High-Precision Isotope-Shift Measurement of $2\,^3S$-$2\,^3P$ Transition in Helium

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The isotope shift for the 2^3S-2^3P transition between ^{3}He and ^{4}He has been measured with a laser-microwave spectroscopy technique to a precision of 1 ppm, or 30 kHz. Our result is 100 times more precise than previous measurements and is by far the most precise isotope-shift measurement ever made for a multielectron system. It provides, for the first time, not only a check for the specific mass shift, but also for the calculations of the nuclear volume shift, the relativistic corrections involving the coupling between electrons, and the QED contributions.

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As a three-body system, helium has attracted the attention of both mathematicians and physicists for many years. Recent new variational techniques[1-4] give very-high-precision calculations for both energy levels and wave functions for this atom. Precision measurement of the isotope shift provides a good test of the theory.

The isotope shift (IS) is the energy difference of atomic levels or transitions for different isotopes. The two major parts are the mass shift (MS) and the volume shift (VS). The MS is due to the motion of the nucleus with finite mass,[5] and the VS (also known as the field shift) is due to the nonzero size and the shape of the electric charge distribution of the nucleus. For light elements the VS is almost negligible compared to the MS, whereas for heavy elements the VS is dominant. The MS is defined to be the sum of the normal mass shift (NMS) and the specific mass shift (SMS). The NMS is the well-known Bohr-reduced mass correction which can be evaluated exactly. The SMS, also called the polarization mass shift and depending on $\langle \sum_{i<j} \mathbf{p}_i \cdot \mathbf{p}_j \rangle$, where $\mathbf{p}_i$ and $\mathbf{p}_j$ are the linear momenta of the electrons, is not zero due to exchange effects and repulsion between the electrons. It is difficult to calculate and requires a good knowledge of the wave functions. A precise determination of the specific mass shift provides experimental investigation of the correlations and is an excellent test of the accuracy of the computed wave functions. Helium is the simplest multielectron system and therefore the best suited for study. The most definitive comparison with theory is provided by the low-lying terms, where $\langle \mathbf{p}_1 \cdot \mathbf{p}_2 \rangle$ is very sensitive to configuration interaction in the computed wave functions.

In this Letter, we report a very-high-precision measurement of the isotope shift in the transition 2^3S-2^3P between ^{3}He and ^{4}He. The total isotope shift can be written as follows:[1,2,4]

$$\mathrm{IS} = [\mathrm{IS}_{\mathrm{nr}} + \mathrm{IS}_{\mathrm{rel}}] + [\mathrm{IS}_M] + [\mathrm{IS}_{\mathrm{nuc}}] + \mathrm{IS}_{\mathrm{RR}} + \mathrm{IS}_{\mathrm{st}} + \mathrm{IS}_{\mathrm{QED}} ,$$

where the brackets denote, in succession, the NMS, SMS, and VS. Here $\mathrm{IS}_{\mathrm{nr}}$ is the nonrelativistic contribution and $\mathrm{IS}_{\mathrm{rel}}$ is the relativistic correction. IS_M includes both first- and second-order terms in μ/M. For this transition, the SMS is very large, 21 253 MHz, whereas the NMS is only 12414 MHz. Thus the measurement provides a sensitive test of the interesting part of the theory. $\mathrm{IS}_{\mathrm{nuc}}$, i.e., VS, is only 0.845 MHz. $\mathrm{IS}_{\mathrm{RR}}$ is the relativistic recoil correction which is on the order of a few MHz. $\mathrm{IS}_{\mathrm{st}}$ is the singlet-triplet mixing which only slightly affects the 2^3P_1 state, by ~ 3 kHz. $\mathrm{IS}_{\mathrm{QED}}$ are the QED corrections including that of the electron anomalous magnetic moment. The two-electron QED calculations for this helium transition still have a large uncertainty (~ 40 MHz). Also, there are some uncalculated terms of order α^4 such as the mixture of 2^3P with the higher levels ($n \geq 3$) that can be as large as 10 MHz. But the uncertainties are reduced by a factor of 4×10^{-5} due to the cancellation when we consider the transition isotope shift. The residual QED contribution is only 135 kHz with an uncertainty of 10%. The total uncertainty of the calculation, quoted by Drake,[4] is 50 kHz.

