ON ONE POSSIBILITY OF MAKING A MEDIUM TRANSPARENT BY MULTIQUANTUM RESONANCE

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It is shown that a certain superposition of coherent electromagnetic waves passing through a resonant medium may suppress the wave absorption completely. This effect of making the medium transparent is the result of vanishing of the probability for multiquantum transition in each atom of the given substance. This is mainly a consequence of the corresponding phase relations between the waves, which are such that the substance remains in its initial state. Thus the "bleaching" discussed here is totally different from the effect due to saturation (population of resonance levels).

1. The behavior of atomic systems in strong electromagnetic fields in the case of single-quantum resonance, when the frequency of the external field ω is sufficiently close to one of the natural frequencies of the atomic transitions, has by now been well investigated. It is known, that in this case the probability of finding the atomic system in one of the resonant states is a kinetic function of the time, the period being inversely proportional to the external field [1]. So long as this period is much smaller than the characteristic relaxation times of the corresponding atomic levels, there are practically no oscillations between the resonant states. What occurs here is usual absorption (scattering) of radiation, a process which turns out to be very strong in the presence of the resonance. But when the period of the oscillations between the atomic states exceeds the relaxation times, then the system is observed, on the average, with equal probability in resonant states and a saturation effect is then realized, namely, the absorption coefficient decreases and the medium becomes more transparent. A "bleaching" of the medium takes place.

Obviously, similar effects take place also in multiquantum resonance, when the natural frequency of the atomic transition is a multiple of ω. In particular, in sufficiently strong fields, an analogous decrease in the absorption coefficient takes place.

However, in the case of multiquantum resonance, the decrease in the absorption coefficient can be the result of other causes, too, which are not at all connected with the equalization of the average populations of the resonant states. The authors have encountered such a phenomenon in an analysis of the problem of third-harmonic generation (generation of a wave with frequency 3ω) under conditions of resonance at double the frequency [2]. It was found, that a coherent superposition of waves with frequencies ω and 3ω takes place in such a medium when a number of conditions is satisfied; this superposition moves in the medium without experiencing absorption, whereas when each wave of frequency ω is taken separately it is attenuated by two-photon absorption. A similar undamped state of coherent waves with frequencies ω and 2ω was obtained in an investigation by Gurevich and Khronopulo [3].

We present here a detailed analysis of phenomena of this type.

2. Let an arbitrary atomic system, processing a definite discrete energy spectrum ϵ_n, be subjected to the action of an external perturbation constituting a superposition of several monochromatic waves. We shall henceforth neglect the broadening of the energy levels due to relaxation processes, and will assume that the reaction of the atomic system to the external perturbation is sufficiently small so that the amplitudes and phases of the waves are fixed. Then the time evolution of such a system can be described in terms of the amplitudes of the quantum states $a_n(t)$, for which the following equation is valid

$$i\hbar\frac{\partial}{\partial t}a_n(t)=\sum_m V_{nm}(t)a_m(t), \tag{1}$$

where the external-perturbation matrix is given by

$$V_{nm}(t)=\sum_\omega V_{nm}(\omega)\exp\{-i\omega t+i\omega_{nm}t\}, \tag{2}$$

the sum being taken here over the positive and negative values of the external-wave frequencies, and where we have by virtue of the hermiticity

$$V_{nm}(\omega) = V_{mn}^*(-\omega).$$

For the interaction with the electromagnetic field we put

$$V_{nm}(\omega) = -\mathbf{d}_{nm}\mathbf{E}(\omega), \tag{3}$$

where $\mathbf{d}_{nm}$ is the matrix of the dipole moment of the atomic system and $\mathbf{E}(\omega)$ is the Fourier component of the electric field of the external wave.

Assume now that there exist among the frequencies of the external waves such that their algebraic sum $\omega_1 + \omega_2 + \ldots + \omega_k$ is close to the frequency of the atomic transition $\omega_{\mu\nu} = (\epsilon_\mu - \epsilon_\nu)/\hbar$. If the external electric field is much weaker than the atomic fields, then the influence of the external field on all the amplitudes a_n, except a_μ and a_ν, is small. We can therefore, within the framework of ordinary perturbation theory, exclude from the system (1) all the amplitudes a_n with $n \neq \mu, \nu$. As a result we arrive at the system of two equations which relate only a_μ and a_ν:

$$\frac{\partial}{\partial t} a_\mu(t) = -ia_\nu(t) \sum_{(\omega_1+\ldots+\omega_k)} f_{\mu\nu}(\omega_1, \ldots, \omega_k) \exp\{i(\omega_1 + \omega_2 + \ldots + \omega_k - \omega_{\mu\nu})t\},$$

