types of glow are observed. Evidence that the observed glows are a solid phenomenon has been given above. The most intense features of the spectra, the α and β lines and the A-bands (Table I and Fig. 3), are quite different in appearance from the usual spectra obtained from gases. However, the NO γ bands (E) and the NH bands (F) show considerable rotational structure and, in addition, indicate a comparatively high rotational "temperature." It seems possible that local heating or "explosions" may form the excited molecules leading to these bands in the gas phase just above the surface. Band heads of the system attributed to NO β (B and C) from the solid are displaced on the average of 9.4 A from the gas NO β system and the system has a very much different structure and intensity distribution in the solid than in the gas. The intensity distribution of the N_2 first positive bands (G) differs in the solid from that in a gas discharge or in a gas afterglow, and the bands also may be shifted to longer wavelengths.

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Molecular Excitation in Beta Decay*

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The theory of molecular excitation in beta decay is developed for the general case of a polyatomic molecule. Calculations of the daughter molecule vibrational excitation are carried through for diatomic molecules, and the results are presented graphically. Finally, the formation of the $(\mathrm{He^3H^3})^+$ molecule ion in the decay of one nucleus of a tritium molecule is studied in detail. It is concluded that the $(\mathrm{He^3H^3})^+$ ground electronic state vibrational spectrum should be observable in the light spontaneously emitted by tritium. For example, neglecting nonradiative transitions, one expects approximately 10^{14} photons per second to be radiated at wave number $\cong 2.35\times 10^3$ cm^{-1} by every gram of tritium in the $1\rightarrow 0$ vibrational transition.

I. INTRODUCTION

When the nucleus of an atom undergoes beta decay, it is possible that the resulting daughter atom will be formed in an electronically excited or ionized state. A number of papers[1–9] have investigated the theory of this process both for electron (±) emission and for electron capture. In addition, the holes produced in the electronic shells and, in the case of K-capture, the ejected orbital electrons have been detected experimentally, with a probability in general agreement with the theoretical predictions.[10–15]

If the nucleus which decays is one of several in a molecule, the physical situation is more complicated. Not only can electronic excitation and ionization occur, but the daughter molecule can be left in an excited vibrational or rotational state. The former effect is due primarily to the sudden change in nuclear charge, while the latter essentially results from the recoil momentum given the decaying nucleus. Dissociation of the daughter molecule can also take place if this molecule is formed either in an unbound electronic state, or in a bound electronic state but in vibrational and rotational states with a combined energy exceeding the depth of the corresponding internuclear potential well.

The purpose of this paper is to present a theoretical treatment of the problem of molecular excitation in beta decay. It is divided into three parts. First, a general discussion is given for polyatomic molecules; time-dependent perturbation theory is used, and the direct interaction between the emitted electron and the molecular electrons is neglected. Second, the special case of diatomic molecules is treated. Here the Born-Oppenheimer[16] approximation gives simple vibrational and rotational wave functions for the nuclear motion so that the problem can be investigated in some detail. Also, a classical distribution of the daughter molecules over their final rotational states is given, and the possibility of experimental detection of the dependence of the final state population on the beta-decay coupling

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† National Science Foundation predoctoral fellow.

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type is considered. Finally, the calculations are carried through for the tritium molecule, which becomes, after the beta-decay of one of the tritium nuclei, the He^3H^3 molecular ion.

It should be added that, particularly in the case of electron (−) emission, the daughter molecule is most often ionized. The general lack of quantitative information about ionized molecules makes it difficult, however, to produce specific theoretical predictions. On the other hand, experimental observations on molecular excitation in beta decay combined with the general theoretical work may provide a valuable tool for the study of previously neglected or unknown ionized molecules.

II. POLYATOMIC MOLECULE CASE

Initial and Final States of the System

The initial and final states of the beta-decaying system for a polyatomic molecule containing $\mathfrak{N}$ nuclei (with one or more of them radioactive) and

$$\mathfrak{Z}=\sum_{N=1}^{\mathfrak{N}} Z_{\text{initial};N}$$

electrons are, neglecting magnetic interactions, described by eigenfunctions of the following Hamiltonian:

$$\begin{aligned}H^{(0)}=\sum_{N=1}^{\mathfrak{N}} H_{\text{nucl};N}(\cdots\mathbf{x}_k,s_k,q_k\cdots)\\ +H_{\text{mol}}(\cdots\mathbf{Y}_\epsilon,s_\epsilon\cdots;\ \cdots\mathbf{R}_N\cdots)+\sum_{N=1}^{\mathfrak{N}}\frac{Z_Nq_le^2}{|\mathbf{R}_N-\mathbf{Y}_l|}\\ -\sum_{\epsilon=1}^{\mathfrak{Z}}\frac{e^2q_l}{|\mathbf{Y}_\epsilon-\mathbf{Y}_l|}+H_{\text{lept}}(\mathbf{Y}_l,s_l,q_l).\end{aligned}\quad(1)$$

Coordinates represented by the capital letters **Y** and **R** refer to axes fixed in space; those represented by the small letters **y** and **r** refer to axes fixed in the center of mass of the molecule; while those represented by **x** refer to the center of mass of a particular nucleus. The coordinates $\mathbf{x}_k$, s_k, q_k are the space, spin, and charge coordinates of the kth nucleon in a given nucleus; $\mathbf{Y}_\epsilon$ and s_ϵ are the space and spin coordinates of the ϵth electron of the molecule; $\mathbf{Y}_l$, s_l, q_l are the space, spin, and charge coordinates of the lepton involved in the beta decay; $\mathbf{R}_N$ is the space coordinate of the center of mass of the Nth nucleus in the molecule. $H_{\text{nucl};N}$ and H_{mol} are respectively the Hamiltonians for the internal motion of the A_N nucleons in the Nth nucleus and for the molecular system of $\mathfrak{Z}$ electrons and $\mathfrak{N}$ nuclei (considered as point charges). H_{lept} is the free-lepton Hamiltonian. Here and in general below our notation follows that of Primakoff and Porter[6] to which reference should be made.

If the interactions between the decay electron and all the other charged bodies except the decaying nucleus (N') are neglected, the Hamiltonian reduces to

$$\begin{aligned}H^{(0)}=\sum_{N=1}^{\mathfrak{N}} H_{\text{nucl};N}(\cdots\mathbf{x}_k,s_k,q_k\cdots)\\ +H_{\text{int mol}}(\cdots\mathbf{y}_\epsilon,s_\epsilon\cdots;\ \cdots\mathbf{r}_N\cdots)\\ +H_{\text{c.m. mol}}(\mathbf{R}_{\text{c.m.}})\\ +\left\{\frac{Z_{N'}e^2q_l}{|\mathbf{r}_{N'}-\mathbf{y}_l|}+H_{\text{lept}}(\mathbf{Y}_l,s_l,q_l)\right\},\end{aligned}\quad(2)$$

where, in addition, the internal molecular motion has been separated from the motion of the center of mass of the molecule. In this approximation, the corresponding eigenfunction can be expressed as a product of functions which are separately eigenfunctions of the individual parts of the Hamiltonian.

