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Anticrossing spectroscopy of the $n = 6, 7, 8\ ^{1,3}D$ states in ^{3}He

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The anticrossing spectra of the $n = 6, 7, 8\ ^1D - ^3D$ states of ^{3}He have been observed. From these measurements precise values of the electrostatic singlet-triplet separation of these states have been obtained and are found to differ systematically from the accepted values of the ^{4}He singlet-triplet separation by approximately 34 MHz. We have investigated mass-dependent contributions to both the energy-level structure and the magnetic-tuning characteristics of the states of interest. We have found no mechanism that can explain such a large difference in the electrostatic-energy separation of the singlet and triplet D states between the two helium isotopes.

INTRODUCTION

A few years ago, the anticrossing of the Zeeman levels of the $n = 7$, ^1D and ^3D states of ^{4}He was first observed and analyzed to yield the zero-field separation of the singlet and triplet manifolds.[1] The experiment was the first application of anticrossing spectroscopy for the determination of the separation in energy of states of different electron spin multiplicity. These relative energies are often not well known due to the optical selection rule forbidding intercombination transitions. This first work was followed up by similar anticrossing observations in ^{4}He for $n = 3-20$.[2-4] Anticrossing spectroscopy has also been extended to molecules to determine multiplicity separations (singlet-triplet or doublet-quartet) in such species as H_2, D_2, He_2, CN, NO, and $O_2{}^+$.[5]

Anticrossing work on ^{4}He has remained active, although the more recent work has shifted to a determination of the separation of states with different angular momentum, L, of the same multiplicity and principal quantum number, n.[6] This has been one part of a much larger effort to determine experimentally, with high precision, the entire electrostatic-energy-level structure of the simplest multielectron atom, helium.[7] This work includes microwave, radio frequency, and laser magnetic resonance spectroscopy of the ^{4}He levels and has met with considerable success. Indeed, so much success that a combination of the raw experimental data and power series formula based upon the data, give us an experimental "determination" of the majority of the ^{4}He levels to an accuracy of a few MHz or better. Extensions of the current techniques promise an essentially complete determination of the ^{4}He electrostatic-energy-level structure in the near future.

It is thus rather remarkable that relatively little attention has been given to the electrostatic structure of the ^{3}He isotope. Because of the relatively large hyperfine interaction between the spin of the ^{3}He nucleus and the 1s inner electron, as the outer electron is promoted to higher values of n and l the inner electron is primarily coupled to the nucleus, and the singlet-triplet designations become progressively less pure. A number of years ago, Fred *et al.*[8] measured the ^{3}He/^{4}He isotope shift for a number of transitions allowed in emission. Similarly there were a few early measurements of magnetic fine structue in ^{3}He.[9] Recently, the technique of Doppler-free intermodulated fluorescence spectroscopy has been used to determine the $2^3P - 3^3D$ isotope shift with a precision of order 10 MHz.[10,11] As a continuation of this work, we decided to apply the anticrossing technique to determine the $n(^1D - ^3D)$ intervals for several n in ^{3}He. When combined with the previous ^{4}He results, one would have high precision isotope shifts for states whose wave functions were, in first order, identical.

We have observed the anticrossing spectra of the $n\,^1D$, $n\,^3D$ states for $n = 6, 7, 8$, and have derived from these measurements precise values for the electrostaic-energy separation between the singlet and triplet manifolds of these states. By electrostatic-energy separation we mean the energy separation after all corrections to the energy levels due to the magnetic coupling of the nuclear and the electronic spin are removed. The surprising result of this work is that we have observed a systematic difference in this electrostatic-energy separation between the ^{3}He value and the accepted value for ^{4}He which, within experimental uncertainty, is constant (~ 34 MHz) over the range of principal quantum numbers we have examined. We have investigated mass-dependent contributions to both the energy-level structure and the magnetic field tuning characteristics of the states of interest and have found no mechanism that can cause such a large difference in the electrostatic-energy separation of the singlet and triplet *D* states.

