TWO QUANTUM TRANSITIONS IN OPTICS

F. V. BUNKIN

P. N. Lebedev Physics Institute, Academy of Sciences, U.S.S.R.

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The problem of stimulated two-quantum emission (absorption) in the optical range is considered. It is assumed that the transition occurs between discrete levels of crystals with impurities or of free atoms. A critical analysis is given of the well-known result due to Kleinman.[1] It is shown that the correct utilization of the "one intermediate level approximation" leads to an expression for the probability of a two-quantum transition which differs significantly from the expression obtained by Kleinman.

In connection with the development of laser physics at present considerable interest is being shown (both by experimentalists and theoreticians) in the problem of multi-quantum processes in optics. It is well known that a theoretical determination of the probabilities (cross sections) of such processes does not involve any difficulties of principle, but, as a rule, it is generally associated with a large amount of calculations (of the compound transition matrix element). The difficulties associated with an exact calculation of the probabilities force us to seek various approximations for them. One such approximation for the probability W_{21} of a stimulated two-quantum transition applicable to impurity levels in crystals, i.e., of a transition from a quantum state 2 of a system (atom) into a state 1 accompanied by the simultaneous emission or absorption of two quanta was proposed by Kleinman.[1] Utilizing the general results of the classical work of Goeppert-Mayer[2] Kleinman obtained the following expression for W_{21}:[1)]

$$W_{21} = f_I^2 c^2 r_0^2 \frac{(2\pi)^3 F_1 F_2}{n_1 n_2 \omega_1 \omega_2 \Delta\omega} \qquad (1)$$

where $F_\alpha = cn_\alpha E_\alpha^2/8\pi\hbar\omega_\alpha$ is the flux of quanta in the α-th electromagnetic wave ($\alpha = 1, 2$; E_α is the amplitude of the wave); ω_1 and ω_2 are the frequencies of the waves ($\omega_1 + \omega_2 = \omega_{21}$); n_1 and n_2 are the indices of refraction of the medium at frequencies ω_1 and ω_2; $\Delta\omega$ is the spectral width of the transition $2 \to 1$; $r_0 = e^2/mc^2$ is the classical electron radius. Formula (1) is obtained[1] in the "one intermediate state approximation," i.e., in deriving it, it was assumed that the greatest contribution to the compound matrix element for the transition M_{21} is made by only one intermediate state of the system k_0 connected to the initial and the final states by the same oscillator strength f_I. Moreover, it is assumed[1] that the state k_0 lies sufficiently far (in energy) from the initial and the final states so that

$$\omega_I \equiv |\omega_{k_0 1}| \sim |\omega_{k_0 2}| \gg \omega_1, \omega_2.$$

The simplicity of formula (1) which contains a single characteristic of the intermediate state—the oscillator strength f_I, apparently, was the reason for its wide use in scientific literature. It is sufficient to cite experimental papers on the observation of two-quantum absorption[3-5] in which the discussion of the results obtained is carried out on the basis of formula (1). The same result is also utilized in the theoretical analysis of processes in the as yet unrealized two-quantum laser,[6,7] and it is also quoted without any critical comments in the review article[8] and in the recently published monograph.[9] We show below that formula (1) is not valid, i.e., it cannot serve as an approximation to the exact expression for W_{21} under any conditions (and, in particular, under those adopted in [1]).

The first suspicion that formula (1) is not valid is associated with the "infrared catastrophe" to which it leads. It is well known that for transitions between states of a discrete spectrum (which are under discussion at present) no "infrared catastrophe" should arise.

