Mass-polarization effects in the 1s2s 1S and 3S states of helium

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From a laser spectroscopic study of isotope shifts in the 1s2s 1S, 3S → 1snp transitions in 3He and 4He and of the hyperfine structure in the 1snp Rydberg states of 3He we have derived highly accurate information on mass-polarization effects in the 1s2s 1S and 3S states. The experimental results, which are in good agreement with recent, high-precision variational calculations, confirm an isotope dependence of \( \langle p_1 \cdot p_2 \rangle \).

Spectroscopic studies of two-electron atoms give more and more insight in the effects of correlations in the motion of both valence electrons. It is well established that electron correlations play an important role in the doubly-excited states of helium and in the ground and low-lying states of alkaline-earth atoms and several models have been developed to treat their influence. One measure of the degree of correlation is the inner product of the linear momenta \( \langle p_1 \cdot p_2 \rangle \) of both electrons. Its measurement provides an important, independent test of model calculations.

The mass-polarization shift is defined as \( \epsilon_M = (m_e/M) \langle p_1 \cdot p_2 \rangle \), with \( m_e \) the electron mass and \( M \) the nuclear mass. This mass-polarization shift is directly related to the so-called specific mass shift (SMS), which gives a substantial contribution to the isotope shift in atomic-level energies. When comparing energies of different isotopes of the same element, shifts arise from differences in nuclear mass and volume, slightly affecting the electronic orbitals. The volume shift is due to the fact that an electron, especially in an orbital with low angular momentum \( l \), experiences different charge distributions close to the nucleus. For light elements, such as helium, the volume effect may be neglected. The mass shift is composed of the normal mass shift (NMS) and the specific mass shift (SMS). NMS accounts for differences in reduced mass and can be calculated exactly. It follows that in helium a determination of level-energy shifts directly yields values for SMS.

The quantity which can be investigated experimentally, however, is the transition isotope shift (TIS), i.e., the difference in energy shifts of the levels involved in the transition. The deduction of level shifts from measured isotope shifts in transitions between low-lying states is a complex problem. When the upper state involved in the transition is a highly excited Rydberg state, the problem simplifies considerably. For helium Rydberg states 1snl, it is easily shown that the inner product \( \langle p_1 \cdot p_2 \rangle \) scales as \( n^{-3/2} \) (approximately), and consequently SMS converges to zero with increasing value of the principal quantum number \( n \) (see, e.g., Ref. 1).

