THE KINETICS OF INDUCED MOSSBAUER RADIATION

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We treat the kinetics of induced Mössbauer radiation in an infinite crystal under conditions where the characteristic time of the process is much less than the line width. It is shown that the maximum of the wave will move with a very low velocity, proportional to the line width.

INTRODUCTION

In recent years there has been an extensive development of generators and amplifiers of electromagnetic waves using the induced radiation from atoms and molecules (masers and lasers, cf., for example, [1]). An important feature of these generators is the extremely high spectral and angular density of the radiation. From a thermodynamic point of view the radiation density depends on the temperature of the source. In lasers the source of the radiation consists of excited atoms in a state with a negative temperature, i.e., one which is above infinite [2] temperature. One can therefore in principle obtain any radiation density. The mechanism of amplification of the electromagnetic wave is connected with the induced radiation of the atoms.

Lasers were first made for the centimeter range (masers) and later for the visible region. The idea naturally suggests itself to extend this same principle to the electromagnetic radiation from nuclei, i.e., to γ rays. Such a proposal seems first to have been made by Rivlin. [3,4] In his report [5] and also in [6] he made a rather detailed quantitative calculation which showed that it was possible in principle to make a γ laser. The basis for this was the fact that the cross section for induced radiation is equal, at resonance, to [1)]

$$\sigma_+ \sim \lambda^2 \sim 10^{-18}\ \text{cm}^2 \quad (\lambda \sim 0.2\text{Å}; \quad \hbar\omega \sim 50\ \text{keV}),$$

and is much greater than the photoabsorption which is the main mechanism for loss of photons in this energy range: $\sigma_- \sim 10^{-22}\ \text{cm}^2$ ($Z \sim 25$).[2)] For the development of the γ wave it is therefore sufficient that the concentration of active nuclei be greater than the critical value

$$\eta > \eta_1 = \sigma_-/\sigma_+ \sim 10^{-4}\ . \qquad (1)$$

Here $\eta = \Delta n/n = (n_2 - n_1)/(n_2 + n_1)$; n_1 and n_2 are the densities of nuclei in the ground and excited states.

The following three main difficulties in producing a γ laser were pointed out: [5,6] a) obtaining excited nuclei in sufficient concentration; b) finding a sufficiently narrow γ line, so that the cross section for induced radiation has the large value cited above; c) reflection of the γ rays to get feedback and increase in the path length of the γ wave in the medium; the need for reflection is assumed in [5,6] in analogy to the optical laser. This last difficulty is most basic.

For reflection of the γ rays it is proposed in [3-6] to use the phenomenon of total internal or Bragg reflection. But this imposes too rigid requirements on the precision of the reflecting surface or crystal ($\sim 10^{-6}$ [7]), which are not attainable. In the case of Bragg reflection the difficulties are intensified by the fact that there must be a definite ratio (to the above accuracy) between the wave length of the γ wave and the period of the crystal lattice, which can only occur by pure chance.

In the present paper we consider the kinetics of one type of γ laser, consisting of a crystal which initially contains only excited nuclei (a pure nuclear isomer). Such a γ laser gives a single pulse of γ radiation. According to the estimates [7] this is the most easily achievable kind of γ laser. Since a rather long time is needed to prepare a crystal consisting of the pure isomer, the lifetime of the isomer must also be long. The characteristic time for development of the γ wave under these conditions is much less than the time for a radia-

[1)] We are considering only a Mössbauer line with the natural width.

[2)] We have taken the optimum values for λ, Z, $\hbar\omega$, according to the estimates in [5,6].

tive transition. Therefore the cross section for induced radiation is not constant, but depends on the whole process of development of the γ wave. This also determines the detailed kinetics of the γ laser. A study of the kinetics shows that although the time for development of the γ wave is relatively long (~ 1 sec), the length over which the γ wave develops is comparatively very small (~ 1 mm). Thus reflection of the γ wave is unnecessary, and we thus eliminate one of the difficulties in producing a γ laser.

Essentially the present paper studies only the initial stage of the development of the γ wave in the crystal. As the flux of γ quanta increases the process becomes very much more complicated: the isomer content changes, there is heating and damaging of the crystal, etc. Some of these questions were treated in [7]. A detailed calculation of these processes and related questions of the use of the γ laser (limiting density of radiation, efficiency, etc) is very complicated and hardly worth doing at present, since the very possibility of making a γ laser is problematical. The first step might be to try to get even a weak amplification of the γ wave in a crystal. The main difficulty in the way of solving this first problem is to find an extremely narrow Mössbauer line (cf. Sec. 4). This difficulty may well be overcome, in view of the extremely active investigation of the Mössbauer effect now going on. Some considerations concerning the minimum width of a Mössbauer line are given in [7], and will also be discussed in a separate paper.