Figure 1(a) shows the 2^3S and 2^3P energy levels for ^{3}He and ^{4}He. The ^{4}He fine-structure intervals are 2^3P_2-2^3P_1, 2291.196(5) MHz,[6] and 2^3P_1-2^3P_0, 29616.844(21) MHz,[7] as measured with an optical-microwave resonance technique by the Yale group. The comparison between experiment and theory for the isotope shift is complicated by the presence of hyperfine structure (HFS) in ^{3}He. By definition, the isotope shift, about 33 668 MHz, is the difference between the ^{3}He and ^{4}He transitions in the absence of the ^{3}He hyperfine interaction. In order to determine the isotope shift, one must know the relationship between the levels of ^{3}He with and without hyperfine structure. To do this, Hinds, Prestag, and Pichanick used a phenomenological Hamiltonian in which the fine-structure intervals are deduced from the experimental data for ^{4}He with theoretical mass-dependent corrections and the hyperfine interactions are expressed by an effective Hamiltonian with coupling constants determined by experiment.[8] This calculation has recently been checked by Drake.[4] The values for the ^{3}He fine-structure intervals without hyperfine interactions, which are the same for both calculations, are[8] 2292.173(9) MHz for 2^3P_2-2^3P_1 and 29616.569(21) MHz for 2^3P_1-2^3P_0. The hyperfine structure constants are slightly different for the two

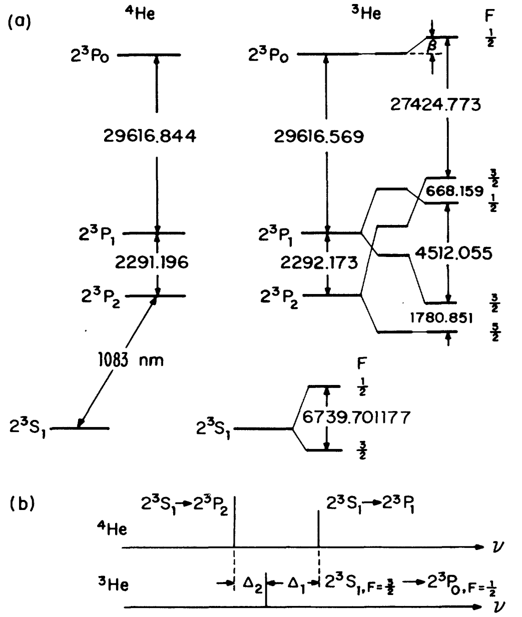

FIG. 1. (a) Energy-level diagram (not to scale) for the 2^3S and 2^3P states of ^{3}He and ^{4}He. Units not indicated are in MHz. For the 2^3P state of ^{3}He, the left-hand column shows the fine-structure intervals in the absence of the hyperfine interaction; the center column shows the hyperfine structure obtained without including off-diagonal terms; the right-hand column shows the real level position obtained with the off-diagonal matrix elements included. The parameter β, which relates the ^{3}He 2^3P_0 level with and without the hyperfine interactions, is 323.977(12) MHz. (b) The transitions of interest for this experiment, which measures Δ_1 and Δ_2.

calculations. For our analysis we used Drake's value for the parameter β appearing in Fig. 1(a), which is 323.977(12) MHz.[4] The value obtained by Hinds, Prestag, and Pichanick differed by only 7 kHz. The measured hyperfine interval for the 2^3S_1 state of ^{3}He is 6739.701 177(16) MHz;[9] the 2^3S_1 level in the absence of the hyperfine interaction is taken to be the center of gravity of the two hyperfine levels. Figure 1(b) shows the relative positions of the transitions of interest in ^{3}He and ^{4}He; the ^{3}He $2^3S_{1,F=3/2} \rightarrow 2^3P_{0,F=1/2}$ transition falls in between the ^{4}He $2^3S_1 \rightarrow 2^3P_1$ and $2^3S_1 \rightarrow 2^3P_2$ transitions. The approximate separations Δ_1 and Δ_2 are 1481 and 811 MHz, respectively. This experiment measures these gaps with the new technique described in the following and thereby obtains a high-precision value of the isotope shift for this transition.