While our work was in progress, we received a preprint of the work of Derouard, Lombardi, and Jost,[12] which describes anticrossing measurements on the $n\,^1D$, $n\,^3D$ intervals of ^{3}He for $n=3-6$. Our experimental techniques are quite similar but our results are complementary, as our measurements determine the $n\,^1D$, ^3D ^{3}He intervals for $n=6-8$. The Derouard *et al.* measurements have the advantage that the individual Zeeman components are at least partially resolved for lower *n*. For our work at higher *n*, the anticrossing lines are narrower for both ^{3}He and ^{4}He and hence a more precise determination of the zero-field separations are possible for each isotope. The data point in common ($n=6$) provides a reference between laboratories on two separate continents, by which one can assay the probability of systematic error.

EXPERIMENTAL

The anticrossing appartus has been described in some detail previously.[1,2] Basically it consists of a source firing electron (100 eV, 1 mA) parallel to the magnetic field. The ^{3}He, at pressures of 15 mtorr, is excited by electron impact. Emission from a given excited state is selected by a monochrometer and monitored as a function of magnetic field. Test runs were made at different current and pressure conditions to check for current or pressure shifts. Absolute magnetic field measurements are made by an NMR probe before and after each data run.

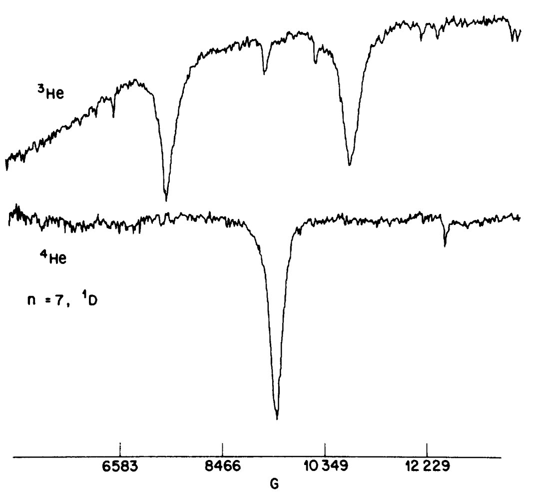

FIG. 1. Anticrossing spectrum for both ^{3}He (top) and ^{4}He *D* states. Spectra were obtained monitoring the 7^1D-2^1P emission. On both the ^{3}He and ^{4}He traces there are weak extraneous anticrossing signals that are not predicted from the magnetic tuning of the $n=7$ levels. These are presumably due to cascade resonances in other *n* levels.

A typical anticrossing spectrum for ^{3}He and ^{4}He is shown in Fig. 1. The corresponding anticrossing fields for $n=6-8$ are listed in Table I for ^{3}He. Separate values are given for the center of the anticrossing signal as observed on the singlet or triplet emmission. This is expected theoretically as each observed line is actually the sum of four components derived from the anticrossing of the individual pairs of Zeeman levels with different m_f values. As the signals from the individual anticrossings are weighted differently in the singlet or triplet emission, the center of the overall signal would be expected to differ. In fact, the observed differences are identical, well within experimental error, to the difference expected theoretically.

The indicated experimental errors in Tables I and II are the standard deviations of the measurements. The statistical error, 5 MHz, quoted on the ^{4}He separations in Table II, is about a factor of 4 smaller than previously quoted[1] for these anticrossings. Some of this difference is attributable to improved experimental techniques in the interim. However, much of the difference is the conscious neglect of systematic error in the present estimate. For example, while a point on the anticrossing curve can be found reproducibly to ±3 G, to quote

TABLE I. Observed ^{3}He anticrossing Fields (Gauss).

n	Anticrossing fields	
	singlet[a]	triplet[b]
6	14782(3)	14783(5)
7	9477(3)	9482(3)
8	6284(5)	. . .

[a] Center of anticrossing curve monitoring the singlet emmission.

[b] Center of anticrossing curve observed monitoring the triplet emmission. For $n=8$ no precision data was taken on the triplet due to the weakness of the signal.

TABLE II. ^{3}He /^{4}He $^1D-{}^3D$ separation, E_0 (MHz).

n	^{3}He	^{4}He	Diff.	^{4}He[a]	Diff.[a]
6	20885(5)	20907(5)	22	20919	34
7	13601(5)	13621(5)	20	13633	32
8	9296(8)	9318(8)	22	9333	37

[a] Currently accepted values for ^{4}He separations.

an absolute uncertainty of this size implies that the center of the anticrossing can be found to within approximately 1% of the observed anticrossing linewidth. (For the ^{3}He results where the linewidths are smaller, this requirement corresponds to 3% of the linewidth.) This is a difficult task and the final determination of the ^{4}He intervals systematically from the currently accepted values by about twice the quoted statistical experimental error.

