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DISSOCIATION OF DIATOMIC MOLECULES DURING BETA DECAY

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A calculation is made of the probability for dissociation of a diatomic molecule during β decay of one of its nuclei. It is shown that the results can be used to determine the interaction constants for the coupling of the electron-neutrino field to nucleons.

1. During the β decay of nuclei there is a change in the nuclear charge; in addition the decaying nucleus receives a recoil momentum. Since the nucleus is surrounded by an electron cloud, the β decay may be accompanied by excitation or ionization of the electron shells. The theory of this phenomenon was first developed by Migdal.[1]

If atoms containing radioactive nuclei are combined into molecules, then in addition to excitation or ionization of the electron shells there is also excitation of vibrational or rotational states of the molecule, or dissociation. It has been shown in many papers[2-4] that the main cause of excitation of molecular vibrations (or dissociation) during β decay is the recoil momentum of the nucleus. These same papers calculate the probability for excitation of vibrational and rotational states of the molecule on the assumption that the molecular vibrations are harmonic. In the paper of Fintak and Wolniewicz[5] the probability for the process is calculated assuming that the atomic interaction is described by the Morse potential.

The present paper calculates the probability for dissociation of a diatomic molecule during β decay of one of its nuclei. It is assumed that the basic mechanism of the dissociation is the excitation of high vibrational and rotational levels because of the recoil momentum (while the electronic state of the molecule is unchanged).

2. The probability for a transition between two states of the molecule, averaged over the β particle distribution, is given by the expression:[4]

$$w = \frac{\iint w(k) P(\varepsilon_e, \vartheta)\, d\varepsilon_e \sin\vartheta\, d\vartheta}{\iint P(\varepsilon_e, \vartheta)\, d\varepsilon_e \sin\vartheta\, d\vartheta}, \tag{1}$$

where $P(\epsilon_e, \vartheta)$ is the distribution of β particles over the energy ϵ_e and the angle ϑ between the directions of the electron and neutrino;

$$w(k) = |V_{if}|^2 = \left|\int \psi_f^*(\mathbf{r}) \exp\left(i \frac{\mu}{M_1} \mathbf{kr}\right) \psi_i(\mathbf{r})\, d\mathbf{r}\right|^2 \tag{2}$$

is the probability for transition between the initial and final states of the molecule for a given recoil momentum $\hbar k$.

The further calculations will be made in the coordinate system fixed to the center of mass of the molecule. It is assumed that before the β decay the molecule was in its ground vibrational and rotational states. Its wave function was then

$$\psi_i(\mathbf{r}) = \left(\frac{2\beta}{\pi r_0^2}\right)^{1/4} \frac{1}{2\sqrt{\pi}\, r} \exp\left[-\frac{\beta}{r_0^2}(r - r_0)^2\right]. \tag{3}$$

Since the final state of the molecule consists of two free atoms, the wave function of the molecule after the β decay can be taken to be the quasiclassical wave function

$$\psi_f(\mathbf{r}) = \left(\frac{2\mu}{\pi\hbar}\right)^{1/2} \frac{1}{r\sqrt{p(r)}} \times \sin\left[\frac{1}{\hbar}\int_{r_1}^{r} p(r)\, dr + \frac{\pi}{4}\right] \frac{\sin[(l + 1/2)\theta + \pi/4]}{\sqrt{\sin\theta}}, \tag{4}$$

where

$$p(r) = \left\{2\mu\left[E - U(r) - \frac{\hbar^2 (l + 1/2)^2}{2\mu r^2}\right]\right\}^{1/2}, \qquad l\theta \gg 1.$$

For $r < r_1$ the wave function of the system is given by the exponentially damped part of expression (4), but with a different normalization factor. It is obvious that this part of the wave function can be neglected in calculating the dissociation probability.

In expressions (2)—(4) the quantity $\mu = M_1 M_2/(M_1 + M_2)$ is the reduced mass of the two nuclei in the molecule; M_1 is the mass of the radioactive nucleus; r is the separation of the nuclei; r_0 is the equilibrium separation; U(r) is the energy of interaction of the atoms in the molecule, and remains unchanged during the process of β decay; E is the internal energy of the final state of the molecule; $\beta = \mu\omega r_0^2/2\hbar \gg 1$; ω is the frequency of vibration of the nuclei; $\mathbf{k} = \mathbf{k}_e + \mathbf{k}_\nu$, $\hbar\mathbf{k}_e$ and $\hbar\mathbf{k}_\nu$ are the momenta of the β particle and the neutrino in the system moving with the radioactive nucleus.

