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Response of a two-level system to a sequence of N radiation pulses

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Abstract. A general expression has been obtained for the polarisation of an assembly of two-level systems irradiated by a sequence of N radiation pulses. The times and amplitudes of the echo-polarisation have been obtained. The method is an extension of the T-matrix method for the exact solution of the problem of interaction of radiation with two-level systems.
The number of polarisation echoes is $3^{N-1} - N$. The echo times are given by

$$t' = (1 + a)\, t_N + (b - a)\, t_{N-1} + (c - b)\, t_{N-2} + \cdots + (q - p)\, t_1$$

where t_k are the pulse times and a, b, c take on values 1, 0, − 1. From the general expression the amplitudes of echoes due to sequences of 2, 3 and 4 pulses are obtained as special cases. Distinct echo sequences determined by time relations among the incident pulses are discussed. The echo sequences exhibit interesting features which are of significance in the application of the phenomenon in holophony, etc.

Keywords. Photon echoes; superradiance; magnetic induction echoes; correlated emission; holophony

1. Introduction

The dynamics of an atom (or an electron in a magnetic field) can be visualised as the motion of a vector $\boldsymbol{r}$ characterising the atom interacting with radiation (Feynman *et al* 1957). The picture represents the action of radiation over a finite interval of time as the tipping of the vector $\boldsymbol{r}$ through an angle θ followed by the free precessional motion of $\boldsymbol{r}$ with angular velocity $w_3 = \dot{\phi}$. If one introduces a pseudo-angular momentum $\boldsymbol{m}$, a pseudo-electric moment μ and a pseudo-field $\boldsymbol{E}$ by the following definitions (Venkatesh and Dixit 1971)

$$\boldsymbol{m} = \hbar \boldsymbol{r}, \quad \mu = \gamma \boldsymbol{r} = \frac{\gamma}{\hbar} \boldsymbol{m}, \quad \boldsymbol{E} = -\frac{\hbar \omega}{\gamma} \tag{1}$$

where ω_0 is the atomic transition frequency ω_0 between the states ψ_a and ψ_b. The other components ω_1, ω_2 are determined by the interaction potential $\hbar$ is Planck's constant (divided by 2π) and $\gamma = (\mu_1 + i\mu_2)_{ab}$, the gyroscopic equations can be recast into the equations of magnetic resonance

$$\frac{d\boldsymbol{m}}{dt} = \mu \times \boldsymbol{E} \tag{2}$$

These c-number relations express precisely the parallelism between the electric and the magnetic case and enable one to treat photon and magnetic induction echoes on a uniform basis.

The gyroscopic equations can be extended to an assembly of two-level systems and the notion of correlation could be defined in terms of $\boldsymbol{r}_j$ of individual atoms. However, the concept of correlation was first introduced by means of general angular momentum operator (Dicke 1954). The components of the angular momentum operator and the gyroscopic vector appear in the density operator ρ or $\mu(t)$ (to be distinguished from magnetic moment μ) which is simply the matrix representation of vector $\boldsymbol{r}$:

$$\mu(t) = 2\rho - 1 = \boldsymbol{r} \cdot \sigma = 2(\hbar)^{-1}\boldsymbol{r} \cdot \boldsymbol{S}_{op} = \begin{pmatrix} r_3 & r_1 - ir_2 \\ r_1 + ir_2 & -r_3 \end{pmatrix} \tag{3}$$

where $\sigma = (\sigma_1, \sigma_2, \sigma_3)$ are Pauli operators and $\boldsymbol{S}_{op}$ is the angular momentum operator of a spin $-\frac{1}{2}$ particle. The initial state corresponds to $\mu(0)$ with $r_1 = r_2 = 0$; $r_3 = 1$.

The expectation value of the polarization is given by

$$\langle \mu_{op} \rangle = \tfrac{1}{2}\gamma \sum_j \boldsymbol{r}_j = \tfrac{1}{2}\gamma p\,(\boldsymbol{r})_{av} \tag{4}$$

where p is the number of two-level systems and $(r)_{av}$ the average value per system.

