

Specific mass shift in the  $1s2s\ ^3S$  and  $1s5p\ ^3P$  states of helium

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We report the first measurement of the specific mass shift of the  $5^3P$  level between  $^3\text{He}$  and  $^4\text{He}$ . A value  $\Delta\nu_s = -1113.1 \pm 5.0$  MHz, twenty times more accurate than the best known theoretical value, was obtained by ultraviolet-infrared double-resonance laser spectroscopy of the  $2^3S$ - $5^3P$ - $13^1,^3D$  transition.

The isotope shift in atomic spectra is generated by a change in either nuclear mass or nuclear volume. The volume shift is the dominant effect in heavy elements and has been investigated to some extent. In light elements, the volume shift is negligible and only the mass shift must be considered. The mass shift results from the additional kinetic energy due to the motion of the nucleus and can be divided into a normal mass shift and a specific mass shift. The normal mass shift is due to the reduced mass correction which can be calculated exactly. The specific mass shift is caused by the correlation between the electrons and is generally difficult to evaluate. A study of the specific mass shift is especially interesting in helium since it is the simplest many-electron system and therefore the best suited for an investigation of the correlation effect, which is caused by electrostatic interaction and electron exchange. In addition, a valuable test of computed helium wave functions is possible.

Two different high-resolution laser-spectroscopic methods have been used to study the optical isotope shift between  $^3\text{He}$  and  $^4\text{He}$ . The isotope shifts in the transitions  $2^3S$ - $n\ ^3S$  ( $n=4-6$ ) and  $2^3S$ - $n\ ^3D$  ( $n=3-6$ ) were measured by Doppler-free two-photon spectroscopy.<sup>1</sup> Results on the specific mass shifts of  $n^3S$  ( $n=2-6$ ) levels were obtained. The nonlinear technique of intermodulated fluorescence spectroscopy was applied to study the  $2^3p$ - $3^3D$  transition,<sup>2</sup> yielding a result on the specific mass shift of the  $2^3P$  level. In both investigations, good agreement was found with shifts calculated by Accad *et al.*<sup>3</sup> and Pekeris.<sup>4</sup> In this Rapid Communication, we report isotope shift measurements in the  $2^3S$ - $5^3P$  and  $5^3$ - $13^3D$  transitions, employing nonlinear uv laser spectroscopy and ultraviolet-infrared double-resonance laser spectroscopy, respectively. The results on the specific mass shifts in the  $2^3S$  and  $5^3P$  levels are compared with theoretical calculations. The result in the  $2^3S$  level is further compared with measurements by two-photon spectroscopy.<sup>1</sup>

We have investigated the  $1s2s\ ^3S$ - $1s5p\ ^3P$  transition of  $^3\text{He}$  and  $^4\text{He}$  at 294.5 nm by the technique of intermodulated fluorescence spectroscopy. The ex-

periments and interpretation of the spectroscopic information are described in detail in Refs. 5 and 6. The ultraviolet radiation is produced by second harmonic generation in a passive ring enhancement cavity, pumped by the output of a ring dye laser. The helium atoms in the metastable  $2^3S$  level are produced by bombardment of ground-state atoms with electrons of 50-eV energy from an electron gun. We have analyzed the energies of the fine and hyperfine components of the  $5^3P$  level of  $^3\text{He}$  and  $^4\text{He}$  relative to the energy value of the spinless electron  $E(5^3P)$ . For  $^4\text{He}$ , the analysis can be done in a simple  $LS$  coupling scheme. In  $^3\text{He}$ ,  $J$  is not a good quantum number due to the strong hyperfine interaction of the  $1s$  electron with the nuclear spin. Therefore, we have done the analysis for  $^3\text{He}$  in an uncoupled representation (see Ref. 5). In Table I, we have listed the energies of the components of the  $5^3P$  state in  $^3\text{He}$  and  $^4\text{He}$ , relative to  $E(5^3P)$ . In addition, we have listed the energies of the  $2^3S$  level. The hyperfine structure of this state was measured by Rosner and Pipkin<sup>7</sup> by optical pumping methods. The isotope shift, which is defined as a shift in the energy values of the spinless electron, can be extracted from a measured frequency shift between transitions in  $^3\text{He}$  and  $^4\text{He}$  with the help of Table I.