Substituting (3) and (4) in (2), after the substitution $r = r_0x$ we get the following expression for the matrix element of the transition:

$$V_{if} = \left(\frac{2\beta}{\pi^7\varepsilon_0^2}\right)^{1/4}\int_0^\pi \sqrt{\sin\theta} \times \sin\left[\left(l+\frac{1}{2}\right)\theta + \frac{\pi}{4}\right]\int_{x_1}^{\infty} \sin\left[\int_{x_1}^{x} p(x)\,dx + \frac{\pi}{4}\right] \times \exp\left[-\beta(x-1)^2 + i\gamma x\cos\theta\right]\frac{d\theta\,dx}{\sqrt{p(x)}}. \tag{5}$$

Here

$$p(x) = \left[\frac{E - U(x)}{\varepsilon_0} - \frac{(l+1/2)^2}{x^2}\right]^{1/2}, \qquad \varepsilon_0 = \frac{\hbar^2}{2\mu r_0^2},$$

$$\gamma = \frac{\mu r_0 k}{M_1} = \left(\frac{\mu E_{\text{rec}}}{M_1\varepsilon_0}\right)^{1/2},$$

$E_{\text{rec}} = \hbar^2k^2/2M_1$ is the recoil energy of the free nucleus, and $k = |\mathbf{k}_e + \mathbf{k}_\nu|$.

The main contribution to the integral over x in (5) comes from the neighborhood of the point x = 1 ($1 - \beta^{-1/2} \lesssim x \lesssim 1 + \beta^{-1/2}$). As we shall see later, the integral is significantly different from zero only when $\gamma\cos\theta \approx \pm p(1)$, where $p(1)$ is the radial momentum at the point x = 1. In the interval $|x - 1| \lesssim \beta^{-1/2}$, the function $U(x)$ can be expanded in powers of $x - 1$, keeping only the first term in the expansion, since $U'(1) = 0$, while the quadratic term is easily seen to be negligible. Then

$$p(x) = \sqrt{\varepsilon - \frac{(l+1/2)^2}{x^2}}, \qquad \varepsilon = \frac{E - U(1)}{\varepsilon_0},$$

$$U(1) = -D - \frac{\hbar\omega}{2},$$

where D is the dissociation energy of the molecule. The quantity $\hbar\omega/2$ can be neglected compared to D, since $\hbar\omega \ll D$.

The turning point is given by $x_1 = (l + 1/2)/\sqrt{\epsilon}$. If $x_1 < 1 - \beta^{-1/2}$, the lower limit of the x integration can be extended to $-\infty$. The values of l corresponding to this case are given by the inequality $l < l_2 = \sqrt{\epsilon}\,(1 - \beta^{-1/2})$. Since the integrand in (5) is essentially different from zero only when $|x - 1| \lesssim \beta^{-1/2}$ we need not consider cases where the turning point $x_1 > 1 + \beta^{-1/2}$. To these values of x_1 there correspond $l > \sqrt{\epsilon}\,(1 + \beta^{-1/2})$, i.e., the upper limit of the l values is given by $l_{\max} = \sqrt{\epsilon}\,(1 + \beta^{-1/2})$. The range of values of l from l_2 to $l_{\max}$ is of order $\sqrt{\epsilon/\beta}$. This quantity is much smaller than the whole range of values of l. Since the transition probability is a slowly varying function of l, it follows that in the summation we can neglect the contribution from values of l close to the upper end of the interval. Thus it is not necessary to know the precise upper limit for l. In addition since the main contribution to the probability comes from $l \gg 1$, we can replace $l + 1/2$ by l throughout.

