

## Influence of Extranuclear Fields on Angular Correlations\*

F. COESTER

*Department of Physics, State University of Iowa, Iowa City, Iowa*

(Received October 5, 1953)

General formulas exhibiting the influence of extranuclear fields on angular correlations of two or three successive  $\gamma$  rays are derived from first principles. They are applicable to polarization correlations as well as to directional correlations. The effect of transitions in the electron shell during the life of the intermediate nucleus is discussed briefly.

### I. INTRODUCTION

THE qualitative picture of  $\gamma$ - $\gamma$  angular correlations is simple enough and well known.<sup>1</sup> The source nuclei decay, each emitting two  $\gamma$  rays in succession. Initially their spins have random orientation, but the ensemble of nuclei having emitted the first  $\gamma$  ray in a definite direction has a nonuniform distribution of spin orientations. The second  $\gamma$  ray is therefore emitted with a nonisotropic angular distribution with respect to the first. The action of extranuclear fields on the magnetic moments and the electric quadrupole moments of the intermediate nuclei may significantly change their spin orientations and thus affect the angular correlation.

In the quantitative theory first given by Hamilton<sup>2</sup> and refined by many authors<sup>3</sup> one does not have, however, a detailed description of the time development corresponding to the picture just sketched. One merely calculates from the time-dependent Schrödinger equation the relative probability for having two  $\gamma$  rays emitted in specified directions during a long time  $t$ . Goertzel's theory<sup>4</sup> of the influence of extranuclear fields follows the same lines. The splitting of the nuclear levels rather than any spin reorientation in time appears to be directly responsible for the influence of extranuclear fields on angular correlations. This theory is necessarily restricted to time independent interactions. Recently Abragam and Pound<sup>5</sup> have given a prescription which modifies Hamilton's formula on the basis of the physical picture in order to describe the influence of time-dependent extranuclear fields as well. Fano<sup>6</sup> has suggested a different approach which promises a consistent quantitative theory in close parallel to the physical picture. The decaying nuclei and the emitted radiation can be described by a density matrix.<sup>7</sup> When extranuclear fields are negligible it suffices to consider only the initial and final density matrix connected by

the  $S$  matrix.<sup>8</sup> In any case the angular correlation can be written in the form

$$W = \text{Tr}(\epsilon\rho), \quad (1)$$

[see Eq. (I2)], where  $\rho$  is the final density matrix. But if extranuclear fields are effective, the final density matrix must be obtained in terms of an explicit description of the intermediate stages.

It is the purpose of this note to provide an explicit description in time of the decaying nuclei along the lines suggested by Fano. While many of our considerations apply to all kinds of cascade decay, we restrict ourselves at first to  $\gamma$  decay of long-lived isomers for the sake of definiteness in the exposition. Gamma cascades preceded by  $\beta$  decay or  $K$  capture are discussed in Sec. IV.

### II. DENSITY MATRICES IN SUCCESSIVE $\gamma$ DECAY

The density matrix  $\rho$  in (1) describes the ensemble of all the nuclei in the source. It can be written in the form

$$\rho = \frac{1}{N} \sum_{\alpha=1}^N \rho^{\alpha},$$

where  $\alpha$  labels the individual nuclei in the source and  $\rho^{\alpha}$  is the density matrix describing the  $\alpha$ th nucleus.

We consider a sequence of nuclear levels  $a, b, c$  with angular momentum  $j_a, m_a, \dots$ . The density matrix  $\rho^{\alpha}$  describes the system containing the nucleus  $\alpha$ , its environment and the emitted radiation. The matrix  $\bar{\rho}^{\alpha}$  is obtained by taking the trace of  $\rho^{\alpha}$  with respect to all photon energies. Since  $\epsilon$  is the unit matrix with respect to the photon energies it is  $\bar{\rho}^{\alpha}$  which we need to calculate the angular correlation. We assume for the sake of simplicity that only transitions between adjacent levels are possible. At the time  $t=0$  all the nuclei are in the level  $a$  and no photons are present. The matrix  $\bar{\rho}^{\alpha}(t)$  then has no off-diagonal elements connecting different nuclear levels and may be written in the form

$$\bar{\rho}^{\alpha} = \rho_a^{\alpha} + \rho_b^{\alpha} + \rho_c^{\alpha} + \dots \quad (2)$$

Each term on the right-hand side of (2) has only matrix elements with respect to the substates of one level  $a, b, c, \dots$ , respectively. If the nucleus is in  $b$  there is always one photon, in  $c$  there are two photons and so

<sup>8</sup> F. Coester and J. M. Jauch, *Helv. Phys. Acta* **26**, 3 (1953) quoted as I in the following.

