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Translated by J. G. Adashko

Ionization broadening of atomic spectra

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 (Submitted July 14, 1976)
 Zh. Eksp. Teor. Fiz. 72, 896–906 (March, 1977)

The spontaneous emission by an atom and the stimulated absorption of a weak test wave are considered with account taken of the process of resonant ionization induced by an intense pumping wave. The spontaneous and stimulated emission (and absorption) spectra are found for atomic transitions in a system of levels coupled by a resonant field, as well as for transitions to some third level. Spectral curves describing two-photon transitions with emission or absorption of a photon having a frequency close to the pumping frequency are obtained.

PACS numbers: 32.80.Bx, 32.80.Kf

1. INTRODUCTION

Earlier investigations of the influence of the intense resonant emission on atoms and molecules dealt with the absorption of a resonant wave by a two-level system with allowance for the saturation effect,^[1,2] and with transitions of such a system to a third level with absorption or emission of photons of a different frequency.^[3] The spontaneous emission by a two-level system in a resonant field and the stimulated absorption of a weak test wave with a frequency near the resonant frequency by this field have been studied by many authors.^[4–7] The resonant interaction between groups of closely spaced levels was the subject of^[8,9], while the resonant interaction in a three-level system was examined in^[10]. In addition to the resonant interaction between two discrete levels, the possibility of ionization was also taken into account.^[11–13]

In studying the emission and absorption spectra of a two-level system,^[1–7] the broadening mechanisms that are frequently studied are collisions between atoms, radiative decay of the levels, and Doppler broadening. It is to be noted that in the interaction between atoms and an intense optical radiation, single-photon and multiphoton ionizations of the atoms can also play a significant role in the formation of the spectra.^[14]

The present study is devoted to an investigation of spontaneous emission, as well as to stimulated transitions in the field of a weak electromagnetic test wave, when an intense resonant pumping wave acts upon the atom, and when account is taken of resonant ionization of the atom.

An interesting methodological feature of the problem

of ionization broadening of atomic spectra is that the corresponding results can be obtained directly from the Schrödinger equation for a single atom, without the use of the concept of the density matrix and without introducing any phenomenological constants into the equations. This is due to the fact that in the case of ionization the irreversibility of the processes is due to the continuity of the spectrum of the ionized atom rather than to its multiparticle character. In this respect ionization broadening resembles greatly the spectral broadening due to radiative decay of levels.^[15]

Resonant ionization of an atom in the field of a strong electromagnetic wave $\mathbf{F}^{(0)} = \mathbf{F}_0 \cos \omega t$ has been studied both in particular cases^[16,17] and in a more general formulation of the problem.^[11–13] The possibility of solving such a problem for a single atom is actually due to a single principal requirement^[12] $F_0 \ll F_{at}$, where $F_{at} = 5 \times 10^9$ V/cm is the field strength inside the atom. A system of quasi-energy wave functions of the atom in the field of a resonant wave (we assume $\hbar = 1$) was determined:

$$\Psi_E = e^{-iEt} \psi_E, \quad (1.1)$$

$$\psi_E = a_E^0 e^{i(n+m)\omega t} \varphi_0 + a_E^1 e^{im\omega t} \varphi_1 + \int b_{E,E'} \varphi_{E'} dE'$$

with the known functions $a_E^{0,1}$, and $b_{E,E'}$, that are independent of time.^[12] Here E is the quasi-energy, $0 \leq E < \infty$, $\varphi_{0,1}$ are the wave functions of the ground and resonant states (with energies $E_{0,1}$), φ_E are the wave functions of the continuous spectrum with energy E , while n and m are the multiplicity of the resonance and the minimum number of quanta required for the transition $\varphi_1 \rightarrow \varphi_E$.

The state of an atom in the field of an intense reso-

nant wave is characterized by three parameters: the detuning $\varepsilon = E_1 - E_0 - n\omega$, the ionization width $\Gamma_i = 2\pi |V_{1E}|^2$, $E = E_1 + m\omega$, and the so-called field width $\Gamma_f = 4|V_{01}|$, $V = -\frac{1}{2}\mathbf{d}\mathbf{F}_0$; \mathbf{d} is the dipole moment of the atom, V_{1E} and V_{01} are the composite matrix elements of orders m and n .

The wave function of the atom corresponding to the instantaneous switching on of the interaction with the strong resonant field and to the initial condition $\Psi(0) = \varphi_0$ is of the form

$$\Psi = \int dE e^{-iEt} \psi_E (a_E^0)^* \quad (1.2)$$

2. ABSORPTION AND EMISSION DURING TRANSITIONS TO THE NON-RESONANT ATOMIC LEVELS

Let the atom be acted upon by an intense pumping wave \mathbf{F}^0 of frequency ω as well as by a weak electromagnetic test wave of different frequency: $\mathbf{F}^{(1)} = \mathbf{F}_1 \cos \Omega t$. We consider first the case when the frequency $n\omega$ of the resonant transition (as well as from other frequencies that are multiples of ω). Let E_k and φ_k be the energy and the wave function of an arbitrary discrete atomic level (different from the ground and the resonant levels), let $\Omega \approx E_k - E_0$, and let the transition $\varphi_0 \rightarrow \varphi_k$ be dipole-allowed. The interaction with the weak field $\mathbf{F}^{(1)}$ can be taken into account by perturbation theory, where Eq. (1.2) must be used for the unperturbed wave function.