$$\begin{aligned}\Psi=\prod_{N=1}^{\mathfrak{N}}\Phi_{\text{nucl};N}(\cdots\mathbf{x}_k,s_k,q_k\cdots)\\ \times\Phi_{\text{int mol}}(\cdots\mathbf{y}_\epsilon,s_\epsilon\cdots;\ \cdots\mathbf{r}_N\cdots)V^{-\frac{1}{2}}\\ \times\exp(i\mathbf{P}\cdot\mathbf{R}_{\text{c.m.}}/\hbar)\mathfrak{U}_{\text{lept}}(\mathbf{Y}_l,s_l,q_l).\end{aligned}\quad(3)$$

For the case of negative electron emission, the lepton in the initial state is a neutrino of negative energy, having as its eigenfunction

$$\begin{aligned}\mathfrak{U}_{\text{lept};i}(\mathbf{Y}_l,s_l,q_l)\\ =V^{-\frac{1}{2}}\exp(-i\mathbf{p}_\nu\cdot\mathbf{Y}_l/\hbar)v_{-s(\nu)}(s_l)w_\nu(q_l)\\ =V^{-\frac{1}{2}}\exp(-i\mathbf{p}_\nu\cdot\mathbf{R}_{\text{c.m.}}/\hbar)\exp(-i\mathbf{p}_\nu\cdot\mathbf{r}_{N'}/\hbar)\\ \times\exp[-i\mathbf{p}_\nu\cdot(\mathbf{y}_l-\mathbf{r}_{N'})/\hbar]v_{-s(\nu)}(s_l)w_\nu(q_l).\end{aligned}\quad(4)$$

After the decay, i.e., in the final state, the lepton is a negative electron. Since only the Coulomb interaction with the emitting nucleus is considered, the space part of the final state lepton eigenfunction referred to the center of mass of the daughter nucleus is $\chi_{\text{lept};f}(\mathbf{y}_l)=V^{-\frac{1}{2}}\exp[i\mathbf{p}_e\cdot(\mathbf{y}_l-\mathbf{r}_{N'})/\hbar]$ times a confluent hypergeometric function of $\mathbf{p}_e\cdot(\mathbf{y}_l-\mathbf{r}_{N'})$. However, referred to axes fixed in space, the lepton eigenfunction computed on the Born-Oppenheimer approximation (i.e., on the assumption that the lepton velocity is large compared with the velocities of nuclei within the molecule) contains an additional and very important exponential factor: $\exp(i\mathbf{p}_e\cdot\mathbf{R}_{\text{c.m.}}/\hbar)\exp(i\mathbf{p}_e\cdot\mathbf{r}_{N'}/\hbar)$. Thus,

$$\begin{aligned}\mathfrak{U}_{\text{lept};f}(\mathbf{Y}_l,s_l,q_l)=\exp(i\mathbf{p}_e\cdot\mathbf{R}_{\text{c.m.}}/\hbar)\exp(i\mathbf{p}_e\cdot\mathbf{r}_{N'}/\hbar)\\ \times\chi_{\text{lept};f}(\mathbf{y}_l)v_{s(e)}(s_l)w_e(q_l).\end{aligned}\quad(5)$$

Transition Probability

The interaction Hamiltonian causing the transition is the beta-decay interaction:

$$\begin{aligned}H^{(1)}=g\sum_{N=1}^{\mathfrak{N}}\sum_{n=1}^{A_N}Q_n\Omega_n^{(\mu)}Q_l\Omega_l^{(\mu)}\delta(\mathbf{r}_N+\mathbf{x}_n-\mathbf{y}_l)\\ +\text{Herm. conj.}\end{aligned}\quad(6)$$

Then the transition probability per unit time is given in first order by time-dependent perturbation theory as

$$P_{i\to f}=\frac{2\pi}{\hbar}\left|\int\psi_f{}^*H^{(1)}\psi_i\right|^2\rho_f, \tag{7}$$

where the density of final states (ρ_f) is in this case

$$\rho_f=V^2(16\pi^5\hbar^6c^3)^{-1}p_e{}^2(E_0-E_e)^2dp_e d\Omega_{e\nu}. \tag{8}$$

The desired total transition probability per unit time is obtained by averaging (7) over the initial states of the system (i.e., of the molecule) using the appropriate Boltzmann statistical weighting factor and then summing it over the final states of the system which are not experimentally distinguished. Except in those special cases where several identical radioactive nuclei are symmetrically located in the molecule, a specification of the type of the final molecule simultaneously tells which nucleus (N') has decayed, so that the interaction Hamiltonian reduces to a single summation. In this form the matrix element can be factorized into a product of integrals. Integration over the center-of-mass coordinates of the molecule simply introduces the requirement of conservation of momentum, so that

$$\begin{aligned}P_{(i)\to(f)}=&\int dp_e\int d\Omega_{e\nu}\sum_{(f)}\mathrm{Av}_iV^2(8\pi^4\hbar^7c^3)^{-1}\\&\times p_e{}^2(E_0-E_e)^2|\mathfrak{M}\mathcal{E}^{(\mu)}{}_{\mathrm{nucl};N'}|^2\\&\times\tfrac{1}{2}\sum_{s_e,s_\nu}|(v_{s(e)}{}^\dagger(s_l)\Omega_l{}^{(\mu)}v_{-s(\nu)}(s_l))|^2\\&\times\bigg|\int\Phi^*{}_{\mathrm{int\ mol};f}\\&\qquad\times\exp[-i(\mathbf{p}_e+\mathbf{p}_\nu)\cdot\mathbf{r}_{N'}/\hbar]\Phi_{\mathrm{int\ mol};i}\bigg|^2,\end{aligned} \tag{9}$$

where

$$\mathrm{Av}_i(\cdots)=\textstyle\sum_i e^{-E_i/kT}(\cdots)/[\sum_i e^{-E_i/kT}]. \tag{10}$$

A considerable simplification is possible if the nuclear matrix element is written as the product of a constant and a function $[M^{(\mu)}(p_e,\theta_{e\nu})]$ which is characteristic of the order of forbiddenness of the decay and of the type of coupling assumed. Then (9) becomes, upon inserting the explicit result of the spin summation and dividing $P_{(i)\to(f)}$ by P_β (the isolated bare nucleus beta-decay probability per unit time).

$$\frac{P_{(i)\to(f)}}{P_\beta}=\frac{\displaystyle\int\int dp_e\sin\theta_{e\nu}d\theta_{e\nu}p_e{}^2(E_0-E_e)^2M^{(\mu)}(p_e,\theta_{e\nu})[1+\lambda(v_e/c)\cos\theta_{e\nu}]\left\{\sum_{(f)}\mathrm{Av}_i\left|\int\Phi^*{}_{\mathrm{int\ mol};f}\exp(i\mathbf{K}\cdot\mathbf{r}_{N'})\Phi_{\mathrm{int\ mol};i}\right|^2\right\}}{\displaystyle\int\int dp_e\sin\theta_{e\nu}d\theta_{e\nu}p_e{}^2(E_0-E_e)^2M^{(\mu)}(p_e,\theta_{e\nu})[1+\lambda(v_e/c)\cos\theta_{e\nu}]}. \tag{11}$$

Here the constant λ is characteristic of the coupling assumed,[17] while $\hbar\mathbf{K}$ is the recoil momentum of the decaying nucleus. It is seen that expression (11) is just the average, over all possible modes of the beta decay, of the term included in the curly brackets. This bracketed term itself can be interpretated as the probability $P_{(f)}(K)$ that a molecule, one of whose nuclei has suffered beta decay and has been given a recoil momentum $\hbar K(p_e,\theta_{e\nu})$, will be found in a specified range of internal molecular final states. Thus

$$\frac{P_{(i)\to(f)}}{P_\beta}=\frac{\displaystyle\int\int dp_ed\theta_{e\nu}G^{(\mu)}(p_e,\theta_{e\nu})P_{(f)}(K)}{\displaystyle\int\int dp_ed\theta_{e\nu}G^{(\mu)}(p_e,\theta_{e\nu})}, \tag{12}$$

where

$$\begin{aligned}G^{(\mu)}(p_e,\theta_{e\nu})\equiv{}&p_e{}^2(E_0-E_e)^2M^{(\mu)}(p_e,\theta_{e\nu})\\&\times[1+\lambda(v_e/c)\cos\theta_{e\nu}]\sin\theta_{e\nu}\end{aligned}$$

is the beta-decay electron distribution function in the variables p_e, $\theta_{e\nu}$.

