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HYPERFINE STRUCTURES OF THE nd $^1D(n = 3-8)$ STATES OF ^3He I

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We have used the beam-foil quantum beat method to measure the hyperfine structure separations F = 3/2 − 5/2 of the 1snd ^1D states (n = 3 − 8) of ^3He I. We observed the single frequency modulated decay curves of the 1s2p ^1P - 1snd ^1D transitions for times after excitation up to 50 ns, corresponding to 4-5 modulation periods. The frequencies obtained (with a precision of 2-5%) are compared with other experiments and theory. The frequencies are determined mainly by the singlet-triplet energy separations and mixing factors for the He I D-states. The results agree with the same parameters obtained from other recent level-crossing measurements in strong magnetic field mixing of the singlet-triplet states.

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1. Introduction

The quantum-beat method of beam-foil spectroscopy is, by now, a well established technique for measuring fine and hyperfine structure in selected atoms and ions [1]. We have employed this method, without external fields, to measure the hyperfine structures of the 1snd ^{1}D states of ^{3}He I for principal quantum numbers, n, from 3 to 8.

It has been known for some time that the hyperfine separation of the F = 3/2 - 5/2, for any of these ^{1}D states, is about two orders of magnitude larger than one might initially estimate [2,3]. This large perturbation of the hyperfine structure is caused by singlet-triplet mixing in the fine-structure segment of the Hamiltonian. Thus, a measurement of the hyperfine splitting yields a sensitive determination of this mixing parameter.

Recently, high precision experiments on ^{3}He have been performed using the magnetic field anticrossing technique [4] and Doppler-free two-photon spectroscopy [5]. These experiments are able to measure accurately many of the intervals in the D manifold for principal quantum numbers 3-6. When such results are combined with the exhaustive information available for the D manifold of ^{4}He [6] a rather complete picture for the eight energy levels comprising this manifold in ^{3}He is able to emerge.

We shall combine the best available results into a semi-empirical calculation of the energy levels of ^{3}He, $\ell = 2$, $n = 3 - 10$. Our quantum-beat measurements will then be compared to the results of this calculation. Close examination of the ^{1}D hyperfine splittings as a function of principal quantum number will reveal a curious feature which our measurements at n = 7 and 8 help to confirm.

2. Theory

One can write the Hamiltonian for neutral helium in a 1snd configuration as

$$H = H_0 + H_{FS} + H_{HF} \tag{1}$$

where H_0 operating upon any state of this configuration yields the mean energy of the ^3D term which will be set to zero.

The fine-structure Hamiltonian, H_{FS}, can be found in the comprehensive review by Miller and Freund [7]. The matrix elements for the fine structure, in the absence of hyperfine structure, are listed in table 1. Note that if the fine structure intervals are measured from their mean value (see fig. 1), the diagonal entries are considerably simplified. The singlet-triplet mixing parameter, α, is the same as that used by others [4,7]. The remaining parameters are defined in fig. 1. Two clarifying points should be made here. The first is that all matrices in the paper shall be restricted to the D manifold; that is, the total orbital angular momentum is taken to be a good quantum number. The second is that the entries for d_2 and D have been adjusted such that after diagonalizing the fine-structure Hamiltonian alone, the best experimental results for these intervals are reproduced. d_1 and d_3 are not affected by singlet-triplet mixing.

The hyperfine-structure Hamiltonian, H_{FS}, has been taken from Lurio, Mandel and Novick [8]. Besides the Fermi contact term (A) which is independent of principal quantum number, the Hamiltonian includes magnetic dipole matrix elements (a). Higher multipole moments are zero. Since the total angular momentum, F, is a good quantum number, the matrix for the total Hamiltonian is block diagonal yielding two single-entry matrices for F = 1/2 and 7/2 and two 3 × 3 matrices for F = 3/2 and 5/2. These matrices are given in table 2. For ℓ = 2, the connection between the hyperfine parameters of

ref. 8 and those of table 2 are: $a_s = 2A$; $a_{3/2} = a$; $a_{5/2} = (3/7)\, a_{3/2}$; $C_1 = (2/5)^{1/2}$; $C_2 = (3/5)^{1/2}$; $\xi = 1$.