Ionization broadening of the spectral absorption line is possible in the considered case only when the periods of the strong and weak pulses exceed the ionization time. The total energy absorbed by an individual atom can be expressed under these conditions in the form

$$\Delta \mathcal{E} = \frac{\Omega \Gamma_f \Gamma_i |W_{k0}|^2}{4^2 |E_a - E_b|^2} \times \left| \frac{1}{E_a'' (E_a - E_b^*) (E_k - E_0 - \Omega - E_a)} + \frac{1}{E_b'' (E_a^* - E_b) (E_k - E_0 - \Omega - E_b)} \right|^2 \quad (2.1)$$

where

$$W = -\frac{1}{2} \mathbf{d} \mathbf{F}_1, \quad E_{a,b}'' = \text{Im } E_{a,b}, \\ E_{a,b} = E_0 + (n+m)\omega + \frac{1}{2} \varepsilon - \frac{1}{2} i \Gamma_i \pm \frac{1}{2} [(\varepsilon - i \Gamma_i / 2)^2 + \Gamma_f^2 / 4]^{1/2}.$$

The absorption coefficient α of the weak wave $\mathbf{F}^{(1)}$ (per unit length) is related to $\Delta \mathcal{E}$ by

$$\alpha = 8\pi N_a \Delta \mathcal{E} / c F_1^2 \Delta t, \quad (2.2)$$

where N_a is the atom concentration, Δt is the interaction period, $\Delta t = \min(\tau, d/v)$, v is the velocity of the atoms, d is the characteristic dimension of the interaction region, and τ is the duration of the pulse $\mathbf{F}^{(0)}$. Equation (2.1) is simplified in the limiting cases of small and large ionization widths. When $\Gamma_i \ll \Gamma_f$ we have

$$\Delta \mathcal{E} = \frac{\Omega |W_{k0}|^2}{R_f^2} \sum_{\pm} \frac{(R_f \mp \varepsilon)^2}{[\Omega - E_k + E_0 + \frac{1}{2} \varepsilon (\pm R_f)]^2 + \frac{1}{16} \Gamma_f^2 (1 \pm \varepsilon / R_f)^2}, \\ R_f = [\varepsilon^2 + \frac{1}{4} \Gamma_f^2]^{1/2}. \quad (2.3)$$

The absorption line at the frequency $\Omega \approx E_k - E_0$ is split by the resonant field into two lines with different ionization widths. This type of splitting into a three-level system was studied in^[3], where, however, the broadening mechanism was not specified, and the width was introduced in the equation for the density matrix by means of a phenomenological parameter.

In the opposite limiting case when $\Gamma_i \gg \Gamma_f$ we have

$$\Delta \mathcal{E} = \Omega |W_{k0}|^2 [(\Omega - E_k + E_0)^2 + (\Gamma_i \Gamma_f^2 / 32 R_f^2)^2]^{-1}, \\ R_f = (\varepsilon^2 + \frac{1}{4} \Gamma_f^2)^{1/2}. \quad (2.4)$$

Consideration of the resonant ionization leads only to a broadening of the absorption line for the transition $\varphi_0 \rightarrow \varphi_k$, where the line width, as was to be expected, is equal to the inverse ionization period of the atom $\Gamma_i \Gamma_f^2 / 16 R_f^2 \sim \frac{1}{4} \Gamma_i^{-1} \Gamma_f^2$.

Equation (2.1) also takes on a comparatively simple form in the case of exact resonance $\varepsilon = 0$ at an arbitrary relation between Γ_i and Γ_f :

$$\Delta \mathcal{E} = \Omega |W_{k0}|^2 \frac{(\Omega - E_k + E_0)^2 + \frac{1}{4} \Gamma_f^2}{[(\Omega - E_k + E_0)^2 - \frac{1}{16} \Gamma_f^2]^2 + \frac{1}{4} \Gamma_f^2 (\Omega - E_k + E_0)^2}. \quad (2.5)$$

It follows from this, in particular, that the shape of the absorption line differs in the general case from a Lorentz line when the resonant ionization is taken into account.

Equations (2.1) and (2.3)–(2.5) are obtained by neglecting the width of the level E_k as well as its ionization width. The last assumption imposes definite limitations on the position of this level. If k is the number of quanta of frequency required for the ionization of the E_k level, then, as can be readily verified, the ionization of this state may be neglected when $k > \max(m, n - m)$.