1. CROSS SECTION FOR INDUCED GAMMA RADIATION

In the first approximation of perturbation theory, the transition amplitude between nondegenerate states of the nucleus under the influence of an external electromagnetic field is:[8]

$$a = -\frac{i}{\hbar}\int_0^t F(t)\, e^{i(\omega_0-\omega)t}\, dt, \qquad (1.1)$$

where ω and ω_0 are the frequencies of the electromagnetic wave and the nuclear transition, respectively, F(t) is the matrix element of the perturbation, whose Hamiltonian has the form

$$\hat{H}_b = \hat{F}e^{-i\omega t} + \hat{F}^* e^{i\omega t}.$$

Let us consider the limiting case when the exponential in (1.1) can be set equal to unity. This assumption is valid if the detuning $\Delta\omega \equiv \omega_0 - \omega \ll t_r^{-1}$, where t_r is the characteristic time for the process. Since in the present case t_r is rather large (~ 1 sec), the last condition means in particular that the width of the line must be extremely small ($\Delta\omega \ll 1$ sec^{-1}). In addition the time for the radiative transition, τ_γ, should be sufficiently large that one can neglect the imaginary part of ω_0 in (1.1):

$$\mathrm{Im}\,\omega_0 = \tau_\gamma^{-1}/2 \ll t_r^{-1}, \text{ i.e., } t_r \ll \tau_\gamma,$$

which is also satisfied in the present case, as indicated in the Introduction.

The amplitude of the vector potential of the electromagnetic wave is A_0; then $F = A_0 F_1$, where F_1 is the matrix element corresponding to unit amplitude. The flux of photons in the wave is

$$q = ck^2 A_0^2/2\pi\hbar\omega, \quad k = \omega/c.$$

From (1.1), using the approximation $(\omega_0 - \omega)t \ll 1$, we find for the cross section for induced radiation:

$$\sigma_i = \frac{1}{q}\frac{d}{dt}|a|^2 = \frac{4\pi|F_1|^2}{k\hbar A_0(t)}\int_0^t A_0(t)\, dt. \qquad (1.2)$$

The quantity $|F_1|^2$ can be related easily to t using the principle of detailed balancing.[7] For unpolarized nuclei and radiation we get

$$|F_1|^2 = \hbar/4kg_1\tau_\gamma, \qquad (1.3)$$

where g_1 is the statistical weight of the state with lower energy. In addition the cross section for the induced transition (1.2) must be multiplied by the statistical weight of the final state g_f. As a result we get:

$$\sigma_i = \frac{\pi}{k^2}\frac{1}{\tau_\gamma A_0(t)}\int_0^t A_0(t)\, dt, \qquad (1.4)$$

since in the case of induced radiation $g_f = g_1$. We note that under these same assumptions the cross section for resonance absorption of photons has the form

$$\sigma_v = \frac{\pi}{k^2}\frac{g_2}{g_1}\frac{1}{\tau_\gamma A_0(t)}\int_0^t A_0(t)\, dt, \qquad (1.5)$$

where $g_2 = g_k$ is the statistical weight of the upper state. Accordingly the maximum cross section at resonance for monochromatic radiation is

$$\sigma_m^{(12)} = 2\pi k^{-2} g_2/g_1 \qquad (1.6)$$

for resonance absorption of photons, and

$$\sigma_m^{(21)} = 2\pi k^{-2} \qquad (1.7)$$

for induced radiation. It is interesting to note that the cross sections (1.4) and (1.7) do not depend directly on the statistical weights of the states.

The lack of symmetry between the direct and inverse cross sections is explained by the fact that the maximum cross section at resonance is proportional to the reciprocal of the width of level 2, i.e., to the time during which coherence of the γ wave is maintained. But the width of the level is determined by the probability for spontaneous transition in a definite direction ($2 \rightarrow 1$), which is thus singled out.

Expression (1.4) shows that the cross section for induced radiation does depend on the kinetics of the process, as was stated above. The physical meaning of this dependence can easily be understood in the simplest case where $A_0 = \text{const}$, i.e., for an electromagnetic wave of constant amplitude acting for a sufficiently short time t. In accordance with the uncertainty relations, the spectral width of a wave of duration t is of order $\Delta\omega \sim t^{-1}$, while the width of the resonance line $\sim \tau_\gamma^{-1}$. Thus the effective (integral) cross section is equal to the resonance value ($\sim \lambda^2$) multiplied by the ratio $\tau_\gamma^{-1}/\Delta\omega \sim t/\tau_\gamma$, which coincides in order of magnitude with (1.4).