DATA ANALYSIS

The positions of the anticrossings were calculated and compared with the observed positions, which resulted in the derived values, given in Table II, for the $n\,^1D$, $n\,^3D$ separations for $n=6$, 7, 8. The basis set of states used for this calculation was the decoupled basis $|L=2, M_L, S, M_S, I=\frac{1}{2}, M_I\rangle$. Both the orbital angular momentum and principal quantum numbers were assumed to be good quantum numbers. The Hamiltonian used to describe the atomic interactions can be written[13]:

$$H=H_0+H_{\rm fs}+H_{\rm hfs}+H_{\rm mag} \tag{1}$$

H_0 consists of the electron-electron and electron-nuclear interactions that give rise to the electrostatic structure in the absence of relativistic fine structure, hyperfine structure, and external field effects. This term is the Coulomb interaction with the inclusion of exchange. For the purpose of our calculation we take H_0 to be diagonal and a simple number, E_0, gives the separation of the $^1D-{}^3D$ levels in the absence of fine, hyperfine, or external field effects.

$H_{\rm fs}$ is the relativistic fine structure Hamiltonian that arises from the interaction of the orbital and electron spin angular momentum. We write this term as[13]

$$H_{\rm fs}=A\vec{\rm L}\cdot\vec{\rm S}+a\vec{\rm L}\cdot\vec{\rm K} \tag{2}$$
$$+b\frac{3(\vec{\rm L}\cdot\vec{\rm S})^2+\frac{3}{2}(\vec{\rm L}\cdot\vec{\rm S})-\vec{\rm L}^2\vec{\rm S}^2}{2S(2S-1)L(2L-1)}\ ,$$

where $\vec{\rm L}$ is the total orbital electronic angular momentum, $\vec{\rm S}$ is the total electronic spin angular momentum, and $\vec{\rm K}$ is the vector difference of the two electron spins. The last term gives the dipolar interaction between the electron's spin in the triplet state. For the purpose of the calculation the relativistic fine structure was assumed to be the same for both ^{3}He and ^{4}He as there is no theoretical reason for these constants to be different for the different isotopes.

The hyperfine interaction can also be written in its parametrized form as

$$H_{\rm hfs}=C\vec{\rm I}\cdot\vec{\rm L}+D\vec{\rm I}\cdot\vec{\rm X}+E\vec{\rm I}\cdot\vec{\rm S}+e\vec{\rm I}\cdot\vec{\rm K}\ , \tag{3}$$

where $\vec{\rm L}$, $\vec{\rm S}$, and $\vec{\rm K}$ have the same meaning as in Eq. (2), $\vec{\rm I}$ is the nuclear spin, and $\vec{\rm X}$ is the vector formed from $\vec{\rm S}$ and $\vec{\rm C}_2[=\sqrt{4\pi/3}\vec{\rm Y}_2]$. The Hamiltonian is written in the parametrized form to make it compatible with the choice of basis states. The most important terms in the hyperfine Hamiltonian are the last terms in Eq. 3 which comprise the diagonal and off diagonal Fermi contact term. These terms represent the interaction of the nuclear spin with the 1*s* core electron. There are good reasons why $E=e$ and we make that assumption: the value of E is independent of principal quantum number for high enough principal quantum number and orbital angular momentum since the interaction is independent of the Rydberg electron (as long as it is not in an *s* state). In fact it was shown in Ref. 10 that even for the 3^3D, E is within a few MHz of the assymptotic value of -4332 MHz.

The first two terms in the hyperfine Hamiltonian represent the interaction of the nuclear spin with the orbital and electronic spin angular momenta of the Rdyberg electron. Thus we expect C and D to have approximately a $(nl)^{-3}$ dependence. It was shown in Ref. 10 that $C=5\pm3$ MHz and $D=0\pm$ MHz for the 3^3D state. Thus for the states we are considering $nD\,(n=6, 7, 8)$ we can set $C=D=0$.

