probability of the inelastic scattering than in the case of a steplike potential,9) as seen, for example, from formula (38). The effect then decreases exponentially with increasing frequency and in real cases it can be noted only at $\omega \sim 10^3$ sec^{-1}, corresponding to small changes of the energy. Therefore the experimental observation of the inelastic interaction of UCN with such potentials is much more complicated. Nonetheless, the effect of heating of the UCN when reflected from an oscillating (as a result of current pulsations) magnetic wall must be taken into account when designing magnetic traps. As follows from the foregoing, the most dangerous here are the low frequencies.

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1) Such a barrier can be realized, for example, by placing a ferromagnetic foil in an alternating magnetic field parallel to the surface of the foil.

2) A particular case of such a potential, namely a rectangular potential threshold, was discussed by us earlier in an analysis of the heating of UCN by reflection from a weakly oscillating wall of a trap.[1] A more general dependence of U_0 on x can be produced for example, with the aid of a magnetic field from specially arranged current-carrying conductors.[2]

3) Vladimirskiĭ has shown[2] that such a potential can be realized in actual constructions of magnetic traps.

4) For this purpose, for example, we can register the absorption of the UCN in a thin foil that perturbs weakly the wave function of the neutron. In order for the registration efficiency to be independent of the UCN momentum, the foil material must have a near-zero coherent neutron scattering length.

5) Inasmuch as in all the examples considered above the momentum distribution of the UCN that are acted upon by the nonstationary potential is subsequently independent of the time, it is possible to use for these measurements, for example, the method of spatial separation of UCN beams moving with different momentum components perpendicular to an equipotential plane and having equal but nonzero momentum components in this plane. It is convenient to direct the primary UCN beam at a grazing angle with the equipotential plane.

6) We use here the customary units. Accordingly, p_0 is replaced by the UCN velocity $v_0 = p_0/m$, and z is multiplied by $(\hbar/m)^{1/2}$.

7) For example, interference filters.

8) Therefore, in particular, the major role may be assumed by effects connected with the non-ideal character of the barrier (finite height and width) and the specific features of its real structure (in the case of a magnetic barrier, for example, by the character of the behavior of the magnetic field near the barrier).

9) This is natural, for the effective time of interaction of the UCN with the wall increases in this case.

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A canonical description of multiquantum resonance interactions of radiation with matter

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It is shown that the response of matter in multiphoton resonance interactions with light involving an arbitrary number of energy levels can be described by the Neumann equation with a certain effective Hamiltonian $\mathscr{V}$ in the right-hand side, and with off-diagonal matrix elements that do not vanish for resonant transitions. The explicit form of this effective Hamiltonian is found. The polarization of the matter can be determined, with allowance for the effects of saturation and the Stark shifts of the levels, with the aid of $\mathscr{V}$ and a generalized dipole moment D that depends on the amplitudes and phases of the interacting fields. As a result, the description of complex resonance interactions of matter with strong fields, including coherent processes, is much simplified. The following matters are treated as examples: induced transition probabilities for an arbitrary number of resonances, the stationary nonlinear susceptibility of a molecule in incoherent and coherent multiphoton processes, and the nonlinear dielectric constant that arises in q-photon scattering of an ultrashort pulse.

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Among the multitude of nonlinear optical phenomena observed in recent years, one can distinguish a large group of resonance processes that take place when conditions of the following type are satisfied:

$$\sum_j n_j\omega_j = \omega_{mn} + \nu_s, \qquad (1)$$

where ω_{mn} are the resonance frequencies of the matter,

ω_j are the frequencies of the interacting fields, ν_s are the deviations from the resonance frequencies (they are much smaller than ω_j and ω_{mn}), and $\sum_j n_j = q_s$ is the order of the s-th resonance. In addition to one-photon interactions, multiphoton absorption and emission, as well as Raman processes of various orders and resonance parametric (i.e., phase dependent) interactions of the fields that take place when several conditions of the type of (1) are satisfied for a single transition, are also to be included among the resonance processes. Even more complicated processes involving resonances of various orders with several transitions in the matter have been observed.[1]

It frequently turns out to be practically impossible to describe resonance interactions by the methods of traditional radiation theory. The difficulties with the theory of resonance interactions are due to the necessity of taking the following circumstances into account (often simultaneously): a) the fields produced by lasers are sometimes so strong as considerably to alter the relative populations of the energy states of a quantum system, even in the case of high-order resonances; b) the interaction time may be very short ($\sim 10^{-12}$ sec), so that the nonstationary aspect of the response of the matter must be taken into account; and c) parametric (i.e., phase dependent) resonance interactions of the fields are possible because of the coherence of the radiation, and this adds the further problem of investigating the effect of the phases of the fields on the dynamics of the resonance interaction. The problem of describing a one-photon resonance was to a considerable extent solved by the introduction of the two-level-molecule model.[2] It was shown in[3] that all the important factors that determine the response of the matter in the case of a two-photon resonance can be taken into account in abridged equations derived by averaging[4] from Neumann's equations for the density matrices. After that similar equations were obtained for processes in which first-, second-, and third-order resonances are simultaneously active.[5,6] These same equations were later derived by other authors.[7–10] It is noteworthy that in all the enumerated situations the abridged equations have the same form as the equations for a two-level system, the difference being that the coefficients contain parameters representing characteristics of all the states of the molecule and the amplitudes of the fields acting on it.

The above circumstance led the authors to the conclusion that a general approach to the description of resonance interactions of arbitrary order taking place between any number of energy levels of the material medium should be possible. We shall see that the response of the matter can be obtained with the aid of the "slow" part of the density matrix, $\bar{\sigma}$. The variation of $\bar{\sigma}$ is described by a time-averaged equation that coincides with Neumann's equation and contains a certain effective Hamiltonian on the right-hand side, whose off-diagonal elements vanish except for resonance transitions. This substantially simplifies the problem of complex resonance interactions of a field with matter; in the case of given fields, at least, this problem becomes quite analogous to the problem of one-photon resonance interactions with the same number of levels as participate in the multiphoton processes. It will be shown that the polarization of the matter can be obtained in the form $\mathrm{Sp}(\mathfrak{D}\bar{\sigma})$, where $\mathfrak{D}$ is a generalized dipole moment that depends on the fields taking part in the multiphoton processes and is simply related to the effective Hamiltonian.