Near x = 1, the argument

$$\sin\left[\int_{x_1}^{x} p(x)\,dx + \frac{\pi}{4}\right]$$

can be expanded in powers of $x - 1$:

$$\int_{x_1}^{x} p(x)\,dx = \int_{x_1}^{1} p(x)\,dx + \sqrt{\varepsilon - l^2}\,(x-1) + \frac{l^2}{\sqrt{\varepsilon - l^2}}\frac{(x-1)^2}{2} + \frac{2l^4 - 3l^2\varepsilon}{(\varepsilon - l^2)^{3/2}}\frac{(x-1)^3}{6}. \tag{6}$$

In this expression the quadratic and later terms can be dropped if the condition $l^2/2\beta\sqrt{\epsilon - l^2} \ll 1$ is satisfied. The maximum value of l given by this condition is of order $\sqrt{\epsilon}\,(1 - \epsilon/8\beta^2)$. The interval of values of l from this value to $l_{\max}$ is of order $\epsilon^{3/2}/8\beta^2$. In practice, the relation $\epsilon^{3/2}/8\beta^2 \sim \sqrt{\epsilon/\beta}$ is satisfied for all molecules. But as we showed earlier, we can neglect the contribution of l values which are separated from the upper limit $l_{\max}$ by an amount $\sim \sqrt{\epsilon/\beta}$. Thus we keep in (6) only the first two terms, and set the upper limit of the summation over l equal to $l_2 = \sqrt{\epsilon}\,(1 - \beta^{-1/2})$.

After integrating over x the expression (5) becomes

$$V_{if} = -\frac{i(8\pi^3\varepsilon_0^2\beta)^{-1/4}}{(\varepsilon - l^2)^{1/4}}\int_0^\pi d\theta\sqrt{\sin\theta}\sin\left(l\theta + \frac{\pi}{4}\right)\exp(i\gamma\cos\theta) \times \left\{\exp\left[-\frac{1}{4\beta}(\gamma\cos\theta + \sqrt{\varepsilon - l^2})^2 + ip_1 + i\frac{\pi}{4}\right] - \exp\left[-\frac{1}{4\beta}(\gamma\cos\theta - \sqrt{\varepsilon - l^2})^2 - ip_1 - i\frac{\pi}{4}\right]\right\}, \tag{7}$$

where $p_1 = \int_{x_1}^{1} p(x)\,dx$. The integral (7) can be calculated by the saddle point method. The points of stationary phase θ_0 and $\pi - \theta_0$ are given by $\sin\theta_0 = \pm l/\gamma$. In the integration over θ we neglect all those terms in (7) for which the saddle point does not fall in the interval $(0, \pi)$; in addition we neglect terms containing rapidly oscillating factors of the form $\cos(p_1 + \sqrt{\gamma^2 - l^2} - l\theta_0 + \pi l)$. Substituting the expression for V_{if} in (2) and summing over l from 0 to l_2, we find for the probability of transition of the molecule to states with energies lying in the interval E to E + dE,

$$dw(k)=\sum_l \frac{l}{2\gamma\sqrt{2\pi\beta}}\,\frac{\exp\left[-\left(\sqrt{\gamma^2-l^2}-\sqrt{\varepsilon-l^2}\right)^2/2\beta\right]}{(\varepsilon-l^2)^{1/2}(\gamma^2-l^2)^{1/2}}\,d\varepsilon. \tag{8}$$

Strictly speaking, the lower limit for the summation over l is not zero, since the expression is valid only when the condition for quasiclassical behavior of the wave function is satisfied: $l\theta \gg 1$. But near the points of stationary phase, θ_0 and $\pi-\theta_0$, this condition is satisfied for sufficiently small l. Actually it follows from the equation $\sin\theta_0 = l/\gamma$ that for sufficiently small l, $\theta_0 \approx l/\gamma$. From the condition $l\theta \gg 1$, we get $l_{\min} \sim \gamma^{1/2} \sim \epsilon^{1/4}$ (since (8) is different from zero only for $\gamma^2 \sim \epsilon$). Since the range of values of l from the exact lower limit $l = 0$ to $l_{\min}$ is much smaller than the whole range of l values, and since furthermore the transition probability is a slowly varying function of l, we can take $l = 0$ for the lower limit.