For the determination of the  $2^3S$ - $5^3P$  isotope shift we have measured the frequency shift between the hyperfine component  $(1)\frac{3}{2}$  -  $(2)\frac{5}{2}$  of  $^3\text{He}$  and the fine-structure component 1-2 of  $^4\text{He}$  (see Ref. 6). The result of  $48\,878.8 \pm 3.0$  MHz is the average of ten measurements. For a frequency scan of about 50 GHz, no adjustment of the temperature of the doubling crystal ammonium dihydrogen arsenate (ADA) had to be made. At the beginning and end of the scan, the uv power dropped to about 80% of the maximum power. The frequency shift was measured with the help of a confocal interferometer with a well-known free spectral range of 151.3777 MHz. From the measured frequency shift of  $48\,878.8 \pm 3.0$  MHz, an isotope shift of  $48\,960.2 \pm 4.0$  MHz was extracted (see Table II, row 1).

The  $1s5p\ ^3P$ - $1s13d\ ^1,^3D$  transition of  $^3\text{He}$  and  $^4\text{He}$  at 2.590  $\mu\text{m}$  was investigated by ultraviolet-infrared

TABLE I. Energy values of the fine-structure and hyperfine-structure components for the  $2^3S$ ,  $5^3P$ , and  $13^1, 3^3D$  levels of  $^3\text{He}$  and  $^4\text{He}$ . The zero energy values are given by the energy of the spinless electron. The accuracy of the energy values is better than or equal to 1 MHz. Numbers are given in MHz.

	$J$	$^4\text{He}$	$(J)$	$F$	$^3\text{He}$
$2^3S$	1	0	1	$\frac{1}{2}$	4493.1
			1	$\frac{3}{2}$	-2246.6
$5^3P$	0	1552.8	0	$\frac{1}{2}$	4872.2
	1	-109.4	1	$\frac{3}{2}$	4103.2
	2	-244.9	1	$\frac{1}{2}$	-1264.8
			2	$\frac{3}{2}$	-2297.0
			2	$\frac{5}{2}$	-2410.1
$13^1D$	2	2269.0	2	$\frac{5}{2}$	7197.3
			2	$\frac{3}{2}$	7180.8
$13^3D$	1	13.4	1	$\frac{3}{2}$	-578.5
	2	-2.7	2	$\frac{5}{2}$	-604.0
	3	-3.9	1	$\frac{1}{2}$	-2153.0
			2	$\frac{3}{2}$	-2161.0
			3	$\frac{5}{2}$	-2169.4
			3	$\frac{7}{2}$	-2170.3

double-resonance laser spectroscopy. The hyperfine measurements, the analysis of the data, and the experimental apparatus are described in Refs. 8 and 9. The  $1s5p\ ^3P$  level was populated with the same uv laser and in the same helium chamber as used in the intermodulated fluorescence experiment. In a second

TABLE II. Experimentally determined isotope shifts  $\Delta\nu_{\text{expt}}$ , Bohr mass shifts  $\Delta\nu_B$ , and specific mass shifts  $\Delta\nu_{s, \text{expt}}$  between  $^3\text{He}$  and  $^4\text{He}$  transitions. Numbers are given in MHz.

Transition	$\Delta\nu_{\text{expt}}$	$\Delta\nu_B$	$\Delta\nu_{s, \text{expt}}$
$2^3S - 5^3P$	$48\,960.2 \pm 4.0$	45\,645.2	$3315.0 \pm 4.0$
$5^3P - 13^3D$	$4077.9 \pm 5.0$	5191.0	$-1113.1 \pm 5.0$
$2^3S - 13^3D$	$53\,038.1 \pm 9.0$	50\,836.2	$2201.9 \pm 9.0$

step, a single-mode infrared cw color center laser was used to excite the  $13d$  level. The experiment yielded spectra free of Doppler broadening since both laser beams were oriented antiparallel in the interaction region. The analysis of the data was done in an uncoupled representation and singlet-triplet mixing was included in the calculations. The good agreement between measured and calculated hyperfine splittings indicates that configuration mixing with other states, e.g., with the  $13g$  state, is negligible. In Table I, we have listed the energies of the  $13^1D$  and  $13^3D$  levels, relative to  $E(13^3D)$ . The energies of  $^4\text{He}$  are taken from Ref. 10.

