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Radio-Frequency Spectrum of $^3He^-$ and $^4He^-$†

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All independent energy intervals have been measured in the metastable $1s2s2p\,^4P$ state of the helium negative ions $^4He^-$ and $^3He^-$. These results are of sufficient precision to provide a critical test of any proposed wave functions for this state and, when adequate wave functions become available, can be used to test relativistic and radiative corrections in three-electron atomic systems.

High-precision spectroscopic measurements on simple atomic systems provide critical tests of our ability to calculate atomic properties from first principles. Here we report on new precision measurements of the fine structure and hyperfine structure of the three-electron system He^- in the metastable autoionizing $1s2s2p\,^4P_J$ state. The experimental technique was described previously, and the fine-structure intervals in $^4He^-$ were reported to accuracies better than 60 MHz.[1] All independent intervals have now been measured to accuracies better than 0.04 MHz in $^4He^-$ and 0.5 MHz in $^3He^-$. These results provide a critical test of any proposed wave functions for this state. At such time as adequate wave functions are obtained, the 5-ppm precision of the $^4P_{1/2}$-$^4P_{3/2}$ interval in $^4He^-$ is sufficient to test relativistic and radiative corrections in three-electron atomic systems.

The relevant energy levels for $^4He^-$ and $^3He^-$ are shown in Fig. 1. This diagram is based on our present experimental results. In $^3He^-$ the F values shown with primes correspond to $J=\frac{3}{2}$ in $^4He^-$. The large hyperfine splitting is due to the two unpaired s electrons.

Three methods were used to obtain zero-field energy intervals. In the first method resonances were obtained with fixed oscillator frequency in magnetic fields of several hundred gauss by sweeping the field. The observed transition frequency was then extrapolated to zero field by cal-

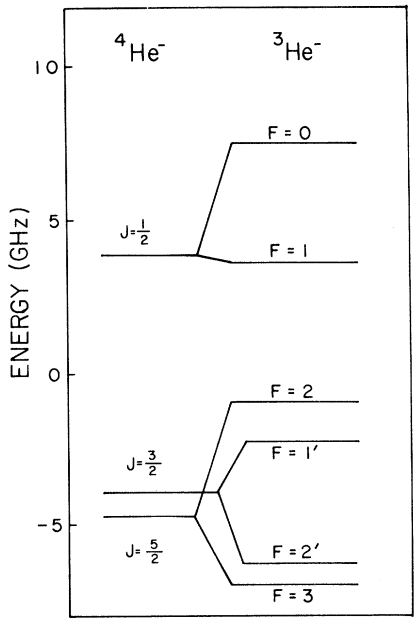

FIG. 1. Energy levels for $^4He^-$ and $^3He^-$ based on present results.

culating the Zeeman effect using

$$\mathcal{H}_z = \mu_B(g_L \vec{L}\cdot\vec{H} + g_s \vec{S}\cdot\vec{H}) - \mu_n g_I \vec{I}\cdot\vec{H}. \quad (1)$$

We assumed $g_s = g_e$, the free-electron g value ($g_e = 2.002\,319$), and $g_L = g_r$, where g_r includes the reduced-mass correction ($g_r = 1 - m_e/M_n$). It is well known that there are further corrections, but simple estimates indicate that they are not needed at the present level of precision.

Unfortunately, there were inhomogeneities in the magnetic field H as great as $\frac{1}{4}$ G because the magnet was designed for lifetime measurements which do not require a precise field. However, the homogeneity was sufficient to obtain intervals accurate to 0.5 MHz or better, depending on the Zeeman effect.

In order to reduce the above uncertainty, some of the resonances were obtained at zero field by varying the oscillator frequency. Because of power variations these resonances were somewhat asymmetrical, with attendant uncertainties in the resonance centers as great as 0.2 MHz.