All of the needed parameters for the matrix elements of the total Hamiltonian for n = 3 - 10 are listed in table 3. The values in parenthesis under d_2 and D are the numbers after diagmalizing the fine-structure Hamiltonian which have been taken from experimental results. Values for the singlet-triplet mixing parameter, α, are based upon a n^{-3} fit to the experimental values given by Derouard et al. [4]. The result is:

$$\alpha = 17527.3 \text{ MHz}/n^3. \qquad (2)$$

3. Experiment

$^3He^+$ ions from Argonne's 120 kV electrostatic accelerator were employed in a standard beam-foil configuration utilizing side-on viewing [9]. The beam energy was adjusted between 60 and 110 kV to insure a minimum of three complete beats for the lowest frequencies measured. The beam velocity was carefully calibrated over the energy range utilized against the well known fine-structure quantum beat of 388.9 nm in ^{4}He I. Thin (5 $\mu g/cm^2$) self-supporting carbon foils were used and the energy loss in the foils was considered for both calibration and data acquisition.

Radiation from the beam first passed through a horizontally oriented polarizer to maximize the ratio of beat amplitude to intensity. It then passed through a 3/4 m Spex monochromator and was detected by one of two cooled photomultipliers used in this experiment. Standard photon counting techniques were employed.

Data were acquired using beam current normalization by summing multiple scans of the foil with each scan composed of approximately 120 points. Figure 2 shows a typical data sample. For analysis, a direct non-linear least-squares procedure which simultaneously adjusted the quantum-beat

parameters (frequency, amplitude and phase) and decay curve parameters (amplitude, lifetime and residual constant) was employed.

Quoted uncertainties of our results are one standard deviation and represent uncertainties arising from the beam-velocity determination, the computer estimated statistical uncertainties of a particular run and the scatter of results from multiple runs.

4. Results

Table 4 shows the results of our calculation for the energy levels of ^{3}He I, 1s nd for n = 3 to 10. The numbers differ somewhat from those of Derouard et al. [4] mostly because of our choice of fine structure experimental results. However, if one compares our intervals to the Doppler-free two-photon results of Biraben et al. [5], agreement is at least as good as those of Derouard et al.

In table 5, our experimental results for the hyperfine splitting of the 1s nd $^1D_2$(3/2 - 5/2) levels are presented along with the theoretical results of several workers. Agreement with the calculation presented here is excellent.

Next, consider the above splitting as a function of principal quantum number. Figure 3 displays a smooth curve drawn through the theoretical values obtained in this work. Note the inflection points at n = 6 and 9. This behavior is unexpected in light of the fact that all but one interaction are decreasing approximately as $1/n^3$. The experimental points closely follow the theoretical curve but clearly high precision results for n > 6 would be desirable.

5. Conclusion

A semi-empirical calculation of the hyperfine structure in the 1snd states of ^{3}He I for n = 3 to 10 has been presented. In addition, direct quantum-beat measurements of the $^1D_2$(3/2 - 5/2) interval have been performed for n = 3 to 8. While our precision for lower n values cannot match that for measurements performed using other techniques, ours are the only such measurements for n = 7 and 8.

We have pointed out the behavior of the hyperfine structure of the singlet level of ^{3}He I as a function of principal quantum number. Recently an experiment has been performed to measure the hyperfine structure of the 5snd singlet and triplet states of ^{87}Sr I [13]. In that experiment a surprising enhancement of the splitting in the singlet states was observed around n = 16. Though singlet-triplet mixing certainly seems to be an important factor, no theoretical explanation for this behavior has yet been advanced. Similarities between that system and the one presented here indicate that a detailed understanding of the hyperfine structure of ^{3}He I may be instrumental in explaining such a newly observed phenomenon.

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Table 1. Fine-structure matrix for 1s nd He I

S J	1 1	1 2	0 2	1 3
1 1	d_1	0	0	0
1 2	0	d_2	$\sqrt{6}\alpha$	0
0 2	0	$\sqrt{6}\alpha$	D	0
1 3	0	0	0	d_3

Table 2. Matrix elements of the total Hamiltonian for 1s nd ^{3}He I

J	F	1 1/2	3 7/2
1	1/2	$d_1 + \frac{A}{2} - \frac{5}{4}a$	0
3	7/2	0	$d_3 + \frac{A}{2} + \frac{15}{28}a$