A more convenient expression results if the variables are changed from p_e and $\theta_{e\nu}$ to p_e and K. The new distribution function $L^{(\mu)}(p_e,K)$ analogous to the previous $G^{(\mu)}(p_e,\theta_{e\nu})$ is then

$$\begin{aligned}L^{(\mu)}(p_e,K)={}&Kp_e(E_0-E_e)M^{(\mu)}(p_e,\theta_{e\nu})\\&\times[1+\lambda(v_e/c)\cos\theta_{e\nu}],\end{aligned} \tag{13}$$

where the $\theta_{e\nu}$ is considered as a function of p_e and K, and the integration over K is performed from $[p_e+(E_0-E_e)/c]/\hbar$ to $|p_e-(E_0-E_e)/c|/\hbar$.

In the special case of allowed decay $M^{(\mu)}(p_e,\theta_{e\nu})$ is independent of $\theta_{e\nu}$, causing the coefficient of λ in the denominator of (11) to vanish. The probability ratio $P_{(i)\to(f)}/P_\beta$ is then a linear function of λ and can be written in the form

$$P_{(i)\to(f)}/P_\beta=D_{(i,f)}+\lambda B_{(i,f)}. \tag{14}$$

III. DIATOMIC MOLECULE CASE

Specialization of Previous Theroy

In the special case of diatomic molecules and with the Born-Oppenheimer approximation, the energy eigenfunctions for the internal molecular motion factor-

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[17] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), p. 716.

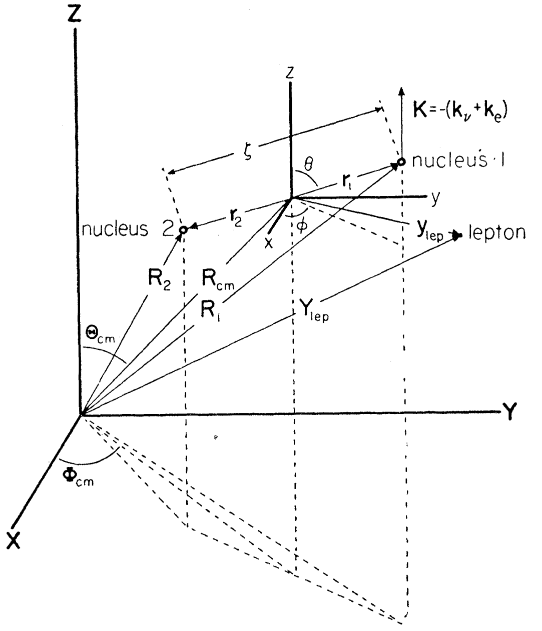

FIG. 1. Coordinate system for diatomic molecule case.

ize in terms of electronic, vibrational, and rotational eigenfunctions as follows[18]:

$$\Phi_{\text{int mol}} \cong \Phi_{\text{el}}(\cdots \mathbf{y}_\epsilon, s_\epsilon \cdots)\Phi_{\text{vib};n}(|\mathbf{r}_1-\mathbf{r}_2|=\zeta) \times\Phi_{\text{rot};J,M}(\theta,\varphi), \quad (15)$$

where

$$\Phi_{\text{vib};n}=\frac{1}{\zeta_0}\left(\frac{\alpha}{\pi^{\frac{1}{2}}2^n n!}\right)^{\frac{1}{2}} H_n[\alpha(\zeta-\zeta_0)] \times\exp[-\tfrac{1}{2}\alpha^2(\zeta-\zeta_0)^2], \quad (16)$$

$$\Phi_{\text{rot};J,M}=\left[\frac{(2J+1)(J-|M|)!}{2(J+|M|)!}\right]^{\frac{1}{2}} \times P_J^{|M|}(\cos\theta)\frac{1}{(2\pi)^{\frac{1}{2}}}e^{iM\varphi}, \quad (17)$$

$E_{\text{int mol}}=E_{\text{vib}}+E_{\text{rot}}+E_{\text{elect}}$,

$\alpha=(\mu k/\hbar^2)^{\frac{1}{4}}=(\mu\omega/\hbar)^{\frac{1}{2}}=(MM'\omega/\hbar(M+M'))^{\frac{1}{2}}$,

M' = mass of decaying nucleus,

ζ_0 = equilibrium internuclear separation.

A diagram of the coordinate system used appears in Fig. 1. Notice that the z-axis has been chosen along the direction $\mathbf{K}/K$ of the nuclear recoil, making the angle between $\mathbf{K}$ and $\mathbf{r}_{N'}$ be θ. The expression which must be evaluated is then

$$P_{(f)}(K)=|\mathfrak{M}\mathcal{E}_{\text{el}}|^2\sum_{(f)}\text{Av}_i\left|\int\int\Phi_{\text{rot};J(f),M(f)}{}^*(\theta,\varphi) \times\mathfrak{F}_{n(i),n(f)}(K,\theta)\Phi_{\text{rot};J(i),M(i)}(\theta,\varphi)d(\cos\theta)d\varphi\right|^2, \quad (18)$$

where

$$\mathfrak{F}_{n(i),n(f)}(K,\theta)=\int_{-\infty}^{\infty}\Phi_{\text{vib};n(f)}{}^*(\zeta)\exp(iK\mu\zeta\cos\theta/M') \times\Phi_{\text{vib};n(i)}(\zeta)\zeta_0{}^2d\zeta, \quad (19)$$

$$\mathfrak{M}\mathcal{E}_{\text{el}}=\int\Phi_{\text{el};f}{}^*(\cdots\mathbf{y}_\epsilon,s_\epsilon\cdots) \times\Phi_{\text{el};i}(\cdots\mathbf{y}_\epsilon,s_\epsilon\cdots). \quad (20)$$

Since the main contribution to the integral over ζ will be in the region of ζ_0, the volume element $\zeta^2d\zeta$ has been replaced by $\zeta_0{}^2d\zeta$, and the integration has been extended to minus infinity. If the experiment performed does not distinguish between individual final rotational states, they must be included in the sum over final states. Use of the closure approximation with respect to these final rotational states yields

$$P_{(f)}(K)=|\mathfrak{M}\mathcal{E}_{\text{el}}|^2\,\text{Av}_i\int\int|\Phi_{\text{rot};J(i),M(i)}(\theta,\varphi)|^2 \times\mathfrak{F}_{n(i),n(f)}{}^*(K,\theta)\mathfrak{F}_{n(i),n(f)}(K,\theta)d(\cos\theta)d\varphi. \quad (21)$$