$$\frac{\partial}{\partial t} a_\nu(t) = -ia_\mu(t) \sum_{(\omega_1+\ldots+\omega_k)} f_{\mu\nu}^*(\omega_1, \ldots, \omega_k) \exp\{-i(\omega_1 + \omega_2 + \ldots + \omega_k - \omega_{\mu\nu})t\}, \tag{4}$$

the summation being carried out over all different resonant combinations of the type $\omega_1 + \omega_2 + \ldots + \omega_k \approx \omega_{\mu\nu}$, and

$$f_{\mu\nu}(\omega_1, \ldots, \omega_k) = \frac{\hbar^{-k}}{(k-1)!} \sum_{n_1,\ldots,n_{k-1}} \sum_P P(\omega_1, \ldots, \omega_k) \times V_{\mu n_1}(\omega_1) V_{n_1 n_2}(\omega_2) \ldots V_{n_{k-1}\nu}(\omega_k) \times [(\omega_1 + \omega_2 + \ldots + \omega_{k-1} - \omega_{n_1\nu}) \times (\omega_2 + \omega_3 + \ldots + \omega_{k-1} - \omega_{n_2\nu}) \ldots (\omega_{k-1} - \omega_{n_{k-1}\nu})]^{-1}, \tag{5}$$

where the symbol $P(\omega_1, \ldots, \omega_k)$ denotes permutation of the frequencies $\omega_1, \ldots, \omega_k$.

The equations in (4) can be solved in general form. The result is particularly simple in the case of strict resonance,

$$\omega_1 + \omega_2 + \ldots + \omega_k = \omega_{\mu\nu}, \tag{6}$$

if the initial conditions are chosen to be $a_\nu(0) = 1$ and $a_\mu(0) = 0$. In this case the probability of finding the atomic system in states with energies ϵ_μ and ϵ_ν during succeeding instants of time are determined by the expressions

$$|a_\mu(t)|^2 = \sin^2 \Omega_k t, \qquad |a_\nu(t)|^2 = \cos^2 \Omega_k t, \tag{7}$$

where the oscillation frequency takes the form

$$\Omega_k = \Big| \sum_{(\omega_1+\ldots+\omega_k)} f_{\mu\nu}(\omega_1, \ldots, \omega_k) \Big|. \tag{8}$$

It follows naturally for single-quantum resonance ($\omega = \omega_{\mu\nu}$) from (5) that

$$f_{\mu\nu}(\omega) = V_{\mu\nu}(\omega)/\hbar,$$

and (7) and (8) coincide with the known expressions.[1]

In a real situation, where the level width is finite, the quantum oscillations will appear only if the period Ω_k greatly exceeds the corresponding relaxation times. If Ω_k is smaller than the relaxation time, then it is possible to introduce in the usual fashion the probability of multiquantum transition per unit time

$$W_k = 2\pi \Big| \sum_{(\omega_1+\ldots+\omega_k)} f_{\mu\nu}(\omega_1, \ldots, \omega_k) \Big|^2 \delta(\omega_1 + \ldots + \omega_k - \omega_{\mu\nu}). \tag{9}$$

It follows from the form of Ω_k and W_k that multiquantum resonance can be realized in a number of ways, provided condition (6) is satisfied. It is important here that the oscillation frequency Ω_k and the transition probability W_k are determined by the sum of the multiquantum-transition amplitudes $f_{\mu\nu}$ from each individual resonant situation. This uncovers a possibility of realizing cases in which all the transition amplitudes $f_{\mu\nu}$ in the sum (8) or (9) are equal to zero (or are close to it). Then the level populations do not change with time and are equal respectively to $|a_\nu(t)|^2 = 1$ and $|a_\mu(t)|^2 = 0$, i.e., the medium remains in its ground state.

We shall determine below, using a concrete example, the conditions that lead to the vanishing of Ω_k.

3. Let the external field consist of four waves with frequencies ω_1, ω_2, ω_3, and ω_4 satisfying the resonance conditions $\omega_1 + \omega_2 = \omega_{\mu\nu}$ and $\omega_3 + \omega_4 = \omega_{\mu\nu}$. Then, taking into account the type of interaction between the medium and the electromagnetic field (3), we find that the frequency of the quantum oscillations is given by

$$\Omega = |f_{\mu\nu}(\omega_1, \omega_2) + f_{\mu\nu}(\omega_3, \omega_4)| = \hbar^{-1} |D_{\mu\nu}(\omega_1, \omega_2) E(\omega_1) E(\omega_2) + D_{\mu\nu}(\omega_3, \omega_4) E(\omega_3) E(\omega_4)|, \tag{10}$$

where

$$D_{\mu\nu}(\omega_1, \omega_2) = \frac{1}{\hbar} \sum_n d^z_{\mu n} d^z_{n\nu} \left(\frac{1}{\omega_1 - \omega_{n\nu}} + \frac{1}{\omega_2 - \omega_{n\nu}} \right).$$

We confine ourselves for simplicity to the case of waves that are linearly polarized in one direction (along the z axis).