The theory makes it possible to take into account all the physical factors that determine the response of the matter for both incoherent and coherent (parametric) multiphoton processes: population saturation, the dynamic Stark shift, and the difference in the linear polarizabilities of the levels, as well as the interference of the probability amplitudes for transitions between the levels due to different resonances. We shall consider the following as examples: 1) induced transition probabilities resulting from an arbitrary number of resonances; 2) the nonlinear susceptibility of the molecules in the case of q-photon incoherent interaction with fields whose energies exceed the energy necessary to saturate the populations and to produce a Stark shift equal to the line width; 3) the behavior of the nonlinear dielectric constant $\varepsilon_{\mathrm{nl}}$ of the matter under q-photon absorption and scattering of ultrashort pulses; and 4) the classification of resonance parametric interactions with respect to the presence of phase locking of the generated waves.

1. AVERAGED MOTION OF A QUANTUM SYSTEM UNDER AN ARBITRARY NUMBER OF MULTIQUANTUM RESONANCE PROCESSES

1. Let us consider a quantum system (a molecule) acted on by the electromagnetic field

$$\mathbf{E} = \sum_j \mathfrak{E}(\omega_j, t)\exp(i\omega_j t), \tag{2}$$

in which the $\mathfrak{E}(\omega_j, t)$ are slowly varying functions of time as compared with the corresponding $\exp(i\omega_j t)$ functions, and some of the frequencies ω_j satisfy the resonance conditions (1).

In the absence of relaxation, the behavior of the system is described by the Neumann equation

$$\dot{\sigma} = -i\hbar^{-1}[V, \sigma], \tag{3}$$

in which σ is the density matrix and V is the Hamiltonian of the system in the interaction representation:

$$V_{mk} = \sum_j V_{mk}^{(j)}(t)\exp[i(\omega_{mk}+\omega_j)t], \tag{4}$$

the $V_{mk}^{(j)}(t)$ being slowly varying functions of time. We shall discuss the phenomenological introduction of relaxation later.

The possibility of simplifying the set of matrix equations (3) is due to the fact that they contain small parameters. To extract these parameters it is sufficient to introduce the dimensionless time $t' = \omega_{mn} t$. Then the ratios $V_{kr}^{(j)}/\hbar\omega_{mn}$ of the amplitudes of the interaction energy to the transition energies appear on the right in Eq. (3); these ratios are much smaller than unity even in fields that can be achieved either by focusing the light from a Q-switched laser or in picosecond pulses from mode-locked lasers (radiant flux density of the order of 10^{11} W/cm^2). Most of the observed nonlinear optical phenomena take place at much lower radiation intensities.

The fact that the $V_{kr}^{(j)}/\hbar\omega_{mn}$ are small gives reason to believe that the density matrix should not only contain fast oscillations with characteristic times of the order of $2\pi/\omega_j$ and $2\pi/\omega_{mn}$, but should also exhibit slow variations with a characteristic time determined by the molecule-field interaction energy. It is well known that separating the "fast" and "slow" variations in equations having small parameters can result in simpler equations. One of the mathematical methods commonly used to do this is the averaging method.[4] We shall apply this method directly to the set of equations (3) without reducing it to the standard form.[4]

Let us find the equations for the variations of the slow part of the density matrix. In accordance with the averaging method we express σ as the sum of the slow ($\bar{\sigma}$) and fast ($\tilde{\sigma}$) parts:

$$\sigma=\bar{\sigma}+\tilde{\sigma}=\bar{\sigma}+\sum_q \tilde{\sigma}^{(q)}, \tag{5}$$

where $\tilde{\sigma}^{(q)}$ is determined with the aid of the equation $\dot{\tilde{\sigma}} = -i\hbar^{-1}[V, \tilde{\sigma}^{(q-1)}]$. The behavior of the slow part of the density matrix is given in the p-th approximation by the equation

$$\dot{\bar{\sigma}}=-i\hbar^{-1}\langle[V, (\bar{\sigma}+\tilde{\sigma}^{(1)}+\ldots+\tilde{\sigma}^{(p)})]\rangle. \tag{6}$$

The angle brackets indicate averaging over a time that is much longer than the characteristic times $2\pi/\omega_{mn}$ and $2\pi/\omega_j$ of the fast variations but much shorter than the "interaction times" $\hbar/|V_{mn}|$ and the times in which the slow amplitudes $V_{mn}^{(j)}(t)$ change significantly. In averaging and calculating $\tilde{\sigma}^{(q)}$, all the slowly varying functions of time ($V_{mn}^{(j)}$, $\bar{\sigma}_{mn}$, and $\exp(i\nu_s t)$) must be taken out from under the integral sign. As usual, it must be assumed that there was no field present at the initial time t_0, i.e., that $V_{mk}^{(j)}(t_0)=0$.

Let us consider the q-th term on the right in Eq. (6). As will be shown below, this term differs from zero in the presence of q-th order resonances. On substituting $\tilde{\sigma}^{(q-1)}$ into (6), it can be seen without difficulty that the latter consists of 2^q terms, which can be divided into groups in which $\bar{\sigma}$ stands respectively in the first, second, ..., m-th place. It can be shown that the group of terms of the commutator $[V, \tilde{\sigma}^{(q-1)}]$ in which $\bar{\sigma}$ stands in the m-th place vanishes unless $m=1$ or $m=q+1$. For this it is sufficient to note that they can be expressed as the average of a derivative with respect to the fast time: $\langle(d/d\tilde{t})(\ldots)\rangle=0$. That this assertion is correct for small q can be easily seen directly from (6) and the expression for $\tilde{\sigma}^{(q-1)}$; then it follows for the $(q+1)$-th term in (6) by induction.