At room temperature and below, the great majority of the molecules will be initially in the lowest vibrational state ($n_i=0$), so that the average need be performed only over initial rotational states. Application of an addition theorem for spherical harmonics,

$$\sum_{M=-J}^{J}|\Phi_{JM}|^2=(2J+1)/4\pi,$$

provides the result that

$$\text{Av}_i|\Phi_{\text{rot};J(i),M(i)}(\theta,\varphi)|^2 =\sum_{M=-J}^{J}|\Phi_{JM}|^2/(2J+1)=1/4\pi. \quad (22)$$

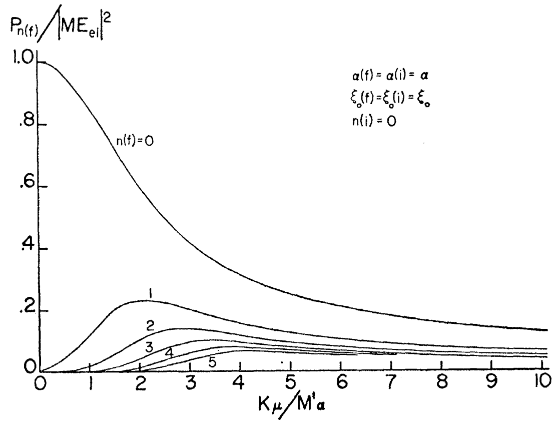

FIG. 2. Vibrational excitation of a diatomic molecule resulting from the recoil with momentum $\hbar K$ (averaged over all directions) of one of its nuclei. The parameter $M'\alpha/\mu$ dividing K in the abscissa is characteristic of the diatomic molecule involved and will occur repeatedly in the following material; it will be noted that $\mu K/M'\alpha$ = (recoil velocity of decaying nucleus) ÷ (zero point vibrational relative velocity in initial molecule).

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[18] L. Pauling and E. B. Wilson, *Introduction to Quantum Mechanics* (McGraw Hill Book Company, Inc., New York, 1935), Chap. 10.

Thus,

$$P_{n(f)}(K)=|\mathfrak{M}\mathcal{E}_{\text{el}}|^2\tfrac{1}{2}\int_{-1}^{1}\mathfrak{F}_{n(i)n(f)}{}^*\mathfrak{F}_{n(i)n(f)}d(\cos\theta). \tag{23}$$

The quantities $\mathfrak{F}_{0,\,n(f)}(K,\theta)$ can be written in general, from the integral in (19),

$$\begin{aligned}\mathfrak{F}_{0,\,n(f)}(K,\theta)=&\left(\frac{\alpha_i}{2^{n_i}n_i!}\right)^{\frac{1}{2}}\left(\frac{\alpha_f}{2^{n_f}n_f!}\right)^{\frac{1}{2}}\left(\frac{2}{\alpha_i^2+\alpha_f^2}\right)^{\frac{1}{2}}\\ &\times\exp\left[-\frac{\alpha_f^2\alpha_i^2\delta^2}{2(\alpha_f^2+\alpha_i^2)}-\frac{K^2\mu^2\cos^2\theta}{2(\alpha_f^2+\alpha_i^2)M'^2}\right.\\ &\left.+i\left(\frac{\alpha_f^2\zeta_{0f}+\alpha_i^2\zeta_{0i}}{\alpha_f^2+\alpha_i^2}\right)\frac{\mu K\cos\theta}{M'}\right]\\ &\times\sum_m\frac{n_f!}{m!(n_f-2m)!}\left(\frac{\alpha_f^2-\alpha_i^2}{\alpha_f^2+\alpha_i^2}\right)\\ &\times\left(\frac{-2\alpha_f\alpha_i^2\delta+i2\alpha_f\mu K\cos\theta/M'}{\alpha_f^2+\alpha_i^2}\right)^{n_f-2m},\end{aligned} \tag{24}$$

where $\delta=\zeta_{0f}-\zeta_{0i}$. For the first two final vibrational states,

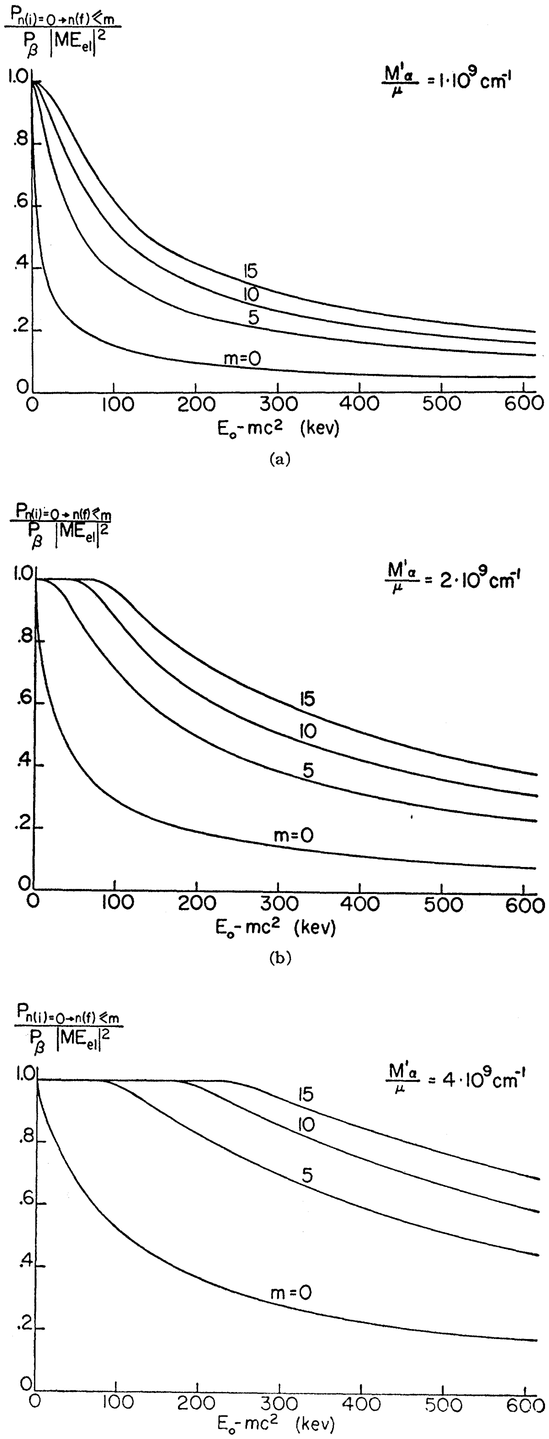
$$\mathfrak{F}_{00}{}^*\mathfrak{F}_{00}(K,\theta)=\frac{2\alpha_i\alpha_f}{(\alpha_i^2+\alpha_f^2)}\exp\left[-\frac{\alpha_f^2\alpha_i^2\delta^2}{(\alpha_f^2+\alpha_i^2)}-\frac{\mu^2K^2\cos^2\theta}{M'^2(\alpha_f^2-\alpha_i^2)}\right], \tag{25}$$

$$\mathfrak{F}_{01}{}^*\mathfrak{F}_{01}(K,\theta)=\mathfrak{F}_{00}{}^*\mathfrak{F}_{00}\left\{\frac{2\alpha_f^2\alpha_i^4\delta^2+2\alpha_f^2K^2\mu^2\cos^2\theta/M'^2}{(\alpha_f^2+\alpha_i^2)^2}\right\}. \tag{26}$$

The integral over θ in (23) is a Gaussian integral, whose values are tabulated. Then the final averaging in (12) and (13) must be performed numerically. Alternatively, the integration over K in (12) and (13) [using (23), (24)] can be performed first. The θ integration remaining is again Gaussian, leaving only the integration over p_e to be carried out numerically.