Under the same conditions, emission at the frequency $\Omega \approx E_1 - E_k$ is possible if the resonant state φ_1 is dipole-coupled with φ_k . The total energy emitted by the atom at the frequency Ω may be determined in a manner fully analogous to the previous case; it is of the form

$$\Delta \mathcal{E} = \frac{\Omega |W_{k1}|^2 \Gamma_f^2 \Gamma_i^2}{4^2 |E_a - E_b|^2} \times \left| \frac{E_a / E_a''}{(E_a - E_b^*) (E_k - E_0 - n\omega + \Omega - E_a)} + \frac{E_b / E_b''}{(E_a^* - E_b) (E_k - E_0 - n\omega + \Omega - E_b)} \right|^2. \quad (2.6)$$

Equation (2.2) describes in this case the amplification coefficient of the weak wave.

In the particular cases of small and large ionization widths, we have in place of (2.3) and (2.4), respectively,

$$\Delta \mathcal{E} = \frac{\Omega \Gamma_f^2 |W_{k1}|^2}{4 R_f^2} \sum_{\pm} \left[\left(E_k - E_0 - n\omega + \Omega - \frac{\varepsilon}{2} \mp \frac{R_f}{2} \right)^2 + \frac{\Gamma_f^2}{16} \left(1 \pm \frac{\varepsilon}{R_f} \right)^2 \right]^{-1}, \\ \Gamma_i \ll \Gamma_f, \quad (2.7)$$

$$\Delta \mathcal{E} = \frac{\Omega \Gamma_f^2 |W_{k1}|^2}{16 R_f^2} \left[(E_k - E_0 - n\omega + \Omega)^2 + \left(\frac{\Gamma_f \Gamma_f^2}{32 R_f^2} \right)^2 \right]^{-1}, \quad \Gamma_i \gg \Gamma_f. \quad (2.8)$$

Equation (2.8) describes the two-stage process $E_0 + n\omega \rightarrow E_1$, $E_1 - \Omega \rightarrow E_k$, and the broadening of the Raman-scattering spectrum is determined, just as in the case of

direct absorption (2.4), by the width of the ground state $\Gamma_i \Gamma_f^2 / 16R_i^2 \sim \Gamma_f^2 / 4\Gamma_i$.

In the case of exact resonance, the expression for $\Delta \mathcal{E}$ takes on the form

$$\Delta \mathcal{E} = \frac{1}{16\Omega} |W_{k_1}|^2 \Gamma_f^2 \left\{ [(E_k - E_0 - n\omega + \Omega)^2 - \frac{1}{16}\Gamma_f^2]^2 + \frac{1}{4}\Gamma_i^2 (E_k - E_0 - n\omega + \Omega)^2 \right\}^{-1}. \quad (2.9)$$

The spontaneous emission by an atom, which is due to the transitions $\varphi_1 \rightarrow \varphi_k$ and is possible when $E_1 > E_k$, has the same spectral properties as the stimulated emission described by Eqs. (2.6)–(2.9). To determine the spectral intensity of emission $d\mathcal{E}/d\Omega$ it is necessary to replace in these expressions the matrix element W_{k_1} by $\mathbf{d}_{k_1} \cdot \mathbf{e}$ (\mathbf{e} is the polarization vector of the emitted photon), to introduce the dimensional factor $\Omega^3/2\pi^2 c^3$, and to integrate over the directions of \mathbf{e} .

3. SPONTANEOUS EMISSION BY AN ATOM WITH TRANSITIONS IN A SYSTEM OF RESONANT LEVELS

We now examine transitions in which both the initial and the final states of the atom in the process of spontaneous emission belong to the class of states ψ_E (1.1), i. e., they remain within the framework of the basis $(\varphi_0, \varphi_1, \varphi_E)$. We first investigate spontaneous emission by an atom in the range of frequencies Ω near the resonant-transition frequency $n\omega$. We use the system of functions ψ_E , (1.1), as the basis, expression (1.2) as the unperturbed wave function, and the interaction operator of the atom with the quantized field as the perturbation.^[15] We find in first-order perturbation theory that up to instant t the spectral intensity of emission by the atom at frequencies near $n\omega$ can be expressed in the form

$$\frac{d\mathcal{E}}{d\Omega} = \frac{2|d_{01}|^2 \Omega^4}{3\pi} \int dE \left| \int_0^t d\tau \int dE' a_{E'}^* a_{E'} a_E^* \exp\{i(\Omega - n\omega + E - E')\tau\} \right|^2. \quad (3.1)$$

In deriving this formula we summed over the polarizations and integrated over the directions of propagation of the emitted photons. The matrix element d_{01} in the simplest case of the $S \rightarrow P$ transition is equal to $\langle \psi_{P0} | d_z | \psi_S \rangle$, where the second subscript in the final-state function labels the zeroth projection of the orbital momentum on the z axis.

