PHYSICAL REVIEW VOLUME 135, NUMBER 4A 17 AUGUST 1964

Calculation of the Hyperfine Splittings of the ^1D Levels of He^3

N. Bessis, H. Lefebvre-Brion, and C. M. Moser
Centre de Mécanique Ondulatoire Appliquée, Paris, France

(Received 30 March 1964)

The He^3 hyperfine splittings of the ^1D levels are shown to arise from the admixture of ^3D states through the total spin-orbit interaction and the magnetic interaction between the nucleus and the 1*s* electron (Fermi contact term). The results of the calculation using hydrogen-like functions are in qualitative agreement with the splittings observed recently by B. Decomps *et al.* for the 4, 5, and 6 ^1D levels (60–80 Mc/sec). The repulsion between ^1D and ^3D levels is also sufficient to explain the "specific" isotope shift observed for ^1D levels of helium.

I. INTRODUCTION

RECENTLY, a magnetic resonance experiment performed on He^3 excited by electron bombardment has revealed the presence of a hyperfine structure for the ^1D levels.[1] This hyperfine structure is rather surprising. In singlets the only interaction which takes place is the magnetic interaction of the nucleus with the outer electron and for these ^1D states this would give splittings of about 1 Mc/sec, whereas the experimental values deduced for zero field are between 60 and 80 Mc/sec.

This hyperfine structure may be due to the fact the observed ^1D levels are not to be represented by pure singlet wave functions but are in fact an admixture of singlet and triplet wave functions. In a study of the triplet intervals of He^4, Araki[2] has already shown that there is an admixture of singlet and triplet states brought about by the spin-orbit coupling operator. In this paper, we shall show that in He^3, admixture is brought about not only by the spin-orbit interaction but also by the Fermi contact term and this makes it possible to explain the observed hfs.

II. CALCULATIONS

The wave functions Ψ_F which represent the F levels ($F=\frac{5}{2},\frac{3}{2}$) of the $n\,^1D$ state are taken to be linear combinations of singlet and triplet wave functions associated with the configuration (*1s*,*nd*). As the hfs splitting of the triplet state is at least of the same order of magnitude as the fine structure separation, J is no longer a good quantum number. For each value of F, the singlet wave function $\psi_{JF}(J=2)$ will admix with the triplet functions φ_{JF}, where J takes the two values $J=F\pm1/2$ corresponding to the two possible orientations of the nuclear spin ($I=1/2$). Thus, we have the following representation of the wave functions of the two levels of the $n\,^1D$ state:

$$\Psi_{5/2}(^1D)=a_0\psi_{2,5/2}+a_1\varphi_{3,5/2}+a_2\varphi_{2,5/2}$$
$$\Psi_{3/2}(^1D)=b_0\psi_{2,3/2}+b_1\varphi_{2,3/2}+b_2\varphi_{1,3/2}.$$

The hyperfine splitting ΔE of the singlet state is given by

$$\Delta E=\langle\Psi_{5/2}|H|\Psi_{5/2}\rangle-\langle\Psi_{3/2}|H|\Psi_{3/2}\rangle, \qquad (1)$$

with $H=H_{L,S}+H_J+H_F$, where $H_{L,S}+H_J$ is the Breit's Hamiltonian, H_J contains the spin-orbit and spin-spin interactions (see Appendix), H_F is the magnetic interaction of the nucleus and the *1s* electron, that is to say the Fermi contact term (the magnetic interaction with the outer electron is negligible).

In calculating ΔE [Eq. (1)], the only approximation we make is to replace the quantity

$$E_0(n\,^1D)-E_0(n\,^3D)=\langle\psi(n\,^1D)|H_{L,S}|\psi(n\,^1D)\rangle -\langle\varphi(n\,^3D)|H_{L,S}|\varphi(n\,^3D)\rangle \qquad (2)$$

by the observed He^4 interval for $^1D-{}^3D$ terms.[3]

The matrix elements of each operator in H_F and H_J are expressed as a product of an angular coefficient and a reduced matrix element (see Appendix). Blume and Watson's[4,5] expressions of the spin-other-orbit and spin-spin interactions have been used to calculate the reduced matrix elements which are, finally, proportional to radial integrals.

As the wave functions used in this paper are linear combinations of Slater determinants built on the single configuration (*1s*,*nd*) the only radial integrals which come into the calculation are:

(a) For the spin-orbit interaction

$$\mathfrak{z}_0(nd)=(\alpha^2/2)Z\langle nd|r^{-3}|nd\rangle,$$

where α and Z are, respectively, the fine structure constant and the charge of the nucleus.