The standard method for measuring the isotope shift is to scan the laser across the spectral lines and use the frequency markers produced by a Fabry-Pérot etalon to measure the separation. Because typically the laser must be scanned over a wide range (several GHz), the useful information (spectral lines < 20 MHz) occupies only a very small part of the scan. As a result it is difficult to locate precisely the centers of the spectral lines and the frequency markers. Furthermore, the precision of this method is limited by other problems, such as nonlinearity of the laser scan and laser jitter. We have developed a new method to measure the isotope shift. A single-mode laser with frequency ν is passed through an electro-optical modulator (EOM) that is driven by a controlled microwave frequency f. The EOM generates two sidebands on the laser light of frequencies $\nu \pm f$. The carrier ν is tuned to the transition for one isotope and f is adjusted so that the sideband ($\nu+f$ or $\nu-f$) is on the transition for the other isotope. Then f is fixed and the laser is scanned across the two isotopes simultaneously. The scan is slow and narrow ranged (just a little broader than the linewidth) so the line centers can be located precisely. Then f is changed slightly and the scan is repeated. This new technique overcomes the shortcomings of the old method and can improve the precision by 2 orders of magnitude. The only requirement for employing this technique is that the two isotopes must not be in the same sample.[10]

In addition, we have developed a technique we call "Doppler-shifted saturation spectroscopy." In Doppler-free saturation spectroscopy, it is well known that if there are two transition lines close enough (approximately the Doppler width) which share the same lower or upper level, a peak will appear when the laser frequency is tuned to the exact middle of the two lines. This is the so-called "crossover," which we refer to here as a "type-I crossover." There is another kind of crossover. When the laser beam contains two frequencies with a fixed separation (e.g., the carrier and the sideband generated by an EOM) and a transition line falls at the exact center of the two frequencies, there will be a signal peak. This is due to one frequency burning a hole in one side of the Doppler profile (rather than at the center) and the other frequency coming from the opposite direction and probing it. We call this situation a "type-II crossover." We actually observed the type-II crossover and used it to take half of our data. Here is how it works: The atoms in the sample have transition frequency ν_0. The laser frequency ν_1 ($\nu_1 = \nu_0 + \Delta$) selects a group of atoms with velocity V and therefore with Doppler shift Δ by hole burning. The laser frequency ν_2 coming from the opposite direction detects this group of atoms with $\nu_2 = \nu_0 - \Delta$. Therefore, $\nu_0 = (\nu_1 + \nu_2)/2$. In our case $\nu_2 = \nu_1 \pm f$, with ν_1 the carrier frequency, ν_2 the sideband frequency, and f the microwave frequency. Therefore, $\nu_0 = \nu_1 \pm f/2$. In the experiment, we set ν_1 on one isotope transition with conventional Doppler-free saturation spectroscopy, and adjust the microwave frequency f to observe ν_0 in the other isotope. The isotope shift is then $f/2$. With this method, we can use the carrier to saturate the transition and use the sideband to probe, which is advantageous when the sideband power is low. Thus the signal-to-noise ratio can be increased significantly.

Figure 2 presents the schematic diagram of the experiment. The ^{3}He and ^{4}He 2^3S_1 metastable states are

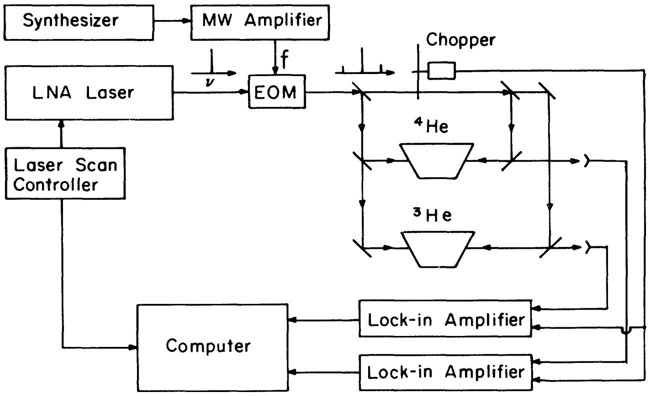

FIG. 2. The experimental arrangement for measuring the helium-isotope shift.