Thus, in each term of (6), only those members in which $\bar{\sigma}$ stands in the first or the last place differ from zero. Making use of the fact that the average of a total derivative with respect to the fast time vanishes, we can finally write the equation for $\bar{\sigma}$ in the form

$$\dot{\bar{\sigma}}=-i\hbar^{-1}[\mathscr{V}, \bar{\sigma}], \tag{7}$$

in which

$$\mathscr{V}=\sum_{q=1}^{p}\mathscr{V}^{(q)}=\langle V\rangle+\frac{i}{\hbar}\left\langle\left(\tilde{\int} V\,dt\right)V\right\rangle+\ldots$$
$$+\left(\frac{i}{\hbar}\right)^{p-1}\left\langle\left(\tilde{\int}\left(\ldots\left(\tilde{\int}\left(\tilde{\int} V\,dt\right)V\,dt\right)\ldots\right)V\,dt\right)V\right\rangle. \tag{8}$$

Here the symbol ~ above the integral signs means that the integration is to be taken over the fast time (i.e., that all the slowly varying functions of time are to be treated as constants) and the fast varying part of the result of the integration is to be excluded. Thus, the motion of the slow part of the density matrix in the case of an arbitrary number of resonances of arbitrary orders is described by an equation having the canonical form (7) with the averaged Hamiltonian (8).

We note that Eq. (7) remains valid in the absence of low-order resonances (for example, if there is no one-photon resonance, then $\langle V\rangle=0$). In fact, the derivation of Eqs. (7) and (8) is based on the assumption that at least some of the matrix elements $\bar{\sigma}_{mn}$ are larger than any of the $\tilde{\sigma}_{mn}$. This condition always obtains for some of the diagonal elements $\bar{\sigma}_{nn}$ (which describe the populations of the levels) because of the condition $\mathrm{Sp}\,\bar{\sigma}=1$.

2. Let us examine the matrix elements of the averaged Hamiltonian. It is easy to see from Eqs. (4) and (8) that the off-diagonal matrix elements of the term $\mathscr{V}^{(q)}$ of the Hamiltonian $\mathscr{V}$ vanish except for transitions for which the conditions for a q-th order resonance are satisfied. For example, the quantities $\mathscr{V}_{12}^{(q)}$ for two levels 1 and 2 between which such a resonance takes place can be expressed in terms of the slow amplitudes of the interaction energy between the system and the electromagnetic field $V_{mk}^{(j)}$ (see Eq. (4)):

$$\mathscr{V}_{12}^{(q)}=v_{12}^{(q)}\exp(i\nu_q t), \tag{9}$$

in which $v_{12}^{(q)}$ coincides with $(K_q)_{12}$—the composite matrix element obtained on calculating the probabilities for a transition between levels 1 and 2 induced by the q-photon process, using ordinary perturbation theory methods.[11,12] The generalization to the case in which there are resonances of order lower than q in addition to the q-th order resonance consists, in accordance with Eq. (8), in excluding terms having resonance denominators from $v_{12}^{(q)}$. In the dipole approximation, $v_{12}^{(q)}$ can be written in the form

$$v_{12}^{(q)}=-\varkappa_{(q)}^{12}\prod_{j=1}^{q}\mathfrak{E}(\omega_j), \tag{10}$$

where $\varkappa_{(q)}^{12}$ is the q-th order polarizability tensor.[12] 1)

It is not difficult to see from Eq. (8) that in case there are several q-th order resonances for the 1–2 transition we have

$$\mathscr{V}_{12}^{(q)}=\sum_s v_{12}^{(qs)}\exp(i\nu_s t), \tag{11}$$

in which s numbers the q-th order resonances and ν_s is the frequency difference corresponding to the s-th resonance.

Unlike the off-diagonal elements, the diagonal elements $\mathscr{V}_{mm}^{(q)}=v_{mm}^{(q)}$ for even q differ from zero not only for the levels that take part in the resonance interactions, but in general for all the levels. The energy $\mathscr{V}^{(2)}_{mm}$ is obviously that of the quadratic Stark shift of the m-th level under the action of the high-frequency field, while the frequencies

$$\Omega_{mn}=\frac{\mathscr{V}^{(2)}_{mm}-\mathscr{V}^{(2)}_{nn}}{\hbar}=\frac{1}{\hbar}\sum_{\substack{a,b\\ j>0}}[\varkappa^{mm}_{ab}(\omega_j)-\varkappa^{nn}_{ab}(\omega_j)]\mathfrak{E}_a(\omega_j)\mathfrak{E}_b(\omega_j) \qquad (12)$$

are those of the Stark shifts of the $m-n$ transitions; here $\varkappa^{mm}_{ab}$ is the linear polarizability tensor for a molecule in the m-th energy state. It follows from Eq. (12) that the change in the frequency of the transition due to the action of the electromagnetic field can be expressed in terms of macroscopic quantities—the refractive indices n^{m}_{ab} of the molecules in the m-th energy state ($n^{m}_{ab}=(1+4\pi N\bar{\sigma}_{mm}\varkappa^{mm}_{ab})^{1/2}$, where $N\bar{\sigma}_{mm}$ is the number density of the molecules in that state). We emphasize that relation (12) between the Ω_{mn} and the polarizabilities obtains for both resonance and nonresonance interactions between the field and the molecule.

In concluding this subsection we note that small additions to the quadratic Stark shift arise not only from the susceptibilities responsible for the incoherent multiphoton processes, but also from those responsible for the coherent ones. For example, $\operatorname{Im}\mathscr{V}^{(3)}_{mm}$ contains a term proportional to the susceptibility χ_{abc}, which is associated with the parametric addition of frequencies in a nonlinear transparent medium.