(b) For the spin-other orbit

$$\mathfrak{z}_1(nd)=(\alpha^2/2)\langle nd(r_1)1s(r_2)|r^{-3}\epsilon(r_1-r_2)|nd(r_1)1s(r_2)\rangle$$
$$\epsilon(x)=1 \quad \text{if} \quad x>0$$
$$=0 \quad \text{if} \quad x<0,$$

$$\mathfrak{z}_2(nd)=-\frac{\alpha^2}{2}\left\langle nd(r_1)nd(r_2)\left|\frac{r_<}{r_>{}^4}r_2\frac{\partial}{\partial r_1}\right|1s(r_1)1s(r_2)\right\rangle. \qquad (3)$$

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[1] (a) B. Decomps, thesis, Paris, 1960 (unpublished); (b) M. Descoubes, B. Decomps, and J. Brossel, Compt. Rend. **258**, 4005 (1964).

[2] G. Araki, Proc. Phys. Math. Soc. Japan **19**, 128 (1937); see also, P. Pluvinage J. Phys. Radium **13**, 405 (1952).

[3] C. E. Moore, *Atomic Energy Levels* (National Bureau of Standards, Washington, D. C., 1949).

[4] M. Blume and R. E. Watson, Proc. Roy. Soc. (London) **270**, 127 (1962).

[5] M. Blume and R. E. Watson (private communication); cf. also H. Horie, Progr. Theoret. Phys. (Kyoto) **10**, 296 (1953).

TABLE I. Matrix elements of H.

$$\langle\psi_{2\,\frac{5}{2}}|H|\psi_{2\,\frac{5}{2}}\rangle=E_0(n\,{}^1D)$$
$$\langle\psi_{2\,\frac{5}{2}}|H|\varphi_{3\,\frac{5}{2}}\rangle=[-(7)^{1/2}/\sqrt{3}]\alpha_s$$
$$\langle\psi_{2\,\frac{5}{2}}|H|\varphi_{2\,\frac{5}{2}}\rangle=(-\sqrt{3}/\sqrt{2})(\mathfrak{z}_0+\mathfrak{z}_1)+(\sqrt{2}/\sqrt{3})\alpha_s$$
$$\langle\varphi_{3\,\frac{5}{2}}|H|\varphi_{3\,\frac{5}{2}}\rangle=E_0(n\,{}^3D)+\mathfrak{z}_0-3\mathfrak{z}_1-(6/5)\mathfrak{z}_2+(2/7)\mathfrak{z}_1-\tfrac{4}{3}\alpha_s$$
$$\langle\varphi_{3\,\frac{5}{2}}|H|\varphi_{2\,\frac{5}{2}}\rangle=[(14)^{1/2}/3]\alpha_s$$
$$\langle\varphi_{2\,\frac{5}{2}}|H|\varphi_{2\,\frac{5}{2}}\rangle=E_0(n\,{}^3D)-\tfrac{1}{2}(\mathfrak{z}_0-3\mathfrak{z}_1)+\tfrac{3}{5}\mathfrak{z}_2-\mathfrak{z}_1+\tfrac{1}{3}\alpha_s$$

$$\langle\psi_{2\,\frac{3}{2}}|H|\psi_{2\,\frac{3}{2}}\rangle=E_0(n\,{}^1D)$$
$$\langle\psi_{2\,\frac{3}{2}}|H|\varphi_{2\,\frac{3}{2}}\rangle=(-\sqrt{3}/\sqrt{2})(\mathfrak{z}_0+\mathfrak{z}_1+\alpha_s)$$
$$\langle\psi_{2\,\frac{3}{2}}|H|\varphi_{1\,\frac{3}{2}}\rangle=(\sqrt{3}/\sqrt{2})\alpha_s$$
$$\langle\varphi_{2\,\frac{3}{2}}|H|\varphi_{2\,\frac{3}{2}}\rangle=E_0(n\,{}^3D)-\tfrac{1}{2}(\mathfrak{z}_0-3\mathfrak{z}_1)+\tfrac{3}{5}\mathfrak{z}_2-\mathfrak{z}_1-\tfrac{1}{2}\alpha_s$$
$$\langle\varphi_{2\,\frac{3}{2}}|H|\varphi_{1\,\frac{3}{2}}\rangle=\tfrac{3}{2}\alpha_s$$
$$\langle\varphi_{1\,\frac{3}{2}}|H|\varphi_{1\,\frac{3}{2}}\rangle=E_0(n\,{}^3D)-\tfrac{3}{2}(\mathfrak{z}_0-3\mathfrak{z}_1)+(9/5)\mathfrak{z}_2+\mathfrak{z}_1-\tfrac{1}{2}\alpha_s$$

(c) For the spin-spin interaction, only $\mathfrak{z}_2(nd)$.