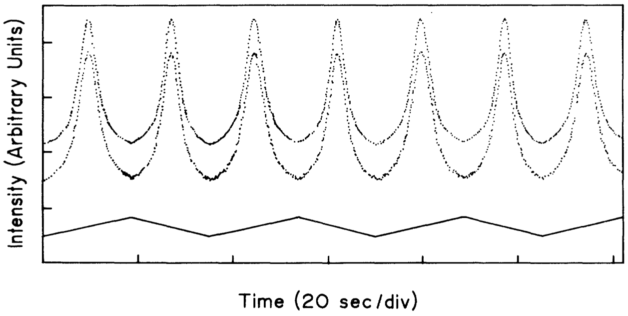

FIG. 3. A portion of a typical scan. The two upper traces are signals from two helium isotopes. The lower trace is the laser frequency scan voltage. The frequency is scanned up and down repeatedly around the line centers.

prepared in two separate low-presure rf discharge cells which are magnetically shielded by Mumetal tubes. The LNA ($La_{1-x}Nd_xMgAl_{11}O_{19}$) laser, whose output wavelength is at 1.083 μm, is a home-made ring laser stabilized to an external cavity.[11] Pumped by the 4-W green line (514 nm) from an Ar^+ laser, it puts out a 100-mW single mode with a bandwidth of 200 kHz. A $LiTaO_3$ crystal EOM generates the sideband frequency which can be precisely tuned by a synthesizer. The laser is then scanned so the carrier and sidebands cross the ^{3}He and ^{4}He transitions, and the saturated absorption signals are detected and recorded simultaneously. Figure 3 shows part of a typical scan. The signal linewidth is about 15 MHz. The laser frequency is scanned up and down around the line center. The scan is repeated for different modulation frequencies. Figure 4 shows a plot of the EOM modulation frequency versus the difference in position between the two isotope line centers. Zero difference means the two line centers appear simultaneously; the corresponding modulation frequency is the isotope shift to be measured.

Cells with pressures of 0.05, 0.2, and 0.6 Torr of both ^{3}He and ^{4}He are used. Pressure shifts are measured with two methods: Cells of two isotopes with the same pressure are measured to determine the differential pressure shift, i.e., the pressure shift for the isotope shift; cells of the same isotope with different pressure are measured to determine the pressure shift for each isotope. Taking the difference of the pressure shift for each isotope we can also get the differential pressure shift. The results of the two methods agree with each other. The measured pressure shifts are $-1.1(2)$ MHz/Torr for ^{3}He, $-0.9(2)$ MHz/Torr for ^{4}He, and 0.14(10) MHz/Torr for the difference between the two isotopes. The minus sign means the frequency is redshifted when the pressure goes up. The differential pressure shift of 0.14(10) MHz/Torr is taken into account in the final result; e.g., the correction due to the pressure shift for the measurement with the 0.2-Torr cells is -28 kHz. To avoid other systematic shifts, care has been taken to use different laser powers, microwave powers, and rf-discharge levels. The laser power has to be kept as low as possible and the two counterpropagating laser beams have to be aligned properly. This is crucial due to the fact that adjacent transitions are fairly close, 6.7 GHz for ^{3}He and 2.3 GHz for ^{4}He, and the pedestal[12] of the signal from one line can extend under the adjacent line shifting its center slightly.[13] Care has been taken to account for the effect of the pedestal.

Each point in Fig. 4 represents 5 to 8 laser scans across the transition. Therefore, one measurement according to such a plot is based on over 100 scans. The final result represents 80 measurements or over 8000 scans. The contribution of the random error of the measurement to the final uncertainty is the same as that from the pressure shift. Uncertainties from other sources are negligible. The final results are $\Delta_1 = 1480.573(30)$ MHz and $\Delta_2 = 810.608(30)$ MHz (see