General Curves

In some cases, it is a good approximation to assume that $\zeta_{0i}=\zeta_{0f}=\zeta_0$ and that $\alpha_f=\alpha_i=\alpha$. This greatly decreases the complexity of the mathematical formulas and makes the theoretical predictions particularly amenable to general graphical presentation. Such approximations have been used in all of the graphs described in this paragraph. Figure 2 is the probability that a diatomic molecule, one of whose nuclei has recoiled with momentum $\hbar K$, will be found in the n_f final vibrational level of a certain specified electronic state, plotted against $\mu K/M'\alpha$ for $n_f=0$, 1, 2, 3, 4, 5 with $n_i=0$. Figure 3 represents probabilities per



(a)

(b)

(c)

FIG. 3. Probability of a diatomic molecule remaining bound after the beta decay of one of its nuclei, plotted against the energy release in the beta decay, for several values of the number of bound vibrational states belonging to that electronic state and for several values of the parameter $M'\alpha/\mu$. The internuclear potential well has been assumed to be parabolic. Nuclear Coulomb field and electron Coulomb field (screening) corrections are neglected.

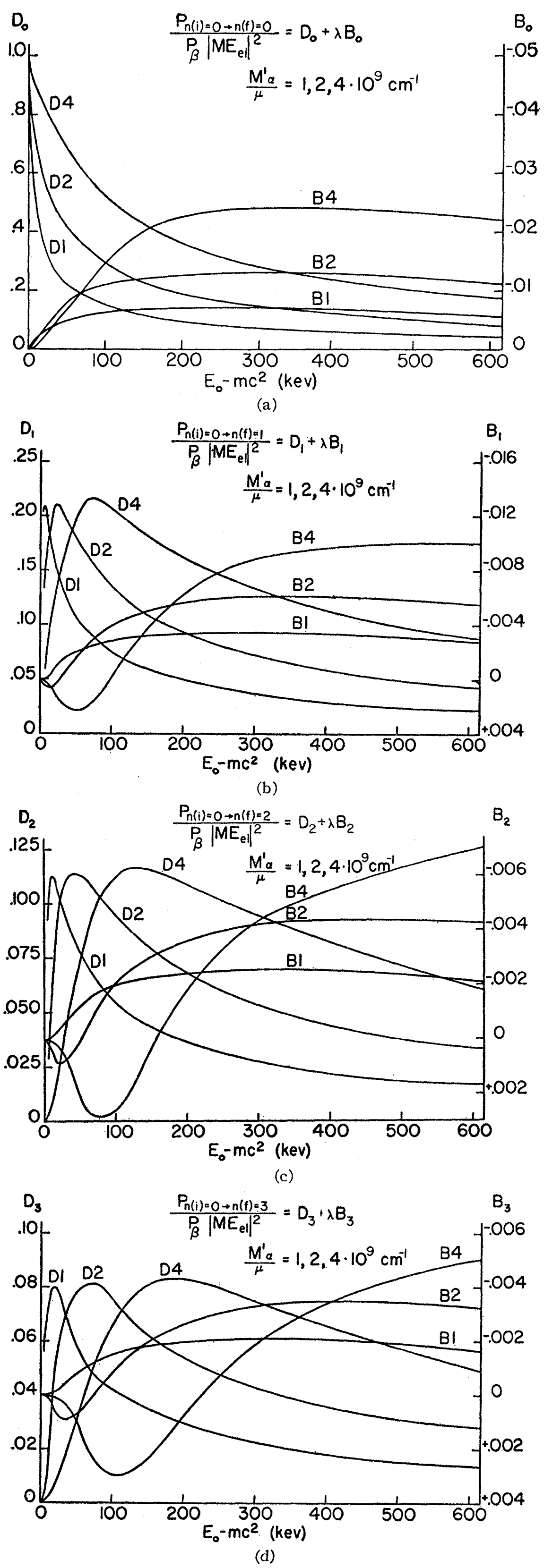


FIG. 4. Probability of a diatomic molecule being found, immediately after beta decay of one of its nuclei, in the n_f vibrational state, for $n_f=0$, 1, 2, 3 and for several values of the parameter $M'\alpha/\mu$. The parameter λ depends on the type of the beta-decay coupling.

(allowed) beta decay of the molecule remaining bound in a given electronic state, plotted against the energy release in the beta decay, for several values of the number of bound vibrational states belonging to that electronic state and for several values of the parameter $M'\alpha/\mu$. Since both the molecular eigenfunctions used and the rotational closure approximation are poor near the top of the internuclear potential well, these curves will be rather inaccurate for shallow wells and for large energy releases. Figure 4 shows plots of the probability per (allowed) beta decay of the daughter molecule being found in the n_f vibrational level of a certain specified electronic state against the beta-decay energy release, for $n_f=0$, 1, 2, 3 and for several values of the parameter $M'\alpha/\mu$. In these last curves (Fig. 4), the dependence on the type of beta-decay coupling (λ) is shown explicitly, while in the previous curves (Fig. 3) it was assumed that $\lambda=0$; in all cases the "probabilities" plotted are actually probabilities divided by the appropriate $|\mathfrak{M}\mathcal{E}_{\text{el}}|^2$.

Measurability of λ

Figure 4 shows that the probability of finding a daughter molecule in a given vibrational state immediately after the beta decay of one of its nuclei may depend by as much as 25% on the beta-decay coupling type (λ). The reason for this dependence is easily seen. If the electron and neutrino tend to be emitted in the same direction, the average recoil momentum given the decaying nucleus will increase, and the daughter molecule will be found more often in a higher vibrational state. Similarly, a tendency toward antiparallel emission is accompanied by a decreased daughter molecule vibrational excitation.

Unfortunately, however, the relative intensities of the infrared lines emitted in the radiative deexcitation of the vibrational states are not nearly so strongly dependent on λ. The intensity of a given line is a function not only of the population probability of the initial state in the radiative process but also of the population probabilities of all states which are energetically above this state and which can decay to it by a chain of (radiative or nonradiative) transitions. If certain simplifying assumptions are made, the problem can be treated quantitatively. Assume that

(1) all transitions are radiative and electric dipole,

(2) higher electronic states, if any, are negligibly populated, and

(3) the vibrational and rotational excitation does not exceed the depth of the internuclear well.

Then λ can be expressed as a function of the ratio of the number of photons emitted in the 2→1 vibrational transition to the number emitted in the 1→0 transition, $\mathcal{R}_{21/10}$, and of the quantities D and B defined in Eq. (14) and given for diatomic molecules as a function of the energy release and of $M'\alpha/\mu$ in Fig. 4:

$$\lambda=[(1-\mathcal{R}_{21/10})(1-D_0)-D_1]/[B_1+B_0(1-\mathcal{R}_{21/10})].$$

Figure 5 is a plot of $\mathcal{R}_{21/10}$ against the beta-decay energy release for $\lambda=\pm1$ and for several values of $M'\alpha/\mu$. The theoretically anticipated small variation of $\mathcal{R}_{21/10}$ with λ would seem to discourage any experimental attempt to measure λ by this means.