In this Rapid Communication we report values for SMS and mass-polarization effects of the 1s2s 1S and 3S states of helium as deduced from a high-resolution laser spectroscopic study of TIS in the transitions 1s2s 1S, 3S → 1snp for 3He and 4He. In the experimental setup an atomic beam of an isotopically enriched mixture of 3He and 4He was used. The gas mixture was expanded from a quartz tube nozzle via a skimmer into the excitation chamber. To populate the metastable 1s2s 1S and 3S states a low-current discharge was maintained between a tantalum needle inside the quartz tube and the skimmer. In the excitation chamber 1snp Rydberg states were excited from these states with a laser beam, perpendicularly intersecting the atomic beam. To eliminate stray electric fields the excitation region was carefully shielded with a metal box. The UV laser radiation required for the excitation was produced by intracavity frequency-doubling of a spectra physics 380D ring dye laser (linewidth < 1 MHz). For the excitation from the 1s2s 1S state, 5 mW of UV light in the wavelength region around 313 nm was generated using the dye Rhodamine B and a temperature-tuned nonlinear rubidium dihydrogen phosphate (RDP) crystal, whereas for the excitation from the 1s2s 3S state, 3 mW in the region 260–263 nm was available from a Coumarine 6 laser frequency doubled in potassium dihydrogen phosphate (KDP). He ions produced from Rydberg states by field ionization were focused into a quadrupole mass filter and detected by an electron multiplier. Calibration of wavelength scans was performed with a Fabry-Perot étalon, length stabilized with an I2-stabilized He-Ne laser. Its free spectral range was 148.9553(6)
MHz, accurately calibrated on the separation of the Na $D_1$ and $D_2$ lines. More details of the setup can be found elsewhere.\textsuperscript{[8,9]} In Fig. 1, as an example, the excitation 1s2s $^1S \rightarrow 1s40p$ is shown. The experimental linewidth of 15 MHz full width at half maximum (FWHM) was entirely due to residual Doppler broadening. This spectrum of 60 GHz width shows the transition to $^1P_1$ in $^4$He (single peak at the high-frequency side) as well as two hyperfine transitions of $^3$He (nuclear spin $I = \frac{3}{2}$). The peak at the low-frequency side contains two hyperfine components with primarily $^3P$ character (with hyperfine quantum numbers $F = \frac{1}{2}$, $\frac{3}{2}$) excited because of hyperfine-induced singlet-triplet mixing. The remaining peak is the regular $^1P_1$ excitation, also with unresolved $F = \frac{1}{2}$, $\frac{3}{2}$ components. Since the hyperfine interaction of the np electron varies approximately as $n^{-3}$ individual hyperfine components could not be observed for high $n$. Although the 1s2s $^1S$ state is populated more efficiently than the 1s2s $^3S$ state in our discharge,\textsuperscript{[9]} we did not observe singlet Rydberg states with $n < 30$ as a consequence of the short radiative lifetime of these states and our detection of He ions produced by field ionization. Field ionization was induced a few mm downstream from the laser-atomic-beam interaction region, so low-$n$ Rydberg atoms with a large $^1P$ fraction in their wave function will decay to the 1s2 $^1S$ ground-state hampering detection. Hyperfine structure of $^3$He is a complicating factor in the determination of TIS. It was studied in detail by Vassen and Hogervorst,\textsuperscript{[8]} who were able to explain their observations, including the effects of hyperfine-induced $n$ mixing, using a three-channel, two-limit (hyperfine energies of the $^3$He $^4$ ground state) quantum-defect analysis. Following the same analysis the center of gravity of the $^3$He hyperfine multiplets was obtained resulting in the determination of the isotope shift in transitions from 1s2s $^1S$ and $^3S$ states. As outlined before, the specific mass shift in the transition may be deduced from TIS by subtracting NMS, neglecting volume effects. To calculate NMS we used $m_e = 5.485803 \times 10^{-4}$ amu, $M_4 =$ 4.001506079 amu, and $M_3 = 3.014932149$ amu.\textsuperscript{[10]} Experimental values of SMS for different values of $n$ for transitions from 1s2s $^1S$ as well as $^3S$ are given in Table I. From this table it can be seen that the specific mass shift converges for high $n$ to a constant value, the SMS of the metastable state. More accurate isotope shift measurements around $n = 50$ are feasible as was already pointed out by Vassen and Hogervorst.\textsuperscript{[9]} As a consequence of the large value of TIS the 1snp peak of $^4$He nearly coincides with the 1s($n+1$)p peaks of $^3$He in the interval $n =$ 50–55. An example is shown in Fig. 2 for the 1s2s $^1S \rightarrow 1s51p \, ^1P_1$ ($^4$He), 1s52p ($^4$He) transitions. In this case the small frequency separations may be measured with higher accuracy. The energy difference between adjacent $n$ levels can in principle be calculated precisely (see, e.g., Ref. 11) resulting in highly accurate values for TIS. We leave this for future studies. Several high-precision variational calculations for the mass-polarization shifts in low-lying states of helium are available.\textsuperscript{[12–14]} Kono and Hattori\textsuperscript{[15]} give values for 1sns
TABLE II. Experimental and calculated values (in MHz) of the SMS \( \epsilon_3 - \epsilon_4 \) of the 1s2s \( 1S \) and \( 3S \) states of helium.

<table>
<thead>
<tr>
<th></th>
<th>SMS (expt.)</th>
<th>SMS (calc.)</th>
<th>SMS (calc.)</th>
<th>SMS (calc.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1s2s ( 1S )</td>
<td>2793.0(40)</td>
<td>2804.9(1)</td>
<td>2790.75(1)</td>
<td>2789.87(1)</td>
</tr>
<tr>
<td>1s2s ( 3S )</td>
<td>2189.6(25)</td>
<td>2196.400(1)</td>
<td>2189.89(1)</td>
<td>2188.68(1)</td>
</tr>
</tbody>
</table>

\( ^{a} \)Reference 13.
\( ^{b} \)Reference 14.
\( ^{c} \)Reference 14 with relativistic corrections of Refs. 15 and 16.

