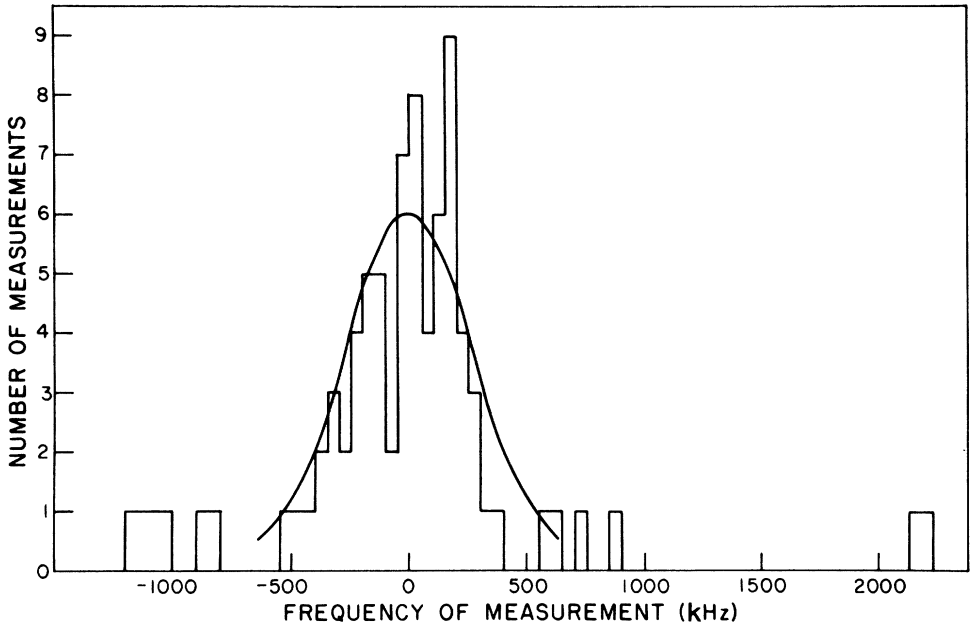

FIG. 4. $XE^{136} - Xe^{136}$ histogram. All reproducibility measurements made with Xe^{136} are included. The Gaussian curve, shown for comparison only, has the same 485-KHz standard deviation and approximately the same normalization as the histogram.

are for later convenience:

$$\begin{aligned} a &= \Delta\nu_1(132-136) = 1623 \pm 88 \text{ kHz}, \\ b &= \Delta\nu_2(132-136) = 2183 \pm 89 \text{ kHz}, \\ c &= \Delta\nu_1(134-136) = 1285 \pm 116 \text{ kHz}, \\ r &= \Delta\nu_2(134-136) = 1982 \pm 127 \text{ kHz}, \\ s &= \Delta\nu_1(132-134) = 737 \pm 542 \text{ kHz}, \\ t &= \Delta\nu_2(132-134) = 1087 \pm 206 \text{ kHz}. \end{aligned} \tag{2}$$

To determine the best value for each isotope shift, it was assumed that the measurements in each set were randomly distributed although they had occasionally occurred in clusters. It was also assumed that systematic errors were the same for each set. Let $\Delta\nu(132-134) = u$ and $\Delta\nu(134-136) = v$ be the true isotope shifts and let w be any constant offset. Then

$$\begin{aligned} &u+v+w=a, \quad v+w=c, \quad u+w=s, \\ &u+v-w=b, \quad v-w=r, \quad u-w=t. \end{aligned} \tag{3}$$

u, v, and w were varied to obtain a minimum value for

$$\delta^2 = \left(\frac{u+v+w-a}{s_a}\right)^2 + \left(\frac{u+v-w-b}{s_b}\right)^2 + \left(\frac{v+w-c}{s_c}\right)^2 + \left(\frac{v-w-r}{s_r}\right)^2 + \left(\frac{u+w-s}{s_s}\right)^2 + \left(\frac{u-w-t}{s_t}\right)^2, \tag{4}$$

where s_a, s_b, etc., are the standard deviations of the means of a, b, c, etc. The minimum value of δ^2 is called χ^2 in the literature.[12]

The results of this calculation are $u = 509$ kHz, $v = 1496$ kHz, and $w = -310$ kHz, leading to $\chi^2 = 8.19$. The probability that a similar set of six measurements related by Eq. (3) could produce a larger value of χ^2 is only 5%,[13] implying that w probably was not constant during the measurements. No standard method exists for determining the best error estimates when χ^2 is so large, but by decreasing χ^2 to 2.37, the probability that similar measurements could produce a larger χ^2 value increases to 50%. This is done by increasing s_a, s_b, etc., by $(8.19/2.37)^{1/2} = 1.86$.