\* Supported in part by the U. S. Atomic Energy Commission.

<sup>1</sup> See, for instance, H. Fraunfelder, *Ann. Rev. Nuclear. Sci.* **2**, 129 (1953).

<sup>2</sup> D. R. Hamilton, *Phys. Rev.* **58**, 122 (1940).

<sup>3</sup> For references see reference 1.

<sup>4</sup> G. Goertzel, *Phys. Rev.* **70**, 897 (1946). See, also, K. Alder, *Helv. Phys. Acta* **25**, 235 (1952).

<sup>5</sup> A. Abragam and R. V. Pound, *Phys. Rev.* **92**, 943 (1953).

<sup>6</sup> U. Fano, *Natl. Bur. Standards Rept.* 1214, 1951 (unpublished); *Phys. Rev.* **90**, 577 (1953).

<sup>7</sup> For a definition see, for instance, R. C. Tolman, *Principles of Statistical Mechanics* (Oxford University Press, London, 1938), Chap. 9.

on. The Hamiltonian of the system is  $H_0 + H_e^\alpha(t) + H$ , where  $H_0$  is the energy of the bare nucleus and the free photons,  $H$  is the interaction of the nucleus with the radiation field, and  $H_e^\alpha(t)$  is the Hamiltonian of the environment in interaction with the nucleus. It has an index  $\alpha$  since different nuclei do not in general have identical environments. In the interaction representation the Schrödinger equation is

$$i\partial\Psi^\alpha/\partial t = [H_e^\alpha + H(t)]\Psi^\alpha, \quad (3)$$

where

$$H(t) = e^{iH_0 t} H e^{-iH_0 t}. \quad (4)$$

$H_e^\alpha$  commutes with  $H_0$ . It is therefore the same in the interaction representation and in the Schrödinger representation. As a consequence of (3)  $\rho^\alpha$  satisfies the equation<sup>9</sup>

$$i\partial\rho^\alpha/\partial t = [(H_e^\alpha + H(t)), \rho^\alpha(t)]. \quad (5)$$

No attempt is made to integrate (5) from  $t=0$  to large  $t$ . In fact this solution would not describe the physical situation since the system is under continuous observation. The emitted photons are at least in principle observed at all times; the system is therefore described by a density matrix  $\rho^\alpha(t)$  which is diagonal in the photon numbers.

In order to get a useful equation for  $\bar{\rho}^\alpha(t)$  from (5), we integrate (5) by perturbation theory between  $t$  and  $t+\delta t$ . The interval  $\delta t$  is so small that  $\langle\delta t H_e^\alpha\rangle \ll 1$ , and  $H_e^\alpha(t)$  is approximately constant in this interval. Further  $\delta t$  is small compared to the lifetimes of the nuclear levels but large compared to the reciprocal frequency of the  $\gamma$  rays. We have then

$$\begin{aligned} \rho^\alpha(t+\delta t) &= \rho^\alpha(t) - i\delta t [H_e^\alpha, \rho^\alpha] - i \int_t^{t+\delta t} dt' [H(t'), \rho^\alpha(t)] \\ &\quad - \int_t^{t+\delta t} dt' \int_t^{t'} dt'' [H(t''), [H(t'), \rho^\alpha(t)]]. \end{aligned} \quad (6)$$

Since the time dependence of  $H(t)$  is known according to (4) we can carry out the integrations in (6) explicitly and then take the trace with respect to the photon energies. The matrix  $\rho^\alpha(t)$  is diagonal with respect to the photon number; therefore the first-order term in  $H$  does not contribute and the result is

$$\bar{\rho}^\alpha(t+\delta t) - \bar{\rho}^\alpha(t) = -\{\gamma [H_e^\alpha, \bar{\rho}^\alpha] + \gamma \bar{\rho}^\alpha - 2\pi H \bar{\rho}^\alpha H\} \delta t, \quad (7)$$

where  $\gamma$  is a diagonal matrix whose eigenvalues  $\gamma_a, \gamma_b, \dots$  are the reciprocal lifetimes of the nuclear levels. For instance,

$$\gamma_a = 2\pi \sum_\xi \sum_M \sum_{m_b} | \langle m_a j_a | H | j_b \xi m_b M \rangle |^2; \quad (8)$$

<sup>9</sup> A similar equation has been discussed recently by R. K. Wangness and F. Bloch, Phys. Rev. **89**, 728 (1953) [Eq. (2.4)]. Our treatment is analogous to theirs in many respects.

