CALCULATION OF THE STEADY-STATE PROBABILITY OF LIGHT ABSORPTION BEYOND THE LIMIT OF APPLICATION OF PERTURBATION THEORY

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1. Introduction

It is usually assumed that radiation acting on an atom induces transitions whose random character leads to exponential equalization of populations:

$$n(t) = N_1 - N_2 = n(0) X(t), \quad (1.1)$$

in which

$$X(t) = \exp(-2Wt) \approx 1 - 2Wt.$$

The probability of induced transitions per unit time is simply a parameter of the exponential decay. However, exponential (purely random) population equalization is not the only possible effect, and it occurs approximately only when perturbation theory is applicable [1, 53. It has been shown [1, 3] that the relaxation in response to intense radiation (outside the limits of application of perturbation theory) may be oscillatory, retaining some of the features of a dynamic process (Fig. 1). It is clear that the transition probability loses its meaning in relation to description of the transfer kinetics.

![Image](image.png)

Fig. 1. Relaxation in response to a phase-modulated wave for: a) $\tau_\theta \ll 1$, b) $\tau_\theta \gg 1$.

This may not be important if the transition probability does not form the basis of evaluation of the steady-state absorption. If the process is certainly exponential, we may expect that the interactions with light and matter will be additive, which gives us a balanced equation via the usual probability scheme for the transitions:

$$\frac{dn}{dt} = -2Wn - (n - n_0) T^{-1}, \quad (1.2)$$

in which $1/T$ is the probability of transition between levels in response to matter and $n_0$ is the equilibrium population of the levels. This readily gives the steady-state population as

$$n_s = n_0 [1 + 2W_s T]^{-1}, \quad (1.3)$$

appearing in the quasi-resonant steady-state absorption

$$I = h \nu_0 W_s n_s, \quad (1.4)$$

The subscript $s$ in this latter formula would not be necessary if (1.2) always applied; but if $W$ loses its meaning, not only does (1.2) become irregular but also the steady-state absorption evaluated from this. It is hopeless to attempt to explain nonexponential decay via an elementary stochastic scheme, but it may be desirable to retain the concept of transition probability for describing the steady-state absorption, merely by retaining (1.3) and (1.4) while assigning the transition probability a more general meaning.
The next section gives a convenient definition of this probability that satisfies the above requirements. The other sections employ this definition in calculating the absorption probabilities for various structures.

2. Evaluation of the Steady-State Probability

Equation (1.3) may be taken as defining the transition probability in the general case if the steady-state population is defined in some independent and exact fashion. Let the atoms collide on the average at intervals $T$, each such collision completely restoring the equilibrium population $n_0$. If the last collision before $t$ occurred at time $t'$, the population will be $R_t = n_0 X(t-t')$; if there were no collisions, the result is $R_0 = n(0) X(t)$. Summation with the appropriate probabilities gives

$$n(t) = R_0 \exp \left(-\frac{t}{T}\right) + \frac{n_0}{T} \int_0^t \exp \left[-\frac{(t-t')}{T}\right] R_t dt.$$

(2.1)

In the steady state ($t \to \infty$) this gives

$$n_s = \bar{n}(\infty).$$

(2.2)

As (1.3) and (2.3) are identical, we have

$$[1 + 2W_s T]^{-1} = \frac{1}{T} \int_0^t \exp \left(-\frac{t}{T}\right) X(t) dt.$$

(2.3)

$X(t)$ alone controls the transition probability if the interaction with the medium is sufficiently weak, and this is due solely to the action of the light on the atom:

$$[2W_s]^{-1} = \int_0^t X(t) dt.$$

(2.4)

For this to be correct, $T$ must be greater than the characteristic times $X(t)$, and we must also have that

$$2W_s T \gg 1.$$

(2.5)

It is clear that even (2.4) is a very general definition, which includes as a special case the exponential result $X(t) = \exp(-2Wt)$, which gives $W_s = W$. If the relaxation is not exponential, we cannot expect, in general, any relation between $W_s$ and $W$. Then any correct definition of $W_s$ requires an exact knowledge of the decay kinetics throughout the time axis, not merely near $t = 0$ where the results from perturbation theory apply.

3. Wave with a Randomly Modulated Phase

We use (2.3) to examine the absorption of radiation having a random and stepwise-varying phase

$$E(t) = E_0 \exp \{i \omega t + i \alpha(t)\},$$

(3.1)

whose amplitude and frequency are considered as constant. If the phase memory is lost in time $\tau_\alpha$, the spectrum of the radiation is [7]

$$g(x) = \frac{\tau_\alpha}{\pi} \left[1 + (\omega - x)^2 \tau_\alpha^2\right]^{-1}.$$

(3.2)

Such radiation acting on a two-level system produces population relaxation described (Fig. 1) by

$$X(t) = \sum_{i=1}^3 C_i \exp z_i t,$$

(3.3)

in which the $z_i$ satisfy the characteristic equation

$$z^3 + \frac{2}{\tau_0} z^2 + \left(\frac{1}{\tau_0} + \omega^2 + \Delta \omega^2\right) z + \frac{\omega^2}{\tau_0} = 0.$$

(3.4)