3. Some remarks are in order concerning the limits of applicability of Eq. (7) with the averaged Hamiltonian (8).

The results obtained above by the averaging method are valid provided no new resonances arise on account of changes in the frequencies of the material system due to the action of the external fields.[13] In other words, not only the given frequency differences ν_α, but also the nonlinear frequency differences arising from the interaction with the field, must be small as compared with all the linear combinations of field frequencies with resonant frequencies of the molecule (excluding combinations that were taken into account in the resonance conditions (1)$^{2)}$). The Stark shift frequencies (12) are obviously nonlinear deviations. Thus, Eq. (7) is valid provided the condition

$$\Omega_{mn}\ll\sum_j l_j\omega_j-\omega_{sr} \qquad (l_j=1,2,\ldots,q_{max}), \qquad (13)$$

where $l_j\neq n_j$ when $s,r=m,n$, is satisfied in addition to the condition $V^{(j)}_{kr}/\hbar\omega_{mn}\ll 1$. We note that condition (13) is actually the same as the condition $V^{(j)}_{kr}/\hbar\omega_{mn}\ll 1$ when $\sum l_j\omega_j-\omega_{sr}\sim\omega_{mn}$.

4. Now let us discuss the phenomenological introduction of relaxation into Eq. (7). In solving problems of quantum radiophysics one ordinarily introduces a relaxation operator of the form

$$(\Gamma\sigma)_{mn}=\begin{cases}T^{-1}_{mn}\sigma_{mn} & n\neq m\\ \sum\limits_{n}(\sigma_{mm}W_{mk}-\sigma_{kk}W_{km}) & n=m\end{cases}. \qquad (14)$$

into Eq. (3). It was shown in[3,5] that taking relaxation into account in the initial equations (3) in this manner leads to the appearance of small additions T^{-1}_{mn} to the line widths in the abridged second-approximation equations for the density matrix. The relative magnitude of these additions is $\sim(V_{mk}/\hbar\omega_{mn})^2\ll 1$, and they can be neglected. A correction to the transition probabilities W_{mk} between the levels also arises on account of the interaction between the matter and the field (2) in the tails of the absorption lines.[3] This correction is negligibly small provided the frequency difference from all the "nonresonance" transitions is much larger than the corresponding line width. In this situation we can obviously take the relaxation into account by introducing the relaxation operator (14) at once into Eq. (7).

2. MOLECULAR POLARIZATION IN RESONANCE PROCESSES; THE GENERALIZED DIPOLE MOMENT

In accordance with (5), the polarization arising in resonance interactions of a molecule with an electromagnetic field is given by

$$\mathrm{P}=\sum_j\mathfrak{P}(\omega_j)\exp(i\omega_j t)=\operatorname{Sp}\{\mathrm{d}(\bar{\sigma}+\tilde{\sigma})\}, \qquad (15)$$

where $\mathbf{d}$ is the dipole moment operator in the interaction representation. The term $\operatorname{Sp}\{\mathbf{d}\bar{\sigma}\}$ represents the polarization component at the frequencies $\omega_{mn}=\omega_\alpha-\nu_\alpha$ of the resonance transitions. The spectral components $\mathbf{P}(\omega_j)$ at frequencies ω_j involved in multiphoton resonance processes are due to the second term in (15), which is proportional to $\tilde{\sigma}$. The rapidly varying part of the density matrix is related to the slowly varying part $\bar{\sigma}$, so the entire polarization (15) can obviously be expressed in terms of $\bar{\sigma}$. Then the proportionality constant, which has the dimensions of a dipole moment, will depend on the fields taking part in the multiphoton process.

Now let us find this "generalized dipole moment" operator and show that its off-diagonal matrix elements, like those of the Hamiltonian (8) for the averaged motion, vanish except for resonance transitions, and that its diagonal matrix elements are connected by the condition $\sum l_j\omega_j=0$. To do this we first find the amplitude of the spectral component of the polarization at one of the frequencies ω_j that occur in the q-th order resonance condition ($q>1$; we first consider the nondegenerate case):

$$\mathfrak{P}^{(q)}(\omega_j)=\operatorname{Sp}\langle\mathrm{d}\exp(-i\omega_j t)\tilde{\sigma}^{(q-1)}\rangle. \qquad (16)$$

Multiplying (16) by $-\mathfrak{E}^*(\omega_j)$ and adding the resulting equation to its complex conjugate, we obtain

$$\mathfrak{P}^{(q)}(\omega_j)\mathfrak{E}^*(\omega_j)+\text{c.c.}=\langle\mathrm{P}^{(q)}(\omega_j)\mathrm{E}(\omega_j)\rangle=-\operatorname{Sp}\langle V^{(j)}\tilde{\sigma}^{(q-1)}\rangle. \qquad (17)$$

Now by substituting the expression for $\sigma^{(q-1)}$ into Eq. (17) and making use of the property of differentiation with respect to the fast time as we did in deriving the averaged Hamiltonian (7), we can show that the polarization at the frequency of any of the fields taking part in a q-photon process is related as follows to $\mathscr{V}^{(q)}$:

$$\langle\mathrm{P}^{(q)}(\omega_j)\mathrm{E}(\omega_j)\rangle=-\operatorname{Sp}\{\mathscr{V}^{(q)}\bar{\sigma}\}=-\Big(\sum_{m>n}\mathscr{V}^{(q)}_{mn}\bar{\sigma}_{nm}+\text{c.c.}+\sum_{m}\mathscr{V}^{(q)}_{mm}\bar{\sigma}_{mm}\Big). \qquad (18)$$

Accordingly, we obtain the following expression for the total polarization $P^{(q)}$:

$$\langle \mathbf{P}^{(q)}\mathbf{E}\rangle = \sum_{j=-q}^{q} \mathfrak{P}^{(q)}(\omega_j)\mathfrak{E}^{\cdot}(\omega_j) = -q\,\mathrm{Sp}\{\mathscr{V}^{(q)}\bar{\sigma}\}. \tag{19}$$

Now we can obviously introduce the generalized dipole moment operator

$$\mathbf{D}^{(q)} = \sum_{j} \mathfrak{D}^{(q)}(\omega_j)\exp i\omega_j t, \tag{20}$$

whose amplitude matrix determines the polarization amplitudes

$$\mathfrak{P}^{(q)}(\omega_j) = \mathrm{Sp}\,\{\mathfrak{D}^{(q)}(\omega_j)\bar{\sigma}\} \tag{21}$$

and can be expressed as follows in terms of the averaged Hamiltonian[3)]:

$$\mathscr{V}_{mn}^{(q)} = -\mathfrak{D}_{mn}^{(q)}(\omega_j)\mathfrak{E}^{\cdot}(\omega_j). \tag{22}$$

In view of the fact that $\mathscr{V}_{mn}^{(q)} = \{\mathscr{V}_{nm}^{(q)}\}^*$, the condition $\mathfrak{D}_{mn}^{(q)}(\omega_j) = \{\mathfrak{D}_{nm}^{(q)}(-\omega_j)\}^*$ is satisfied. Equation (22) admits of a simple physical treatment: the averaged interaction energy for any of the fields taking part in a q-photon interaction with the $m-n$ transition is analogous to the interaction energy of a field with a two-level system having the dipole moment $\mathfrak{D}_{mn}^{(q)}$, the quantity $\mathscr{V}_{mn}^{(q)}$ being the same for all the fields.