The integrals in Eq. (3.1) can be calculated fairly easily, but the resulting expressions are quite cumbersome. We therefore present the calculation results only in the limiting cases $\Gamma_i \ll \Gamma_f$ and $\Gamma_i \gg \Gamma_f$, when the pulse duration and observation time exceed the ionization time of the atom, as necessary for the ionization broadening of the spontaneous emission.

If the ionization width is small, $\Gamma_i \ll \Gamma_f$, then in the limit $\Gamma_i t \gg 1$ the spectral density of atom emission takes the form

$$\frac{d\mathcal{E}}{d\Omega} = \frac{|d_{01}|^2 \Gamma_f^2 \Omega^4}{24\pi R_i^2} \left\{ \sum_{\pm} \frac{R_{i\pm E}}{(\Omega - n\omega)^2 + \frac{1}{4}\Gamma_i^2 (1 \mp \varepsilon/R_i)^2} + R_i \sum_{\pm} \frac{1}{(\Omega - n\omega \pm R_i)^2 + \frac{1}{4}\Gamma_i^2} \right\}. \quad (3.2)$$

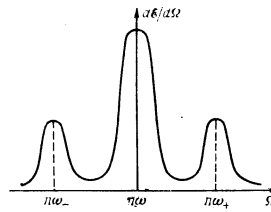


FIG. 1. Spectral intensity of spontaneous emission by an atom in the vicinity of the frequency $\Omega \sim n\omega$ of the resonant transition at $\Gamma_i \ll \Gamma_f$, $n\omega_{\pm} = n\omega \pm [\varepsilon^2 + (\Gamma_f/2)^2]^{1/2}$.

The first term in the curly brackets describes the shape of the emission line near the central frequency $n\omega$, which is a superposition of two Lorentz contours. The second term corresponds to emission at shifted frequencies $n\omega \pm R_f$. The dependence of $d\mathcal{E}/d\Omega$ on the frequency is illustrated qualitatively in Fig. 1. As a consequence of the condition $\Gamma_i \ll \Gamma_f$, the shift is significantly greater than the width of the maxima. It is interesting to note that, in the case of large detuning $|\varepsilon| \gg \Gamma_f$, a narrow peak of width $\Gamma_i \Gamma_f / 2^{3/2} |\varepsilon|$ appears in the vicinity of the central frequency, on top of the background of a broad line (width $\sim \Gamma_i$).

Equation (3.2), just as the results (2.3) and (2.7) of the preceding section, admits of the following illustrative interpretation. In the approximation $\Gamma_i \ll \Gamma_f$ one may assume that each of the levels E_0 and E_1 is split under the influence of resonant emission, into two levels $E_{0,1}^{\pm}$ with ionization widths Γ_{\pm} :

$$E_0 \rightarrow E_0^{\pm} = E_0 + \frac{1}{2}\varepsilon \pm \frac{1}{2}R_i, \quad E_1 \rightarrow E_1^{\pm} = E_1 - \frac{1}{2}\varepsilon \pm \frac{1}{2}R_i, \quad (3.3)$$

$$\Gamma_{\pm} = \frac{1}{2}\Gamma_i (1 \pm \varepsilon/R_i).$$

The level scheme (3.3) describes correctly transitions to a third level (Eqs. (2.3), (2.7)) as well as transitions within the system of levels E_0 and E_1 (Eq. (3.2)), if the width of the emission (absorption) line is defined as the sum of the widths of the initial and final levels $E_{0,1}^{\pm}$. If the ionization width is large, $\Gamma_i \gg \Gamma_f$, only one clearly pronounced maximum appears in the spontaneous-emission spectrum of the atom in the vicinity of the frequency n

$$\frac{d\mathcal{E}}{d\Omega} = \frac{|d_{01}|^2 \Gamma_f^2}{12\pi R_i^2} \frac{1}{(\Omega - n\omega)^2 + (\Gamma_f/2 + \Gamma_i/16R_i^2)^2}. \quad (3.4)$$

The width of the spectral line is determined by the reciprocal of the time of the atomic ionization. Equation (3.4) describes the broadening of the resonant fluorescence spectrum,^[15] under monochromatic pumping, on account of the finite width of the ground state, equal to $\Gamma_i \Gamma_f^2 / 16R_i^2$ in this case. Processes of higher order, corresponding to nonresonant absorption or emission of a certain number of strong-wave quanta with emission of a photon having a frequency that is a multiple of ω , are also of the same type as the examined transitions (in the basis $(\varphi_0, \varphi_1, \varphi_E)$). To describe these processes it is necessary, according to perturbation theory, to take into account the nonresonant coupling of the states ψ_E with the remaining atomic states φ_k . This leads in second-order perturbation theory to a spectral intensity of the spontaneous emission in the form