J	F	1 3/2	2 3/2	2' 3/2
1	3/2	$d_1 - \frac{A}{4} + \frac{5}{8}a$	$\frac{3}{4}A - \frac{15}{32}a$	$-\sqrt{6}\left(\frac{A}{4} - \frac{15}{32}a\right)$
2	3/2	$\frac{3}{4}A - \frac{15}{32}a$	$d_2 - \frac{A}{4} - \frac{15}{16}a$	$\sqrt{6}\left(\frac{A}{4} + \frac{5}{64}a + \alpha\right)$
2'	3/2	$-\sqrt{6}\left(\frac{A}{4} - \frac{5}{64}a\right)$	$\sqrt{6}\left(\frac{A}{4} + \frac{5}{64}a + \alpha\right)$	$D - \frac{15}{16}a$

J	F	2 5/2	3 5/2	2' 5/2
2	5/2	$d_2 + \frac{A}{6} + \frac{5}{8}a$	$\frac{\sqrt{14}}{2}\left(\frac{A}{3} - \frac{15}{112}a\right)$	$\frac{-1}{\sqrt{6}}\left(A + \frac{5}{16}a - 6\alpha\right)$
3	5/2	$\frac{\sqrt{14}}{2}\left(\frac{A}{3} - \frac{15}{112}a\right)$	$d_3 - \frac{2}{3}A - \frac{5}{7}a$	$\frac{\sqrt{21}}{6}\left(A - \frac{5}{56}a\right)$
2'	5/2	$\frac{-1}{\sqrt{6}}\left(A + \frac{5}{16}a - 6\alpha\right)$	$\frac{\sqrt{21}}{6}\left(A - \frac{5}{56}a\right)$	$D + \frac{5}{8}a$

Primes denote singlet states.

Table 3. Parameters used for matrix evaluation

n	d_1	d_2	d_3	D	α	a
3	1095.5	-205.3(-230.0)	-305.2	102091.3(102116)	649.2	-1.61
4	460.8	-86.4(-94.0)	-130.4	59009.4(59017)	273.9	-0.68
5	235.8	-44.1(-47.5)	-67.1	34022.5(34026)	140.2	-0.35
6	136.4	-25.5(-27.4)	-38.9	20917.3(20919.2)	81.1	-0.20
7	85.9	-16.0(-17.2)	-24.6	13632.2(13633.3)	51.1	-0.13
8	57.6	-10.7(-11.5)	-16.5	9331.9(9332.7)	34.2	-0.08
9	40.4	-7.5(-8.1)	-11.6	6650.0(6650.5)	24.0	-0.06
10	29.5	-5.5(-5.9)	-8.4	4897.8(4898.1)	17.5	-0.04

All values in MHz.

Numbers in parenthesis are after diagonalizing fine-structure matrix (see text).

Table 4. Hyperfine energy levels for ^{3}He I 1s nd states

3d	4d	5d	6d	F
102315.4	59315.1	34531.1	21765.3	5/2
102176.3	59210.4	34435.1	21673.2	3/2
4765.4	4332.8	4020.1	3633.9	3/2
3843.9	3912.0	3764.7	3450.3	5/2
-1068.9	-1704.7	-1930.2	-2029.7	1/2
-1791.7	-1992.1	-2074.2	-2112.2	3/2
-2413.0	-2268.4	-2218.3	-2196.3	5/2
-2472.5	-2297.1	-2233.7	-2205.4	7/2

7d	8d	9d	10d	F
14992.7	11370.8	9441.4	8394.9	5/2
14906.7	11297.3	9384.7	8353.6	3/2
3094.0	2391.2	1614.7	889.1	3/2
2950.5	2279.3	1531.1	828.2	5/2
-2080.3	-2108.8	-2125.9	-2136.9	1/2
-2132.0	-2143.3	-2150.1	-2154.5	3/2
-2185.2	-2179.0	-2175.2	-2172.8	5/2
-2191.0	-2182.9	-2178.0	-2174.9	7/2

All values in MHz.

Table 5. Comparison of theory and experiment for ^{3}He I nd ^{1}D hyperfine splittings

n	Theory					This Expt.
	Bessis et al.[a] (corrected)	Descoubes[b]	Liao[c] et al.	Derouard[d] et al.	This Work	
3	144	138	138.4	139.2	139.1	140.9 ± 3.2
4	105	102	102.6	104.7	104.7	105.2 ± 2.0
5	96	93.8	93.8	95.8	96.0	96.5 ± 2.5
6	92	89.7	90.8	92.0	92.1	88.9 ± 2.0
7	86	118	85.3	—	86.0	84.2 ± 2.3
8	75	—	—	—	73.5	77.5 ± 2.8
9	—	—	—	—	56.7	—
10	—	—	—	—	41.3	—

All values in MHz.