Let us compare this result with the result from classical mechanics: the total probability for dissociation is w = 0 for $\mu E_{\mathrm{rec}}/M_1 < D$, and w = 1 for $\mu E_{\mathrm{rec}}/M_1 > D$. Let us integrate (8) over ϵ and replace the summation over l by an integration. Noting that the main contribution to the integral comes from $\epsilon \sim \gamma^2$ (since $\gamma^2/8\beta = (\mu/2M_1)E_{\mathrm{rec}}/\hbar\omega \sim D/\hbar\omega \gg 1$), we expand the exponential function in powers of $\epsilon-\gamma^2$. Then the total probability for dissociation is

$$w=\frac{1}{4\gamma\sqrt{2\pi\beta}}\int\limits_{D/\varepsilon_0}^{\infty} d\varepsilon\int\limits_0^{\gamma}\frac{dl^2}{\gamma^2-l^2}\exp\left[-\frac{(\varepsilon-\gamma^2)^2}{8\beta(\gamma^2-l^2)}\right]$$

or, after the substitutions $\epsilon-\gamma^2 = z$ and $(\gamma^2-l^2)^{-1} = t$,

$$w=\frac{1}{4\gamma\sqrt{2\pi\beta}}\int\limits_{D/\varepsilon_0-\gamma^2}^{\infty} dz\int\limits_{1/\gamma^2}^{\infty}\frac{dt}{t}\exp\left\{-\frac{z^2t}{8\beta}\right\}. \tag{9}$$

Then (to exponential accuracy)

$$w=\begin{cases}0, & \gamma^2<D/\varepsilon_0-4\sqrt{\beta D/\varepsilon_0}\\ 1, & \gamma^2>D/\varepsilon_0+4\sqrt{\beta D/\varepsilon_0}\end{cases}. \tag{10}$$

After substitution of the values of ϵ_0 and β, the last inequalities take the form

$$\mu E_{\mathrm{rec}}/M_1 \lesseqgtr D\left(1\mp 2\sqrt{\hbar\omega/D}\right).$$

In the intermediate range of values of E_{rec}, the integral (9) can easily be computed numerically. As we see from (10), the limiting transition $\hbar \to 0$ gives the formulas of classical mechanics.

3. Now we average expression (8) over the distribution of β particles. We shall consider only allowed β transitions. For such transitions the distribution of the β particles over energy and over the angle between the directions of the electron (positron) and neutrino are given by the following expression:[6]

$$P(\varepsilon_e,\vartheta)=\mathrm{const}\cdot F(\varepsilon_e)(E_0-\varepsilon_e)^2\varepsilon_e k_e\left(1+\alpha\frac{v_e}{c}\cos\vartheta\right). \tag{11}$$

Here $\epsilon_e = c\sqrt{\hbar^2k_e^2+m^2c^2}$, m and v_e are the energy, mass and velocity of the β particle; E_0 is the β decay energy; α is a dimensionless constant which is characteristic of the particular nucleus;

$$F(\varepsilon_e)=2(1+\delta)\left[(2\delta)!\right]^{-2}(2k_eR)^{2\delta-2}e^{\pi\eta}\left|(\delta-1+i\eta)!\right|^2$$

is the function which takes account of the effect of the Coulomb field on the emerging β particle; $\delta = \sqrt{1-(Ze^2/\hbar c)^2}$; $\eta = \pm Ze^2/\hbar v_e$ (the signs ± refer to positron and electron); Z is the charge of the radioactive nucleus after the decay; R is the nuclear radius.

We replace ϵ_e and ϑ by new integration variables s and γ:

$$s=\frac{\varepsilon_e}{mc^2},\qquad s_0=\frac{E_0}{mc^2},\qquad \gamma^2=(k_e^2+k_\nu^2+2k_ek_\nu\cos\vartheta)\frac{\mu^2r_0^2}{M_1^2}.$$

Substituting (8) and (11) in (1) and replacing the sum over l by an integral, we find for the probability per unit energy range (averaged over the β particle distribution)

$$\frac{dw}{d\varepsilon}=\frac{(2\pi\beta)^{-1/2}\varepsilon_0}{N\varepsilon D}\int\limits_1^{s_0}ds\int\limits_0^{\sqrt{\varepsilon}}dl^2\int\limits_{\gamma_1}^{\gamma_2}d\gamma F(s)\left\{2s(s_0-s)\right.$$
$$\left.+\alpha\left[\frac{\gamma^2\varepsilon_0}{\varkappa^2D}-(s_0-s)^2-s^2+1\right]\right\}$$
$$\times\frac{\exp\left[-\left(\sqrt{\gamma^2-l^2}-\sqrt{\varepsilon-l^2}\right)^2/2\beta\right]}{(\varepsilon-l^2)^{1/2}(\gamma^2-l^2)^{1/2}};$$
$$N=16\varkappa^2\int\limits_1^{s_0}F(s)\sqrt{s^2-1}\,(s_0-s)^2\,s\,ds,$$
$$\varkappa^2=\frac{\mu m^2c^2}{2DM_1^2},\qquad \gamma_{1,2}=\frac{\mu r_0mc}{M_1\hbar}\left|\sqrt{s^2-1}\mp(s_0-s)\right|. \tag{12}$$