\( 1S \) and \( 1snp \) \( 1S \) \( P \) series up to \( n = 8 \). Assuming no mass dependence of \( \langle p_1 \cdot p_2 \rangle \), the SMS for these levels may be calculated directly from SMS \( \epsilon_3 = (M_4 - M_3) / M_3 \).

From isotope shift calculations for the 1snp levels in between \( n = 3 \) and 8 we calculated the contribution of the 1snp levels with \( n > 8 \) to the specific mass shift of the transition using the extended scaling law \( a(n) = b(n)^{-3} + c(n)^{-7} \) (Ref. 8) where higher-order odd powers are neglected. It follows that the contribution of the 1snp level in the triplet case is, e.g., \( -2.0 \) MHz for \( n = 40, -1.0 \) MHz for \( n = 51 \), and only \( 0.5 \) MHz for \( n = 60 \), whereas in the singlet case it is \( +1.9 \) MHz for \( n = 40 \) and \( +0.7 \) MHz for \( n = 56 \). Correcting the transition SMS of Table I with these 1snp contributions, the SMS of the 1s2s \( 1S \) and \( 3S \) states are obtained. Averaged values are given in Table II. It is important to note that the SMS of these states is the difference in the mass-polarization shifts \( \epsilon_3 - \epsilon_4 \).

From the calculations of Kono and Hattori, \( ^{13} \) assuming again no mass dependence of \( \langle p_1 \cdot p_2 \rangle \), the SMS of the 1s2s \( 1S \) and \( 2S \) states may be evaluated. These values are included in Table II. A systematic difference between experiment and these calculations is obvious. The calculated value for 1s2s \( 3S \) state is 6.8 (2.5) MHz higher than the experimental value, whereas for the 1s2s \( 1S \) difference is 11.9 (4.0) MHz. Recent theoretical calculations of Drake \( ^{14} \) for the \( 1s^2 \) and 1s2s levels in \( ^3 \)He and \( ^4 \)He using the mass-polarization operator \( (\mu / M) p_1 \cdot p_2 \) \( \mu \) is the reduced electron mass \( (m_e M) / (m_e + M) \), however, show a small mass dependence of \( \langle p_1 \cdot p_2 \rangle \):

\[
\epsilon_M(1s2s \ 3S) = 0.007442130706(\mu / M) - 0.0574958(\mu / M)^2,
\epsilon_M(1s2s \ 1S) = 0.009503564419(\mu / M) - 0.135277(\mu / M)^2,
\]

in units of \( 2R_M \). Here \( R_M = (1 - \mu / M) / R_m \), with \( R_m \) the Rydberg constant. [Note: in Ref. 8 the relation for \( \epsilon_M(1s2s \ 1S) \) of \( ^3 \)He is not correctly transformed into units of \( cm^{-1} \) as \( R_M \) for \( ^4 \)He was substituted. This only slightly affects the calculated value in that work.] From these relations \( \epsilon_3 \) and \( \epsilon_4 \) are calculated. The values of \( \epsilon_3 - \epsilon_4 \) (equal to SMS) for the 1s2s \( 1S \) and \( 3S \) states are also shown in Table II. They are in agreement with experimental values, thus confirming the mass dependence of the inner product \( \langle p_1 \cdot p_2 \rangle \). This mass dependence is negligible for the SMS of highly excited 1snp states. Finally, relativistic corrections have to be taken into account. Drake and co-worker \( ^{15, 16} \) calculated the correction for the SMS of the 1s2s \( 3S \) state to be \(-1.21 \) MHz. Following the same procedure we find a value of \(-0.88 \) MHz for the relativistic correction for SMS of the 1s2s \( 1S \) state. In the last column of Table II the relativistically corrected values of SMS of 1s2s \( 1S \) and \( 3S \) states are given.

The conclusion is that the measured differences in mass-polarization shifts of the isotopes \( ^3 \)He and \( ^4 \)He both for the 1s2s \( 1S \) and \( 3S \) states are in good agreement with the recent calculations of Drake. A mass dependence of the inner product \( \langle p_1 \cdot p_2 \rangle \) of the momenta of the two electrons is confirmed. The present results support the validity of these variational calculations for two-electron atoms.

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\( ^{15} \)G. W. F. Drake (private communication).