The micromechanism underlying superradiance is the correlation of the pseudomoments of atoms of the assembly, and was considered by Venkatesh and Roy (1971) to derive correlated states. The phenomenon of pulse-like radiations in spontaneous radiative processes, analogous to magnetic induction echoes first observed by Hahn (1954), is essentially the enhanced spontaneous emission from an assembly of two-level systems which momentarily is in a superradiant state as was pointed out by Abella *et al* (1966). The superradiance originally discussed by Dicke, however, corresponds to the circumstances of free induction decay observed by Hahn in the magnetic case. *Two* radiation pulses are required for the production of a pulse-like radiation called a photon echo, the first one to tip the correlated momenta directed along the static field in an equilibrium state on the plane transverse to the field, and the second to drive the precessing individual moments back to the ' line ' where they are in step again after a definite interval of time.

The general problem considered here is the response of an assembly of similar atoms with an isolated pair of spectral levels or electrons in a magnetic field to a sequence of N radiation pulses when the centre frequency of the pulse is equal to the transition frequency ω_0. The response is in the form of a sequence of photon (or magnetic induction) echoes bearing definite time relations to each other within the relaxation time T_2. The formalism is applicable uniformly to magnetic induction and photon echoes.

The dynamics of the atom interacting with radiation and the radiative process are completely described in a rotating coordinate system Σ' (interaction picture) by the following pulse matrix representing a rotation in spin space (Venkatesh and Dixit 1971):

$$T^{(k)} = \begin{pmatrix} T_{11}^{(k)} & T_{12}^{(k)} \\ T_{21}^{(k)} & T_{22}^{(k)} \end{pmatrix} = \begin{pmatrix} \cos\frac{\theta_k}{2}\exp(i\phi_k/2 & -i\sin\frac{\theta_k}{2}\exp(i\phi_k/2) \\ -i\sin\frac{\theta_k}{2}\exp(-i\phi_k/2) & \cos\frac{\theta_k}{2}\exp(-i\phi_k/2) \end{pmatrix} \tag{5}$$

The index k labels the quantities corresponding to the k-th pulses. The matrix elements can be written as a single expression:

$$T_{ab}^{(k)} = f_{ab} \sin \tfrac{1}{2} (\theta_k + \pi\delta_{ab}) \exp [- \tfrac{1}{2} \mathrm{i} (-1)^a \phi_k] \tag{6}$$

where

$$\begin{aligned} f_{ab} &= 1 \quad \text{for} \quad a = b \\ &= -\mathrm{i} \quad \text{for} \quad a \neq b \end{aligned} \tag{7}$$

θ and ϕ are determined by the incident pulse strength and the incremental precessional velocity of a random nature due to a weak inhomogeneous field superposed on the steady field. They are given by the relations:

$$\begin{aligned} \theta_k &= \frac{\mu_{ij}}{\hbar} E \,.\, \triangle t_k \\ \phi_k &= \delta\omega\, (t_{k+1} - t_k) \end{aligned} \tag{8}$$

t_k is the time of application of the k-th pulse, $\triangle t_k$ the duration of the k-th pulse assumed to be very much smaller than the time intervals between the successive pulses. δw is the incremental precessional velocity having a spread $\triangle w$ related to the relaxation time T_2:

$$\triangle \omega = \frac{1}{T_2} \tag{9}$$

The process initiated by the sequence of pulses is described by the unitary transformation of the matrix μ (0) at $t = 0$ by the product matrix $T = \pi_k T^{(k)}$. The relvant physical quantities are given by

$$\begin{aligned} r_1 - \mathrm{i} r_2 &= -2 T_{11} T_{12} \\ r_3 &= | T_{11} |^2 - | T_{12} |^2 \end{aligned} \tag{10}$$

Sequence of N pulses

For a sequence of N pulses the matrix elements in question are:

$$T_{11} = \sum_k \sum_j \ldots \sum_q \sum_p T_{1k}^{(N)} T_{kj}^{(N-1)} T_{jm}^{(N-2)} \ldots T_{qp}^{(2)} T_{p1}^{(1)}$$

$$T_{12} = \sum_{k'} \sum_{j'} \ldots \sum_{q'} \sum_{p'} T_{1k'}^{(N)} T_{k'j'}^{(N-1)} T_{j'm'}^{(N-2)} \ldots T_{q'p'}^{(2)} T_{p'2}^{(1)}$$

$$T_{22} = \sum_a \sum_b \ldots \sum_r \sum_s T_{2a}^{(N)} T_{ab}^{(N-1)} T_{bc}^{(N-2)} \ldots T_{rs}^{(2)} T_{s2}^{(1)}$$

with $k, j, \ldots = 1, 2,$

The polarisation and energy are given by:

$$\begin{aligned} & -\tfrac{1}{2} (r_1 - \mathrm{i} r_2) \\ &= T_{11} T_{12} = \sum_k \ldots \sum_{p'} T_{1k}^{(N)} T_{kj}^{(N-1)} T_{jm}^{(N-2)} \ldots T_{qp}^{(2)} T_{p1}^{(1)} \,.\, T_{1k'}^{(N)} T_{k'j'}^{(N-1)} T_{j'm'}^{(N-2)} \\ & \qquad \ldots T_{q'p'}^{(2)} T_{p'2}^{(1)} \\ & \qquad = \sum_k \ldots \sum_{p'} T_{1k}^{(N)} T_{1k'}^{(N)} T_{kj}^{(N-1)} T_{k'j'}^{(N-1)} \ldots T_{qp}^{(2)} T_{q'p'}^{(2)} T_{p1}^{(1)} T_{p'2}^{(1)} \end{aligned}$$

and

$$\begin{aligned} r_3 = & \left| \sum \ldots \sum T_{1k}^{(N)} T_{kj}^{(N-1)} T_{jm}^{(N-2)} \ldots T_{qp}^{(2)} T_{p1}^{(1)} \right|^2 - \left| \sum \ldots \sum T_{1a}^{(N)} T_{ab}^{(N-1)} \right. \\ & \left. \times T_{bc}^{(N-2)} \ldots T_{ks}^{(2)} T_{s2}^{(1)} \right|^2 \end{aligned}$$

On expansion:

$$-\tfrac{1}{2}(r_1 - ir_2) =$$

$$= \sum_k \sum_{k'} \ldots \sum_q \sum_{q'} f_{1k} f_{1k'} \sin\tfrac{1}{2}(\theta_N + \pi\delta_{1k}) \sin\tfrac{1}{2}(\theta_N + \pi\delta_{1k'})$$
$$\times \exp[-\tfrac{1}{2}\mathrm{i}\,\{(-1)^1 + (-1)^1\}\phi_N]$$
$$\times f_{kj} f_{k'j'} \sin\tfrac{1}{2}(\theta_{N-1} + \pi\delta_{kj}) \sin\tfrac{1}{2}(\theta_{N-1} + \pi\delta_{k'j'})$$
$$\times \exp[-\tfrac{1}{2}\{(-1)^k + (-1)^{k'}\}\phi_{N-1}]$$
$$\times f_{jm} f_{j'm'} \sin\tfrac{1}{2}(\theta_{N-2} + \pi\delta_{jm}) \sin\tfrac{1}{2}(\theta_{N-2} + \pi\delta_{j'm'})$$
$$\times \exp[-\tfrac{1}{2}\mathrm{i}\,\{(-1)^j + (-1)^{j'}\}\phi_{N-2}]$$

....

$$\times f_{pq} f_{p'q'} \sin\tfrac{1}{2}(\theta_2 + \pi\delta_{qp}) \,.\, \sin\tfrac{1}{2}(\theta_2 + \pi\delta_{p'q'})$$
$$\times \exp[-\tfrac{1}{2}\mathrm{i}\,\{(-1)^p + (-1)^{p'}\}\phi_2]$$
$$\times f_{q1} f_{q'2} \sin\tfrac{1}{2}(\theta_1 + \pi\delta_{q1}) \sin\tfrac{1}{2}(\theta_1 + \pi\delta_{q'2})$$
$$\times \exp[-\tfrac{1}{2}\mathrm{i}\,\{(-1)^q + (-1)^{q'}\}\phi_1] \qquad (11)$$

Apart from the factor $f = f_{1k} f_{1k'} \ldots f_{q1} f_{q'2}$, each term of the summation is a product of two functions: the amplitude function $F(\theta_N \ldots \theta_1)$ containing the pulse strengths $\theta_N \ldots \theta_1$, and the exponential function Φ containing the pulse time $t_N, \ldots, t_1$ in ϕ_k's. Thus $-\frac{1}{2}(r_1 - ir_2) = \Sigma \ldots \Sigma\, f \,.\, F(\theta_1, \ldots, \theta_N)\, \Phi(\phi_1, \ldots, \phi_N)$.