In view of the additivity of $\mathscr{V}$, both for resonances of different orders (see Eq. (8)) and for several resonances of the same order (see Eq. (11)), it follows from Eqs. (18) and (15) that the generalized dipole moment for the case of an arbitrary number of resonances has the form

$$\mathbf{D}_{mn} = \sum_{q_s, j} \mathfrak{D}_{mn}^{(q_s)}(\omega_j)\exp(i\omega_j t) = \sum_{j} \mathfrak{D}_{mn}(\omega_j)\exp(i\omega_j t), \tag{23}$$

where the index s is included to take account of the possibility that there may be several q-th order resonances. Correspondingly, the total polarization is given by

$$\mathbf{P} = \sum \mathbf{P}^{(q)} = \mathrm{Sp}(\mathfrak{D}\bar{\sigma}). \tag{24}$$

It is not difficult to show that all the amplitudes $\mathfrak{P}^{(q)}(\omega_j)$ (and hence all the polarizations $\mathbf{P}$) consist of two parts: a resonance part $\mathfrak{P}^{(q)\mathrm{res}}(\omega_j)$, associated with the q-th order resonance condition, and a nonresonance part $\mathfrak{P}^{(q)\mathrm{nonres}}(\omega_j)$, which may differ from zero even when there are no resonances. In fact, it was shown in Sec. 1 that the off-diagonal matrix elements $\mathscr{V}_{mn}^{(q)}$ vanish except in the presence of q-th order resonances; hence the first two terms in (18) determine the resonance part of the polarization:

$$\mathfrak{P}^{(q)\,\mathrm{res}}(\omega_j) = \sum_{m\neq n} \mathfrak{D}_{mn}^{(q)}(\omega_j)\bar{\sigma}_{nm}. \tag{25}$$

Formulas (10) and (22) enable us to express the $\mathfrak{D}_{mn}^{(q)}(\omega_j)$ in terms of the components of the q-th order polarizability tensor:

$$\mathfrak{D}_{mn}^{(q)}(\omega_j) = \varkappa_{(q)}^{mn}\mathfrak{E}^{\cdot}(\omega_1)\ldots\mathfrak{E}^{\cdot}(\omega_{j-1})\mathfrak{E}^{\cdot}(\omega_{j+1})\ldots\mathfrak{E}^{\cdot}(\omega_q)\exp i\nu_q t. \tag{26}$$

All the formulas obtained above are valid when there is no degeneracy. Using Eq. (19), it is not difficult to show that for an n_j-fold degenerate resonance of frequency ω_j we have

$$\mathfrak{P}^{(q)}(\omega_j)\mathfrak{E}^{\cdot}(\omega_j) = -n_j\,\mathrm{Sp}(\mathscr{V}^{(q)}\bar{\sigma}) \tag{27}$$

and consequently,

$$\mathscr{V}_{mn}^{(q)} = -n_j^{-1}\mathfrak{D}_{mn}^{(q)}(\omega_j)\mathfrak{E}^{\cdot}(\omega_j), \qquad (m>n). \tag{28}$$

The third term on the right in Eq. (18), which is proportional to $\mathscr{V}_{mm}$, may differ from zero if the field contains frequency components whose frequencies ω_j satisfy the condition $\sum_j l_j\omega_j = 0$. Hence it is the nonresonance part of the polarization that corresponds to this term; its amplitudes can be written in the form

$$\mathfrak{P}^{(q)\,\mathrm{nonres}}(\omega_j = \omega_1 + \ldots + \omega_{j-1} + \omega_{j+1} + \ldots + \omega_q) = \sum_{n} \mathfrak{D}_{nn}^{(q)}(\omega_j)\bar{\sigma}_{nn}, \tag{29}$$

the projections $\mathfrak{D}_{nn}^{(q)}$ being expressible in terms of the diagonal components of the polarizability tensors, $\varkappa_{a_1\ldots a_q}^{nn}$, with the aid of Eq. (26) with $m = n$ and $\nu_q = 0$.[4)]

As examples let us consider the $\mathfrak{P}^{(q)\mathrm{nonres}}$ with $q = 1$, 2, and 3. There is obviously no nonresonance polarization in the first order. When $q = 2$, the nonresonance polarization is determined by the linear susceptibility:

$$\{\mathfrak{P}^{(2)\mathrm{nonres}}(\omega_j)\}_a = \chi_{ab}\mathfrak{E}_b(\omega_j)$$

The part of the polarization $\mathbf{P}^{(3)\mathrm{nonres}}$ that is quadratic in the field coincides with the corresponding component of the polarization as calculated with second-order perturbation theory (see Sec. 13 in[14]) provided terms having resonance denominators are excluded from the susceptibility tensor χ_{abc}:

$$\{\mathfrak{P}^{(3)\mathrm{nonres}}(\omega_k = \omega_s + \omega_l)\}_a = \sum_{n} \{\mathfrak{D}_{nn}^{(3)}(\omega_k)\}_a\bar{\sigma}_{nn}$$

$$= \sum_{n,b,c} \bar{\sigma}_{nn}\varkappa_{abc}^{nn}(\omega_l, \omega)\mathfrak{E}_b(\omega_l)\mathfrak{E}_c(\omega_c) = \sum_{b,c} \chi_{abc}(\omega_l, \omega_s)\mathfrak{E}_b(\omega_l)\mathfrak{E}_c(\omega_s). \tag{30}$$

As is known, it is the polarization (30) that is responsible for three-frequency nonresonance parametric interactions such as frequency adding and doubling in nonlinear transparent media.