The propagation of errors from the original measurements to u, v, and w was computed, and the results were increased by 1.86 to yield

$$\begin{aligned} u &= 500 \pm 200 \text{ kHz}, \\ v &= 1500 \pm 180 \text{ kHz}, \\ w &= -300 \pm 130 \text{ kHz}, \end{aligned} \tag{5}$$

where the results are rounded off. These errors represent an accuracy of almost 1 part in 10^9 of the laser transition frequency. However, since measurements made with Xe^{136} in both lasers were not this good, this high compatibility of the measurements may have been somewhat fortuitous. For this reason, the more conservative estimate of 2 parts in 10^9 was chosen, and the final isotope-shift estimates were

$$\begin{aligned} &\Delta\nu(132-134) \\ &\quad = 500 \pm 300 \text{ kHz } (0.017 \pm 0.01 \times 10^{-3} \text{ cm}^{-1}), \\ &\Delta\nu(134-136) \\ &\quad = 1500 \pm 300 \text{ kHz } (0.050 \pm 0.01 \times 10^{-3} \text{ cm}^{-1}). \end{aligned} \tag{6}$$

V. DISCUSSION

The normal mass effect for $\Delta\nu(132-134)$ and $\Delta\nu(134-136)$ is 9.06 MHz, where the heavier isotope has the higher frequency. In addition to being much smaller than this, the values given in (6) have the opposite sign. The 600 ± 1500 kHz measurement by Vetter *et al.*[7] for $\Delta\nu(134-132)$ has the same sign as the normal mass effect, but the value in (6) is compatible with their measurement due to their much larger error estimate. It might be possible for isotope shifts of the opposite sign from the normal mass effect to arise entirely from specific mass effects, but in this case $\Delta\nu(132-134)$ and $\Delta\nu(134-136)$ must be equal. However, if each is in error by one standard deviation, they still differ by more than one standard deviation. Therefore it appears unlikely that only mass effects contribute.

The spreading of the nuclear electrostatic charge upon the addition of neutrons raises the atomic energy levels so that the upper level of a transition contributes shifts in the same direction as the normal mass effect, while the lower level contributes shifts in the opposite direction. Unless the size and direction of the specific mass effect is known, little can be learned about the field shifts. Nevertheless, estimates can be made of the magnitudes possible due to various contributions.

Isotope shifts involving the xenon $6s$ states are all in the neighborhood of 30 MHz (1×10^{-3} cm^{-1}).[7,9] Configuration mixing of $6s$ levels with $5d$ levels could produce some isotope effect for the 2.02-μ line, and it would be difficult to determine its size without good estimates of the amount of mixing. The field effect due to a $p_{1/2}$ state is about 24 times smaller than that for an s state of the same principal quantum number,[11] so that the maximum shift of the lower level of the transition would be near 1 MHz if it were due entirely to a $p_{1/2}$ state. In the limit where the electrostatic interaction between the $6p$ state and the $5p^5$ core vanishes, this level is due wholly to a $p_{3/2}$ state,[14] so that any significant contribution from the $6p$ levels appears unlikely.

It is also possible for the core electrons to produce isotope shifts due to the change in the amount

the optical electron screens the nucleus from the core electron before and after the transition. The field effect in an atomic level is proportional to n^{-3}, where n is the principal quantum number,[11] so that the unscreened shift due to a 1s level could be as large as 30×6^3 or 7000 MHz (200×10^{-3} cm^{-1}). Only a 1-part-in-10^3 change in the screening during the transition would be sufficient to produce a significant effect. The sign of this effect would depend on whether the $5d$ or $6p$ electrons spend more time inside the orbits of the 1s electrons. In any case, it is likely that screening contributes to the nuclear field effects which occur in this transition.

VI. CONCLUSION

Xenon isotope shifts were measured for the transition

$$(^2P^0_{3/2})\,5d\,[3/2]^0_1 \rightarrow (^2P^0_{3/2})6p[3/2]_1$$

at 2.0268 μ between isotopes Xe^{132}, Xe^{134}, and Xe^{136}. The results of the measurements were

$$\Delta\nu(132-134) = 500\pm300 \text{ kHz } (0.017\pm0.01\times10^{-3} \text{ cm}^{-1}), \quad (6')$$

$$\Delta\nu(134-136) = 1500\pm300 \text{ kHz } (0.050\pm0.01\times10^{-3} \text{ cm}^{-1})$$

which represent an accuracy of 2 parts in 10^9 of the transition frequency. The shifts were found to differ in direction from the normal mass effect, and the magnitude of the two were sufficiently different to indicate the probable contribution of significant nuclear field effects. The screening of the core electrons from the nucleus by the optical electrons is probably sufficient to account for these contributions, but configuration mixing between 6s and $5d$ levels could also be significant.

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