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NONLINEAR POLARIZABILITY IN RESONANT INTERACTIONS OF AN ELECTROMAGNETIC FIELD WITH MATTER

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A theory of nonlinear polarizability (χ_{nl}) of matter in single- and two-photon absorption (emission) and stimulated Raman scattering is developed. Account is taken of the simultaneous action of all the factors leading to the formation of χ_{nl}, namely the difference between the linear polarizabilities of the matter in the ground and excited states, κ_1 and κ_2 (which is allowance for the influence of the multilevel nature of the system), saturation of the populations of these states, and the high-frequency Stark effect. The behavior of χ_{nl} is investigated as a function of the deviation from resonance and of the energy of the fields interacting with the matter, up to fields exceeding the saturating ones and the characteristic Stark field that shifts the absorption line by an amount equal to its half-width. A connection is established between the Stark constant and $\kappa_2 - \kappa_1$; it is shown that this connection makes it possible to determine this constant from measurements of the nonlinear polarizability of the matter in fields smaller by several orders of magnitude than the Stark field, even for strongly broadened and weakly resolved transitions. Expressions are obtained for χ_{nl} following the action of short pulses (including picosecond ones) on the matter. The estimates show that the resonant interactions can exert a strong influence on the self-action of light, and in many cases they can determine it; thus, for example, SRS can determine the self-focusing in the propagation of picosecond pulses in gases.

INTRODUCTION

MOST nonlinear-optics investigations involving the study of the nonlinearity of the refractive index are devoted to the nonresonant interaction of light with matter. However, an investigation of the nonlinear polarizability of matter under resonant interactions, such as single-photon absorption and emission, two-photon absorption (TPA), stimulated Raman scattering (SRS), etc. is also of interest for the following reasons:

a) Such a polarizability can influence the character of the self-action of light in cases when the contribution to the refractive index of the medium, due to the Kerr effect, striction, and heating is small or of the same order. It can also be assumed that the resonant interactions listed above can determine effects of self-action in vapors and gases, where there may be no other mechanisms of varying the polarizability. In addition, self-action of light resulting from resonant processes may appear in the case of amplification or generation (in lasers, amplifiers, in SRS).

b) Measurements of the nonlinear polarizability (χ_{nl}) can give additional information on the parameters of the material. Thus, it will be shown below that from the dependence of χ_{nl} on the light intensity it is possible to determine the linear polarizability of a substance in excited states, as well as the constants characterizing the high-frequency Stark shift of the levels.

The influence of strong resonant fields on the polarizability of matter χ is connected with three phenomena. First, the polarizability saturates, since saturation occurs in the population of the levels for which resonance occurs. Second, the linear polarizability of matter in the excited state differs in the general case from the polarizability of the ground state, and therefore the excitation of matter under the influence of the field causes a change of χ. Finally, the shift of the absorption, due to the Stark effect, can lead to a noticeable change of χ or the same field, if its frequency (or the algebraic sum of the frequencies of the fields acting on the substance) is close to resonance.

The first two causes, which are connected with the changes of the level populations in the substance, were discussed qualitatively many times in the literature[1-4], but there is still no consistent theory capable of taking into account simultaneously all the processes listed above and of explaining their interrelationship in concrete cases. We know of two papers[5,6] in which a quantitative calculation was made of the nonlinear polarizability in two particular cases. In[5] they considered the polarizability of a two-level system, due only to saturation of the populations. We shall show below in the calculation of χ_{nl} in the general case one cannot confine oneself to the purely two-level approximation; this can lead to errors, particularly in the calculation of χ_{nl} of gases and of the working media of lasers, for which the calculation was carried out in[5]. In[6] we obtained and discussed an expression for the polarizability resulting from SRS when the difference of the pump frequency and the Stokes component coincides with the center of the shifted Stark line of the transition. The conditions for the applicability of this expression are discussed in Sec. 3.

We now proceed to calculate general expressions for the polarizability of matter under the resonant processes indicated above.