[a] Rev. 3; corrected for more recent ^{1}D - ^{3}D separation.

[b] Ref. 11

[c] Ref. 12

[d] Ref. 4

Figure Captions

Fig. 1 Energy level diagram of 1s nd ^{3}He I. Scale is approximate.

Fig. 2 Quantum-beat data of 1s 6d $^1D_{3/2 - 5/2}$ ^{3}He I.
Measured frequency of this run is 90.4 MHz.

Fig. 3 Hyperfine frequency of 1s nd $^1D_{3/2 - 5/2}$. Smooth curve is a graphical interpolation of theoretical results presented here.

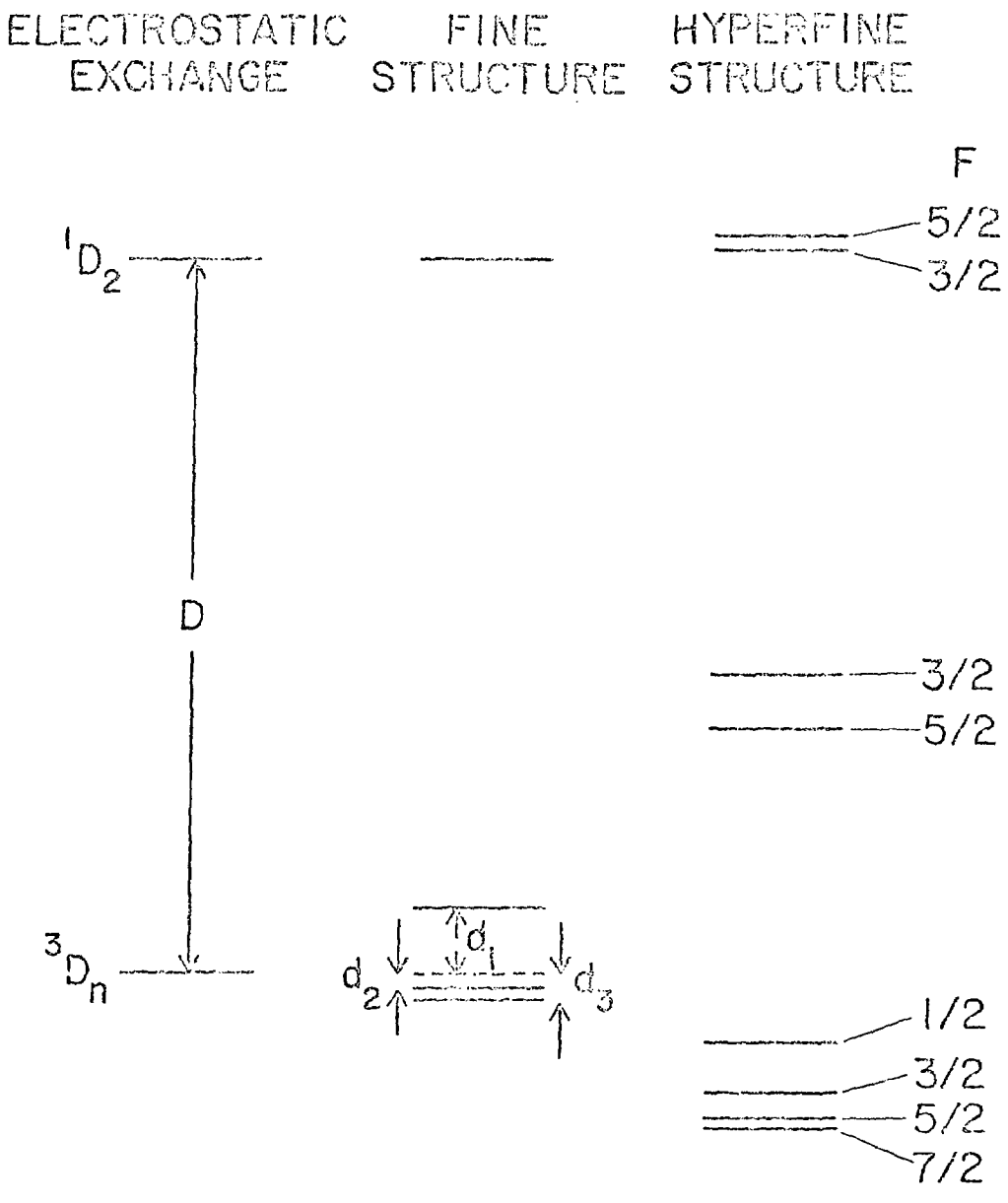
ELECTROSTATIC EXCHANGE
FINE STRUCTURE
HYPERFINE STRUCTURE
F
$^1D_2$
5/2
3/2
D
3/2
5/2
^3D_n
d_1
d_2
d_3
1/2
3/2
5/2
7/2