Thus, Eq. (24) describes the response of matter both to multiphoton resonance interactions, and to nonresonance interactions of the frequency components of the field (2). Since $\bar{\sigma}$ satisfies Eq. (13), the resonance part of (24) contains terms proportional to products of field components that take part in different multiphoton processes, including processes of different orders. This part of the polarization depends on the phase relations between the $\mathfrak{E}(\omega_j)$ and is due to their resonance parametric interactions.

3. THE GENERALIZED TWO-LEVEL SYSTEM

Interactions in which resonances of different orders take place for just one transition play an important part both in clarifying the basic laws governing resonance interactions of radiation with matter and in practical applications. If the populations of the levels that are not involved in the resonance transition can be neglected, then, with the relaxation operator (14) and the quadratic Stark shift (12) taken into account, Eq. (7) takes the form

$$\frac{d\bar{\sigma}}{dt}+(T^{-1}-i\Omega)\bar{\sigma}=\frac{i}{\hbar}\mathscr{V}\eta, \tag{31a}$$

$$\frac{d\eta}{dt}+\frac{\eta-\eta_0}{\tau}=-\frac{4}{\hbar}\operatorname{Im}(\bar{\sigma}\mathscr{V}^*). \tag{31b}$$

Here the subscripts 1 and 2 have been dropped from the quantities $\bar{\sigma}_{12}$, T_{12}, Ω_{12}, and $\mathscr{V}_{12}$; $\eta=\bar{\sigma}_{11}-\bar{\sigma}_{22}$ is the difference between the populations of the resonance levels; and τ is the time constant for relaxation of η to its equilibrium value η_0.

Thus, it turns out to be possible to generalize the equations for a two-level system to the case of multiphoton resonance interactions. Hence we shall call Eqs. (31a) and (31b) the equations of the generalized two-level system.5) [15]

As in the case of an ordinary two-level system, the populations $\bar{\sigma}_{11}$ and $\bar{\sigma}_{22}$ satisfy the normalization condition

$$\operatorname{Sp}\sigma=\operatorname{Sp}\bar{\sigma}=\bar{\sigma}_{11}+\bar{\sigma}_{22}=1. \tag{32}$$

This follows from the form of expression (5) for the density matrix since all the terms of the rapidly varying part $\tilde{\sigma}=\sum_q\tilde{\sigma}^{(q)}$ of the density matrix satisfy the condition $\operatorname{Sp}\tilde{\sigma}^{(q)}=0$.

When the conditions for the applicability of the equations of the generalized two-level system are satisfied it is convenient to use the following expression for the polarization amplitudes in terms of the variables $\bar{\sigma}$ and η in place of Eqs. (25) and (29):

$$\begin{aligned}\mathfrak{P}(\omega_j)=\mathfrak{P}^{\text{nonres}}(\omega_j)+\mathfrak{P}^{\text{res}}(\omega_j)=\tfrac{1}{2}\{\mathfrak{D}_{11}(\omega_j)+\mathfrak{D}_{22}(\omega_j)\\+[\mathfrak{D}_{11}(\omega_j)-\mathfrak{D}_{22}(\omega_j)]\eta\}+\mathfrak{D}_{21}(\omega_j)\bar{\sigma}.\end{aligned} \tag{33}$$

In the case of a quasistationary interaction6) the population difference satisfies the balance equation

$$\dot{\eta}+(\eta-\eta_0)\tau^{-1}=-2\eta W_{\text{ind}}, \tag{34}$$

in which W_{ind} is the probability for an induced transition between levels 1 and 2:

$$W_{\text{ind}}=2|v|^2T\hbar^{-2}[1+(\nu-\Omega)^2T^2]^{-1}. \tag{35}$$

It is easy to see from Eq. (34) that the time constant for changes in the population difference under the action of the fields—the "induced relaxation time"—is given by

$$\tau_{\text{ind}}=(w_{12}+w_{21})^{-1}=\tau(1+2\tau W_{\text{ind}})^{-1}, \tag{36}$$

where the total probabilities for transitions between these levels are

$$w_{12}=W_{12}+W_{\text{ind}},\quad w_{21}=W_{21}+W_{\text{ind}}.$$

Using Eqs. (35) and (36), we can find the condition for the interaction to be quasistationary; when $\tau\gg T$, this condition has the form

$$4|v|^2T^2\hbar^{-2}\ll 1+(\nu-\Omega)^2T^2. \tag{37}$$

In the case of a quasistationary interaction, the state of the molecule affects the polarization amplitudes only via the population difference:

$$\mathfrak{P}^{\text{res}}(\omega_j)=\hbar^{-1}Tv\mathfrak{D}^*(\omega_j)\eta\frac{i+(\nu-\Omega)T}{1+(\nu-\Omega)^2T^2} \tag{38}$$

(here $\mathfrak{D}^*(\omega_j)=\mathfrak{D}_{21}(\omega_j)=\mathfrak{D}^*_{12}(\omega_j)$). We note that the amplitudes and phases of the interacting fields may turn out to be such that the following conditions are satisfied:

$$v=e^{i\nu t}\sum_s v^{(s)}=0,$$

$$\mathfrak{D}(\omega_j)=e^{i\nu t}\sum_{s'}\mathfrak{D}^{(s')}(\omega_j)=0$$

(see Eqs. (11) and (28)). It is easy to see from Eq. (35) that in such cases $W_{\text{ind}}=0$ and the level populations assume their equilibrium values: $\eta=\eta_0$ (also see (31)). Then the resonance polarization also vanishes, while the nonresonance polarization has the same form as in the absence of resonance. Since field energy is not absorbed by the matter in such situations, we may say that we have parametric brightening of the matter. When $v=0$, transfer of energy from some frequencies to others can take place only as a result of the presence of nonresonance susceptibility.