Rotational Distribution

The distribution of the beta-decay excited daughter molecules over their final rotational states (in a given final vibrational state) differs from that encountered in other methods of excitation. If the radiative lifetime for vibrational deexcitation is short compared to the lifetime for nonradiative rotational transitions, the characteristic rotational distribution is reflected in the fine structure of the lines emitted in vibrational transitions, thus providing an identifying characteristic of the lines sought. This distribution could, of course, be obtained quantum-mechanically by eliminating the sum over final rotational states in (18). However, the integrals then become difficult to evaluate. Hence a classical calculation, valid for high rotational quantum numbers, has been performed using the recoil momentum distribution given by Kofoed-Hansen[19] for allowed transitions and assuming $\lambda=0$. Figure 6 gives the unnormalized probability of finding the daughter molecule with rotational quantum number J_f as a function of a quantity proportional to

$$[2n_f(M'\alpha/\mu)^2+((M+M')/M\zeta_0)^2J_f^2]^{\frac{1}{2}}.$$

Notice that, for $n_f=0$, the abscissa is proportional to J_f. The values of the J_f which correspond to the maxima of $P(J_f)$ (the most probable J_f) vary from about $\frac{1}{2}$ to about $\frac{2}{3}$ of $(J_f)_{\max}$ as E_0-mc^2 varies from 0 kev to 560 kev.

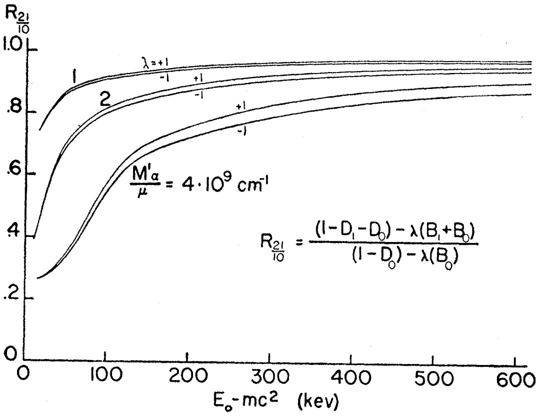


FIG. 5. Dependence of the ratio of the number of photons emitted in the 2→1 vibrational transition to the number emitted in the 1→0 vibrational transition on the beta-decay energy release, the beta-decay coupling type, and the parameter $M'\alpha/\mu$ characteristic of the particular diatomic molecule involved in the decay.

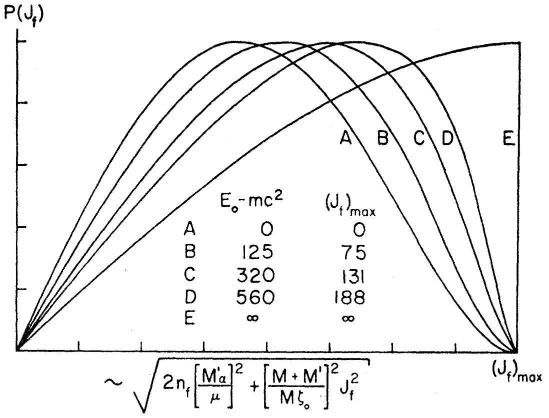


FIG. 6. Semiclassical distribution of daughter molecules over their rotational states immediately following beta decay. The values given for E_0-mc^2, the beta-decay energy release, are in kev. The $(J_f)_{\max}$ listed are for $n_f=0$, $M=M'$, and $\zeta_0=1.5$ Bohr radii.

If, on the other hand, the radiative lifetime for vibrational deexcitation is long compared to the lifetime for nonradiative rotational transitions, the fine structure of the vibrational lines corresponds to a rotational distribution which is Boltzmann. For the case of comparable lifetimes, the rotational distribution characterizing the vibrational fine structure will be intermediate between the Boltzmann and the above described characteristic distributions.

IV. TRITIUM MOLECULE EXAMPLE

One particularly favorable case for both theoretical discussion and experimental investigation is that of the tritium molecule, which can beta decay according to the scheme

$$T_2\equiv H^3H^3\rightarrow(He^3H^3)^++e^-+\nu.$$

The He^3H^3 molecule ion is electronically almost identical with the He^4H molecule ion,[20–23] which, first detected mass spectrographically in the early 1920's, is known to have at least one stable electronic state (the ground state). The lowest electronic energy eigenfunction and eigenvalue of $(HeH)^+$ have been the subject of several papers,[24–28] as have the corresponding quantities in the H_2 case.[29]

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[19] O. Kofoed-Hansen, Phys. Rev. **74**, 1785 (1948).

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[22] K. T. Bainbridge, Phys. Rev. **44**, 57 (1933).

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[24] G. Glockler and D. L. Fuller, J. Chem. Phys. **1**, 886 (1933).

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[27] S. Toh, Proc. Phys.-Math. Soc. Japan **22**, 119 (1940).

[28] A. A. Evett, Phys. Rev. **98**, 1191(A) (1955).

[29] H. M. James and A. S. Coolidge, J. Chem. Phys. **1**, 825 (1933).

TABLE I. Ground electronic state constants of various molecules. Data on $(HeH)^+$ type molecules are from the work of Evett.[31] Vibrational levels have been calculated from the equation $W/hc = \omega_e(n+\frac{1}{2}) - \omega_e\chi_e(n+\frac{1}{2})^2$.

	T_2	$(He^3H)^+$	$(He^3D)^+$	$(He^3T)^+$	$(He^4H)^+$	$(He^4D)^+$	$(He^4T)^+$
μ (mass units)	1.5085	0.7556	1.2080	1.5085	0.8054	1.3403	1.7205
$10^{-9}\alpha$ (cm^{-1})	1.07	0.886	0.996	1.053	0.900	1.022	1.088
ζ_0 (Bohr radii)	1.402	1.432	1.432	1.432	1.432	1.432	1.432
$\hbar^2/2I$ (cm^{-1})	19.8	38.8	24.3	19.5	36.4	21.9	17.1
ω_e (cm^{-1})	2553.8	3716	2939	2630	3600	2790	2463
$\omega_e\chi_e$ (cm^{-1})	43.872	214.2	134.0	107.3	201.0	120.8	94.1
W_0/hc (cm^{-1})	1266	1804	1436	1288	1750	1365	1208
$(W_1-W_0)/hc$ (cm^{-1})	2466	3288	2671	2352	3198	2549	2274
$(W_2-W_1)/hc$ (cm^{-1})	2378	2859	2403	2012	2796	2307	2086
$(W_3-W_2)/hc$ (cm^{-1})	2291	2431	2135	1671	2394	2066	1898
$(W_4-W_3)/hc$ (cm^{-1})	2203	2002	1867	1330	1992	1824	1710

The electronic overlap integral in Eq. (20) was calculated using simple wave functions of the form suggested by James and Coolidge.[29] To facilitate the integration the equilibrium internuclear distances in the lowest electronic states were assumed to be 1.4 Bohr radii for both molecules.