Fig. 1

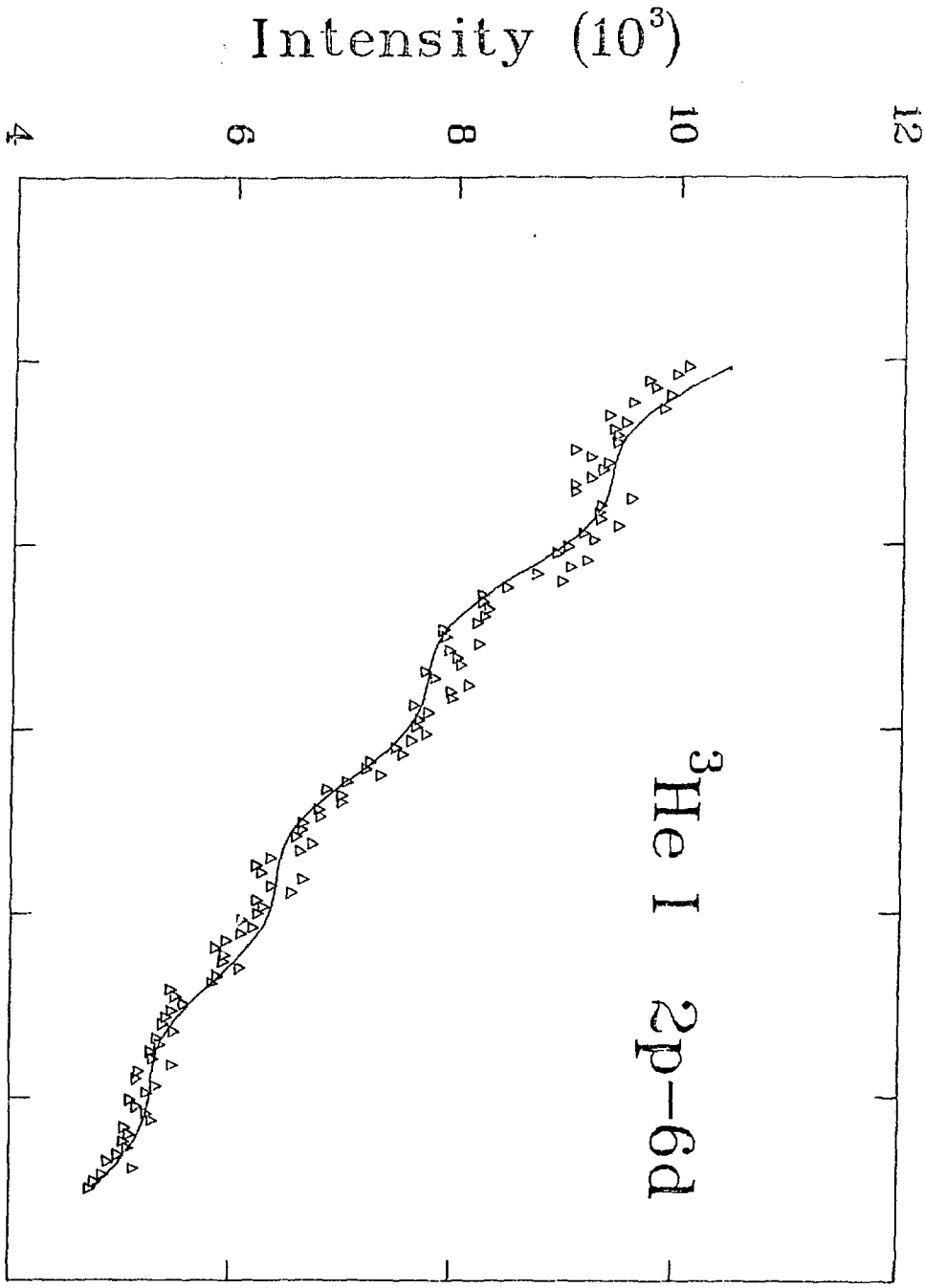

Fig. 2

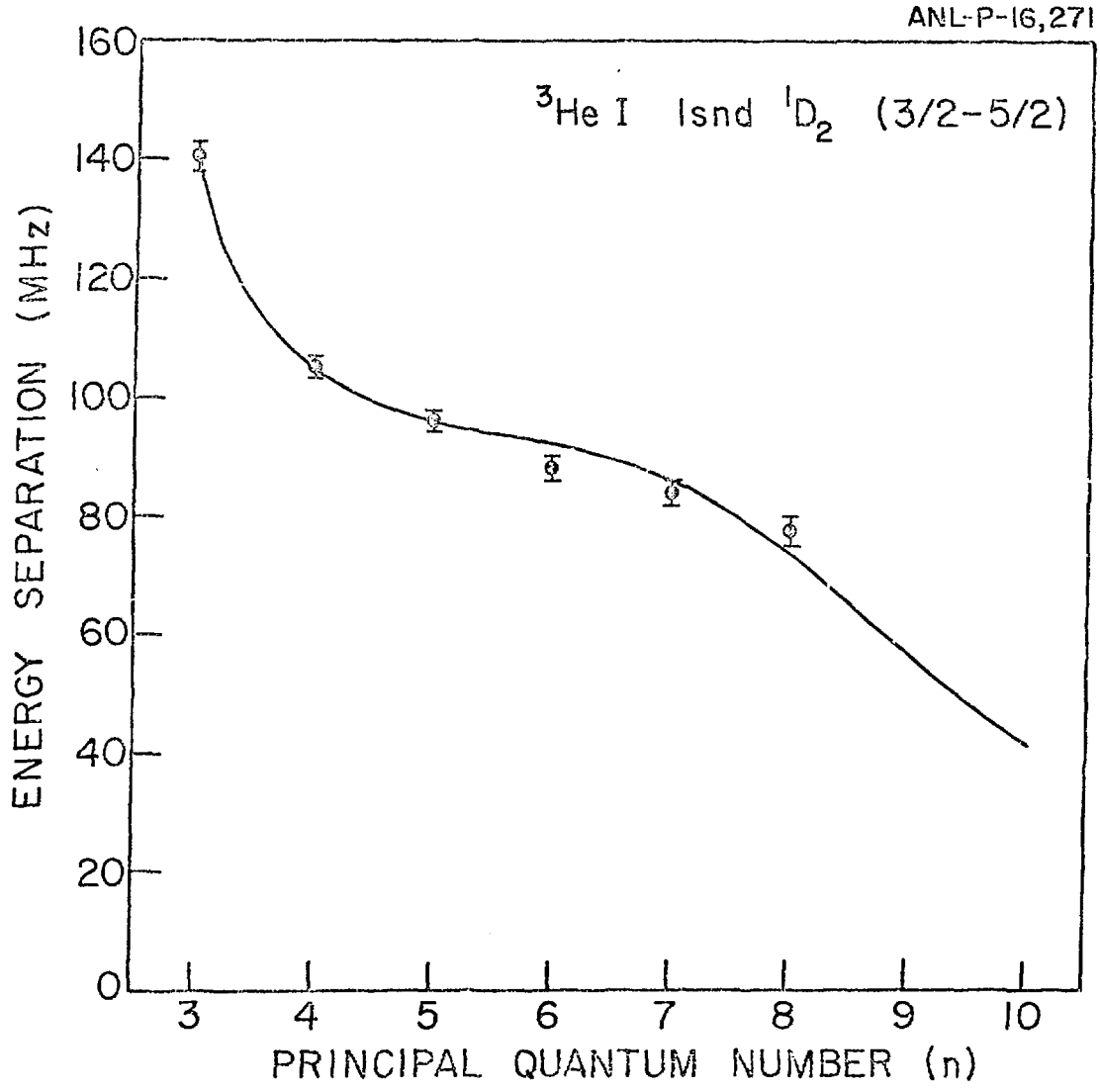
ANL-P-16,271
^{3}He I 1snd $^1D_2$ (3/2-5/2)
ENERGY SEPARATION (MHz)
160
140
120
100
80
60
40
20
0
3
4
5
6
7
8
9
10
PRINCIPAL QUANTUM NUMBER (n)

Fig. 3