$\xi$  and  $M$  are photon quantum numbers.<sup>10</sup> Since

$$\langle j_a m_a | H | j_b \xi m_b M \rangle = \langle j_b L m_b M | j_a m_a \rangle H_{ba}(\xi), \quad (9)$$

$\gamma_a$  is independent of  $m_a$ . More explicitly, (7) may be written in the form

$$\partial\rho_a^\alpha/\partial t = -i[H_e^\alpha, \rho_a^\alpha] - \gamma_a \rho_a^\alpha, \quad (10a)$$

$$\partial\rho_b^\alpha/\partial t = -i[H_e^\alpha, \rho_b^\alpha] - \gamma_b \rho_b^\alpha + 2\pi \mathbf{H}_{ba} \rho_a^\alpha \mathbf{H}_{ab}. \quad (10b)$$

Similar equations hold for  $\rho_c^\alpha$  and further terms if any. One can easily verify from (10) that the trace of  $\bar{\rho}^\alpha = \rho_a^\alpha + \rho_b^\alpha + \dots$  is constant in time since

$$2\pi \text{Tr}(\mathbf{H}_{ba} \rho_a^\alpha \mathbf{H}_{ab}) = \gamma_a \rho_a^\alpha. \quad (11)$$

The solution of (10a), (10b),  $\dots$  can easily be given in terms of a matrix  $U^\alpha(t, t_0)$  which satisfies the equation

$$i\partial U^\alpha(t, t_0)/\partial t = H_e^\alpha U^\alpha(t, t_0), \quad (t > t_0), \quad (12)$$

and the initial condition  $U^\alpha(t_0, t_0) = 1$ . From (10a) and (12) we find

$$\rho_a^\alpha(t) = e^{-\gamma_a t} U^\alpha(t, 0) \rho_a^\alpha(0) U^\alpha(t, 0)^+. \quad (13a)$$

Since  $\rho_a^\alpha(t)$  is known, (10b) can be integrated. The result is

$$\rho_b^\alpha(t) = 2\pi \int_0^t dt' e^{-\gamma_b(t-t')} U^\alpha(t, t') \mathbf{H}_{ba} \rho_a^\alpha(t') \mathbf{H}_{ab} U^\alpha(t, t')^+. \quad (13b)$$

In the same way we obtain the density matrix at any level. For instance we get  $\rho_c^\alpha(t)$  by substituting  $a \rightarrow b$ ,  $b \rightarrow c$  in Eq. (13b).

### III. EXPLICIT ANGULAR CORRELATION FUNCTIONS

If the level  $c$  is the ground level ( $\gamma_c = 0$ ), the correlation of the two  $\gamma$  rays is, according to (1),

$$W = \text{const Tr}\{\epsilon(\Omega_1 \Omega_2) \rho_c(t)\}, \quad (14)$$

where  $\epsilon$  is the unit matrix with respect to all variables except those describing the observed photons,

$$\rho_c = \frac{1}{N} \sum_\alpha \rho_c^\alpha, \quad (15)$$

and  $t$  is in (14) by definition the total duration of the experiment. We shall see later that the angular correlation is actually independent of  $t$ , as expected. From (14), (15), and (13c) (not written out) it follows that

$$W = \text{const Tr}\left\{ \epsilon(\Omega_1 \Omega_2) \int_0^t dt' \mathbf{H}_{cb} \rho_b(t') \mathbf{H}_{bc} \right\}. \quad (16)$$

We assume that for every environment there is a large number of nuclei uniformly distributed over the sub-

<sup>10</sup> We follow the notation of I.

states of the level  $a$  and find from (13a) and (13b)

$$\rho_a(t) = \frac{1}{N} \sum_{\alpha} 2\pi \int_0^t dt' e^{-\gamma_b(t-t')} e^{-\gamma_a t'} \rho_a(0) \times U^{\alpha}(t, t') \mathbf{H}_{ba} \mathbf{H}_{ab} U^{\alpha}(t, t')^{\dagger}. \quad (13b')$$