We see therefore that by suitable choice of the phases and amplitudes of the external waves we can cause Ω to vanish. For a nondegenerate atomic system when the matrix $d_{\mu\nu}$ is real, it is necessary in this case to satisfy the following conditions:

$$D_{\mu\nu}(\omega_1, \omega_2) R_1 R_2 = D_{\mu\nu}(\omega_3, \omega_4) R_3 R_4, \qquad (11)$$

$$\varphi_1 + \varphi_2 - \varphi_3 - \varphi_4 = \pi, \qquad (12)$$

$$\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3 + \mathbf{k}_4, \qquad (13)$$

where it was assumed that

$$E(\omega_j) = R_j \exp\{i\mathbf{k}_j\mathbf{r} + i\varphi_j\}, \quad j = 1, 2, 3, 4.$$

Equations (11) and (12) are the conditions imposed on the intensities and phases of the external waves. Equation (13) is the well known spatial synchronism condition of nonlinear optics.

The foregoing example includes the particular case when two waves with frequencies ω and 3ω are incident on the medium, and the resonance is realized at the frequency 2ω. It follows from (10)—(12)—that such an aggregate of coherent waves will go through the substance without absorption if the condition of spatial synchronism $3\mathbf{k}_1 = \mathbf{k}_3$ is satisfied, as well as the conditions

$$a = 1, \quad \varphi = 0,$$

$$a = \frac{R_3}{R_1}\frac{D_{\mu\nu}(-\omega, 3\omega)}{D_{\mu\nu}(\omega, \omega)}, \quad \varphi = \varphi_3 - 3\varphi_1 - \pi. \qquad (14)$$

Thus, the process of suppression of two-photon absorption depends to an essential degree on the phase relations of the waves passing through the medium. When conditions (14) are violated the probability of the quantum oscillations Ω will be given by

$$\Omega = |1 - a\exp\{i\varphi + i\Delta\mathbf{k}\mathbf{r}\}|\,\Omega_0, \qquad (15)$$

and the probability W of two-photon transition will be given by

$$W = |1 - a\exp\{i\varphi + i\Delta\mathbf{k}\mathbf{r}\}|^2\, W_0, \qquad (16)$$

where $\Delta\mathbf{k} = \mathbf{k}_3 - 3\mathbf{k}_1$ and Ω_0 and W_0 pertain to two-photon resonance for the wave of frequency.

In this case the medium becomes absorbing. When $\Delta\mathbf{k} \neq 0$ the absorption varies periodically as the waves pass through the medium, from a minimum $(1 - a)^2 W_0$ to a maximum $(1 + a)^2 W_0$. Such a change in absorption will take place at distances $l = \pi/\Delta k$.

We did not take into account above the reaction of the medium to the external radiation; this can be done for sufficiently weak fields, in sufficiently rarefied media, or at small distances. Allowance for the reaction of the medium leads to a nonlinear electrodynamic problem and will not be considered here. For the particular case of third-harmonic generation, such an analysis was presented by us in [2], where it was shown that the conditions (14) are realized automatically under conditions of strict resonance at frequency 2ω and in the presence of spatial synchronism, and such a state of the interacting concrete waves is stable.

As to the effect of suppression of two-photon absorption, this phenomenon can apparently be observed in the radio band. On the one hand, we actually do not encounter the spatial-synchronism problem here, since the corresponding wavelengths greatly exceed the dimensions of the investigated samples. On the other hand, the choice of the medium is an exceedingly simple problem. We can use a paramagnetic material placed in a magnetic field. By varying the latter, we can readily produce the corresponding resonance.

We note in conclusion that there is a very close analogy between the "bleaching" effect considered in this paper and the effect of suppression of inelastic channels of nuclear reactions in resonant nuclear scattering in crystals [4]. In the latter case, the medium can become analogously bleached as a result of the presence of a coherent superposition of two plane waves (incident and diffracted), synchronized in such a way that the amplitude for the production of a compound nucleus, which is a sum of the amplitudes of each of the waves, turns out to be equal to zero.

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