The most precise zero-field intervals were measured with a novel "minimum-width" method. In this method the oscillator frequency f is kept fixed at a value near the estimated position of the zero-field interval Δ, and the magnetic field is swept through zero from negative to positive values. With sufficiently low power the width of the resonant structure is independent of power level. The width is a minimum when $f = \Delta$ since all the transitions occur at the same field value, $H = 0$. When f departs from Δ the width increases because the magnetic sublevels must be Zeeman shifted to bring them into resonance, and different sublevels have different g values.

This method was used for the measurement of $^4\Delta_{35}$, the interval $J = \frac{3}{2} \rightarrow \frac{5}{2}$ in $^4He^-$. The dependence of width on oscillator frequency is shown in Fig. 2. Four such observations were made under a variety of experimental conditions to reveal possible systematic errors. The parameters include ion-beam velocity, rf power level, and ion-beam focusing. No dependence of the zero-field interval on these parameters was detected. It is well known that rf spectroscopy does not yield the sign of an individual difference. However, the relative signs of contiguous intervals are established by measurements made in a field that couples states of the same M. In our previous work it was shown that $^4\Delta_{35}$ and $^4\Delta_{13}$ have the same sign.[1] Choosing a positive sign in accordance with existing theory,[2] we have

$$^4\Delta_{35} = 825.02 \pm 0.03 \text{ MHz}.$$

The uncertainty is estimated from signal-to-noise considerations and the observed oscillator drift.

To measure $^4\Delta_{13}$ a cylindrical TE_{011}-mode cavity was used. The minimum width of the resonance curve was found to lie at 7819.42 ± 0.04 MHz. The interval was also measured with a direct frequency scan shown in Fig. 3. The central dip is due to interference between transi-

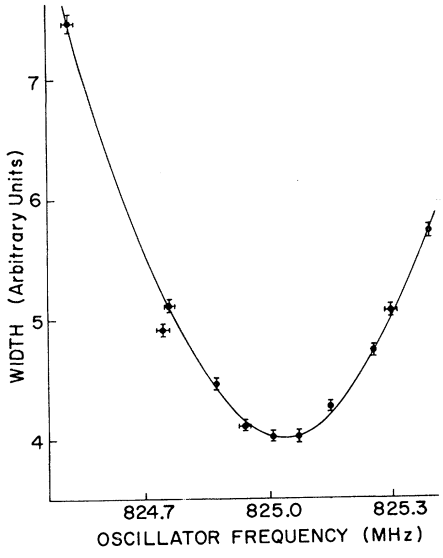

FIG. 2. Resonance width versus frequency for the $J = \frac{5}{2} \rightarrow \frac{3}{2}$ transition in $^4He^-$.

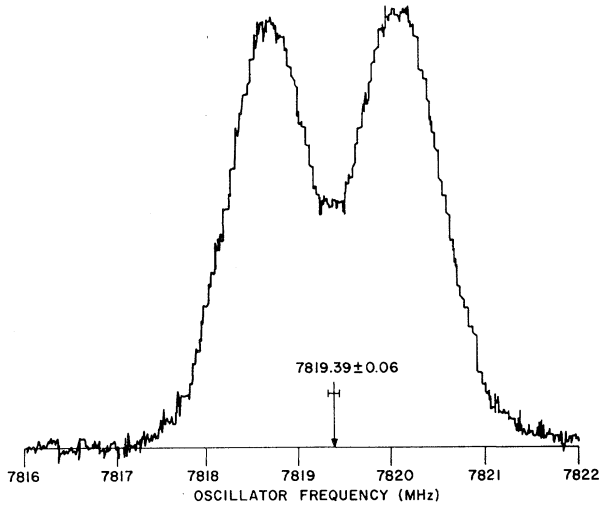

FIG. 3. Resonance curve for the $J = \frac{3}{2} \rightarrow \frac{1}{2}$ transition in $^4He^-$ obtained by sweeping the oscillator frequency with $H = 0$. The dip is due to interference between transitions with $\Delta M = \pm 1$ and $\Delta M = 0$.

TABLE I. Observations in $^3He^-$.