$\rho_a(0)$  is here merely a normalization factor. Inserting (13b') in (16) we write  $W$  in the form

$$W = \text{const} \int_0^t dt' \int_0^{t'} dt'' e^{-\gamma_b(t-t'')} e^{-\gamma_a t''} \times \frac{1}{N} \sum_{\alpha} \text{Tr} \{ \epsilon(\Omega_1 \Omega_2) \mathbf{H}_{cb} U^{\alpha}(t', t'') \times \mathbf{H}_{ba} \mathbf{H}_{ab} U^{\alpha}(t', t'')^{\dagger} + \mathbf{H}_{bc} \}. \quad (17)$$

If  $c$  is not the ground level, but further decays are not observed we must replace (14) by

$$W = \text{const} \text{Tr} \{ \epsilon(\Omega_1 \Omega_2) (\rho_c + \rho_d + \dots) \}. \quad (18)$$

Equation (18) leads again to (17) since by (10) and (11)

$$\partial \text{Tr} \{ \epsilon(\rho_d + \dots) \} / \partial t = \gamma_c \text{Tr}(\epsilon \rho_c). \quad (19)$$

In order to evaluate the trace in (17) we use a representation specified by the quantum numbers  $jm$  for the nucleus,  $\xi M$  for the photons, and  $\kappa$  for the atomic environment. The efficiency matrix is in this representation:

$$\begin{aligned} & (\kappa m_c \xi_2 M_2 \xi_1 M_1 | \epsilon(\Omega_1 \Omega_2) | \kappa' m_c' \xi_2' M_2' \xi_1' M_1') \\ &= \delta(\kappa, \kappa') \delta(m_c, m_c') \sum_{k_1 n_1} (-1)^{L_1' - M_1'} \\ & \times (L_1 L_1' M_1 - M_1' | k_1 n_1) (\xi_1 | \epsilon(k_1, n_1; \Omega_1) | \xi_1') \\ & \times \sum_{k_2 n_2} (-1)^{L_2' - M_2'} (L_2 L_2' M_2 - M_2' | k_2 n_2) \\ & \times (\xi_2 | \epsilon(k_2, n_2; \Omega_2) | \xi_2'). \quad (20) \end{aligned}$$

The matrix elements of  $U^{\alpha}$  describing the influence of the extranuclear fields are combined in the expression

$$\begin{aligned} & (\bar{k} \bar{n} \bar{\kappa} \bar{\kappa}' | G^{\alpha}(t', t'') | k n \kappa \kappa') \\ &= \sum_{m_b m_b'} \sum_{\bar{m}_b \bar{m}_b'} (\bar{m}_b \bar{\kappa} | U^{\alpha}(t', t'') | m_b \kappa) \\ & \times (\bar{m}_b' \bar{\kappa}' | U^{\alpha}(t', t'') | m_b' \kappa') (-1)^{i_b - m_b'} \\ & \times (j_b j_b m_b - m_b' | k n) (-1)^{i_b - \bar{m}_b'} \\ & \times (j_b j_b \bar{m}_b - \bar{m}_b' | \bar{k} \bar{n}). \quad (21) \end{aligned}$$

The matrix elements of the Hamiltonian are given by (9) and a similar expression for the second transition. All sums over magnetic quantum numbers are carried

out using Eq. (IB6).<sup>11</sup> The result is

$$W(\Omega_1 \Omega_2) = \text{const} \sum_{k n} \sum_{\bar{k} \bar{n}} (00 | F_{cb}(\Omega_2) | \bar{k} \bar{n}) \times (\bar{k} \bar{n} | G | k n) (k n | F_{ba}(\Omega_1) | 00), \quad (22)$$

where

$$(\bar{k} \bar{n} | G | k n) = \frac{1}{N} \sum_{\alpha} \sum_{\kappa \bar{\kappa}} \int_0^t dt' \int_0^{t'} dt'' \gamma_b e^{-\gamma_b(t-t'')} \times \gamma_a e^{-\gamma_a t''} (\bar{k} \bar{n} \bar{\kappa} \bar{\kappa}' | G^{\alpha}(t', t'') | k n \kappa \kappa'), \quad (23)$$

and

$$\begin{aligned} & (k n | F_{ba}(\Omega_1) | k' n') \\ &= \sum_{k_1 n_1} H_{ba}(\xi_1) H_{ba}(\xi_1') (\xi_1 | \epsilon(k_1, n_1; \Omega_1) | \xi_1') \\ & \times (k k_1 n n_1 | k' n') (j_a j_a | \Gamma(j_b j_b L_1 L_1' k') | k k_1). \quad (24) \end{aligned}$$