2. THE KINETIC EQUATION

We shall restrict our treatment to the simplest case of a plane wave with intensity I(x, t). The equation for I obviously has the form

$$\frac{\partial I(x, t)}{\partial x} = I(x, t)\,(f\sigma_i\,\Delta n - n\sigma_-), \qquad (2.1)$$

where Δn and n are the densities of the active nuclei and of all the nuclei, respectively; f is the fraction of Mössbauer photons. Since $I \sim A_0^2$, we find after substitution, dividing by $2A_0$ and differentiating with respect to t:

$$\frac{\partial^2 A_0}{\partial x \partial t} + \frac{1}{2l_-}\frac{\partial A_0}{\partial t} = KA_0, \qquad (2.2)$$

where

$$l_- = 1/n\sigma_-, \quad K = \pi\eta n f/2k^2\tau_\gamma, \quad \eta = \Delta n/n. \qquad (2.3)$$

From now on we shall assume that the quantities l_- and K are constant. This means, in particular, that we are neglecting the decrease in the concentration η of active nuclei due to both the spontaneous decay ($t \ll \tau_\gamma$) and the induced transitions (since we may assume that the γ wave develops within a narrow solid angle and that its total intensity is negligible).

Equation (2.2) must be solved with the boundary condition

$$A_0(0, t) = A_{sp} = 1, \qquad (2.4)$$

where A_{sp} is the amplitude of the spontaneous radiation, which we shall assume to be constant during the course of the whole process ($t \ll \tau_\gamma$). We take the initial condition in the form

$$A_0(x, 0) = \exp(-x/2l_-). \qquad (2.5)$$

This condition corresponds to a damping of the spontaneous wave, since the cross section for the induced radiation is equal to zero at t = 0 (the point x = t = 0 corresponds to the initiation of the spontaneous wave).

Condition (2.5) is approximate, since at t = 0 the spontaneous wave can have gone only through a path x = ct = 0. This means that (2.5) must be considered for some $t \neq 0$ and $x < ct_1$, where t_1 should be much smaller than the characteristic time for development of the γ wave. The condition for validity of this approximation will be considered later (Sec. 4).

3. SOLUTION

It is easily verified by direct substitution that the solution of Eq. (2.2) with the conditions (2.4), (2.5) has the form

$$A_0(x, t) = J_0(2i\sqrt{Kxt})\exp\{-x/2l_-\}, \qquad (2.6)$$

where J_0 is the Bessel function of order zero.

To analyze the solution, we consider the most interesting limiting case of $A_0 \gg 1$. Using the asymptotic form of the Bessel function, we find

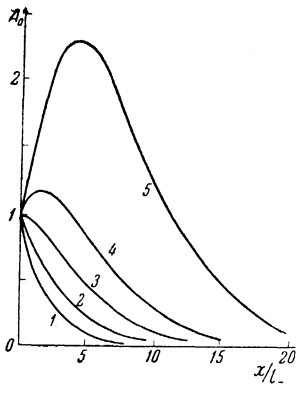
$$A_0^2 = \frac{\exp(4\sqrt{Kxt} - x/l_-)}{4\pi\sqrt{Kxt}}. \qquad (2.7)$$

A graph of $A_0(x)$ at different times, giving a picture of the development of the γ wave, is shown in the figure.

The maximum of the γ wave at each time occurs at a distance

$$x_m \approx 4Ktl_-^2. \qquad (2.8)$$

The maximum intensity of the wave as a function



Amplification of γ wave by induced radiation: dependence of amplitude on distance (in units of l_-). 1 – at the initial time, 2 – after 1 sec, 3 – after 2 sec, 4 – after 3 sec, 5 – after 5 sec; $\tau_\gamma = 10^4$ sec; $t_r = 1$ sec.

of time, and the distance to the maximum, are given by the formulas

$$A_{0m}^2 = \frac{\exp(4Kt l_-)}{8\pi K t l_-} \approx \frac{\exp(x/l_-)}{2\pi x/l_-}. \qquad (2.9)$$

The characteristic rise time of the γ wave is

$$t_r \approx (4kl_-)^{-1} = \tau_\gamma \sigma_- k^2/2\pi\eta f = \tau_\gamma \eta_1/f\eta, \qquad (2.10)$$

where $\eta_1 = \sigma_-/\sigma_+ = \sigma_- k^2/2\pi$ is the critical concentration of the nuclear isomer (1). The characteristic distance over which the γ wave develops is l, i.e., the mean free path of the quantum before absorption.