1. INITIAL EQUATIONS

We represent the electromagnetic wave in the form

$$\sum_s \mathbf{E}_s \exp(i\omega_s t) + \text{c.c.}$$

We assume here that in the case of single-photon resonance there is only a wave of frequency $\omega_S = \omega_0$, and the resonance condition $\omega_0 = \omega_{21} + \delta$ is satisfied, whereas for TPA and SRS we have s = 1, 2, and $\omega_1 \pm \omega_2 = \omega_{21} + \delta$, where $\delta \ll \omega_S, \omega_{21}$; ω_{21} is the frequency of the transition between the levels 1 and 2 in the medium. To calculate the polarizability of the medium, resulting from the action of the field, we use the procedure described in[7,8]. As a result we find that the polarizability at the frequency ω_S is equal to $N\chi(\omega_S)$, where N is the number of particles per unit volume, and the polarizability of one particle is

$$\chi(\omega_s) = \varkappa_1^{(s)}\sigma_{11} + \varkappa_2^{(s)}\sigma_{22} + \mathrm{Re}(\Phi_{21}^{(s)}\sigma_{12}E_s^{-1}e^{-i\delta t}). \quad (1)$$

Here $\chi_j^{(s)}$ is the linear nonresonant part of the polarizability of the medium in states 1 and 2. For single-photon resonance we have

$$\varkappa_j^{(0)} = \frac{2}{\hbar}\sum_{n>2}\frac{|p_{jn}^{(0)}|^2\omega_{nj}}{\omega_{nj}^2 - \omega_0^2} - \frac{(-1)^j|p_{21}^{(0)}|^2}{\hbar(\omega_{21}+\omega_0)}, \quad (2)$$

and for TPA and SRS

$$\varkappa_j^{(s)} = \frac{2}{\hbar}\sum_{n>1}\frac{|p_{jn}^{(s)}|^2\omega_{nj}}{\omega_{nj}^2 - \omega_0^2}. \quad (3)$$

In (2) and (3), j = 1 and 2, and the summation is over all levels of the particle.

The quantity $\Phi_{21}^{(s)}$ for single-photon resonance, TPA, and SRS is given, respectively, by

$$\Phi_{21}^{(0)} = p_{21}^{(0)}, \qquad \Phi_{21}^{(1)} = r_{12}^* E_2^* \hbar^{-1}, \qquad \Phi_{21}^{(1)} = r_{12}^* E_2 \hbar^{-1}; \quad (4)$$

$\Phi_{21}^{(2)}$ is obtained from (4) by making the substitution $E_3^* \to E_1^*$ in the case of SRS. Let us determine also the quantity r_{12} in these formulas; this quantity characterizes the efficiency of the TPA:

$$r_{12} = \sum_{(s)}\left(\frac{p_{1n}^{(1)}p_{n2}^{(2)}}{\omega_{n2}+\omega_2} + \frac{p_{1n}^{(2)}p_{n2}^{(1)}}{\omega_{n2}+\omega_1}\right); \quad (5)$$

and in the case of the SRS the constant r_{12} is obtained from (5) by replacing ω_2 by $-\omega_2$. In formulas (2)–(5), ω_{jn} is the frequency of the transition between the states j and n, and $p_{jn}^{(s)}$ is the projection of the matrix element of the dipole moment of this transition on the interaction of the field of frequency ω_S.

Equation (1) contains the populations of the ground (σ_{11}) and excited (σ_{22}) states of the particle, and σ_{12} is the slow amplitude of the off-diagonal element of the density matrix, which couples these states. According to the results of[7] and[8], these quantities satisfy the following equations:

$$\dot\sigma_{12} + (T_{12}^{-1} - i\Omega_{12})\sigma_{12} = \begin{cases} i\hbar^{-1}p_{12}^{(0)}E_0(\sigma_{22}-\sigma_{11})e^{i\delta t}, & (6a)\\ i\hbar^{-2}r_{12}E_1E_2(\sigma_{22}-\sigma_{11})e^{i\delta t}, & (6b)\\ i\hbar^{-2}r_{12}E_1E_2^*(\sigma_{22}-\sigma_{11})e^{i\delta t}; & (6c)\end{cases}$$

$$\dot\sigma_{11} + W_{12}\sigma_{11} - W_{21}\sigma_{22} = \begin{cases} 2\hbar^{-1}\,\mathrm{Im}(\sigma_{12}p_{21}^{(0)}E_0^*e^{-i\delta t}), & (7a)\\ 2\hbar^{-2}\,\mathrm{Im}(\sigma_{12}r_{12}^*E_1^*E_2^*e^{-i\delta t}), & (7b)\\ 2\hbar^{-2}\,\mathrm{Im}(\sigma_{12}r_{12}^*E_1^*E_2e^{-i\delta t}) & (7c)\end{cases}$$

and the normalization condition $\sigma_{11} + \sigma_{22} = 1$. In the right-hand sides of (6) and (7) it is necessary to take respectively the first, second, or third line for the single-photon resonance, TPA, and SRS. T_{12} is the reciprocal line width of the transition 2—1 (we shall henceforth omit the indices of the transition for T_{12}, r_{12}, and p_{12}); W_{21} and W_{12} are the probabilities of relaxation of the populations between levels 1 and 2 in the absence of the field: $W_{21} = \sigma_{11}^0/\tau$, $W_{12} = \sigma_{22}^0/\tau$, where σ_{11}^0 and σ_{22}^0 are the populations of the levels in the absence of a field and τ is the lifetime of the excited state.