The matrix element  $H_{ba}(\xi)$  is real.<sup>12</sup>  $F_{cb}$  is obtained by substituting  $a \rightarrow b$ ,  $b \rightarrow c$ , and  $1 \rightarrow 2$  in (24). For directional correlations,<sup>13</sup>

$$(\xi | \epsilon(k, n; \Omega) | \xi') = [4\pi / (2k+1)]^{\frac{1}{2}} C_k(\xi, \xi') Y_k^n(\Omega)^*. \quad (25)$$

In the absence of any interaction of the nucleus with its environment  $U^{\alpha}$  is the unit matrix. Equation (22) reduces then to the unperturbed correlation:

$$W(\Omega_1 \Omega_2) = \text{const} \sum_{k n} (00 | F_{cb}(\Omega_2) | k n) (k n | F_{ba}(\Omega_1) | 00). \quad (26)$$

If  $H_e^{\alpha}$  is time-independent, the eigenvalues of  $U^{\alpha}(t', t'')$  are of the form  $e^{i\omega(t'-t'')}$ .  $U^{\alpha}(t', t'')$  and therefore  $G^{\alpha}(t', t'')$  depend only on the difference  $t' - t''$  and not on  $t'$  and  $t''$  separately. If on the other hand  $H_e^{\alpha}$  is the interaction with randomly fluctuating fields such as occur in liquids,  $G(t', t'') = N^{-1} \sum_{\alpha} G^{\alpha}(t', t'')$  again depends only on the difference  $t' - t''$ . This is a consequence of the ergodic hypothesis which says that the ensemble average  $N^{-1} \sum_{\alpha}$  is equivalent to a time average. Equation (23) may then be simplified somewhat. The duration of the experiment is always long compared to the lifetime of the intermediate state  $\gamma_b t \gg 1$ . If it is also long compared to the lifetime of the source  $\gamma_a t \gg 1$  we may replace  $t$  in (23) by  $\infty$  and use the

<sup>11</sup> There is a misprint in the last line of (IB6). Instead of  $W(acb'f; b\kappa)$  read  $W(acb'f; b\kappa)$ . Delete the equality sign at the end of the previous line.

*Note added in proof.*—Coefficients closely related to the  $\Gamma$  coefficients of I have been introduced independently by many authors in unpublished papers. Several of these definitions and their relations are given by H. A. Jahn and J. Hope [Phys. Rev. **93**, 318 (1954)]. The  $\Gamma$  coefficients are related to the Wigner  $9j$  symbol according to

$$(cc' | \Gamma(aa'bb'd) | ef) = \left\{ \begin{matrix} a'e & a \\ b'f & b \\ c'd & c \end{matrix} \right\}.$$

The Wigner  $9j$  symbol is identical with Fano's  $\alpha$  coefficient (Nat. Bur. Standards Report 1214, 1951, unpublished).

<sup>12</sup> S. P. Lloyd, Phys. Rev. **81**, 161 (1951); F. Coester, Phys. Rev. **89**, 619 (1953).

<sup>13</sup> See Eqs. (IC2) (IC3), and (IC3).

relation

$$\int_0^\infty dt' \int_0^{t'} dt'' \dots = \int_0^\infty dt'' \int_0^{t''} dt' \dots, \quad (27)$$

which gives

$$(\bar{k}\bar{n}|G|kn) = \sum_{\kappa\bar{\kappa}} \int_0^\infty dt \gamma_b e^{-\gamma_b t} (\bar{k}\bar{n}\bar{\kappa}\bar{\kappa}|G(t)|kn\kappa\kappa). \quad (28)$$

At this point we establish the connection with the work of Abragam and Pound<sup>5</sup> by the relation

$$\sum_{\kappa\bar{\kappa}} (k_2\mu_2\bar{\kappa}\bar{\kappa}|G(t)|k_1\mu_1\kappa\kappa) = \frac{[(2k_1+1)(2k_2+1)]^{\frac{1}{2}}}{2j_b+1} \times \text{III}(k_1, k_2, \mu_1, \mu_2, t). \quad (29)$$