$$\psi((HeH)^+) = (2\pi)^{-1}e^{-1.25(\xi_1+\xi_2)}\{24.8\},$$

$$\psi'((HeH)^+) = (2\pi)^{-1}e^{-1.25(\xi_1+\xi_2)}\{22.2+14.8(\eta_1+\eta_2)\},$$

$$\psi(H_2) = (2\pi)^{-1}e^{-0.75(\xi_1+\xi_2)}\{3.40\},$$

$$\psi'(H_2) = (2\pi)^{-1}e^{-0.75(\xi_1+\xi_2)}\{3.04+0.564(\eta_1^2+\eta_2^2)\}.$$

The coordinates ξ_1, ξ_2, η_1, η_2 are the elliptical coordinates of James and Coolidge.[29] The absolute squares of the overlap integrals are tabulated below.[30]

	$\psi((HeH)^+)$	$\psi'((HeH)^+)$
$\psi(H_2)$	0.73	0.59
$\psi'(H_2)$	0.72	0.58

Whereas little improvement is obtained by increasing the number of terms in the hydrogen molecule function, the addition of one term to the helium-hydride function has appreciable effect, indicating that a more accurate calculation in this case would probably be desirable.

Table I gives various ground electronic state constants for the molecules of interest. Figures given for helium-hydride type molecules have been calculated from the internuclear potential predictions of Evett,[28,31] who has done the most accurate theoretical work on this molecule. Figure 7 is a graph of $P_{n(f)}(K)/|\mathfrak{M}\mathcal{E}_{el}|^2$ against K calculated from Eqs. (23) and (24) for final vibrational quantum numbers of 0, 1, and 2. The nonzero values of $P_1(0)$ and $P_2(0)$ are manifestations of the Franck-Condon principle and occur because of the different well shapes and equilibrium internuclear distances in He^3H^{3+} and in (diatomic) tritium. The probabilities of the He^3H^3 molecule ion being formed in the ground or first- or second-excited vibrational states of the ground electronic state (all rotational states) is then the average of $P_{n(f)}(K)$ over p_e and K with the distribution function of Eq. (13). Since tritium decay is an allowed transition, the $M^{(\mu)}(p_e)$ used is just the Coulomb-field Fermi function[32]

$$F(E,Z) = \frac{2\pi(Z/137)(c/v_e)}{1-\exp[-2\pi(Z/137)(c/v_e)]}, \tag{28}$$

with $Z=2$. The results obtained from Eqs. (12), (13), (23), and (24) are

$$P_0/2P_\beta = |\mathfrak{M}\mathcal{E}_{el}|^2(0.671-0.003\lambda)$$
$$= (0.389-0.002\lambda), \tag{29}$$

$$P_1/2P_\beta = |\mathfrak{M}\mathcal{E}_{el}|^2(0.192+0.0005\lambda)$$
$$= (0.111+0.0003\lambda), \tag{30}$$

$$P_2/2P_\beta = |\mathfrak{M}\mathcal{E}_{el}|^2(0.078+0.0007\lambda)$$
$$= (0.045+0.0004\lambda), \tag{31}$$

where $P_n/2P_\beta$ is the probability per decay of the parent T_2 molecule of finding the daughter $(He^3T)^+$ molecule ion in the nth vibrational state of the ground electronic state.

Equations (29)–(31) indicate that, if only radiative

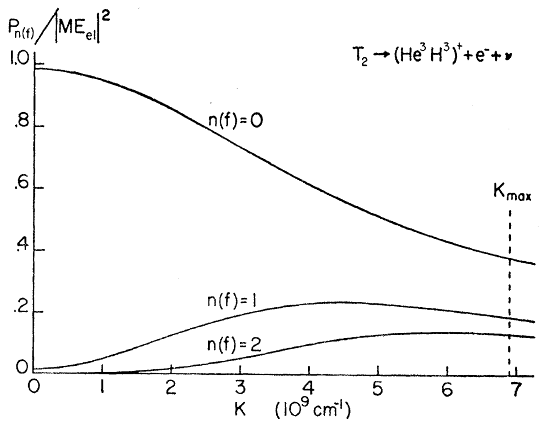

FIG. 7. Vibrational excitation of daughter $(He^3T)^+$ resulting from recoil momentum $\hbar K$ (averaged over all directions) given to one nucleus in the beta decay of the parent tritium molecule.

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[30] H. M. Schwartz (private communication) has calculated, using the functions of S. Toh and of James and Coolidge, that the absolute square of the overlap integral is 0.93.

[31] A. A. Evett (private communication). The author is indebted to Dr. Evett for permission to use his $(HeH)^+$ internuclear well data before its publication.

[32] Reference 17, p. 680.

deexcitation of the excited vibrational states occurs, about 10^{14} photons per second or 10^{-5} watt at wave number $\cong 2.35\times 10^3$ cm^{-1} (Table I) will be radiated by every gram of tritium in the 1→0 vibrational transition. However, this intensity will be greatly decreased if nonradiative deexcitation processes are dominant. Of particular importance are

(1) de-excitation of the $(\mathrm{He^3T})^+$ by collision,

(2) dissociation of $(\mathrm{He^3T})^+$ by capture of a free electron (HeH is not bound), and

(3) disappearance of $(\mathrm{He^3T})^+$ in collision reactions with T_2 or with impurities.

If it is assumed that the effective dipole moment of $(\mathrm{He^3T})^+$ has the form

$$\mu(\zeta)\cong\tfrac{1}{2}e\zeta=\tfrac{1}{2}e\zeta_0+\tfrac{1}{2}e(\zeta-\zeta_0),$$

where e is the proton charge, the calculated radiative lifetime of the nth vibrational level is $0.008(n+1)^{-1}$ second. With regard to (1), no predictions can be made about the lifetime for vibrational deexcitation of $(\mathrm{He^3T})^+$ in collision with T_2, since the interaction potential is unknown. However, in T_2-T_2 collisions the method of Schwartz, Slawsky, and Herzfeld[33] yields a lifetime for vibrational deexcitation of about 0.005 second at $T=300$°K and atmospheric pressure. For $T_2-(\mathrm{He^3T})^+$ collisions the range of the attractive interaction is larger, tending to increase the rate of deexcitation, but the repulsive hard core is smaller, causing a decrease in the rate. With regard to (2), for the same conditions of temperature and pressure, a crude calculation of the lifetime of $(\mathrm{He^3T})^+$ for dissociation by capture of the free electrons arising from ionization by the tritium decay beta particles gives a lower limit of 0.002 second. Thus it would seem probable that at atmospheric pressure and room temperature radiative de-excitation of the excited vibrational levels of the daughter $(\mathrm{He^3T})^+$ molecules competes favorably with the nonradiative processes. Lowering the pressure decreases the proportion of nonradiative processes but, for a given volume, also decreases the source strength. Finally, Table II, which lists collision processes and their initial and final state energies (from Table III), reveals three possible reactions which can destroy the helium-hydride molecule ion. Two of these are very improbable since in both cases the concentration of the second initial state molecule (i.e., of the H or $\mathrm{He^+}$) in tritium should be very small. The third process poses a possible threat to the observability of the optical spectrum of the helium-tritide ion.[34]

TABLE II. Reactions by which $(\mathrm{HeH})^+$ may dissociate nonradiatively. Initial and final state energies given do not include molecular vibrational and rotational energies.