The high-frequency Stark effect enters in these equations in the form of the quantity Ω_{12}, an expression for which is given in[8] (formula (1)). Comparing this formula with (2) and (3), we can easily verify that

$$\Omega_{12} = \hbar^{-1}\sum_s(\varkappa_1^{(s)} - \varkappa_2^{(s)})|E_s|^2. \quad (8)$$

It is easy to show that relation (8) is satisfied not only in the cases of single- and two-photon resonances; an analogous relation holds also between the Stark shift of any level under the influence of a field of arbitrary frequency and the polarizability of matter in these states at the field frequency.

2. POLARIZABILITY OF MATTER AT THE RESONANT-FIELD FREQUENCY

Let us consider first the stationary case. Substituting the stationary solutions (6a) and (7a) in (1), we can represent the polarizability of one particle in the form

$$\chi = \chi_0 + \chi_1 = \frac{1}{2}[\varkappa_1 + \varkappa_2 - (\varkappa_2 - \varkappa_1)\eta_0] + \frac{|p|^2T}{\hbar}\eta_0\frac{mk\vartheta - (\Delta + mk)}{1 + (\Delta + mk)^2 + 2\vartheta m}. \quad (9)$$

The index 0, which denotes that all the quantities pertain to a field of frequency ω_0, will henceforth be omitted in this section. We also introduce the following notation: $\eta_0 = \sigma_{11}^0 - \sigma_{22}^0$; $m = |p|^2T^2|E|^2/\hbar^2$—dimensionless quantity proportional to the flux intensity, $k = \hbar(\kappa_2 - \kappa_1)/|p|^2T$—ratio of the difference of the linear polarizabilities in the excited and in the ground states through the change of the resonant part of the linear polarizability in the region of anomalous dispersion.

Obviously, when $m \to 0$ Eq. (9) describes the anomalous dispersion of the linear polarizability in the resonance region; at larger deviations $\delta \sim \omega$, with allowance for (2), Eq. (9) goes over into a formula describing the normal dispersion.

The field-dependent terms of (9) have a clear physical meaning. The term $mk\vartheta$ in the numerator describes the change of χ as a result of the redistribution of the populations of the levels 1 and 2 with unequal κ_1 and κ_2; the characteristic field for this process is $\tilde E$:

$$|\tilde E|^2 = \hbar/2\tau(\varkappa_2 - \varkappa_1), \quad \text{i.e.,}\ \tilde m = (k\vartheta)^{-1}. \quad (10)$$

At $E = \tilde E$, the null point of the function χ_1 is on the edge of the absorption line (with allowance for its Stark shift); thus, when $E > \tilde E$ the quantity χ_1 has the same sign in the entire absorption (or emission) band.

The product mk, which is contained in Δ, is due to the high-frequency Stark-effect shift of the center of the absorption line; this effect can be characterized by means of the field E_{St}:

$$|E_{st}|^2 = \hbar/T|\varkappa_2 - \varkappa_1|, \quad \text{i.e.,}\ m_{st} = k^{-1}. \quad (11)$$

Under the influence of this field, the center of the absorption line shifts by an amount equal to its half-width. Finally, $2m\vartheta$ in the denominator of χ_1 describes the polarizability saturation connected with the population saturation; the saturating field is

$$|E_{\text{sat}}|^2 = \hbar^2/4|p|^2\tau T, \text{ i.e., } m_{\text{sat}} = (2\vartheta)^{-1}. \quad (12)$$

We shall show below that the behavior of the polarizability depends significantly on the ratio of these two fields.

Let us discuss the properties of the expression obtained for the polarizability. We note first that the formula for χ_1 coincides with the well known expressions for the polarizability of a two-level system[1)] only for such states 1 and 2 for which $\kappa_1 = \kappa_2$ (k = 0). In this case the dependence of χ on the field is determined only by the effect of saturation of the populations of these states. If $2\tau = T$ ($\vartheta = 1$),[2)] the change of the numerator of χ_1, due to the difference between κ_2 and κ_1, is exactly cancelled by its change due to the Stark effect. Then χ_1 practically coincides with the polarizability of the two-level system, so long as $E \ll E_{St}$.