The assumption  $\gamma_a t \gg 1$  which led to (28) is in general not justified. On the other hand the lifetime of the source is large compared to that of the intermediate state,  $\gamma_a \ll \gamma_b$ . We may therefore use in (22) the transformation

$$\int_0^t dt' \int_0^{t'} dt'' \dots = \int_0^t dt'' \int_0^{t''} dt' \dots - \int_0^t dt'' \int_{t-t''}^{t''} dt' \dots \quad (30)$$

and neglect the second term on the right-hand side, since it is small of order  $\gamma_a/\gamma_b$  compared to the first one. Since the factor  $1 - e^{-\gamma_a t}$  does not affect the correlation, we may still use (28) in (22).

So far we have assumed that the resolving time of the coincidence circuit is large compared to the lifetime of the intermediate nucleus. If this is not the case and delayed coincidences are counted, we have, instead of (28):

$$(\bar{k}\bar{n}|G|kn) = \sum_{\kappa\bar{\kappa}} \int_{\tau_1}^{\tau_2} dt e^{-\gamma_b t} \gamma_b (\bar{k}\bar{n}\bar{\kappa}\bar{\kappa}|G(t)|kn\kappa\kappa), \quad (31)$$

where  $\tau_1$  and  $\tau_2$  are the minimum and maximum delay times; that means the second quantum is delayed with respect to the first by a time  $t$  between  $\tau_1$  and  $\tau_2$ .<sup>14</sup>

Triple correlation functions can easily be derived with the same methods. In analogy to (17), we get

$$W = \text{const} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt''' e^{-\gamma_c(t'-t''')} \times e^{-\gamma_b(t''-t''')} e^{-\gamma_a t'''} \frac{1}{N} \sum_{\alpha} \text{Tr} \{ \epsilon(\Omega_1 \Omega_2 \Omega_3) \times \mathbf{H}_{ac} U^{\alpha}(t', t'') \mathbf{H}_{cb} U^{\alpha}(t'', t''') \mathbf{H}_{ba} \mathbf{H}_{ab} \times U^{\alpha}(t'', t''') + \mathbf{H}_{bc} U^{\alpha}(t', t'') + \mathbf{H}_{ca} \}. \quad (32)$$

<sup>14</sup> For a time-independent interaction L. C. Biedenharn and M. E. Rose [Revs. Modern Phys. 25, 729 (1953)] have derived a formula for delayed coincidence angular correlations [Eq. (125)]. Integrating this formula over  $t$  from  $\tau_1$  to  $\tau_2$  we obtain the same angular correlation formula as (22) with (31). See also S. P. Lloyd, Phys. Rev. 82, 277 (1951).

The evaluation of the trace gives, in analogy to (22),

$$W = \text{const} \sum_{\kappa\bar{\kappa}} \sum_{\kappa'\bar{\kappa}''} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt''' e^{-\gamma_c(t'-t''')} \times e^{-\gamma_b(t''-t''')} e^{-\gamma_a t'''} \frac{1}{N} \sum_{\alpha} \sum_{\bar{k}_c \bar{n}_c} \sum_{k_c n_c} \sum_{\bar{k}_b \bar{n}_b} \sum_{k_b n_b} \times (00|F_{dc}(\Omega_3)|\bar{k}_c \bar{n}_c)(\bar{k}_c \bar{n}_c \bar{k}_c \bar{k}_c|G_c^{\alpha}(t', t'')|k_c n_c \kappa' \kappa'') \times (k_c n_c|F_{cb}(\Omega_2)|\bar{k}_b \bar{n}_b) \times (\bar{k}_b \bar{n}_b \kappa' \kappa''|G_b^{\alpha}(t'', t''')|k_b n_b \kappa \kappa) \times (k_b n_b|F_{ba}(\Omega_1)|00), \quad (33)$$

where  $G_b^{\alpha}$  is given by (21) and  $G_c^{\alpha}$  is the corresponding expression for the level  $c$ . For time independent interactions,  $G_b^{\alpha}$  and  $G_c^{\alpha}$  are functions of  $t''-t'''$  and  $t'-t''$ , respectively. For liquid sources with randomly fluctuating fields,  $\sum_{\alpha} G_c^{\alpha}(t', t'') G_b^{\alpha}(t'', t''')$  is a function of  $t'-t''$  and  $t''-t'''$  only. In both cases we can simplify (33) by changing the time integration according to

$$\int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt''' \dots \rightarrow \int_0^t dt''' \int_0^{\infty} dt'' \int_0^{\infty} dt' \dots \times \int_0^{\infty} dt'' (t''-t''') \dots \quad (34)$$

This is not rigorous but only terms which are small, of the order  $\gamma_a/\gamma_b$  or  $\gamma_a/\gamma_c$ , are neglected.

