

It can be stated with some assurance that Li^8 is present as a residual nucleus, that the splitting of carbon into three alpha-particles does occur with 90-Mev neutrons, and that carbon does split into two heavy fragments (Li, Li or α , Be).

The cross sections measured by Hadley and York have been verified and extended to encompass lower energy singly charged particles.

The author wishes to express his gratitude to Dr. Burton J. Moyer for his help in suggesting the problem and carrying out the work; to Mr. James Vale and the cyclotron crew for their cooperation, and to Mr. Charles Godfrey for help in operation of the rotating disk apparatus. He is especially indebted to Dr. Wilson Powell and members of his group who organized, equipped and helped operate the cloud-chamber run.

Spin Interactions of Accelerated Nuclei in Molecules

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(Received December 31, 1952)

The contributions to the spin-rotational magnetic interaction of a nucleus in a molecule of terms dependent on the acceleration of the nucleus are calculated. One of these arises from the fact that the acceleration in molecules is electrical and the moving nuclear magnetic moment interacts with the electric field. The other is the Thomas precession similar to that which occurs for electrons in atoms. The effects of both centripetal acceleration and zero-point vibration acceleration are considered; the latter produces the largest effect. A general expression for the combined effect of these terms is given. When it is averaged over the zero point vibration of the H_2 and D_2 molecules, the contribution to the spin-rotational interaction constant c is 1059 cps for H_2 and 30.6 cps for D_2 .

I. INTRODUCTION

IN his theory of the spin-rotational magnetic interaction of a nucleus in a molecule, Wick¹ assumes that the interaction arises exclusively from the magnetic field which results from the circulation of the other charges in the rotating molecule. Although this assumption is approximately valid, an appreciable correction is required when the effects of the acceleration of the nucleus are considered. One of these effects is that the acceleration of the nucleus in the molecule is caused by an electric field and the moving nuclear magnetic moment interacts with this field. The other is the Thomas precession. The accelerations producing these effects arise from two sources: the rotation of the molecule and the zero-point vibration in the molecule. Although for each of these the average acceleration is zero, they nevertheless contribute to the spin-rotational magnetic interaction as is discussed in the following paragraphs.

II. INTERACTIONS

Since the acceleration of a nucleus in the molecule is caused by an electric field, a nucleus of mass M whose acceleration is $d\mathbf{v}/dt$ must be acted on by an electric field, \mathbf{E} , which is given by

$$M d\mathbf{v}/dt = Ze\mathbf{E}. \quad (1)$$

However, to the nucleus moving with velocity \mathbf{v} through this electric field, there will appear to be a magnetic field

$$\mathbf{H}_E = \mathbf{E} \times \mathbf{v}/c. \quad (2)$$

If the nucleus has a gyromagnetic ratio γ , it will precess in this magnetic field with the angular frequency

$$\omega_E = -\gamma \mathbf{H}_E = -\gamma \mathbf{E} \times \mathbf{v}/c. \quad (3)$$

In addition, there will be the purely kinematical Thomas precession^{2,3}

$$\omega_T = (d\mathbf{v}/dt) \times \mathbf{v}/2c^2 = (Ze/2Mc) \mathbf{E} \times \mathbf{v}/c, \quad (4)$$

where the last step comes from Eq. (1). The total precessional frequency, ω_A , due to the nuclear acceleration is therefore

$$\omega_A = -\gamma [1 - (Ze/2Mc\gamma)] \mathbf{E} \times \mathbf{v}/c. \quad (5)$$

Since the discussion of nuclear precession frequencies is usually given in terms of the Hamiltonian of the system which gives rise to these precession frequencies, it is convenient to express the result in the form of a term \mathcal{H}_A of the nuclear Hamiltonian which will give rise to the precession frequency of Eq. (5). If the nucleus has the spin angular momentum \mathbf{I} , in units of \hbar , this precessional frequency will result if

$$\mathcal{H}_A = \hbar \mathbf{I} \cdot \omega_A = -\hbar \gamma [1 - (Ze/2Mc\gamma)] \mathbf{I} \cdot (\mathbf{E} \times \mathbf{v}/c). \quad (6)$$

From this it is apparent that even though \mathbf{E} is zero on the average, it makes a resultant contribution since it is the average value of $\mathbf{E} \times \mathbf{v}/c$ and not of \mathbf{E} that is important. When \mathbf{E} results from centripetal stretching it is clear that $\mathbf{E} \times \mathbf{v}/c$ has a net average value since it

² L. H. Thomas, *Phil. Mag.* **3**, 1 (1926).

³ H. C. Corben and P. Stehle, *Classical Mechanics* (John Wiley and Sons, Inc., New York, 1950).

¹ G. C. Wick, *Phys. Rev.* **73**, 51 (1948).

is always in the same direction. When \mathbf{E} results from the restoring forces on the nucleus in its zero point vibration, $\mathbf{E} \times \mathbf{v}/c$ reverses its sense when \mathbf{E} reverses and it is less apparent that $\mathbf{E} \times \mathbf{v}/c$ does not average to zero. However, $\mathbf{E} \times \mathbf{v}/c$ does not average to zero in this case either since the direction of \mathbf{E} and the magnitude of \mathbf{v} are related as a result of the molecule rotating more rapidly when the nuclei are closer together in the course of the vibration.