(d) For the Fermi contact term

$$\alpha_S=(8\pi/3)\beta_e\beta_N(1s)^2_{r=0},$$

where β_e and β_N are, respectively, the magnetic moments of the electron and of the nucleus.

An automatic program has been written for the IBM-704 computer by one of us (N.B.) to calculate $\mathfrak{z}_1$ and $\mathfrak{z}_2$.

The orbitals used in the calculation are the hydrogen-like $1s$ function with $Z=2$ and d functions with $Z=1$. The expressions of the matrix elements of H in terms of the radial integrals and the numerical values of these integrals are given, respectively, in Tables I and II.

TABLE II. Numerical values of parameters (in Mc/sec).

n	α_s[a]	$\mathfrak{z}_0$[b]	$\mathfrak{z}_1$	$\mathfrak{z}_2$	$E_0(n\,{}^1D)-E_0(n\,{}^3D)$
3	−2166	866	430	14	10^5
4	−2166	365	181	8	5.64×10^4
5	−2166	187	93	5	3.18×10^4
6	−2166	108	54	3	1.95×10^4
7	−2166	68	34	...	0.93×10^4
8	−2166	46	23	...	0.93×10^4

[a] $(8\pi/3)\beta_e\beta_N=-850.5773a_0^3$ Mc/sec.
[b] $\alpha^2/2=175\,184.7a_0^3$ Mc/sec.

In Table III, we give the values of $\Delta E(n\,{}^1D)$ obtained from Eq. (1) by diagonalizing matrices for $F=5/2$ and $F=3/2$. [The numerical values for $\Delta E(3\,{}^1D)$ and $\Delta E(4\,{}^1D)$ are not modified if the diagonal matrix elements of H_J are replaced by the He^4 observed triplet intervals.[6]]

The hyperfine splittings of the ^1D levels thus appears because the $F=3/2$ and $F=5/2$ levels are unequally pushed apart by the corresponding levels of the triplet. The level $F=5/2$ is shifted more than the level $F=3/2$.

TABLE III. Hyperfine splittings of ^1D states (in Mc/sec).

n	Calculation without admixture	Calculation with admixture	Experiment[a]
3	2	146±3[b]	...
4	1	110±3	|60|±20
5	~0	103±5	|65|±10
6	~0	98±5	|80|±?
7	~0	110±8	...
8	~0	75±5	...

[a] See Ref. 1(b).
[b] This error has been calculated in assuming an error of 0.05 mK on the experimental value of $E_0(n\,{}^1D)-E_0(n\,{}^3D)$.

From the experimental results now known[1] only the absolute value of ΔE has been determined. The values which we have calculated are in reasonably good qualitative agreement with experiment.

The shifts of each of the ^1D levels are given in Table IV as the difference

$$\langle\Psi_F|H|\Psi_F\rangle-E_0(n\,{}^1D),$$

where $E_0(n\,{}^1D)$ is the observed height of the singlet in He^4. The shifts increase very rapidly with n as the distance $E_0(n\,{}^1D)-E_0(n\,{}^3D)$ diminishes.

TABLE IV. Shift of ^1D states.

	Calculated shift by admixture in				"Specific" isotope shift	
	Mc/sec		(mK)[a]		(mK)	
n	$F=\frac{5}{2}$	$F=\frac{3}{2}$	$F=\frac{5}{2}$	$F=\frac{3}{2}$	[b]	[c]
3	233	86	7	3	2	−2
4	321	211	10	7	7	7
5	548	445	16	15	14	14
6	918	820	31±3	28±2	24	39
7	2061	1951	69±10	65±10	41	36
8	2046	1971	68±10	66±10	53	53

[a] Cf. Ref. b of Table III for the assumed error.
[b] See Ref. 7.
[c] L. C. Bradley and H. Kuhn, Proc. Roy. Soc. (London) **A209**, 325 (1951).