#### IV. $\gamma$ CASCADES PRECEDED BY $\beta$ DECAY OR $K$ CAPTURE

In many instances the initial state of a  $\gamma$  cascade is not a long-lived isomeric state, but a short-lived excited state reached by  $\beta$  decay or  $K$  capture from a long-lived state, labeled 0 in the following.  $\beta$  decay or  $K$  capture leaves the electron shell in an excited state which may decay by x-ray or Auger emission during the nuclear decay. We must then find a way to describe simultaneous decay of the nuclei and their electron shells. Actually the electron shell decays through a sequence of excited states which begins with very short-lived states and may proceed through intermediate states with lifetimes comparable to those of the nuclear levels. For the sake of simplicity, however, we assume only one excited electron state.

The symbols  $a, b, c$  label the three nuclear levels with the electrons in the ground state;  $a', b', c'$  are the same nuclear levels with the electrons in the excited state.  $\mathbf{H}'$  is the matrix for the electronic transition.  $\gamma$  is the reciprocal lifetime of the excited electron state. The correlation formula corresponding to (16) is now

$$W = \text{const} \text{Tr} \left\{ \epsilon(\Omega_1 \Omega_2) \int_0^t dt' \mathbf{H}_{cb} (\rho_b(t') + \rho_{b'}(t')) \mathbf{H}_{bc} \right\} \quad (35)$$

where  $\rho_b(t)$  and  $\rho_{b'}(t)$  are to be determined from the following set of equations corresponding to (10):

$$\begin{aligned} \partial \rho_{a'}^\alpha / \partial t = & -i[H_e^\alpha, \rho_{a'}^\alpha] - (\gamma_a + \gamma) \rho_{a'}^\alpha \\ & + 2\pi \mathbf{H}_{a0} \rho_0^\alpha \mathbf{H}_{0a}, \end{aligned} \quad (36a')$$

$$\partial \rho_a^\alpha / \partial t = -i[H_e^\alpha, \rho_a^\alpha] - \gamma_a \rho_a^\alpha + 2\pi \mathbf{H}' \rho_{a'}^\alpha \mathbf{H}', \quad (36a)$$

$$\begin{aligned} \partial \rho_{b'}^\alpha / \partial t = & -i[H_e^\alpha, \rho_{b'}^\alpha] - (\gamma_b + \gamma) \rho_{b'}^\alpha \\ & + 2\pi \mathbf{H}_{ba} \rho_{a'}^\alpha \mathbf{H}_{ab}, \end{aligned} \quad (36b')$$

$$\begin{aligned} \partial \rho_b^\alpha / \partial t = & -i[H_e^\alpha, \rho_b^\alpha] - \gamma_b \rho_b^\alpha + 2\pi \mathbf{H}' \rho_{b'}^\alpha \mathbf{H}' \\ & + 2\pi \mathbf{H}_{ba} \rho_{a'}^\alpha \mathbf{H}_{ab}. \end{aligned} \quad (36b)$$

In the average over  $\alpha$ ,  $\rho_0(t)$  is proportional to the unit matrix and its time dependence is given by the factor  $e^{-\gamma_0 t}$ . Since the decay  $0 \rightarrow a$  is not observed and  $\gamma_0 \ll \gamma_a$ , this is still true for  $\rho_{a'}(t)$ . The desired solutions of the Eqs. (36b') and (36b) are then

$$\begin{aligned} \rho_{b'}(t) = \text{const} \frac{1}{N} \sum_\alpha \int_0^t dt' e^{-(\gamma_a + \gamma)(t-t')} e^{-\gamma_0 t'} \\ \times U^\alpha(t, t') \mathbf{H}_{ba} \mathbf{H}_{ab} U^\alpha(t, t') + \end{aligned} \quad (37)$$