$F(\theta_N \ldots \theta_1)$ is again a product of N factors each referring to a definite pulse. Each such factor may be expressed as follows:

$$F_s(\theta_s) = \sin\tfrac{1}{2}(\theta_s + \pi\delta_{\alpha\beta}) \sin\tfrac{1}{2}(\theta_s + \pi\delta_{\alpha'\beta'})$$
$$= \tfrac{1}{2}(1 - \cos\theta_s)(1 - \delta_{\alpha\beta})(1 - \delta_{\alpha'\beta'}) + \tfrac{1}{2}(1 + \cos\theta_s)\,\delta_{\alpha\beta}\delta_{\alpha'\beta'} +$$
$$+ \tfrac{1}{2}\sin\theta_s\,(\delta_{\alpha\beta} + \delta_{\alpha'\beta'} - 2\delta_{\alpha\beta}\delta_{\alpha'\beta'}) \qquad (12)$$

Further for any given set of values α, β, α', β' only one of these terms is selected. For $\alpha = \beta$, $\alpha' = \beta'$; $\alpha = \beta$, $\alpha' \neq \beta'$; $\alpha \neq \beta$, $\alpha' \neq \beta'$, the functions are $\frac{1}{2}(1 + \cos\theta)$; $\frac{1}{2}\sin\theta$ and $\frac{1}{2}(1 - \cos\theta)$ respectively.

The corresponding multiplying factor has the values

$$\begin{array}{llll} f_{\alpha\beta} f_{\alpha'\beta'} = 1 & \alpha = \beta; & \alpha = \beta' & \\ \phantom{f_{\alpha\beta} f_{\alpha'\beta'} =} -\mathrm{i} & \alpha = \beta, & \alpha' \neq \beta' & \\ \phantom{f_{\alpha\beta} f_{\alpha'\beta'} =} -1 & \alpha \neq \beta & \alpha' \neq \beta' & \qquad (13) \end{array}$$

so that $f = \pm 1, \pm \mathrm{i}$.

The exponential function Φ is a product of exponentials:

$$\exp\big[-\tfrac{1}{2}\mathrm{i}\,[\{(-1)^1 + (-1)^1\}\phi_N + \{(-1)^k + (-1)^{k'}\}\phi_{N-1} +$$
$$+ \{(-1)^j + (-1)^{j'}\}\phi_{N-2} \ldots + \{(-1)^q + (-1)^{q'}\}\phi_1\,]\big]$$

Note that ϕ_k's refer to an individual atom and are given by:

$$\phi_N = \delta w\,(t - t_N), \quad \phi_{N-1} = \delta w\,(t_N - t_{N-1}), \ldots, \phi_2 = \delta w\,(t_3 - t_2),$$
$$\phi_1 = \delta w\,(t_2 - t_1) \qquad (14)$$

This exponential has the form

$$\Phi = \exp[-\tfrac{1}{2}\,\mathrm{i}\,(t - t')\,\delta\omega]$$

where

$$t' = 1 + \tfrac{1}{2}[\{(-1)^k\} + (-1)^{k'}]\, t_N + \tfrac{1}{2}[(-1)^j + (-1)^{j'} - (-1)^k - (-1)^{k'}]\, t_{N-1} + \ldots + \tfrac{1}{2}[(-1)^r + (-1)^{r'} - (-1)^q (-1)^{q'}]\, t_1$$

Since k. k', ... take on values 1, 2 only the echo time may be written as

$$t' = [1 + (-1)^{kk'}\,\delta_{kk'}]\, t_N - [(-1)^{kk'}\,\delta_{kk'} - (-1)^{jj'}\,\delta_{jj'}]\, t_{N-1} - \ldots - [(-1)^{qq'}\,\delta_{qq'} - (-1)^{rr'}\,\delta_{rr'}]\, t_1 \ldots \tag{15 a}$$

or

$$t' = (1 + a)\, t_N - (a - b)\, t_{N-1} - (b - c)\, t_{N-2} - \ldots - (x - y)\, t_1 \tag{15 b}$$

where constants $a, b, \ldots, x, y$ take on the values $1, 0, -1$. The coefficient of t_N takes the values 2, 1, 0 and the coefficients of other pulse times take the values $2, 1, 0, -1, -2$.

For a given set of coefficients the values of $k, k', j, j', \ldots$ in (15 *a*) can be obtained and hence the amplitude F in (11) can be determined.