For the magnetic interaction we use the standard contributions:

$$H_{\rm mag}=\mu_0\vec{\rm B}\cdot(g_l\vec{\rm L}+g_s\vec{\rm S})-g_I\vec{\rm I}\mu_0\vec{\rm B}+\sum_i e^2\frac{(\vec{\rm B}\times\vec{\rm r}_i)^2}{8mc^2}\ , \tag{4}$$

where

$$\mu_0=\textit{Bohr magneton}=1.399\,60\ {\rm MHz/G}\ ,$$

$$g_l=(1-m/M)=0.999\,86(^4{\rm He}),0.999\,82(^3{\rm He})\ ,$$

$$g_s = 2.002\,32\ ,$$

$$g_I = 0.000\,162\ .$$

The final term is the diamagnetic interaction.

Using the above Hamiltonian and the basis states the energy structure of the n,D manifold was calculated as a function of the fine and hyperfine structure parameters and magnetic field using a computer program that diagonalized the complete matrix of the Hamiltonian including all basis states of a given L, n and spin multiplicity. Figure 2(a) illustrates the behavior of the $m_f = 0.5$, $n = 7$ D states as the magnetic field is varied. In the Hamiltonian in Eq. (4) there are no terms that mix states with different values of m_f (i.e., m_f remains a good quantum number) and thus we can make a similar energy diagram for each allowed value of m_f $(=-3.5,-2.5,-1.5,-0.5,0.5,1.5,2.5,3.5)$ with different values of m_f occurring at a slightly different magnetic field. Interestingly enough, for fields larger than 2 kG, the magnetic field has decoupled the angular momenta and each state is well represented by one of the states in the original basis set (except near an anticrossing).

At relatively low fields a wide anticrossing occurs which is due to the $\vec{I}\cdot\vec{K}$ interaction between the singlet and triplet states. Due to the excessive width of this anticrossing, it is not spectroscopically very useful.

At higher magnetic fields three narrower anticrossings are predicted. We immediately see from Fig. 1 that the ^{3}He anticrossing in the middle is much narrower than the other two, and it is this single anticrossing that we use for the purpose of obtaining a value for the electrostatic energy separation. The reason for this is that all parametrized constants other than E_0 have been measured in ^{3}He (Refs. 10 and 11) or measured[13] in ^{4}He and do not involve constants that change in going from one isotope to the other. Additionally these constants were used in explaining the results of a laser spectroscopy experiment in ^{3}He (Refs. 10 and 11) and did not cause any inconsistencies in the data analysis. In fact, the one parameter that the laser experiments of Refs. (10 and 11) were not sensitive to was E_0.

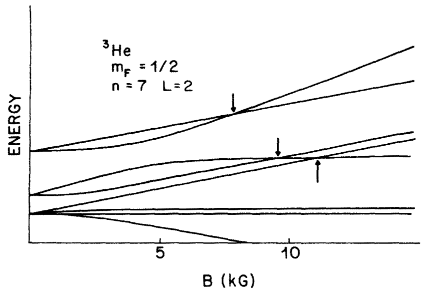

FIG. 2. Calculated energy-level structure of the $n = 7$, $L = 2$, $m_f = \frac{1}{2}$ states in ^{3}He. The arrows indicate the three high-field anticrossings that can be seen in Fig. 1.

In the anticrossing spectroscopy of ^{4}He described in Refs. 1 and 2 the spectrum was much simpler. ^{4}He has no hyperfine structure (the nucleus has zero spin) thus there is one anticrossing for each set of states with a particular value of m_j. It was this work that led to the determination of the $n(^3D - {}^1D)$ separation in ^{4}He and the associated measured values of the singlet-triplet mixing coefficient, a.

The anticrossing of interest is composed of four components with $m_f = -1.5, -0.5, 0.5$, and 1.5. Because the states are decoupled at the large magnetic fields required, each state involved in the anticrossing has a definite value of m_l (except at the crossing). The weighting factors from Table II in Ref. 2 are used for each of the components. The predicted position for each individual component is given by minimizing the energy separation between the two states involved in the anticrossing as a function of magnetic field.