Transition[a] F_i	M_i	F_f	M_f	Magnetic Field[b] (kHz)	f_{osc} (MHz)	f_{fit}[c] (MHz)	Difference (MHz)
2	-2	1'	-1	1434.4	778.9 ± 0.3	778.9	0.0 ± 0.3
2	-	1'	-	0	1238.97 ± 0.06	1239.06	- 0.09 ± 0.06
2	-2	1	-1	1820.0	5953.0 ± 0.7	5953.1	- 0.1 ± 0.7
2	-	1	-	0	4615.1 ± 0.2	4615.1	0.0 ± 0.2
3	-	2'	-	0	733.45 ± 0.06	733.56	- 0.11 ± 0.06
0	0	1	-1	1482.9	4621.9 ± 0.5	4621.4	+ 0.5 ± 0.5
2	-1	2'	0	1247.0	4621.5 ± 0.5	4621.7	- 0.2 ± 0.5
3	2	1'	1	1074.9	4621.7 ± 0.2	4621.7	0.0 ± 0.2
3	-1	2	-1	1595.7	5951.01 ± 0.12	5951.07	- 0.06 ± 0.12

[a]The "initial" state is taken as the one with the longest lifetime.
[b]Measured in units of the proton nuclear resonance frequency.
[c]Frequencies calculated using Eqs. (1) and (2) and varying C_{so}, C_{ss}, a_c, and C_{md} to obtain a least-squares "best fit." All observations are given the same weight.

tions with $\Delta M = 0$ and those with $\Delta M = \pm 1$. Taking the resonance center as the midpoint of the two peaks, we find a value of 7819.39 ± 0.06 MHz, where the error is estimated from the noise and the slight asymmetry of the curve. Combining the above, we obtain

$$^4\Delta_{13} = 7819.41 \pm 0.04 \text{ MHz},$$

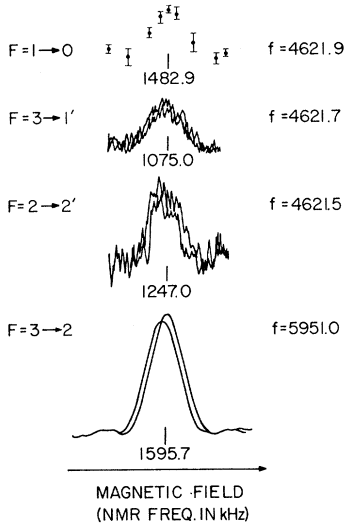

FIG. 4. Zeeman resonance curves for $^3He^-$. The magnetic field is shown in units of the proton nuclear magnetic resonance frequency, and the oscillator frequency f is shown in megahertz.

with the sign established according to the above argument.

The data for $^3He^-$ are given in Table I. In Fig. 4 we show a sample of resonances observed at a few hundred gauss. The interval $F = 2 \rightarrow 1'$ was measured by the method of minimum width, and $F = 3 \rightarrow 2'$ and $F = 2 \rightarrow 1$ were measured with frequency scans at zero field. As above, we choose the sign of one interval and infer the signs of the remainder from observations made in a finite field. With a positive sign for $^3\Delta_{01}$, the interval $F = 0 \rightarrow 1$ in $^3He^-$, we have observed the zero-field intervals given in Table II. The first five values form a complete set of independent intervals from which any further intervals may be derived. Thus, the direct observation of the intervals 2

TABLE II. The observed zero-field intervals in $^3He^-$.

Zero-field interval	Frequency (MHz)
$^3\Delta_{01}$	3929.0 ± 0.5
$^3\Delta_{12}$	4615.1 ± 0.2
$^3\Delta_{23}$	6008.89 ± 0.12
$^3\Delta_{21'}$	1238.97 ± 0.06
$^3\Delta_{2'3}$	733.45 ± 0.06
$^3\Delta_{22'}$	5275.2 ± 0.5
$^3\Delta_{1'3}$	4769.9 ± 0.2

TABLE III. Structure parameters (values in megahertz).