$$\frac{d\mathcal{E}}{d\Omega} = \frac{\Omega^4}{4\pi^2} \int d\omega_0 \int dE \left\{ (\bar{d}_{01} \cdot \mathbf{e})^2 \sum_{\pm} \left| \int dE' \int_0^t d\tau a_{E'}^* a_{E'} a_E^* \times \right. \right.$$

$$\times \exp[i(E-E'+\Omega-n\omega\pm\omega)\tau] \Big|^2 + \left| \int dE' \int d\tau a_{E'}^{*0} (a_E^0 a_E^{*0} \bar{d}_{00} e + a_{E'}^1 a_E^{*1} \bar{d}_{11} e) \exp[i(E-E'+\Omega-\omega)\tau] \right|^2 \Big\}, \quad (3.5)$$

where \bar{d}_{ij} are composite matrix elements of second order and d_{0e} is the element of solid angle in the direction of the polarization vector e of the emitted photon.

The first term in (3.5) describes spontaneous emission of an atom at frequencies near $(n \mp 1)\omega$, $n \neq 1$. Comparison of this term with Eq. (3.1) shows that the emission spectrum at the frequencies $\Omega \sim (n \mp 1)\omega$ duplicates the spectral curves that determine the emission of an atom in the vicinity of the resonant frequency $n\omega$ (Eqs. (3.2), (3.4)). In the quantitative sense, the emission lines in the vicinity of the frequencies $(n \mp 1)\omega$ are weaker than the resonant emission because $|\bar{d}_{01}/d_{01}|^2 \sim F_0^2/F_{at}^2$.

The second term in (3.5) describes spontaneous emission of an atom at frequencies near the pump frequency ω , i. e., it describes the Rayleigh-scattering spectrum of the atom with account taken of the splitting and broadening by the resonant wave. In the case of small ionization widths Γ_i , Eq. (3.5) yields

$$\left(\frac{d\mathcal{E}}{d\Omega} \right)_\omega = \frac{\Omega^4}{8\pi^2 R_i^3} \int d\omega_e \left\{ \frac{4}{\Gamma_i^2} \sum_{\pm} (R_i \pm \epsilon) \frac{[\bar{d}_{11} e \Gamma_i^2/4 + \bar{d}_{00} e (R_i \mp \epsilon)]^2}{(\Omega - \omega)^2 + \Gamma_i^2 (1 \pm \epsilon/R_i)^2/4} + R_i |\bar{d}_{00} - \bar{d}_{11}| e|^2 \sum_{\pm} \frac{(R_i \pm \epsilon)^2}{(\Omega - \omega \pm R_i)^2 + \Gamma_i^2/4} \right\}. \quad (3.6)$$

In the spectral sense, this result is similar to Eq. (3.2), but the emission lines are weaker than those in the vicinity of the resonant frequency $n\omega$ because of the small parameter $(F_0/F_{at})^2$.

Finally, in the case of a large ionization width $\Gamma_i \gg \Gamma_f$, we find from Eq. (3.5)

$$\left(\frac{d\mathcal{E}}{d\Omega} \right)_\omega = \frac{\Omega^4}{\pi^2} \int d\omega_e \frac{|\bar{d}_{00} e|^2}{(\Omega - \omega)^2 + (\Gamma_i \Gamma_f^2/16R_i^2)^2}. \quad (3.7)$$

The interpretation of this result (in analogy with (3.4)) is that Eq. (3.7) describes nonresonant Rayleigh scattering whose spectrum is broadened as a consequence of the finite width $\Gamma_i \Gamma_f^2/16R_i^2$ of the ground state. Quantitative comparison of Eqs. (3.4) and (3.7) (which are valid when $n > 2m$) shows that in the case $n = 2m + 1$

$$(d\mathcal{E}/d\Omega)_\omega \sim (d\mathcal{E}/d\Omega)_{n\omega},$$

and when $n > 2m + 1$

$$(d\mathcal{E}/d\Omega)_\omega \gg (d\mathcal{E}/d\Omega)_{n\omega}.$$

Thus, whereas in the case of a small ionization width $\Gamma_i \ll \Gamma_f$ the most intense spontaneous emission lines of the atom are located in the region of the resonant frequency $n\omega$, in the opposite case $\Gamma_i \gg \Gamma_f$ the emission at the pump frequency may be comparable in magnitude (at $n = 2m + 1$) with the emission at the frequency $n\omega$, or may even exceed it (at $n > 2m + 1$).

4. STIMULATED ABSORPTION AND EMISSION FOR TRANSITIONS IN A SYSTEM OF RESONANT LEVELS

In contrast to transitions to nonresonant atomic levels (Sec. 2), stimulated transitions in the basis of functions ψ_E (1.1) require a separate investigation, for in these cases the energy conservation laws admit of both absorption and emission of a photon of frequency Ω . This leads under certain conditions to cancellation of the direct and inverse processes, and to qualitatively different new features of the stimulated transitions in comparison with the spontaneous ones.