The general expression for the probability of a stimulated two-quantum emission can be repre-

[1)] In contrast to [1], we give the expression for W_{21}, and not for the cross section $\sigma_{1,2} = W_{21}/F_{1,2}$ for the emission (absorption) of a quantum $\hbar\omega_{1,2}$ when the flux of the quanta $F_{2,1}$ is given. Moreover, here a nondegenerate case is assumed, i.e., ω_1 and ω_2, generally speaking, do not coincide.

sented on the basis of [2] in the form

$$W_{21} = \frac{(2\pi)^3}{c^2\hbar^2}\frac{\omega_1\omega_2}{n_1 n_2 \Delta\omega} F_1 F_2 |M_{21}|^2, \qquad (2)$$

where the compound matrix element M_{21} can be written in two mathematically equivalent forms:[2)]

$$M_{21} = \frac{\hbar e^2}{m}\sum_k \frac{f_k^{(21)}}{(\omega_{k1}-\omega_1)(\omega_{k1}-\omega_2)} = \frac{\hbar e^2}{m}\sum_k \frac{\omega_{1k}\omega_{k2}}{\omega_1\omega_2}\frac{f_k^{(21)}}{(\omega_{k1}-\omega_1)(\omega_{k1}-\omega_2)} \qquad (3)$$

In formulas (3) the summation is meant to be taken over all possible intermediate states k and we have introduced the notation

$$f_k^{(21)} = (m/\hbar e^2)[\mu_{1k}^{(2)}\mu_{k2}^{(1)}(\omega_{k2}+\omega_2) + \mu_{1k}^{(1)}\mu_{k2}^{(2)}(\omega_{k1}-\omega_2)]. \qquad (4)$$

Here $\mu_{ij}^{(\alpha)} = (\mu\mu_\alpha)_{ij}$ is the matrix element of the component of the dipole moment of the quantum system along the direction of the polarization u_α of the field of the α-th wave. The quantity $f_k^{(21)}$ has the structure of the usual oscillator strength and can be called "the oscillator strength for a two-quantum transition."

The quantities $f_k^{(21)}$ in analogy with the usual oscillator strengths satisfy definite sum rules the principal one of which has the form

$$\sum_k f_k^{(21)} = 0. \qquad (5)$$

The proof of this rule is similar to the proof of the Thomas-Reiche-Kuhn theorem and is based on the commutation properties of the operators $\mu^{(\alpha)} = (\mu\mu_\alpha)$ and $\mu^{(\alpha)}$:

$$\mu^{(2)}\mu^{(1)} - \mu^{(1)}\mu^{(2)} = 0;$$

$$\mu^{(2)}\dot{\mu}^{(1)} - \dot{\mu}^{(1)}\mu^{(2)} = i\hbar\frac{e^2}{m}(\mathbf{u}_1\mathbf{u}_2). \qquad (6)$$

The equivalence of the two forms (3) of the expression for M_{21} follows from rule (5): addition of the left hand side of (5) to the second part of formula (3) transforms it into the first part. From rule (5) there also follows the obvious conclusion: *if in the system only one intermediate level k is present (to which single-quantum transitions from states 1 and 2 are allowed) two-quantum transitions in it (Raman scattering included) are impossible*, i.e., the probability W_{21} is strictly equal to zero.[3)] We note that in the case of one intermediate level $\mu_{1k}^{(2)}\mu_{k2}^{(1)} = \mu_{1k}^{(1)}\mu_{k2}^{(2)}$ and, therefore, from the condition $f_k^{(21)} = 0$ we obtain $\omega_{k2} = -\omega_{k1} = -(\omega_1+\omega_2)/2$, i.e., the level k should lie halfway between levels 1 and 2. From here it follows, in particular, that two-quantum transitions between the levels $m = \pm 1$ of a spin $S = 1$ in a magnetic field are impossible.

Thus, the "one intermediate level approximation" with the condition $\omega_I \gg \omega_{1,2}$ adopted in [1] is, strictly speaking, internally contradictory. This is the explanation of the origin of the erroneous formula (1): it is formally obtained if in the second expression for M_{21} (cf., (3)) we take only the single term with $k = k_0$ under the condition $|\omega_{k_0 1}| \sim |\omega_{k_0 2}| \gg \omega_1, \omega_2$, and set $f_I^2 \sim |f_{k_0}^{(21)}|^2$. We note that the first expression (3) in this case leads to quite a different result for M_{21} (cf., below). Later we shall show to what result the correct utilization of the "single intermediate level approximation" leads.