From the comparison between the predicted and observed anticrossing positions we are able to determine a value of the electrostatic-energy separation, E_0, between the singlet and triplet states for either ^{3}He or ^{4}He. Of course the inference of the electrostatic-energy difference relies on our knowledge of the fine and hyperfine structure constants for helium. The previous information about the fine structure constants comes from measurements in ^{4}He.[1-4] The hyperfine structure constants for the 2^3P and the 3^3D states have been previously measured using a Doppler-free laser spectroscopic technique.[10] In that experiment, the ^{4}He fine structure constants and electrostatic-energy separation were used for the determination of the hyperfine constants of ^{3}He, and the observed spectroscopic line centers were fit with errors well within experimental uncertainty. The one parameter that the experiment was not particularly sensitive to was the elecrostatic singlet-triplet separation. The present experiment, in contrast, is very sensitive to this energy difference and thus the logical parameter to fit is the electrostatic singlet-triplet separation.

We determine this separation, E_0, by minimizing the absolute value of the difference between the calculated and the observed anticrossing position as a function of the electrostatic singlet-triplet separation. In the course of doing the experiment ^{4}He data was retaken for the same values of principal quantum number as was the ^{3}He data ($n=6$, 7, 8). This served several purposes; it tested the algorithm for fitting the data and it allowed a relative measurement between the ^{3}He and the ^{4}He systems to be made on the same experimental apparatus. The results of these measurements are shown in Table II.

The conclusion drawn from Table II is that the ^{3}He E_0 values are consistently measured to be 20 MHz less than those of ^{4}He for all three principal quantum numbers. Of some concern is that our ^{4}He values for the $^1D-{}^3D$ separation is approximately 12 MHz less than the previously reported numbers. The value for $n=7$ has, in fact, been measured to better than one MHz using the microwave-optical technique.[7] As we noted in the previous section, we believe that the ^{4}He value may well suffer from a systematic shift a factor of 2 larger, i.e., about 10 MHz, than the quoted experimental error. Nevertheless, we believe the systematic shifts in the apparatus to be identical for ^{3}He and ^{4}He so that the difference between the values for E_0 for ^{3}He and ^{4}He is significant. Finally we note that our derived values for the ^{3}He interval agrees extremely well (within 2 MHz) with that of Derouard *et al.* for $n=6$.

DISCUSSION

It is important here to emphasize that we are discussing the electrostatic-energy separation between the nominal ^1D and ^3D manifolds. This separation is what is left after all normal spin-dependent corrections to the energy levels have been removed. This is in contrast to recent theoretical calculations of so-called electrostatic-energy separations where the effects of nuclear spin on the energy levels is explicitly ignored.[14] Surprisingly enough, this anticrossing technique, in several important aspects, can be a more sensitive measurement of E_0 than optical or microwave methods that may have an intrinsic experimental precision $10-100$ times better. To illustrate this point we will consider the problem in second-order perturbation theory. The separation between a singlet state and a triplet state can be written as:

$$\Delta E = E_1 + \frac{H_{\text{od}}^2}{E_1} \, . \tag{5}$$

In Eq. (5) E_1 now contains E_0, the electrostatic-energy separation and the diagonal (within a particular spin multiplicity manifold) fine structure and hyperfine structure effects. The second term in Eq (5) gives the contribution of the off-diagonal interactions between the singlet and triplet manifolds. Note that it is the sum of the Hamiltonian terms that is squared and not the sum of the squares of the contributing interactions. The existence of cross terms was actually pointed out in Ref. 10 and was shown to give rise to the hyperfine structure in the ^1D states of ^{3}He.

For ^{3}He, $H_{\text{od}}=a\vec{\mathrm{L}}\cdot\vec{\mathrm{K}}+e\,\vec{\mathrm{I}}\cdot\vec{\mathrm{K}}$. The crucial point of this argument is that these off-diagonal matrix elements are not well measured (if they are measured at all) and a $1-5\%$ error in their value is not at all unreasonable. Although the effect of the fine and hyperfine structure within a particular spin multiplicity may be precisely accounted for to obtain E_0 from E_1, due to the uncertainty of the off-diagonal term is not so easy to extract E_1, or E_0 from the measured zero-field interval, ΔE. For instance, for $n=7$, a 1% uncertainty in the value of e ($=40$ MHz) can give rise to a 25 MHz in the second-order correction required to derive a value for E_0. This uncertainty is comparable to the measured ^{3}He-^{4}He shift in E_0.