The singlet-triplet repulsion, which had been alluded to by Fred, Tomkins, Brody, and Hamermesh,[7] also gives an explication to the disagreement which exists between the experimental value of the isotope shift of the ^1D levels and the value which is calculated only considering the normal isotope shift. This difference, shown in columns 6 and 7 of Table IV, which certain authors[8] have attributed to an anormal specific isotope shift, is of the same order of magnitude as the calculated shift (columns 4 and 5).

The singlet-triplet admixture of course will have an effect on the calculated hyperfine structure of the $n\,{}^3D$

TABLE V. Hyperfine structure of ^3D states (in mK).

Levels	Calculation without admixture[a]	Calculation with admixture	Experiment[a]
$3\,{}^3D_{1\frac{3}{2}-1\frac{1}{2}}$	193	194	191.7±2
$3\,{}^3D_{1\frac{3}{2}-3,7/2,}$	241	241	243.5±10
$4\,{}^3D_{1\frac{3}{2}-1\frac{1}{2}}$	204	201	~202

[a] See Ref. 6. Note that $\langle{}^3D_{1,1/2,}|H_F|{}^3D_{1,1/2,}\rangle=\langle{}^3D_{3,7/2,}|H_F|{}^3D_{3,7/2,}\rangle=\alpha_s$.

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[6] H. Chantrel, Ann. Phys. (Paris) 4, 965 (1959).

[7] M. Fred, F. S. Tomkins, J. K. Brody, and M. Hamermesh, Phys. Rev. **82**, 406 (1951).

[8] G. Araki, K. Mano, and M. Ohta, Phys. Rev. **115**, 1222 (1959).

states. But as the hfs has only been measured optically[6] the experimental error is of the same order of magnitude as the difference between the hyperfine structure calculated with and without admixture of the ^1D states (cf. Table V).

ACKNOWLEDGMENTS

Our thanks are due to Professor J. Brossel and Professor A. Kastler for bringing to our attention the experimental work. We are particularly grateful to B. Decomps for communicating to us the results of his experiments before publication and for many stimulating discussions.

APPENDIX

The factorization of the matrix elements of different operators is given by the general formula[9]:

$$\langle LS'J'IF|\mathcal{T}^{(K)}\cdot[T^{(k_L)}\times T^{(k_S)}]^{(K)}|LSJIF\rangle_{M_F=F}$$

$$=(-1)(2K+1)^{1/2}(2F+1)^{1/2}\begin{Bmatrix} J' & J & K \\ I & I & K \\ F & F & O \end{Bmatrix}$$

$$\times[(2J+1)(2J'+1)(2K+1)]^{1/2}\begin{Bmatrix} L & L & k_L \\ S' & S & k_S \\ J' & J & K \end{Bmatrix}$$

$$\times\langle I||\mathcal{T}^{(K)}||I\rangle\langle LS'||T^{(k_L)}\times T^{(k_S)}||LS\rangle.$$

$\mathcal{T}^{(K)}$ notes a tensor operator of rank K working only on nuclear coordinates; $T^{(k_L)}$ and $T^{(k_S)}$ are tensor operators of rank k_L and k_S working, respectively, on space and spin electron coordinates.

The $9-j$ coefficients are given in tables.[10]

$$H_J(K=0)=\frac{\alpha^2}{2}Z\sum_i \frac{\mathbf{l}_i\mathbf{s}_i}{r_i^3}$$

direct spin-orbit coupling ($k_L=1,k_S=1$)

$$-\frac{\alpha^2}{2}\sum_{i\neq j}\left(\frac{\mathbf{r}_{ij}}{r_{ij}^3}\times\mathbf{p}_i\right)(\mathbf{s}_i+2\mathbf{s}_j)$$

spin-other-orbit coupling ($k_L=1,k_S=1$)

$$+\frac{\alpha^2}{2}\sum_{i\neq j}\frac{1}{r_{ij}^3}(\mathbf{s}_i\mathbf{s}_j)-\frac{3(\mathbf{s}_i\cdot\mathbf{r}_{ij})(\mathbf{s}_j\cdot\mathbf{r}_{ij})}{r_{ij}^2},$$

spin-spin interaction ($k_L=2,k_S=2$)

$$H_F(K=1)=\mu_N\frac{8\pi}{3}2\beta_e\sum_i\delta(\mathbf{r}_i)\cdot\mathbf{s}_i$$

Fermi contact operator ($k_L=0,k_S=1$).

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[9] A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton University Press, Princeton, New Jersey, 1960).

[10] K. M. Howell, *Tables of 9j-Symbols* (University of Southampton, Research Report, 1959).