For the determination of the  $5^3P - 13^3D$  isotope shift, some changes in the experimental apparatus had to be made. The frequency shift between two components in  $^3\text{He}$  and  $^4\text{He}$  cannot be measured by a simple scan of the color center laser across the isotopic lines. Simultaneously, the uv laser frequency must be changed from the  $^3\text{He}$  uv line to the  $^4\text{He}$  uv line, i.e., by about 50 GHz. Furthermore, it is not sufficient to stabilize the uv laser in the center of the Doppler profile of either isotope. For an exact determination of the isotope shift, the uv laser must be stabilized on known components of the  $2^3S - 5^3P$  transition. Therefore, we have used a combined experimental setup for intermodulated fluorescence and for double-resonance spectroscopy.

The isotope shift of the  $5^3P - 13^3D$  line was determined by measuring the frequency shift between the hyperfine component  $(1)\frac{3}{2} - (2)\frac{5}{2}$  (component of the  $5^3P - 13^1D$  transition) of  $^3\text{He}$  and the fine-structure component 2-3 of  $^4\text{He}$ . The uv laser was stabilized either to the  $(1)\frac{1}{2} - (1)\frac{3}{2}$  component of the  $2^3S - 5^3P$  transition in  $^3\text{He}$  or to the 1-2 component of  $^4\text{He}$ . The frequency shift was measured to be  $1224.2 \pm 3.0$  MHz. From the result, an isotopic shift of  $4077.3 \pm 5.0$  MHz was extracted when corrected with respect to  $E(13^3D)$ . In Ref. 2, it was suggested to measure the isotopic shift directly between the components  $(2)\frac{5}{2} - (3)\frac{7}{2}$  of  $^3\text{He}$  and 2-3 of  $^4\text{He}$ . The two transitions are not affected by singlet-triplet mixing and should be identical within experimental error. We have corrected the measured frequency shift of  $1224.2 \pm 3.0$  MHz with respect to these two transitions, yielding an isotope shift of  $4078.5 \pm 5.0$  MHz. We adopt the average of both values as the isotope shift of the  $5^3P - 13^3D$  transition. The result of  $4077.9 \pm 5.0$  MHz is listed in row 1 of Table II. The isotopic shift of the  $2^3S - 13^3D$  transition, which is also listed in Table II, is given by the sum of the results in the  $2^3S - 5^3P$  and  $5^3P - 13^3D$  lines.

For a two-electron atom, the isotopic mass shift is given by the mean value of

$$\frac{(p_1 + p_2)^2}{2M} = \frac{p_1^2 + p_2^2}{2M} + \frac{\bar{p}_1 \cdot \bar{p}_2}{M}, \quad (1)$$

where  $M$  is the nuclear mass and  $p_1, p_2$  are the mo-

menta of the two electrons. The first term on the right-hand side of Eq. (1) is the Bohr or normal mass shift. The second term, which is called the specific mass shift, describes the correlation between the electrons. The Bohr mass shift  $\Delta\nu_B$  between  $^3\text{He}$  and  $^4\text{He}$  is determined only by the reduced masses of the isotopes and can be calculated exactly.<sup>2</sup> We have obtained  $\Delta\nu_B$ , as listed in row 2 of Table II, using atomic masses tabulated in Ref. 11. The third row in Table II lists the experimentally determined specific mass shifts  $\Delta\nu_{s, \text{expt}}$  between  $^3\text{He}$  and  $^4\text{He}$  for the three investigated transitions. The values are given simply by the difference of  $\Delta\nu_{\text{expt}} - \Delta\nu_B$ , since the volume shift in helium is negligible at an uncertainty level of a few MHz.

The specific mass shift term  $\bar{p}_1 \cdot \bar{p}_2 / M$  cannot be calculated exactly and must be treated as a perturbation. The calculated shift depends strongly on the assumed wave functions and is, therefore, a sensitive indicator of their suitability. Hughes and Eckart<sup>12</sup> evaluated the specific mass shift for two-electron atoms using hydrogenlike wave functions with different effective nuclear charges for each electron. These calculations resulted in zero shifts for  $s$  and  $d$  terms, in contradiction to experimental results.<sup>1</sup> More exact calculations have been carried out by Accad *et al.*<sup>3</sup> They determined the specific mass shift and other properties of the low-lying  $s$  and  $p$  states for two-electron atoms up to  $Z = 10$ . The employed wave functions include the interelectronic distance explicitly and are particularly appropriate for the low-lying states. The theoretical values  $\Delta\nu_{s, \text{theor}}$  for the  $2^3S$  and  $5^3P$  levels are listed in row 1 of Table III as obtained from Ref. 3. The convergence interval for the specific mass shift of the  $2^3S$  level is about 1 kHz, indicating the effectiveness of the method employed in Ref. 3 when applied to low-lying states. For this computation, a 364-term expansion was used. A convergence interval of about 90 MHz was obtained for the  $5^3P$  level although a 560-term expansion was used in this case. The effectiveness of

the method apparently diminishes with higher principal quantum numbers, as was pointed out by Accad *et al.*<sup>3</sup>