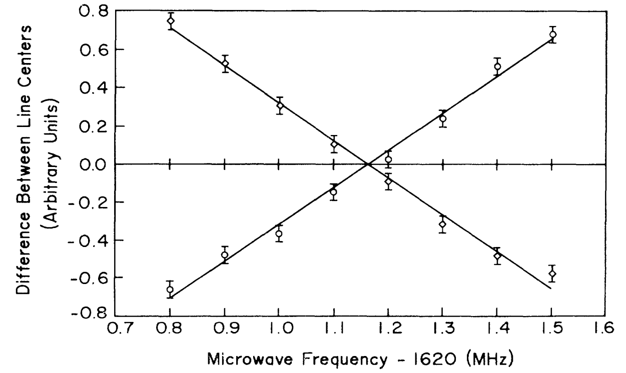

FIG. 4. The EOM modulation frequency vs the difference between the two isotope line centers. The two sets of data are for the laser scanning up and down, respectively. The Doppler-shifted saturation spectroscopy technique is used to measure Δ_2. The frequency f is the intersection of a least-squares fit to the data with the horizontal axis, and $\Delta_2 = f/2$. The fact that the two sets of data intersect the axis at the same point is a tribute to the quality of the data.

TABLE I. The isotope shifts in the 2^3S_1-$2^3P_{0,1,2}$ transitions between ^{3}He and ^{4}He. Here $\mathrm{IS_{expt}}$ are our results; $\mathrm{IS_{theory}}$, NMS, and SMS are calculated by Drake (Ref. 4); $\Delta_{\mathrm{expt}} = \mathrm{IS_{expt}} - \mathrm{NMS} - \mathrm{SMS}$; $\Delta_{\mathrm{theory}} = \mathrm{IS_{nuc}} + \mathrm{IS_{st}} + \mathrm{IS_{RR}} + \mathrm{IS_{QED}}$. Units are in MHz.

Transition	$\mathrm{IS_{expt}}$	$\mathrm{IS_{theory}}$	NMS	SMS	Δ_{expt}	Δ_{theory}
2^3S_1-2^3P_0	33667.968(38)	33667.856(50)	12416.792	21256.922	−5.746(38)	−5.858(50)
2^3S_1-2^3P_1	33667.693(38)	33667.571(50)	12415.467	21256.922	−4.696(38)	−4.818(50)
2^3S_1-2^3P_1	33668.670(38)	33668.555(50)	12415.359	21256.922	−3.611(38)	−3.726(50)

Fig. 1). The result for Δ_2 agrees well with the previous less precise measurement of 811(3) MHz.[14] No previous measurement for Δ_1 has been reported. A crucial test of the reliability of our results is to compare the ^{4}He fine structure 2^3P_2-2^3P_1 deduced from our data, 2291.181(42) MHz, with the known high-precision measurement of this splitting, which is 2291.196(5) MHz.[6] The agreement is very good. The isotope shifts are deduced according to Fig. 1 and listed in the first column of Table I. The last two columns compare the small corrections $\mathrm{IS_{nuc}} + \mathrm{IS_{RR}} + \mathrm{IS_{st}} + \mathrm{IS_{QED}}$. Almost all previous isotope-shift experiments have been unable to test these small corrections due to their large uncertainties. Even though our results differ with the theory by about 1.6 standard deviations, they still support the theory to the 100-kHz level. This is by far the best confirmation of the two-electron theory.

The uncertainties in this experiment are mainly due to the spectral linewidth and the pressure shift. If this experiment were carried out in an atomic beam, where the linewidth is much narrower and the pressure shift is negligible, the uncertainties should be easily reduced by another order of magnitude. With current high-precision-measurement techniques, the determination of the absolute value for an optical resonance frequency is basically limited by the precision of the optical frequency standard used; the best available standard is the $^{127}\mathrm{I}_2$-stabilized He-Ne laser with a precision of 1.6×10^{-10}.[15] For the helium $2^3S \rightarrow 2^3P$ transition, this limit is 44 kHz. The new spectroscopy technique we have developed does not need any optical frequency reference. With this technique, the differences of optical frequencies can be measured much more precisely than the frequencies themselves.

In conclusion, the isotope shift between ^{3}He and ^{4}He of the $2^3S \rightarrow 2^3P$ transition has been measured to a precision of 1 ppm. With a hundredfold improvement in precision over the previous experiment, this is the most precise isotope-shift measurement ever made for a multielectron system. This dramatic improvement is made possible with the laser-microwave spectroscopy technique (including Doppler-shifted saturation spectroscopy) we developed, which is generally applicable and should be a very useful experimental method.

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