Initial state			Final state	
Approximate energy (ev)				Approximate energy (ev)
-112.733	$(\mathrm{HeH})^++\mathrm{H_2}$	$\nrightarrow$	$\mathrm{He+H+H_2^+}$	-108.959
		$\nrightarrow$	$\mathrm{He+H^++H_2}$	-110.915
		$\rightarrow$	$\mathrm{He+H_3^+}$	-117.46
-97.182	$(\mathrm{HeH})^++\mathrm{H_2^+}$	$\nrightarrow$	$\mathrm{He+H^++H_2^+}$	-95.364
		$\nrightarrow$	$\mathrm{He^++H_2+H^+}$	-86.335
		$\nrightarrow$	$\mathrm{He^++H_2^++H}$	-84.379
		$\nrightarrow$	$\mathrm{He^{++}+H_2+H}$	-45.530
		$\nrightarrow$	$\mathrm{He^++H_3^+}$	-92.88
-94.393	$(\mathrm{HeH})^++\mathrm{H}$	$\nrightarrow$	$\mathrm{He+H+H^+}$	-92.575
		$\rightarrow$	$\mathrm{He+H_2^+}$	-95.364
		$\nrightarrow$	$\mathrm{He^++H_2}$	-86.335
-80.798	$(\mathrm{HeH})^++\mathrm{H^+}$	$\nrightarrow$	$\mathrm{He^++H_2^+}$	-70.784
		$\nrightarrow$	$\mathrm{He+H^++H^+}$	-78.98
-217.378	$(\mathrm{HeH})^++\mathrm{He_2^+}$	$\nrightarrow$	$\mathrm{He_2^++He^++H}$	-204.575
		$\nrightarrow$	$\mathrm{He_2^++He+H^+}$	-215.56
-159.778	$(\mathrm{HeH})^++\mathrm{He}$	$\nrightarrow$	$\mathrm{He+He+H^+}$	-157.96
		$\nrightarrow$	$\mathrm{He_2^++H}$	-150.175
-135.198	$(\mathrm{HeH})^++\mathrm{He^+}$	$\nrightarrow$	$\mathrm{He+He^++H^+}$	-133.38
		$\nrightarrow$	$\mathrm{He^++He^++H}$	-122.395
		$\rightarrow$	$\mathrm{He_2^++H^+}$	-136.58

TABLE III. Total energies of atoms and molecules present in the tritium. Molecular energies given are measured to the bottom of the internuclear potential well from a zero of energy which corresponds to complete dissociation into nuclei and electrons.

Molecule or atom	Energy (ev)
$\mathrm{H_2}$	-31.935
$\mathrm{H_2^+}$	-16.384
$\mathrm{H_3^+}$	-38.48[a]
H	-13.595
$\mathrm{H^+}$	0
$(\mathrm{HeH})^+$	-80.798
$\mathrm{He_2^+}$	-136.58
He	-78.98
$\mathrm{He^+}$	-54.40
$\mathrm{He^{++}}$	-0

[a] Barker, Giddings, and Eyring, J. Chem. Phys. **23**, 344 (1955).

Since the beta decay of tritium is an allowed transition with spin change zero, both scalar and tensor interactions are present. The value of λ is given by[35]

$$\lambda=\frac{\frac{1}{3}g_T^2\left|\int\sigma\right|^2-g_S^2\left|\int 1\right|^2}{g_T^2\left|\int\sigma\right|^2+g_S^2\left|\int 1\right|^2},\qquad(32)$$

where g_S and g_T are the tensor and scalar coupling constants and $\int\sigma$ and $\int 1$ are the Gamow-Teller and the Fermi nuclear matrix elements. Also $g_T^2\cong g_S^2$, and for tritium $|\int\sigma|^2\cong 3|\int 1|^2\cong 3$; thus $\lambda\cong 0$. In any

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[33] Schwartz, Slawsky, and Herzfeld, J. Chem. Phys. **20**, 1591 (1952).

[34] The rate of this process is proportional to $\exp[-E/kT]$, where E is the activation energy (unknown) for the reaction. Thus the rate can be greatly decreased by reduction of the temperature. See Glasstone, Laidler, and Eyring, *The Theory of Rate Processes* (McGraw-Hill Book Company, Inc., New York, 1941).

[35] O. Kofoed-Hansen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **28**, No. 9 (1954).

event, the dependence of the population of the daughter $(\mathrm{He^3T})^+$ molecule states on λ is so small that experimental verification of the effect seems impossible.

If the vibrational spectrum of $(\mathrm{He^3T})^+$ is too far in the infrared (Table I) to be conveniently observed, TD or TH may be used instead as the parent molecule. The vibrational spectra of the corresponding daughter molecules, namely $(\mathrm{He^3D})^+$ or $(\mathrm{He^3H})^+$, lie nearer to the visible region (Table I) and may possibly be detectible with higher efficiency. However, the intensity of the lines will be less in these cases because of the decreased probability of beta decay per molecule and of the presence of large amounts of T_2, and H_2 or D_2. Equations (29)–(31) for P_0, P_1, and P_2 do not hold here exactly because α depends on the reduced mass μ $(\alpha\sim\mu^{\frac{1}{4}})$.

V. CONCLUSION[36]

Another example of molecular excitation in beta decay is provided by $\mathrm{C^{14}O}$, which becomes, after the decay, the nitrous oxide molecule ion.

$$\mathrm{C^{14}O}\rightarrow(\mathrm{N^{14}O})^+ + e^- + \nu.$$

Here the rather large energy release and the favorable value of $M'\alpha/\mu$ $(4\times10^9\ \mathrm{cm}^{-1}$, assuming the daughter to have the same α as the parent) provide a much greater dependence on the beta-decay coupling type (λ) than in the tritium case (cf Fig. 4). Since the transition involves a spin change of unity, only the tensor interaction contributes, so that $\lambda=+\frac{1}{3}$. Unfortunately, there are very little data on the $(\mathrm{NO})^+$ molecule, and no precise quantitative predictions can be made; however, Eq. (20) and Fig. 4 indicate an appreciable probability of the $(\mathrm{N^{14}O})^+$ being formed in various low vibrational states of its ground electronic state.

The appendix of Herzberg's book[37] on diatomic molecules lists thirty diatomic molecule ions. Little is known in detail about such molecule ions because of the difficulties in maintaining a high ion concentration and of the relatively inefficient methods of excitation. It would seem, though, from a consideration of the tritium results and from Figs. 3 and 4, that production of such ions by low energy beta-decay is a rather efficient process. Thus, in those cases where the decay process is rapid enough to provide the necessary ion concentration, useful information on the daughter molecule ion should be obtained. Also a study of the molecular excitation in beta decay provides (in principle at least) a method for determining the type of the beta-decay interaction through its determination of the λ value, and promises to increase the detailed understanding of radiochemical processes.

ACKNOWLEDGMENTS

This investigation was originally begun by H. Primakoff and F. T. Porter. The author wishes to thank H. Primakoff for his continued guidance.

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[36] *Note added in proof.*—See also H. M. Schwartz, J. Chem. Phys. **23**, 400 (1955); J. phys. radium **16**, 497 (1955).

[37] G. Herzberg, *Spectra of Diatomic Molecules* (D. Van Nostrand and Company, Inc., New York, 1951).