Let us examine anew, as in Sec. 2, the interaction of an atom in a field $F^{(0)}$ with a weak test pulse $F^{(1)} = F_1 \cos \Omega t$, assuming this time that the frequency Ω can be close to $k\omega$, where $k = 1, 2, \dots, n$. We assume that the duration of the test pulse $F^{(1)}$ does not exceed the time of action of the strong field $F^{(0)}$ on the atom. We present the results of an investigation based on a semiclassical approach, by determining the rate of the energy dissipation at the frequency Ω in the form

$$\frac{d\mathcal{E}}{dt} = F^{(1)} \frac{d}{dt} \langle d \rangle, \quad (4.1)$$

where $\langle d \rangle$ is the average dipole moment of the atom in the fields $F^{(0)}$ and $F^{(1)}$, and the bar denotes averaging over the rapid oscillations. The physical sense of this equation is evident, since the right side of (4.1) describes the work performed per unit time by the field $F^{(1)}$ on the atom.

In computing the average dipole moment $\langle d \rangle$ we again use the system of functions ψ_E (1.1) as the basis. We take into account the interaction with the field $F^{(1)}$ by perturbation theory, using Eq. (1.2) as the zeroth-order approximation function. If the fields $F^{(0)}$ and $F^{(1)}$ are completely uncorrelated, then averaging over their phase differences must be carried out in the final results. For correlated fields, the need for this averaging arises in the summation over the different atoms if the dimensions of the interaction region are sufficiently large, as is assumed in the following. The results obtained in this manner agree fully with the results obtained by the quantum-electrodynamical method.

It can be easily verified that under the foregoing assumptions a non-zero contribution to $d\mathcal{E}/dt$ (4.1) comes from the dipole moment of first order (in F_1). The expression for the total energy dissipated at the frequency Ω in the time t can be expressed in this approximation in the form

$$\Delta\mathcal{E} = n\omega |W_{01}|^2 \int dE \left\{ |a_E^1|^2 \left| \int_0^t d\tau \int dE' |a_{E'}^0|^2 \exp[i(E-E'+n\omega-\Omega)\tau] \right|^2 - |a_E^0|^2 \left| \int_0^t d\tau \int dE' a_{E'}^{*0} a_{E'}^1 \exp[i(E-E'-n\omega+\Omega)\tau] \right|^2 \right\}. \quad (4.2)$$

We note that the second term in the curly brackets of this equation agrees, apart from a normalization factor, with Eq. (3.1), i. e., it describes the emission process, whereas the first term describes absorption of a photon of frequency Ω .

If the ionization width $\Gamma_i \ll \Gamma_f$ is small, no ionization broadening occurs if the duration of the test pulse is short in comparison with the ionization time of the atom Γ_i^{-1} . Introducing in this case the switching function of the test wave $f(t)$, and assuming that $f_{\max} = f(t_0) = 1$, we obtain from (4.2)

$$\Delta \mathcal{E} = \frac{\pi^2 n \omega |W_{01}|^2}{2R_f^3} \left\{ \sum_{\pm} (R_f \pm \varepsilon) \exp \left[-\frac{\Gamma_i t_0}{2} \left(1 \mp \frac{\varepsilon}{R_f} \right) \right] \right\} \times \left\{ \sum_{\pm} (R_f \pm \varepsilon)^2 |f_{\pm}|^2 \right\}, \quad (4.3)$$

$$v = \Omega - n\omega \mp R_f,$$

where

$$f_{\pm} = (2\pi)^{-1} \int e^{i v t} f(t) dt$$

is the Fourier transform of the switching function. The exponential factors in the first sum of (4.3) describe the change in the populations of the states $\varphi_{0,1}$ during the process of resonant ionization.

If the durations of the test pulse and of the pumping pulse exceed the ionization time Γ_i^{-1} , and if $\varepsilon \neq 0$, then the total energy dissipated at the frequency Ω is

$$\Delta \mathcal{E} = 4 \frac{|W_{01}|^2 \varepsilon n \omega}{\Gamma_f^2 R_f} \left\{ \sum_{\pm} \frac{\varepsilon \pm R_f}{(\Omega - n\omega \mp R_f)^2 + 1/4 \Gamma_f^2} \right. \\ \left. + \frac{(n\omega - \Omega) \Gamma_f^2}{4R_f^2} \sum_{\pm} \frac{R_f \pm \varepsilon}{(\Omega - n\omega)^2 + 1/4 \Gamma_f^2 (1 \mp \varepsilon/R_f)^2} \right\}. \quad (4.4)$$