III. DIATOMIC MOLECULES

Although Eq. (6) is generally applicable to any molecule, we shall for simplicity limit the discussion from here on to diatomic molecules. Then the \mathbf{v} in Eq. (6) can be simply expressed in terms of the rotational angular momentum $\hbar\mathbf{J}$ of the molecule. Let μ be the reduced mass of the molecule, ω its rotational angular velocity, and \mathbf{R} the radius vector from the other nucleus in the molecule to the nucleus concerned. Then

$$\mathbf{v} = \omega \times \mathbf{R} (\mu/M), \quad (7)$$

and

$$\omega = \hbar\mathbf{J}/\mu R^2. \quad (8)$$

Therefore,

$$\mathbf{v} = (\hbar/MR)\mathbf{J} \times \mathbf{R}/R, \quad (9)$$

and, as \mathbf{E} is parallel to \mathbf{R} or equal to $\mathbf{E}\mathbf{R}/R$,

$$\mathbf{E} \times \frac{\mathbf{v}}{c} = \left(\frac{\hbar E}{MRc} \right) \frac{\mathbf{R}}{R} \times \left(\mathbf{J} \times \frac{\mathbf{R}}{R} \right) = \left(\frac{\hbar E}{MRc} \right) \mathbf{J}. \quad (10)$$

In the analysis of experiments on spin-rotational interactions the results are usually^{4,5} expressed in terms of a parameter c_N defined as the coefficient of $-\hbar\mathbf{I} \cdot \mathbf{J}$ in the molecular Hamiltonian. Therefore, if c_{NA} is the contribution to c_N owing to the acceleration of the nucleus, and if ${}_v\langle c_{NA} \rangle_J$ is its expectation value⁵ for the molecule x in vibrational state v and rotational state J ,

$$\begin{aligned} {}_v\langle c_{NA} \rangle_J &= -{}_v\langle 3c_A/\hbar\mathbf{I} \cdot \mathbf{J} \rangle \\ &= (\gamma/2\pi)(\hbar/Mc)(1 - Ze/2Mc\gamma) {}_v\langle E/R \rangle_J. \end{aligned} \quad (11)$$

The potential in which the nucleus moves can be satisfactorily approximated by a Morse potential,⁶ V , whose parameters are determined⁵ to give the best fit for the vibrational state concerned. Then

$$\begin{aligned} \left\langle \frac{E}{R} \right\rangle &= - \left\langle \frac{1}{ZeR} \frac{dV}{dR} \right\rangle = - \left\langle \frac{1}{ZeR} \frac{d}{dR} \{ D \exp[-2a(R-R_e)] - 2D \exp[-a(R-R_e)] \} \right\rangle \\ &= \frac{2Da}{ZeR_e} \left\langle \frac{\exp[-2a(R-R_e)] - \exp[-a(R-R_e)]}{1 + (R-R_e)/R_e} \right\rangle. \end{aligned} \quad (12)$$

The quantity to be averaged in Eq. (12) may be expanded in a power series of the small quantity $(R-R_e)/R_e$. Therefore, only integral powers of $(R-R_e)/R_e$ need be averaged. Expressions for such averages have been given in an earlier paper⁵ for the zero-point vibration in a Morse potential. When these expressions are combined with Eqs. (11) and (12), one obtains

$$\begin{aligned} {}_0\langle c_{NA} \rangle_J &= 2\pi\gamma e(\hbar c/e^2)(\mu/M)(1/Z)(1 - Ze/2Mc\gamma) \\ &\times \{ B_e\omega_e + B_e^2[-(aR_e)^2 - (aR_e) + 3 - 4J(J+1)] \}, \end{aligned} \quad (13)$$

where B_e , ω_e , and a are the molecular quantities usually designated by these symbols.^{5,7}

IV. MOLECULAR HYDROGEN AND DEUTERIUM

If numerical values^{5,7,8} are substituted in Eq. (13) in the case of molecular H_2 in the first rotational state, one finds

$${}_0^{\text{H}}\langle c_{HA} \rangle_1 = 1059 \text{ cps.} \quad (14)$$

⁴ N. F. Ramsey, Phys. Rev. **85**, 60 (1952).

⁵ N. F. Ramsey, Phys. Rev. **87**, 1075 (1952).

⁶ P. M. Morse, Phys. Rev. **34**, 57 (1929).

⁷ G. Herzberg, *Spectra of Diatomic Molecules* (D. Van Nostrand Company, Inc., New York, 1950).

⁸ J. W. M. DuMond and E. R. Cohen, Phys. Rev. **82**, 556 (1951).

For D_2 , on the other hand,

$${}_0^{\text{D}}\langle c_{DA} \rangle_1 = 30.6 \text{ cps.} \quad (15)$$

Since the experimental value^{9,10} for ${}_0^{\text{H}}\langle c_H \rangle_1$ is $113\,904 \pm 30$ cps, the correction in the case of H_2 is quite important since it is much larger than the experimental error. On the other hand for D_2 , the experimental value is 8773 ± 25 cps, in comparison to which the acceleration contribution is almost negligible.

Before the experimental constants c are interpreted in terms of Wick's theory^{1,5} the above acceleration contributions to c should be deducted.

Although, for the experiments so far completed on H_2 and D_2 , the above theory merely gives a correction to be made to the spin-rotational interaction constant, experiments with HD now in progress should yield sufficient data to check experimentally that the spin interaction of accelerated nuclei is in accordance with the present theoretical prediction.

The author wishes to thank Professor Van Vleck, Professor Furry, and Mr. V. Culler for stimulating discussions of subjects related to this paper.

⁹ Kolsky, Phipps, Ramsey, and Silsbee, Phys. Rev. **87**, 395 (1952).

¹⁰ Harrick, Barnes, Bray, and Ramsey, private communication.