From (4) the polarisation of the assembly is obtained by taking the appropriate distribution function for the random variable δw and integrating over the width w. Let us take the Gaussian distribution for δw as an example:

$$P(\delta w) = \frac{T_2}{(2\pi)^{\frac{1}{2}}} \exp\left[-\frac{1}{2}(\delta w T_2)^2\right] \tag{16}$$

The polarisation per molecule is then given by

$$\tfrac{1}{2}(r_1 - \mathrm{i}r_2)_{\mathrm{av}} = -\int_w \Sigma f F(\theta_N \ldots \theta_1)\, P(\delta w) \exp[-\tfrac{1}{2}\,\mathrm{i}\,(i - t')\,\delta\omega]\, d\delta w$$

After integration we have for the ensemble average

$$\tfrac{1}{2}(r_1)_{\mathrm{av}} = \Sigma\, f F(\theta_1, \ldots \theta_N)\, h(\triangle w, t - t') \tag{17}$$

where

$$h(\triangle w, t - t') \equiv h(t') = \exp[-(t - t')^2/2T_2^2] \tag{18}$$

The total polarisation is found by multiplying (17) by the number of two-level systems p in the assembly so that the intensity of emitted radiation is proportional to p^2.

With time the successive terms of the series (17) become dominant when the condition $t = t'$ is satisfied and $h(t')$ in (18) assumes the maximum value. These large transient polarisations give rise to echoes. These echoes correspond to different sets of coefficients in (15 *b*) which determine the echo times as well as the echo amplitudes. Definite time relations exist among the echoes within the relaxation time T_2 of the system. Time coherence of the echoes arises essentially from the two microscopic processes—atomic transitions under the action of the first pulse and then the free precessional motion followed by its reversal under the action of the next pulse. Excluding the N instants for the N pulses the number of echoes is given by

$$n = 3^{N-1} - N \tag{19}$$

The details of procedure in this calculation have been omitted. Of these echoes some are eliminated by causality principle.

The analytical theory leads to a straight-forward prescription for the construction of the echo " spectrum " with its quantitative features for any number of pulses. The spectrum could also be visualised geometrically in terms of reflections and displacements of echoes relative to the incident pulses on the time axis. The relative positions of the incident pulses and echoes on the time axis are indicated in a typical diagram (figure 1).

Our analysis would apply to the primary phenomenon of photon echoes from two-level systems contained in a thin slab of material. The transformation formalism is equally applicable to propagation of a particle through a sequence of N-square wells. Furthermore, in view of the parallelism between optical polarisation as a two-level problem and the problem of electronic or atomic transitions presented here the formalism can also be applied to the transmission of light through multilayer dielectric (or a series of dielectric media separated from each other) and the study of polarisation forms. The complete analysis of photon echoes would include consideration of K-vectors and the angular dependence of emission from $(r_1)_{av}$ as well as damping of $\boldsymbol{r}$ due to echo emission for a thick slab. These analyses will be carried out elsewhere.

The following considerations, however, would strictly apply to magnetic induction echoes.

Sequence of four incident pulses

The comparatively simple case of a four-pulse sequence will be taken to illustrate the formal procedures outlined above. We shall discuss this example in detail since it brings out the most important features of echo sequences. The cases of 2 and 3 pulses follow as special cases but they are much more easily treated by direct multiplication of matrices (Venkatesh and Sarkar 1974). The echo times are found from the expression

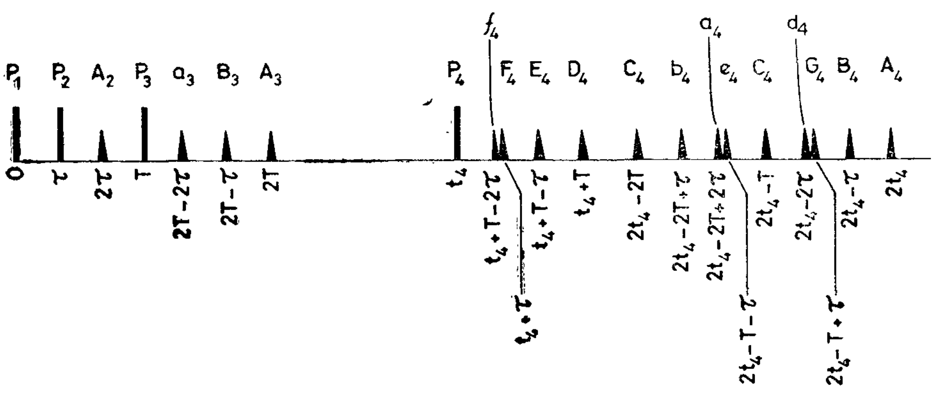

Figure 1. Relative positions of incident pulses and echoes for a sequence of four pulses ($t_4 > 2T > 4\tau$). Not strictly to scale.