For concreteness we assume that all the intermediate levels k lie with respect to energy above the levels 1 and 2, with the first of these levels k_0 making a significant contribution to the sum (3). Such a level can stand for a whole band of states, as assumed in [1]. Introducing the notation: $\omega_I = \omega_{k_0 1}$, $\omega_I' = \omega_{k_0 2}$ (we do not assume now that $\omega_{k_0 1} \gg \omega_1, \omega_2$) and

$$f_I^{(21)} = \frac{1}{g_I}\sum_\gamma f_{k_0\gamma}^{(21)},$$

where g_I is the statistical weight of the level k_0, while the summation is taken over all the states γ of the level k_0, we obtain for M_{21} on the basis of (3) two equivalent expressions:

$$M_{21} = \frac{\hbar e^2}{m}\frac{g_I f_I^{(21)}}{(\omega_I-\omega_1)(\omega_I-\omega_2)} \times\left\{1+\sum_k{}'\left(\frac{\omega_I}{\omega_{k1}}\right)^2\frac{f_k^{(21)}}{g_I f_I^{(21)}}\frac{(1-\omega_1/\omega_I)(1-\omega_2/\omega_I)}{(1-\omega_1/\omega_{k1})(1-\omega_2/\omega_{k1})}\right\}$$
$$= -\frac{\hbar e^2}{m}\frac{g_I f_I^{(21)}}{\omega_1\omega_2}\frac{\omega_I'}{\omega_I}\left(1-\frac{\omega_1}{\omega_I}\right)^{-1}\left(1-\frac{\omega_2}{\omega_I}\right)^{-1} \times\left\{1+\sum_k{}'\frac{\omega_I}{\omega_I'}\frac{\omega_{k2}}{\omega_{k1}}\frac{f_k^{(21)}}{g_I f_I^{(21)}}\frac{(1-\omega_1/\omega_I)(1-\omega_2/\omega_I)}{(1-\omega_1/\omega_{k1})(1-\omega_2/\omega_{k1})}\right\}. \qquad (7)$$

The summations in formula (7) are assumed to be taken over all the intermediate states with the exception of k_0.

[2)] We note that the first form is directly obtained in the case when the operator for the interaction with the field is chosen in the form $V = -\mu(E_1 + E_2)$, while the second form is obtained in the case when $V = -ep(A_1 + A_2)/mc$ (E_α, A_α are respectively the field strength and the vector potential). It is well known that in the dipole approximation, which is the only one considered by us at present, both forms of the operator V are equivalent.

[3)] Naturally, within the limits of the dipole approximation considered by us.

The "one intermediate level approximation" consists of neglecting the sum in the first formula (7) compared to unity. Such neglect can be justified if the frequencies ω_{k1}, for which $f_k^{(21)}$ are not small, are large compared to ω_I. Formally this approximation corresponds in formulas (7) to the transition to the limit $\omega_{k1} \to \infty$. In this case we obtain for M_{21}

$$M_{21} = \frac{\hbar e^2}{m} \frac{g_I f_I^{(21)}}{(\omega_I - \omega_1)(\omega_I - \omega_2)}. \quad (8)$$

The second formula (7) differs by the fact that the sum appearing in it makes a significant contribution under all conditions and, therefore, cannot be neglected. The unjustified neglect of this sum leads to formula (1); but if in it we go to the limit $\omega_{k1} \to \infty$ and utilize rule (5) then we again obtain the correct formula (8).

In conclusion we give the final expression for the probability of a stimulated two-quantum transition obtained in the "one intermediate level approximation:"

$$W_{21} = g_I^2 |f_I^{(21)}|^2 r_0^2 c^2 \frac{(2\pi)^3}{n_1 n_2 \Delta\omega} \frac{\omega_1 \omega_2 F_1 F_2}{(\omega_I - \omega_1)^2 (\omega_I - \omega_2)^2}. \quad (9)$$

In this case the coefficient of amplification (absorption) $\alpha_{1,2}$ at the frequency $\omega_{1,2}$ is evaluated by means of the formula

$$\alpha_{1,2} = \sigma_{1,2} \Delta n = \frac{W_{21} \Delta n}{F_{1,2}} \ (\text{cm}^{-1}),$$

where Δn is the density of the difference in the population of levels 2 and 1.

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203