The possibility of establishing equilibrium populations at definite nonvanishing field strengths in certain parametric resonance processes was pointed out in[7,16–18]. Here we have become convinced that parametric brightening of matter is a common characteristic of all such processes.

4. STATIONARY SUSCEPTIBILITY AND POPULATION DIFFERENCE FOR A RESONANCE TRANSITION UNDER INCOHERENT INTERACTIONS

By incoherent interactions of light with matter we mean those that take place when one of the resonance conditions (1) is satisfied: one- and many-photon absorption (emission) and Raman processes (in the latter, some of the frequencies occurring in (1) will have different signs). The theory developed above makes it very simple to find the population difference between the resonance levels and the susceptibility of the matter for resonances of any order in fields that are strong enough to saturate the populations and to shift the absorption line substantially. If the fields act on the molecule for a time longer than τ_{ind}, the stationary populations

$$\eta_{\text{st}}=\eta_0\{1+4\tau T\hbar^{-2}|v|^2[1+(\nu-\Omega)^2T^2]^{-1}\}^{-1}. \tag{39}$$

will be established. For incoherent interactions, η_{st} depends only on the field intensities $I_j=|\mathfrak{E}(\omega_j)|^2$:

$$\eta_{\text{st}}=\eta_0\left\{1+I_{\text{sat}}^{-q}\prod_{j>0}I_j^{n_j}\left[1+\left(\Delta+\sum_{j>0}K_jI_j\right)^2\right]^{-1}\right\}^{-1}. \tag{39a}$$

Here we have introduced the saturation intensity $I_{\text{sat}}=\hbar^{2/q}(4\tau T|\varkappa^{12}_{(q)}|^2)^{-1/q}$ and the dimensionless frequency mismatch $\Delta=\nu T$. The constant

$$K_j=\hbar^{-1}[\varkappa_{jj}^{22}(\omega_j)-\varkappa_{jj}^{11}(\omega_j)]T$$

determines the Stark shift of the transition frequency under the action of the field $\mathfrak{E}(\omega_j)$.

Now let us introduce the nonlinear susceptibility of the generalized two-level system at frequency ω_j by means of the relation

$$\{\mathfrak{P}(\omega_j)\}_a=\chi_{ab}(\omega_j)\mathfrak{E}_b(\omega_j). \tag{40}$$

This susceptibility consists of resonance and nonresonance parts (in accordance with Eq. (33)).

Using Eqs. (39a), (33), and (40), we can easily find the stationary susceptibility for a resonance of arbitrary order q. Let us write down its diagonal component for polarization of the field $\mathfrak{E}(\omega_j)$:

$$\chi_{jj}(\omega_j)=\chi_0(\omega_j)+\frac{\eta_0 T}{\hbar}|\varkappa_{(q)}|^2\left\{1+\left(\Delta+\sum_l K_l I_l\right)^2+I_{\text{sat}}^{-q}\prod_l I_l^{n_l}\right\}^{-1}$$
$$\times\left[2K_j I_j\vartheta-\left(\Delta+\sum_l K_l I_l\right)n_j-in_j\right]I_j^{n_j-1}\prod_{l\neq j}I_l^{n_l}, \tag{41}$$

where

$$\chi_0(\omega_j)={}^1/_2\{\varkappa_{jj}^{11}(\omega_j)+\varkappa_{jj}^{22}(\omega_j)+[\varkappa_{jj}^{11}(\omega_j)-\varkappa_{jj}^{22}(\omega_j)]\eta_0\} \tag{42}$$

and $\vartheta=2\tau/T$. The imaginary part of expression (41) determines the absorption (emission) of energy by the matter from (into) the field component $\mathfrak{E}(\omega_j)$, while the real part determines the refractive index of the matter for this field component.

To each of the physical factors that affect the response of the susceptibility to variations of the fields there correspond specific terms in (41). The term $K_j I_j \vartheta$ in the square brackets describes the effect on the susceptibility of the redistribution of populations between two levels 1 and 2 whose linear polarizabilities $\varkappa_{jj}^{11}$ and $\varkappa_{jj}^{22}$ differ from one another. The presence of the products $K_1 I_1$ as terms added to Δ leads to a change in the susceptibility resulting from the Stark shift of the center of the resonance line. Finally, the quantity

$$I_{\text{sat}}^{-q}\prod_l I_l^{n_l}$$

in the curly brackets in (41) is responsible for the saturation of $\chi_{jj}(\omega_j)$ associated with saturation of the populations in a q-photon process.

5. THE REAL PART OF THE NONLINEAR SUSCEPTIBILITY ARISING UNDER THE ACTION OF LIGHT PULSES

Investigation of the real part of the nonlinear susceptibility of matter arising under the action of light pulses is of interest in connection with the observed self-focusing of picosecond pulses in gases,[19,20] where the contributions to the nonlinear dielectric constant ε_{nl} from the Kerr effect and electrostriction are negligibly small.

Here we consider the ratio of the contributions to the nonlinear dielectric constant from the excitation of the matter and the dynamic Stark effect for various resonance processes that take place in the action of an ultrashort pulse on matter. It is usually assumed[20] that in such a case it is always the Stark effect that makes the main contribution to the magnitude and sign of ε_{nl}. Using the equations of the generalized two-level system, we can show that this assumption is not always correct for all processes.