The uncertainty of the off-diagonal contribution has no consequence in our anticrossing determination of E_0. To justify this statement we present the following simplified picture: Consider a two-level atomic system whose zero-field separation in the absence of any off-diagonal interaction is E_0. Assuming the two states tune relative to one another at a rate of $\mu_0 B$, (B is the magnetic field value) then the value of B at which the two states would cross is E_0/μ_0. Now we consider the additional interaction, H_p, between the two states; The separation of the pair of levels as a function of the magnetic field is just $[(E_0-\mu_0 B)]^2+4H_p^2]^{1/2}$. Taking the derivative of this expression with respect to the magnetic field and setting that quantity equal to zero to obtain the minimum value of the level separation shows that the minimum, i.e., the anticrossing field, is still at $B=E_0/\mu_0$. While the value of H_p determines the width of the avoided crossing it does not affect the position of the avoided crossing. (This argument is of course only true if H_p is independent of magnetic field.) Thus, since we measure the position of the anticrossing, our measurement is insensitive to uncertainties in the values of the off-diagonal matrix elements.

The largest contribution and most obvious term to examine for systematic differences between values of E_0 for the two isotopes, ^{3}He and ^{4}He, is the reduced mass corrections to the energy-level structure. This correction is simply known as the normal mass shift and depends only on the reduced mass of the particular atomic system and not on the electronic wave functions. We can write the change in energy separation between any two electronic states of a given isotope, due to the finite mass of the nucleus as[11]

$$\Delta E_{nms} = \frac{m}{M}\Delta T \,, \tag{6}$$

where m is the electron mass, M is the nuclear mass, and ΔT is the energy separation between the two states of interest. For ^{4}He and ^{3}He ${}^1D$ and ${}^3D$ states, we can write specifically,

$$\Delta E_{nms}({}^4\text{He}) = \frac{m}{M_4}\Delta T_4 \tag{7a}$$

and

$$\Delta E_{nms}({}^3\text{He}) = \left[\frac{m}{M_4}\right]\left[\frac{m+M_4}{m+M_3}\right]\Delta T_4 \,, \tag{7b}$$

where ΔT_4 is the ^{4}He ${}^1D - {}^3D$ energy separation and ΔT_3 is related to ΔT_4 through the finite mass dependence of the Rydberg. For the $n=6$, ${}^1D - {}^3D$ energy separation, we find ΔE_{nms} (^{4}He)$=2.8$ MHz and ΔE_{nms} (^{3}He)$=3.7$ MHz so the relative normal mass shift between the two isotopes is less than one MHz.

An additional isotope shift can arise from terms in the kinetic energy of the form $\sum_{i,k} \vec{p}_i \cdot \vec{p}_k / M$. These corrections must be determined using the calculated wave functions and give rise to what is called the specific mass shift. In the Hughes-Eckhart approximation this term gives a zero result for $1snl$ configurations of the helium atom. Better calculations are possible when more precise wave functions that include the effects of configuration mixing are used. To our knowledge these precise wave functions have not been calculated for helium D states,[15] but it seems unlikely that this correction would ever be more than ΔE_{nms}.

Other mass-dependent effects that can impact our results are in the linear Zeeman tuning g factor. Clearly a change in the g factor would effect how we extrapolate back to zero field and thus alter the derived value of E_0. The largest of these effects is the simple $(1-m/M)$ mass correction to g_l which is incorporated in our analysis [Eq. (4)]. Moreover, the difference in linear tuning between ^{3}He and ^{4}He $=[g_1({}^4\text{He}) - g_l({}^3\text{He})]\mu_0 B$ is only 0.5 MHz at 10 000 Gauss and thus is small compared to the discrepancy described in the last section.

In conclusion, we have measured an anomolous shift between the derived values for the electrostatic-energy separation of the $n=6$, 7, $8\,{}^1D - {}^3D$ states in ^{3}He and ^{4}He. We have considered known mass-dependent effects in both the energy structure and the magnetic field tuning parameters of these atomic isotopes. These effects do not appear to be large enough to account for the observed differences.

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