$$t' = (1 + a)\, t_4 + (b - a)\, T + (c - b)\, \tau \quad (t_1 = 0,\, t_2 = \tau;\; t_3 = T) \qquad (20)$$

by taking possible combinations of the terms in the expansion of $(a_1 + a_2 + a_3)$ $(b_1 + b_2 + b_3)(c_1 + c_2 + c_3)$. Note that the combinations $a_1b_2c_3$, $a_3b_1c_2$, for instance enable one to write with reference to the formula (20) with $a_1 = b_1 = c_1 = 1$, $a_2 = b_2 = c_2 = 0$, $a_3 = b_3 = c_3 = -1$ *in a purely formal manner*:

$$(1 + a_1) = 2;\; (b_2 - a_1) = 1 - 2 = -1;\; (c_3 - b_2) = 2 - 3 = -1;$$
$$t' = 2t_4 - T - \tau$$
$$(1 + a_3) = 0;\; (b_1 - a_3) = 3 - 1 = 2;\; (c_2 - b_1) = 1 - 2 = -1;$$
$$t' = 2T - \tau.$$

The echo times can thus be readily written out with the aid of the terms of the expansion. The echoes identified by their amplitudes are labelled and listed in an array in table 1 along with their times of occurrence:

Table 1. Echoes identified by their amplitudes

$A_2 : 2\tau$	$p_2 : \tau$	$p_1 : 0$
$A_3 : 2T$	$B_3 : 2T - \tau$	$a_3 : 2T - 2\tau\,(T > 2\tau)$
$C_3 : T + \tau$	$p_3 : T$	$x_3^* : T - \tau$
$D_4 : t_4 + T$	$E_4 : t_4 + T - \tau$	$f_4 : t_4 + T - 2\tau\,(T > 2\tau)$
$F_4 : t_4 + \tau$	$p_4 : t_4$	$x_4^* : t_4 - \tau$
$g_4 : t_4 - T + 2\tau\,(T < 2\tau)$	$y_4^* : t_4 - T + \tau$	$z_4^* : t_4 - T$
$A_4 : 2t_4$	$B_4 : 2t_4 - \tau$	$d_4 : 2t_4 - 2\tau\,(t_4 > 2\tau)$
$G_4 : 2t_4 - T + \tau$	$C_4 : 2t_4 - T$	$e_4 : 2t_4 - T - \tau\,(t_4 > T + \tau)$
$a_4 : 2t_4 - 2T + 2\tau\,(t_4 > 2T - 2\tau)$	$b_4 : 2t_4 - 2T + \tau\,(t_4 > 2T - \tau)$	$c_4 : 2t_4 - 2T(t_4 > 2T)$

The echoes due to the first two pulses are entered in the first block and those due to the third and fourth pulses are entered in the second, third and fourth blocks. Those labelled by capital letters are primary echoes requiring no special time conditions and those labelled by small letters require special conditions on time to be satisfied. The asterisks indicate echoes which do not occur at all since they violate causality principle. There are altogether 19 echoes after excluding 4 pulses and 4 ghosts.

From the echo times the indices $k, k', \ldots$ in the amplitudes can be constructed from (11) and (12). The echo amplitudes are given in the appendix.

Echo sequences and their applications

Two sets of echoes can be distinguished. The primary echoes which imply no conditions on time apart from the primary conditions:

$$t_4 > T > \tau > 0$$

There are also conditions on relative times of the incident pulses for the occurrence of *secondary* echoes. For three incident pulses, for example, there is only one conditional echo and for four incident pulses there are eight indicated by small letters in table 1.

Partial sequences of echoes involving primary conditions for the time sequence can be written out. The longest possible sequences are obtained by matching all these bits of sequences, and are characterised by the time conditions of secondary echoes and the matching conditions on time.

In the case of four incident pulses, for example, a possible long sequence is (in the order of increasing time)

$$\text{I}\quad A_2a_3B_3A_3\ f_4c_4e_4b_4a_4d_4\ C_4E_4D_4G_4\dot{B}_4A_4.\quad (t_4 > 2T > 4\tau)$$

With extended time conditions, the echoes F_4 and C_3 will appear

$$\text{I}'\ldots\ldots\ldots\ldots\ldots A_2a_3C_3B_3\ldots\ldots c_4F_4e_4.\quad (2T+\tau > t_4 > 3T-2\tau > T + 2\tau).$$

The echoes g_4 and f_4 cannot be accommodated at all in this sequence.