The first term describes stimulated absorption and amplification at frequencies shifted with respect to $n\omega$ by $\pm R_f$. The second term describes the structure of the absorption spectrum in the vicinity of the central frequency (Fig. 2a). As can be readily seen, it is of higher order of smallness in the parameter Γ_i/Γ_f than the first term. In the case of exact resonance $\varepsilon = 0$, Eq. (4.4) vanishes. In this case, however, absorption in the vicinity of frequencies $n\omega \pm R_f$ becomes possible in first order of Γ_i/Γ_f . The spectrum $\Delta \mathcal{E}(\Omega)$ (Fig. 2b) is then given by

$$\Delta \mathcal{E} = 2 \frac{\Omega |W_{01}|^2}{\Gamma_f} \sum_{\pm} \frac{(\Omega - n\omega \mp 1/2 \Gamma_f) (\pm 1)}{(\Omega - n\omega \mp 1/2 \Gamma_f)^2 + 1/4 \Gamma_f^2}. \quad (4.5)$$

It follows from Eq. (4.4) that amplification of the test signal is possible at the frequency $n\omega - \varepsilon R_f/|\varepsilon|$. This result, as well as the form of the spectral curves in the case $\Gamma_i \ll \Gamma_f$ under consideration (Fig. 2), resembles in many respects the situation of stimulated transitions in a two-level system with phenomenologically introduced parameters that describe the level widths.^[4-7] The structure of the spectral lines described by Eq. (4.4) can be interpreted in the same manner as in the case of spontaneous emission (Eq. (3.2)) in terms of the transitions between the split and broadened levels $E_{0,1}^{\pm}$ (3.3).

In the case of a large ionization width $\Gamma_i \gg \Gamma_f$, and at pulse durations exceeding the ionization time of the atom ($\sim \Gamma_i \Gamma_f^{-2}$), calculation based on Eq. (4.2), with allowance for smallness of the parameter Γ_f/Γ_i , gives

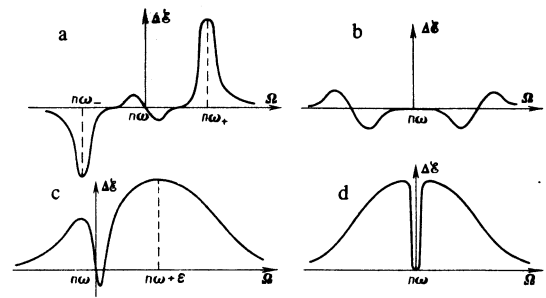


FIG. 2. Test-wave energy $\Delta \mathcal{E}$ dissipated by an atom at the frequency $\Omega \sim n\omega$ in the cases: a) $\Gamma_i \ll \Gamma_f \sim \varepsilon$; b) $\Gamma_i \ll \Gamma_f$, $\varepsilon = 0$; c) $\Gamma_f \ll \Gamma_i \sim \varepsilon$; d) $\Gamma_f \ll \Gamma_i$, $\varepsilon = 0$.

$$\Delta \mathcal{E} = 4n\omega |W_{01}|^2 \left\{ -\frac{1}{R_f^2} \frac{\varepsilon(\Omega - n\omega) + \Gamma_f^2 \Gamma_f^2 / 32 R_f^2}{(\Omega - n\omega)^2 + (\Gamma_f \Gamma_f^2 / 16 R_f^2)^2} + 4 \frac{R_f^2 / \Gamma_f^2}{(\Omega - n\omega - \varepsilon)^2 + \Gamma_f^2 / 4} \right\}. \quad (4.6)$$

The first term in (4.6) arises in consequence of the superposition of the two-stage transitions $E_0 + n\omega - E_1$, $E_1 - \Omega - E_0$ and $E_0 + \Omega - E_1$, $E_1 - n\omega - E_0$. The broadening of the spectrum is connected with the ionization width of the ground state. The second term in Eq. (4.6) describes direct absorption of a photon of frequency Ω in the transition $\varphi_0 \rightarrow \varphi_1$. The width of this line is determined by the ionization width of the resonant state. A plot of $\Delta \mathcal{E}(\Omega)$ is shown in Fig. 2c. The case of exact resonance (Fig. 2d) is distinct, since at $\varepsilon = 0$ the positions of the extrema of the individual terms in Eq. (4.6) coincide.

If the duration t of the test pulse is smaller than the atomic ionization time, but larger than the ionization time of the resonant level, a narrow spectral line of width $\sim \Gamma_f^2 \Gamma_i^{-1}$ does not have sufficient time to form, and we obtain instead of Eq. (4.6)

$$\Delta \mathcal{E} = \frac{\Omega |W_{01}|^2 \Gamma_f t}{(\Omega - n\omega - \varepsilon)^2 + 1/4 \Gamma_f^2} \exp \left(-\frac{\Gamma_f \Gamma_f^2 t_0}{16 R_f^2} \right), \quad (4.7)$$

where the exponential term denotes the degree of depletion of the ground state of the atom through ionization by the instant t_0 when the test pulse is turned on. Consequently, in stimulated absorption of the test wave at $\Gamma_i \gg \Gamma_f$, ionization broadening of the spectral line becomes possible even at interaction times shorter than the ionization time of the atom.