To our knowledge, no precise calculations concerning the specific mass shift in the  $13^3D$  level have been carried out. Since the  $13d$  Rydberg electron in helium has virtually no correlation with the  $1s$  electron, we assume a zero specific mass shift for this state. This assumption is supported by the measurements described in Ref. 1, where a constant specific mass shift  $\Delta\nu_s$  is obtained for the  $2^3S - n^3D$  ( $n = 3-6$ ) transitions. A nonzero specific shift in  $nd$  states should result in an  $n$ -dependent variation of  $\Delta\nu_s$ , so that a zero specific shift in  $d$  states must be assumed. Consistently, the  $\Delta\nu_s$  obtained for these transitions coincide despite a relativistic correction with a theoretically determined specific mass shift in the  $2^3S$  level.

The experimentally determined specific mass shifts obtained for the transitions listed in Table II can easily be transformed into level shifts with the well-founded assumption of a zero specific mass shift in the  $13d$  state.  $\Delta\nu_{s, \text{expt}}$  for the  $2^3S$  and  $5^3P$  are given simply by the results in the  $2^3S - 13^3D$  and  $5^3P - 13^3D$  transitions, respectively. Both shifts are listed in row 2 of Table III. For comparison, we have also listed the result of Ref. 1 for the  $2^3S$  level. Up to this point, we have not considered relativistic corrections  $\Delta\nu_{s, \text{rel}}$  to the mass shift. One theoretical approach to these relativistic corrections has been formulated by Stone.<sup>13</sup> Stone's theory was used in Ref. 1 to calculate  $\Delta\nu_{s, \text{rel}}$  for  $ns$  ( $n = 2-6$ ) levels. In Table III, row 3, we list the result of  $\Delta\nu_{s, \text{rel}} = -11.0$  MHz obtained for the  $2^3S$  level. The  $\Delta\nu_{s, \text{rel}}$  for  $5^3P$  and  $13^3D$  are zero. The experimental result of Ref. 1 for the specific mass shift of the  $2^3S$  level coincides with theory when Stone's relativistic corrections are included. Our result in the  $2^3S$  does not confirm Stone's theory. We have not yet found an explanation for the difference of about 17 MHz between the two experimental results in the  $2^3S$  level. This difference exceeds the uncertainty intervals by 4 MHz.

The specific mass shift value  $\Delta\nu_{s, \text{expt}} = -1113.1 \pm 5.0$  MHz obtained for the  $5^3P$  state is the first accurate experimental value for a higher excited  $p$  state. The theoretical result of  $\Delta\nu_{s, \text{theor}} = -1079.1 \pm 90.0$  MHz coincides with the experimental value within the uncertainty intervals. However, the latter is about 20 times more accurate. We hope that this experimental result will aid future refinement of model calculations.

In conclusion, we have shown that our experimental result for the specific mass shift in the  $2^3S$  state of helium does not confirm Stone's theory for the relativistic mass shift correction. The first accurate experimental specific mass shift was obtained for the  $5^3P$  level. This value is about 20 times more accurate than the best known theoretical result.

TABLE III. Experimentally and theoretically determined specific mass shifts  $\Delta\nu_s$  between  $^3\text{He}$  and  $^4\text{He}$  levels.  $\Delta\nu_{s, \text{rel}}$  is a predicted relativistic mass shift correction. Numbers are given in MHz.

Level	$\Delta\nu_{s, \text{theor}}$	$\Delta\nu_{s, \text{expt}}$	$\Delta\nu_{s, \text{rel}}$
$2^3S$	$2196.4 \pm 0.001^a$	$2201.9 \pm 9.0^b$ $2185.1 \pm 4.0^c$	$-11.0^c$
$5^3P$	$-1079.1 \pm 90.0^a$	$-1113.1 \pm 5.0^b$	0.0
$13^3D$	0.0	0.0	0.0

<sup>a</sup>Reference 3.

<sup>b</sup>This work.

<sup>c</sup>Reference 1.

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