The integration over γ is easily done by expanding the exponential function near its maximum $\gamma = \sqrt{\epsilon}$ and removing the value of the slowly varying function at that point from under the integral sign. After integrating (12) over γ and introducing new variables $\sigma = (E+D)/D \equiv \epsilon\epsilon_0/D$ and $y = l^2/\epsilon \equiv l^2\epsilon_0/\sigma D$, we get

$$\frac{dw}{d\sigma}=\frac{1}{2N}\int\limits_1^{s_0}ds\,F(s)\left\{2s(s_0-s)+\alpha\left[\frac{\sigma}{\varkappa^2}-(s_0-s)^2\right.\right.$$
$$\left.\left.-s^2+1\right]\right\}\int\limits_0^1\frac{dy}{\sqrt{1-y}}\left[\Phi\left(\frac{u_2}{\sqrt{1-y}}\right)-\Phi\left(\frac{u_1}{\sqrt{1-y}}\right)\right];$$

$$\Phi(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2}\, dt,$$

$$u_{1,2} = \left(\frac{2D}{\hbar\omega}\right)^{1/2} [\varkappa \,|\, \sqrt{s^2 - 1} \mp (s_0 - s) \,| - \sqrt{\sigma}].$$

This expression can be transformed to a single integral. We finally have

$$\frac{dw}{d\sigma} = \frac{1}{N} \int_1^{s_0} ds\, F(s) \Big\{ 2s(s_0 - s) + \alpha \Big[\frac{\sigma}{\varkappa^2} - (s_0 - s)^2 - s^2 + 1 \Big] \Big\} \times \Big\{ \Phi(u_2) - \Phi(u_1) - \frac{u_2}{\sqrt{\pi}} \operatorname{Ei}(-u_2^2) + \frac{u_1}{\sqrt{\pi}} \operatorname{Ei}(-u_1^2) \Big\}$$

$$\operatorname{Ei}(-x) = -\int_x^\infty \frac{e^{-t}}{i}\, dt. \tag{13}$$

The remaining integration in (13) can be done numerically.

4. Obviously not every molecule containing a β-active nucleus can dissociate during β decay. Dissociation is possible only when the energy of the β decay exceeds some definite value. From (12) we see that the probability of dissociation for a given recoil momentum $\hbar k$ is substantially different from zero only when $\gamma^2 \approx \epsilon$. Setting $\epsilon = \epsilon_{\min} = D/\epsilon_0$ in this equation, we find the minimum recoil energy of the β-active nucleus (mass M_1) for which dissociation becomes energetically possible: $E_{\text{rec}} \geq D(M_1 + M_2)/M_2$.

Expressing E_{rec} in terms of the energy E_0 liberated in the β decay ($E_0 = E_\beta + mc^2$; E_β is the maximum kinetic energy of the β particle), we get the required relation:

$$E_0 \geqslant \Big[2\frac{M_1}{M_2}(M_1 + M_2)c^2 D + m^2c^4 \Big]^{1/2}. \tag{14}$$

When the reverse inequality is satisfied, there is only excitation of vibrational and rotational levels in the region of the discrete spectrum ($E < 0$). In this connection we note that formula (13) for the probability density of the transition to the state with energy $E \equiv D(\sigma - 1)$ can be used for other purposes than calculating the dissociation probability ($\sigma > 1$). It is obvious that it is also applicable for finding the probability of excitation of high vibrational and rotational levels ($\sigma < 1$), so long as the quasiclassical approximation for the wave function is still valid.