We point out that the interpretation of the results pertaining to a large ionization with $\Gamma_i \gg \Gamma_f$ (Eqs. (2.4), (2.8), (3.4), (3.7), (4.6), (4.7)) is based on the premise that the levels E_0 and E_1 of the free atom are shifted on account of the nonresonant Stark effect and are broadened by the ionization. The widths of the levels E_0 and E_1 are respectively equal to the reciprocal ionization time $\sim \Gamma_i^{-1} \Gamma_f^2$ of the atom and to the reciprocal ionization time $\sim \Gamma_i$ of the resonant state.

Finally, let us examine the stimulated absorption and emission caused by two-quantum transitions through non-resonant atomic levels. As in the case of spontaneous emission, such transitions turn out to be possible primarily at frequencies Ω near $(n \pm 1)\omega$ or near ω . The energy dissipated at the frequencies $\Omega \sim (n \pm 1)\omega$ is deter-

mined by formulas such as (4.2)–(4.6), where it is necessary to replace the frequency $n\omega$ by $(n \pm 1)\omega$ and the matrix element W_{01} by the composite second-order matrix element \bar{W}_{01} . The spectral characteristics of the stimulated absorption do not differ in this case from those examined above, and the absorbed (emitted) energy is lower than in the vicinity of the resonant frequency by a factor $(F_0/F_{at})^2$. When $\Omega \sim \omega$, the dissipated energy is determined by an expression of the form

$$\Delta \mathcal{E} = \omega \int dE \left\{ \left| \int dE' \int d\tau a_{E'}^{*0} (a_E^{*0} a_{E'}^0 \bar{W}_{00} + a_{E'}^{*1} a_E^1 \bar{W}_{11}) \right|^2 \right. \\ \times \exp[i(E - E' + \omega - \Omega)\tau] \left. - \left| \int dE' \int d\tau a_{E'}^{*0} (a_E^{*0} a_{E'}^0 \bar{W}_{00} \right. \right. \\ \left. \left. + a_{E'}^{*1} a_E^1 \bar{W}_{11}) \exp[i(E - E' - \omega + \Omega)\tau] \right|^2 \right\}, \quad (4.8)$$

where \bar{W}_{00} and \bar{W}_{11} are the diagonal composite matrix elements.

In the limiting cases $\Gamma_i \ll \Gamma_f$ and $\Gamma_i \gg \Gamma_f$ this expression yields respectively

$$(\Delta \mathcal{E})_0 = \frac{\omega \mathcal{E}}{R_i} (\bar{W}_{00} - \bar{W}_{11})^2 \sum_{\pm} \frac{(\pm 1)}{(\Omega - \omega \mp R_i)^2 + \Gamma_i^2/4}, \quad \Gamma_i \gg \Gamma_f, \quad (4.9)$$

$$(\Delta \mathcal{E})_0 = 4 \frac{\bar{W}_{00} \bar{W}_{11} \mathcal{E} \Omega (\omega - \Omega)}{R_i^2 [(\Omega - \omega)^2 + (\Gamma_i \Gamma_f^2 / 16 R_i^2)^2]}, \quad \Gamma_i \gg \Gamma_f, \quad (4.10)$$

Comparison of these expressions with Eqs. (4.4) and (4.6) shows that in both cases $(\Delta \mathcal{E})_\omega \sim (F_0/F_{at})^2 (\Delta \mathcal{E})_{n\omega} \ll (\Delta \mathcal{E})_{n\omega}$. As applied to the case $\Gamma_i \gg \Gamma_f$, this conclusion differs from the corresponding result for spontaneous emission. This is due to the cancellation, in stimulated transitions, of the fundamental process responsible for the spontaneous emission by atoms at frequencies near the pump frequency ω .

We express our deep gratitude to V. A. Khodovoi who

stimulated our interest in the problem of the ionization broadening of atomic spectra, as well as to V. P. Makarov for many useful discussions.

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Translated by J. G. Adashko

Multiphoton processes in the radiation field of a multimode laser

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(Submitted June 15, 1976)
Zh. Eksp. Teor. Fiz. **72**, 907–917 (March 1977)

The probabilities of multiphoton ionization by single-mode and multimode laser pulses of given mean field strength have been measured. For a power-law five-photon process, the probability in the field of the multimode laser is greater by a factor of 5! For an eleven-photon process, the corresponding factor has been found experimentally to be much less than 11! This reduction is explained by a departure from the power-law variation in the region approaching tunneling.

PACS numbers: 42.60.He, 42.65.Bp

§1. INTRODUCTION

One of the main problems in studying elementary nonlinear optical processes is the determination of the absolute probabilities of bound-bound and bound-free tran-

sitions.^[1,2] For technical reasons, the experimental data are usually more readily obtained by using the radiation from multimode lasers. For a given mean intensity in a radiation pulse, the transition probability is determined essentially by the mode content of the laser