and

$$\begin{aligned} \rho_b(t) = \text{const} \left\{ \frac{1}{N} \sum_\alpha \int_0^t dt' \frac{\gamma}{\gamma_a} e^{-\gamma_b(t-t')} e^{-\gamma_0 t'} U^\alpha(t, t') \right. \\ \times \mathbf{H}_{ba} \mathbf{H}_{ab} U^\alpha(t, t') + \frac{1}{N} \sum_\alpha \int_0^t dt' \int_0^{t'} dt'' \\ \times 2\pi e^{-\gamma_b(t-t')} e^{-(\gamma_a + \gamma)t' - t''} e^{-\gamma_0 t''} U^\alpha(t, t') \\ \left. \times \mathbf{H}' U^\alpha(t', t'') \mathbf{H}_{ba} \mathbf{H}_{ab} U^\alpha(t', t'') + \mathbf{H}' U^\alpha(t, t') + \right\}. \end{aligned} \quad (38)$$

Inserting (37) and (38) in (35), we have clearly three terms in the correlation function. The first term in (38) describes those cases where the electron shell decays while the nucleus is still in the level  $a$ ; the second term describes decays of the electrons during the life of the state  $b$ ; (37) describes the nuclei whose electrons remain in the excited state during the life of the state  $b$ . For large  $\gamma$ , the first term in (38) is predominant; for small  $\gamma$ , (37) is predominant. In both cases we have the same correlation function as in Sec. III.

I wish to thank Dr. H. Frauenfelder for several stimulating discussions.

## Gamma Radiation from Proton Bombardment of $\text{Li}^7$ †

ALFRED A. KRAUS, JR.\*

*Kellogg Radiation Laboratory, California Institute of Technology, Pasadena, California*

(Received December 7, 1953)

The capture  $\gamma$  rays from the reaction  $\text{Li}^7(p, \gamma)$  have been investigated by measuring excitation functions and angular distributions. In addition to the well-known resonance at 441 kev, the excitation curve exhibits resonance at 1030 kev in proton bombarding energy corresponding to an excited state at 18.14 Mev in  $\text{Be}^8$ . Near this resonance the  $\gamma$  rays have a nonisotropic angular distribution with fore-and-aft asymmetry. The yield integrated over this resonance corresponds to a radiation width given by  $\omega\Gamma_\gamma = 2$  ev.

IN the bombardment of  $\text{Li}^7$  by protons,  $\gamma$  rays of 15- and 18-Mev energy are produced in the capture reaction,  $\text{Li}^7(p, \gamma)$ . In addition, the first excited state of  $\text{Li}^7$  is produced by inelastic scattering of the protons and the decay of this state results in the emission of 478-kev  $\gamma$  rays, the over-all process being indicated by  $\text{Li}^7(p, p' \gamma)$ . The excitation curve for the 478-kev  $\gamma$  rays exhibits resonance<sup>1</sup> at 1030-kev bombarding energy and the behavior of the inelastically scattered protons near this resonance has recently been studied in this laboratory.<sup>2</sup> In addition, the cross section for the protons

elastically scattered by  $\text{Li}^7$  shows a strong anomaly near this resonance.<sup>3</sup> The present investigation was conducted to determine whether or not the capture  $\gamma$  rays for  $\text{Li}^7(p, \gamma)$  are resonant at this energy.

Evaporated lithium targets were bombarded with protons from the 2-Mev electrostatic accelerator of the Kellogg Radiation Laboratory. The  $\gamma$  rays were detected with a scintillation counter made of a  $\text{NaI}(\text{Tl})$  crystal  $1\frac{1}{2}$  in. in diameter and 2 in. long, cemented to a 5819 photomultiplier tube. The output was fed into a linear amplifier and to two discriminators, each having its output pulses counted on decade scalars. The system was unable to discriminate between the 15- and 18-Mev  $\gamma$  rays from the  $\text{Li}^7(p, \gamma)$  reaction. One discriminator was set to count all events over 5 Mev, and the second was set to count the 478-kev (soft)  $\gamma$  rays from the

† This work was supported by the joint program of the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.

\* G. E. Fellow in Physics, 1952–1953 academic year. Now at the Department of Physics, The Rice Institute, Houston, Texas.

<sup>1</sup> Brown, Snyder, Fowler, and Lauritsen, *Phys. Rev.* **82**, 159 (1951).

<sup>2</sup> Mozer, Fowler, and Lauritsen, *Phys. Rev.* **93**, 829 (1954).

<sup>3</sup> Waters, Fowler, and Lauritsen, *Phys. Rev.* **91**, 917 (1953).