As we see, formula (13) can be written in the form $dw/d\sigma = A(\sigma) + \alpha B(\sigma)$. The quantity α depends on the constants in the interaction of the electron-neutrino field with the nucleons:[6]

$$\alpha = \frac{C_V^2 \,|\int 1|^2 - {}^1/_3\, C_A^2 \,|\int \sigma|^2}{C_V^2 \,|\int 1|^2 + C_A^2 \,|\int \sigma|^2}; \qquad -1 \leqslant \alpha \leqslant 1.$$

This quantity can be found experimentally from measurements of the electron-neutrino angular correlation. In those rare cases where α is known, $dw/d\sigma$ is easily calculated if we know the molecular parameters D and ω. In general one can calculate only the quantities $A(\sigma)$ and $B(\sigma)$ and give limits on $dw/d\sigma$ corresponding to the values $\alpha = \pm 1$. As an example we calculated the quantities $A(\sigma)$ and $B(\sigma)$ for three molecules: $I^{127}I^{130*}$, $Ca^{40}I^{133*}$, and $Sn^{120}O^{19*}$ (the asterisk denotes the radioactive nucleus).

Figure 1 shows the variation of $A(\sigma)$ and $B(\sigma)$ for the dissociation of the I_2 molecule ($E \equiv D(\sigma - 1)$ is the relative energy of the two atoms of the molecule). The actual values of $dw/d\sigma$ should be contained between the dashed curves I and II. In this and the following two figures, the curves I and II correspond to $\alpha = \pm 1$.

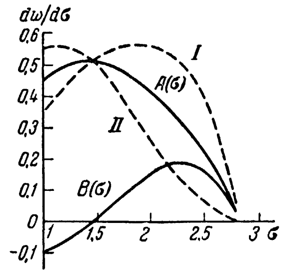

FIG. 1

Figures 2 and 3 give similar graphs for CaI and SnO. For the case of CaI we also calculated the probability of transition of the molecule to highly excited states ($\sigma < 1$). In view of the large decay energy of O^{19} ($E_0 = 5$ MeV), there is practically no excitation of vibrations of the SnO molecule with $E < 0$. The total probability for all processes (mainly dissociation) is

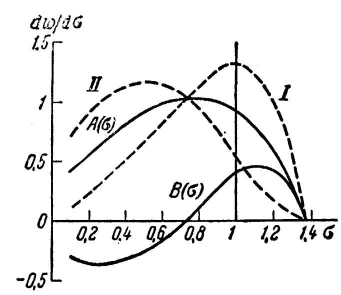

FIG. 2

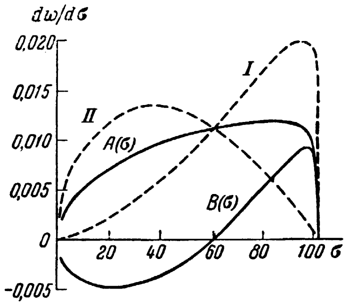

FIG. 3

$$w = \int_0^\infty \frac{dw}{d\sigma} d\sigma = 1,$$

as we see from Fig. 3. Here

$$\int_0^\infty A(\sigma)\, d\sigma = 1, \qquad \int_0^\infty B(\sigma)\, d\sigma = 0,$$

which means that the normalization condition is satisfied for any value of α.

We note that these results can be used for finding the value of α. In fact, by measuring the total dissociation probability w_{exp}, from the relation

$$w_{\text{exp}} = \int_1^\infty A(\sigma)\, d\sigma + \alpha \int_1^\infty B(\sigma)\, d\sigma,$$

one can determine α. In cases like that shown in Fig. 3, one should measure the dissociation probability either for a given E or over some range of values of E, since the total probability for dissociation is here practically equal to unity. In this case the distribution of the dissociated atoms over mass M_1 and energy E_1 in the lab system and the distribution of these same atoms with energy E in the system of the center of mass of the molecule practically coincide, since $M_1 \ll M_2$ and the energy of the motion of the center of the molecule is small compared to $E_1 = EM_1/\mu \approx E$.

One might expect that the proposed method for determining α is simpler than the usual method, which requires the measurement of the angle of emergence of the radioactive nucleus and the β particle. In various cases similar to those given in Figs. 1 and 2, where the recoil energy is not very large, there is no need to find the energy distribution of the atoms from the dissociated molecules. To determine α one need only measure the total probability for dissociation of the molecule during the β decay of one of its nuclei.

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