We get a result which at first glance seems paradoxical: the greater the absorption, the higher the intensity of the γ wave at a given distance from the source. This is explained qualitatively as follows. The cross section for induced radiation is equal in order of magnitude to $\sigma_+ t_r/\tau_\gamma$, where t_r is the characteristic time for the development of the γ wave, or the time delay until the γ line narrows. For the γ wave to develop it is necessary that $\sigma_+ t_r/\tau_\gamma > \sigma_-$. Consequently the greater the absorption, the longer the delay time [cf. also (2.10)], the greater the cross section for induced radiation, and the faster the increase in intensity of the γ wave with distance. In the opposite limiting case ($t_r \gg \tau_\gamma$), which occurs in optical lasers, the growth of the electromagnetic wave would occur over a distance $l_+ = (n\sigma_+)^{-1}$, which is a factor $\sigma_+/\sigma_- = \eta_1^{-1} \sim 10^4$ times smaller than l_-. Thus the detailed kinetics of the γ laser give a relative increase in characteristic length by a factor η_1^{-1}. The reason why this increased length is still much less than for the optical laser is mainly because the γ laser we are considering contains the pure isomer, whereas in the optical laser one can produce only a very small excess of excited atoms.

The motion of the maximum of the γ wave, which is easily seen in the figure occurs at a speed of

$$v_m \approx 4Kl_-^2 = f\eta l_-/\eta_1\tau_\gamma \approx l_-/t_r. \qquad (2.11)$$

In order for the approximate boundary condition (2.5) to be valid, this velocity must be much less than the velocity of light (cf. Sec. 4).

The width (2.9) of the γ lines is of order t_r^{-1}, i.e., a factor η_1^{-1} times larger than the radiation width τ_γ^{-1} from (2.10). This is connected with the nonstationarity of the γ wave. According to (2.11), the speed with which the maximum moves is proportional to the width of the γ line.

The theory developed here can also be applied to the resonance absorption of γ rays by unexcited nuclei, for example to the study of the time dependence of the Mössbauer radiation. This effect has been studied experimentally,[9] and has also been calculated by another method.[9,10] For the case of absorption it is sufficient to replace σ_i by $\sigma_v = -\sigma_i g_2/g_i$ and K by $K' = -Kg_2/g_1$ in the formulas: we then have from (2.6)

$$A_0^2 = J_0^2(2\sqrt{K'xt})\exp(-x/l_-), \qquad (2.12)$$

which coincides with the result of the computations in [9,10] for $\omega = \omega_0$, as assumed in the present paper, and in the absence of photoabsorption ($l_- \to \infty$) as assumed in [9,10]. It should however be noted that the experiment[9] gives twice as large an absorption cross section as is calculated from our assumptions. The cause of this discrepancy is still not known.

4. SOME ESTIMATES

To get rough numerical estimates to supplement our picture of the development of the Mössbauer γ wave in the crystal, we set $\eta \sim 10^{-4}$ (1) and $\tau_\gamma \sim 10^4$ sec. This last quantity is determined, as pointed out in the Introduction, by the time for preparation of the pure isomer. According to (2.10), the characteristic time for buildup of the γ wave is $t_r \sim 1$ sec ($f \sim \eta \sim 1$). After this time the γ wave has traversed a path $\sim 3 \times 10^{10}$ cm. But the maximum, and with it the region of strongest induced radiation, moves with the relatively low velocity (2.11), and during the characteristic time t_r covers only a distance $\sim l \sim 1$ mm ($n \sim 10^{23}$ cm^{-3}; $\sigma_- \sim 10^{-22}$ cm^2). From this it follows first of all, that the approximation in Sec. 3 ($v/c \sim 0.1/3 \times 10^{10} \sim 3 \times 10^{-12} \ll 1$) is satisfied. Since the characteristic distance for development of the γ wave is ~ 1 mm, a crystal which is several centimeters long can already be regarded as an infinite medium, so that reflection of the γ rays is not necessary for developing the reaction. The directionality of the induced radiation will be determined by the geometry of the crystal.

The investigation in the present paper of the kinetics of induced γ radiation is based on the assumption that during the whole of the characteristic time for development of the γ wave (t_r) its interaction with the atoms remains strictly resonant. This means that the width of the γ line in the crystal should be much smaller than $t_r^{-1} \sim 1$ sec^{-1}. This is much larger than the natural width $\tau_\gamma^{-1} \sim 10^{-4}$ sec^{-1}, but is still five orders of magnitude less than the width of the narrowest line found up to the present time, for Zn^{67} ($\Delta\omega \sim 10^5$ sec^{-1}[11]).

The effect of dynamic broadening of the line on the kinetics of the induced radiation can also occur in optical lasers. In the present theory of the pulsed laser[12] this effect is not taken into account, since at present the line width of a laser is relatively large because of imperfection of the crystal. But in the future, as the characteristics of the laser improve, this effect may become significant.

We remark in conclusion that an x-ray laser, operating on electronic transitions is hardly feasible, because of the absence of metastable levels, and the consequent difficulties in producing a significant concentration of excited atoms.

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318