Const	Experiment[a] $^4He^-$	Experiment[a] $^3He^-$	Theory[b]
C_{so}	−1550.740 ±0.010	−1550.77 ± 0.09	−1840
C_{ss}	406.9111 ± 0.0026	406.96 ± 0.04	342
a_c	···	−2955.0 ±0.3	−3030
C_{md}	···	−8.4 ±0.3	−5.6

[a] $1\text{-}\sigma$ uncertainties are given for the experimental results.
[b] Ref. 2.

$\rightarrow 2'$ and $1' \rightarrow 3$ provides a test of the internal consistency of these measurements. In the case of the former, the difference between the value derived from the first five measurements and the directly measured interval is 0.3 ± 0.5 MHz, and in the latter it is 0.0 ± 0.2 MHz, when the errors are added quadratically.

In order to compare the experimental results for the two isotopes and to compare the published theoretical estimates with our experimental results, we use the energy parameters of Manson.[2] Assuming LS coupling, he writes the fine- and hyperfine-structure Hamiltonians as

$$\mathcal{H}_{fs} = C_{so}\vec{L}\cdot\vec{S} + C_{ss}[\tfrac{3}{2}(\vec{L}\cdot\vec{S}) + 3(\vec{L}\cdot\vec{S})^2 - L(L+1)S(S+1)], \tag{2a}$$

and

$$\mathcal{H}_{hfs} = a_c\vec{I}\cdot\vec{S} + C_{md}\{\vec{I}\cdot\vec{L} + \tfrac{2}{15}[(\vec{I}\cdot\vec{S})L(L+1) - \tfrac{3}{2}(\vec{I}\cdot\vec{L})(\vec{L}\cdot\vec{S}) - \tfrac{3}{2}(\vec{L}\cdot\vec{S})(\vec{I}\cdot\vec{L})]\}, \tag{2b}$$

and predicts the values shown in Table III for the spin-orbit coefficient C_{so}, spin-spin coefficient C_{ss}, Fermi contact term a_c, and magnetic-dipole hyperfine interaction C_{md}. (The sign given for C_{md} in Ref. 2 is incorrect; it should be negative.[3])

The experimental coefficients for $^3He^-$ given in Table III were obtained by fitting Eqs. (1) and (2) to the observations in Table I, where the agreement shown between observed and calculated transition frequencies indicates that the Hamiltonian given is adequate to characterize the present results. In $^4He^-$ the two fine-structure coefficients are obtained from the two measured zero-field intervals with no degrees of freedom remaining to test the assumptions on which the given Hamiltonian is based. Manson's values for the spin-orbit and spin-spin coefficients agree with experiment to 18% and the Fermi term to 3%. The small magnetic dipole term agrees to 30%. This overall level of agreement is expected since Manson's variational calculation of the total energy (as a fraction of the ionization potential for the state) is 5% higher than the measured energy. Unfortunately, the more complex wave function of Weiss[4] which has given a total energy within 0.3% of that measured has not yet been used for the evaluation of the fine and hyperfine structure. Comparing the experimental results for the two isotopes, we see that the spin-orbit and spin-spin coefficients are in essential agreement and that no isotopic shift has been detected at the present level of precision.

Clearly better theoretical calculations are required before we can hope to use the present results to test fundamental atomic properties. It is, of course, desirable that Weiss's wave function be used for this purpose, but it is the hope of the authors that the present results will stimulate efforts to calculate the structure parameters with a relative precision of 10^{-5} to 10^{-6}. On this level our results will provide a critical test of the theory of relativistic and radiative corrections for three-electron systems.

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[1] D. L. Mader and R. Novick, Phys. Rev. Lett. 29, 199 (1972), and in *Atomic Physics 3*, edited by S. J. Smith and G. K. Walters (Plenum, New York, 1973), pp. 169–180.

[2] S. T. Manson, Phys. Rev. A 3, 147 (1971).

[3] S. T. Manson, private communication.

[4] A. Weiss, private communication (see Ref. 2).