Let us consider an ultrashort pulse whose length t_p satisfies the conditions

$$t_p \ll T;\ \tau;\ T\left|\Delta+\sum K_l I_l\right|^{-1}.$$

The last of these conditions means that the field frequency differs from the frequency at the center of the Stark-shifted line by less than the frequency width of the pulse. In this case the equation for $\overline{\sigma}$ and η can be solved by iteration. Using Eqs. (33) and (40), we obtain

$$\operatorname{Re}\chi_{jj}(\omega_j)-\chi_0(\omega_j)=\frac{\eta_0 T|v|^2}{\hbar I_j}\left\{K_j I_j-\frac{n_j}{2}\left(\Delta+\sum_l K_l I_l\right)\right\}\frac{t^2}{T^2}. \tag{43}$$

From this it follows that in the case of one-photon absorption ($l=j,\ n_j=1$) it is not the Stark effect that plays the principal part in forming ε_{nl}, but the excitation of the matter (the term $K_j I_j$). In induced Raman scattering ($n_j=1$) the sign of ε_{nl} at the pumping frequency is also determined by the excitation of the matter provided less than half of the absorbed pumping energy goes into the Stokes component; at the frequency of the latter, however, the sign of ε_{nl} is determined in this case by the Stark effect. It is interesting that in another two-photon process—degenerate two-photon absorption ($n_j=2$)—these two factors cancel one another out and the sign of ε_{nl} depends only on the frequency mismatch Δ. In third- and higher-order resonances, the Stark effect always makes the main contribution to ε_{nl}.

6. FEATURES OF THE RESPONSE OF MATTER IN PARAMETRIC RESONANCE INTERACTIONS

Parametric resonance interactions (PRI) are now already finding application in the construction of frequency-tunable sources of coherent light in the relatively unaccessible regions of the infrared and ultraviolet.[21,22] The most promising PRI from this point of view are those in which phase locking of the generated waves is possible. This enables one to achieve efficient generation even when there are considerable frequency differences between the waves.[23] A classification of PRI from the point of view of the possible occurrence of this phenomenon would therefore be of interest.

In order to obtain such a classification let us consider the susceptibility $\chi_{jj}^{\text{res}}(\omega_j)$ responsible for the PRI. We express $\chi_{jj}^{\text{res}}(\omega_j)$ in terms of the real wave amplitudes $\mathbf{A}_l$ and the slow phases φ_l defined by the equation

$$\mathfrak{E}(\omega_l)=\mathbf{A}_l\exp(-ik_l z-\varphi_l). \tag{44}$$

Using Eqs. (38), (11), (9), and (26), we can write

$$\chi_{jj}^{\text{res}}(\omega_j)=-\frac{\eta}{\hbar}\frac{i+(\nu-\Omega)T}{1+(\nu-\Omega)^2T^2}\sum_{s,s'}\varkappa_{(q_s)}\varkappa^{*}_{(q_{s'})}n_j^{(s')}$$

$$\times\exp\left\{i\sum_{l}(n_l^{(s')}+n_l^{(s)})(\varphi_l+k_lz)\right\}A_j^{m_j-2}\prod_{l\neq j}A_l^{m_l};\qquad(45)$$

$$m_{j,l}=n_{j,l}^{(s)}+n_{j,l}^{(s')}.$$

From this it follows that the parametric resonance processes can be divided into three groups as regards their relationship to the field component $\mathfrak{E}(\omega_j)$. The first group consists of the processes for which there is at least one of the resonances (1) that does not involve the frequency ω_j (i.e., for which $n_j^{(s)}=0$), while among the resonances that do involve ω_j there is at least one that is not degenerate in this frequency (i.e., for which $n_j^{(s')}=1$). Then when the amplitude A_j is small enough, $\chi_{jj}^{\text{res}}(\omega_j)$ will be proportional to A_j^{-1} and will tend to infinity as $A_j\to 0$ with the other field amplitudes remaining finite. Hence at the entrance face (or at $t=0$) the phase φ_j must suddenly assume a value such that the derivative φ_j', which is proportional to $\operatorname{Re}\chi_{jj}(\omega_j)$, will vanish, i.e., the phase of the weak field will be locked. The phase will obviously remain locked until the intensity grows to such an extent that $I_j=A_j^2$. Hence the spatial (or temporal) scale of the changes in the phase φ_j will not be determined by the deviations of the wave vectors of the interacting fields, but by the rate of growth of A_j characteristic of the given process.

The second group consists of the processes for which the lowest value that $n_j^{(s)}+n_j^{(s')}$ can assume is two. In this case $\chi_{jj}^{\text{res}}(\omega_j)$ is independent of A_j when the latter is small, and the variation of the phase will be determined by both the resonance and nonresonance parts of the susceptibility. The phase may become locked in a region in which $\operatorname{Re}\chi_{jj}(\omega_j)\approx 0$ provided there is a resonance that does not involve the frequency ω_l $(n_l^{(s)}=0)$ and the other frequency components of the field are strong enough.

The third group consists of the processes for which the lowest value that can be assumed by $n_j^{(s)}+n_j^{(s')}$ is greater than two. Since

$$\lim_{A_j\to 0}\chi_{jj}^{\text{res}}(\omega_j)=0,$$

in this case, there will be no phase locking, the behavior of the phase being determined by the nonresonance part of the susceptibility.

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[1] To avoid errors the following must be borne in mind: The frequency ω_j must be taken with the same sign in the expression for the polarizability and in $v_{12}^{(q)}$ as it has in the resonance condition (1).

[2] Of course it is sufficient that this condition be satisfied for combinations of no more than $q_{\max}$ frequencies, where $q_{\max}$ is the highest of the orders of the resonances (1) that are present in the linear approximation.

[3] It must be pointed out that the frequency ω_j occurs with the same sign in the quantity $\mathfrak{D}_{mn}^{(q)}(\omega_j)$ as determined by Eq. (22) as it has in the resonance condition $_{j=1}\,\omega_j=\omega_{mn}$. Thus, if $\omega_1-\omega_2=\omega_{21}$, then $\mathfrak{D}_{21}^2(\omega_1)$ is the amplitude for $\exp(i\omega_1 t)$, and $\mathfrak{D}_{21}^2(-\omega_2)$, that for $\exp(-i\omega_2 t)$.

[4] We call that the resonance terms must be excluded from the expressions for the $\varkappa_{a_1\ldots a_q}^{nn}$.

[5] When there are several resonant transitions, one can, with the aid of Eqs. (7), (8), and (14), easily write down the equations for a generalized n-level system, where n is the number of levels taking part in resonance interactions with the field.

[6] We are considering the situation, which is the one most frequently encountered in experiments, in which either there is only one resonance, or all the deviations from the corresponding resonance frequency are the same $(\nu_s=\nu)$.

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