The following sequence is essentially opposite to I:

$$\text{II.}\quad A_2B_3C_3A_4\ g_4F_4c_4C_4E_4D_4e_4b_4d_4\ a_4G_4B_4A_4\ (\tfrac{3}{2}\,\tau < T < 2\,\tau;\ t_4 > 2T+\tau)$$

in which the echoes f_4, a_3 cannot be accommodated since they are conditional with $T > 2\tau$. When the time conditions are extended in the sense of I′ the primary sequence of course remains unaltered while the secondary echoes are displaced with respect to each other and with respect to the primary echoes. In addition one observes the reversal of subsequences as well as dispersal of bits of subsequences when the time conditions are altered. Many such sequences can be obtained by more detailed matching conditions on pulsetimes and the possibilities are greatly enriched by a sequence of 5 or more pulses.

The experience of the two-level system previously subjected to a sequence of N pulses can be reconstructed by the application of the $(N+1)$ th pulse. For instance, the observation of three or more echoes (d_4, e_4 and c_4) would enable one to determine t_4, T and τ if the pulse strengths θ_K are known for the identification of the individual echoes. It may be noted that an echo due to the Nth pulse can occur earlier than an echo due to a previous sequence of $N-1$ pulses or may coincide with it. Such sequences can be identified by conditions of the type:

$$(1+a)\,t_4 \lesseqgtr (\triangle a - \triangle b)\,T + (\triangle b - \triangle c)\,\tau\ (\triangle a, \triangle b, \triangle c = 2, 1, 0, -1, -2) \tag{21}$$

Echoes can be cut off not only by the precise timing of the pulses but also by a suitable choice of the states of polarisation of the pulses and the pulse strengths (or simply the pulse durations). For instance in the case of three pulses the echo occurring at $t' = 2T$ can be eliminated by choosing $\theta_2 = \tau$. The relaxation time T_2 may range from 10^{-3} sec to 10^{-6} sec and can of course be lengthened within limits by working the crystal at very low temperatures reducing field inhomogeneity or by using a gas at low pressures. However, the relaxation time can provide a useful time range for finite echo sequences and serve as a full stop to meaningful sequences.

The basic facts underlying important applications of the echo phenomenon have been outlined above. The use of the two-level system as a memory device has been considered by Watson (1971) with particular reference to magnetic induction echoes in the radio frequency and microwave ranges. The considerations can be extended to photon echoes. The memory is preserved in the relative motions of the quasimoments of the atoms. Long or short conditional sequences could be exploited for miniaturisation of temporal sequences. A speech lasting for an hour (6000 spoken words) could be compressed into a time interval of 6 milliseconds if the relaxation time is of the order of a microsecond.

Essentially in echo production the number of pulses is amplified. For two applied pulses there is one echo, for three pulses there are five echoes, for four 19 echoes and so on. The number of input pulses can be increased by the echo phenomenon itself by using an aligned series of ruby rods of short length. With three incident pulses and two ruby rods one can obtain 76 echoes at most in a suitable experimental arrangement. The essential requirement is a laser-type axial emission of the five echo pulses from the first ruby rod although it may be noted that *no population inversion is required for the production of echoes and the amplification of their number.*

Summary and discussion

The T-matrix formalism is most appropriate for dealing with two-level problems, in particular the echo phenomenon, in regard to the ease and directness with which it can be applied to any number of pulses as well as the generality of the results which apply to magnetic induction as well as photon echoes. A more quantitative understanding of the echo production, the amplitudes and times of the echoes, in particular the conditions under which echoes can be attenuated or completely cut out, and the predetermination of sequences of incident pulses for the required sequences of echoes, are basic to all applications of this phenomenon. The echo times are simple linear combinations of the pulse times and the regularities are easily seen when they are written in an array as in table 1. The echoes can be constructed algebraically according to a definite prescription once the laws of their formation are known by the analytical theory. The linear combinations can be realised more easily as reflections and displacements relative to the incident pulses.

The most important applications of the echo phenomenon utilize the echo sequences which are entirely characterised by detailed time conditions on the applied pulses. The sequences can be systematically constructed by matching the primary sequences. Time conditions result from this matching as well as from the conditional echoes of the sequence.

It might perhaps be pointed out here that there are two ways in which enhanced emission of radiation can occur with intensity proportional to the square of the number of emitters. The underlying processes however are entirely different. The one is a spontaneous emission process in which the spins are momentarily in step at definite times. This does not require population inversion necessarily. The other is stimulated emission requiring population inversion in laser-like devices.

Brewer and Shoemaker (1971) have observed the photon echo with two pulses obtained by the use of CW radiation from CO_2 laser and pulsed Stark fields for pulsing the molecular level splitting in $C^{13}H_3F$ and NH_2D. A simple experimental method is, therefore, feasible for obtaining many radiation pulses by Stark modulation of the transition frequency of a suitable two-level system. One can thus obtain very long conditional sequences. The general theory of echoes due to a sequence of N pulses therefore becomes meaningful,

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Appendix

Amplitudes of echoes for a four pulse sequence

$$\begin{aligned}(r_1)_{\text{ar}} = {} & \tfrac{1}{8}(1-\cos\theta_4)(1+\cos\theta_3)(1+\cos\theta_2)\sin\theta_1 h(2t_4) + \tfrac{1}{4}(1-\cos\theta_4) \\ & \times (1+\cos\theta_3)\sin\theta_2\cos\theta_1 h\,(2t_4-\tau) - \tfrac{1}{8}(1-\cos\theta_4) \\ & \times (1+\cos\theta_3)(1-\cos\theta_2)\sin\theta_1 h\,((2t_4-2\tau) \\ & -\tfrac{1}{4}(1-\cos\theta_4)(\sin\theta_3\sin\theta_2\sin\theta_1 h\,(2t_4-T+\tau) + \tfrac{1}{2}(1-\cos\theta_4) \\ & \times \sin\theta_3\cos\theta_2\cos\theta_1 h\,(2t_4-T) - \tfrac{1}{4}(1-\cos\theta_4)\sin\theta_3\sin\theta_2 \\ & \times \sin\theta_1 h\,(2t_4-T-\tau) \\ & +\tfrac{1}{8}(1-\cos\theta_4)(1-\cos\theta_3)(1-\cos\theta_2)\sin\theta_1 h\,(2t_4-2T+2\tau) \\ & -\tfrac{1}{4}(1-\cos\theta_4)(1-\cos\theta_3)\sin\theta_2\cos\theta_1 h\,(2t_4-2T+\tau) \\ & -\tfrac{1}{8}(1-\cos\theta_4)(1-\cos\theta_3)(1+\cos\theta_2)\sin\theta_1 h\,(2t_4-2\tau) \\ & +\tfrac{1}{4}\sin\theta_4\sin\theta_3(1+\cos\theta_2)\sin\theta_1 h\,(t_4+T) + \tfrac{1}{2}\sin\theta_4\sin\theta_3\sin\theta_2 \\ & \times \cos\theta_1 h\,(t_4+T-\tau) - \tfrac{1}{4}\sin\theta_4\sin\theta_3(1-\cos\theta_2)\sin\theta_1 h \\ & \times (2t_4+T-2\tau) \\ & +\tfrac{1}{2}\sin\theta_4\cos\theta_3\sin\theta_2\sin\theta_1 h\,(t_4+\tau) \\ & -\tfrac{1}{4}\sin\theta_4\sin\theta_3\cos\theta_2\sin\theta_1 h\,(t_4-T+2\tau) \\ & +\tfrac{1}{8}(1+\cos\theta_4)(1-\cos\theta_3)(1+\cos\theta_2)\sin\theta_1 h\,(2T) + \tfrac{1}{4}(1+\cos\theta_4) \\ & \times (1-\cos\theta_3)\sin\theta_2\cos\theta_1 h\,(2T-\tau) - \tfrac{1}{8}(1+\cos\theta_4)(1-\cos\theta_3) \\ & \times (1-\cos\theta_2)\sin\theta_1 h\,(2T-2\tau) \\ & +\tfrac{1}{4}(1+\cos\theta_4)\sin\theta_3\sin\theta_2\sin\theta_1 h\,(T+\tau) \\ & +\tfrac{1}{8}(1+\cos\theta_4)(1+\cos\theta_3)(1-\cos\theta_2)\sin\theta_1 h\,(2\tau)\end{aligned}$$

Note that the pulse strengths to be put into the amplitude of an echo depend on the incident pulse sequence to which the echo belongs. For instance for the echo at $t' = 2\tau$ (last term above) $\theta_4 = \theta_3 = 0$, so that the amplitude is really $\tfrac{1}{2}(1-\cos\theta_2)\sin\theta_1 h\,(2\tau)$.