Let us proceed to discuss the general case when $k \neq 0$ and $\vartheta \neq 1$. The dependence of χ_1 on Δ is shown in Fig. 1 for several values of m at $\eta_0 > 0$ and $k > 0$. The corresponding curves for $\eta_0 < 0$, $k < 0$; $\eta_0 > 0$, $k < 0$; $\eta_0 < 0$, $k > 0$ can be obtained from the plots of Fig. 1 by recognizing that

$$\chi_1(\eta_0, -k, -\Delta) = -\chi_1(\eta_0, k, \Delta), \qquad \chi_1(-\eta_0, k, \Delta) = -\chi_1(\eta_0, k, \Delta) \quad (13)$$

at the same value of m. The positions of the extremal points on the curves of Fig. 1 are determined by the formula

$$\Delta_{1,2} = mk(\vartheta - 1) \mp \sqrt{1 + 2m\vartheta + (mk\vartheta)^2}, \quad (14)$$

and χ_1 at these points is equal to

$$\chi_1(\Delta_{1,2}) = \pm |p|^2 T \eta_0 [2\hbar(\sqrt{1 + 2m\vartheta + (mk\vartheta)^2} \mp mk\vartheta)]^{-1}. \quad (15)$$

We can easily see that when $k > \vartheta/(\vartheta - 1)$ and the field increases, Δ_1 first increases, and then, reaching a value $\Delta_1 = -(k\vartheta)^{-1}[\vartheta - 1 + \sqrt{(k^2 - 1)(2\vartheta - 1)}]$, begins to decrease and approaches as before the center of the line[3)], which shifts under the influence of the high-frequency Stark effect. For strong fields ($mk^2\vartheta \gg 1$)[4)] we have $\Delta_1 \approx -(mk + k^{-1})$ and the total polarizability at this detuning is $\chi(\Delta_1) = (\kappa_1 + \kappa_2)/2$. We note that this is also the value of the polarizability of a medium when the populations of states 1 and 2 become equalized not as a result of the resonant field, but in some other manner. This obvious result cannot be obtained if we confine ourselves, in the description of the nonlinear resonant polarizability, to the two-level approximation; in fact,

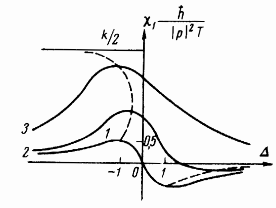

FIG. 1. Plot of χ_1 against the detuning Δ (k = 5, ϑ = 10): 1—m = 0, 2—m = 0.037; 3—m ≐ 0.2. The dashed lines show curves along which the maximum and minimum of the function $\chi_1(\Delta)$ move with increasing field.

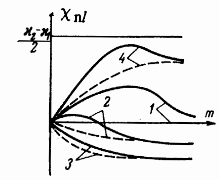

FIG. 2. Dependence of the nonlinear part of the polarizability on the intensity of the field in single-photon resonance (see (16)), 1—Δ = 0; 2—$\Delta < 0$; $D = (\vartheta + 1)\Delta^2 + 2\vartheta\Delta/k + \vartheta - 1 > 0$; 3—$\Delta < 0$; $D < 0$; 4—$\Delta > 0$. The dashed lines show plots of $\chi_{nl}(m)$ in the two-level model for the same values of Δ.

in this approximation $\lim_{m\to\infty} \chi_1 = 0$ and $\lim_{m\to\infty} \chi = \chi_0 = \kappa_2\sigma_{22}^0 + \kappa_1\sigma_{11}^0 \approx \kappa_1$ (see (9)).

If $k < \vartheta/(\vartheta - 1)$, then Δ_1 decreases monotonically with increasing field; the asymptotic behavior of Δ_1 in strong fields and of the limiting value of $\chi(\Delta_1)$ are then the same as in the preceding case.

The second extremum at all $k > 0$ moves farther away from the line center with increasing field; when $mk^2\vartheta \gg 1$ we have $\Delta_2 \approx mk(2\vartheta - 1) + k^{-1}$ and $\lim_{m\to\infty} \chi(\Delta_2) = \chi_0$. The description of the behavior of the extrema of $\chi(\Delta)$ in the case $k < 0$ can be easily obtained from the picture presented above for $k > 0$, using relations (13).

For certain applications, particularly in the analysis of self-action of electromagnetic waves, a decisive role is played by the nonlinear part of the total polarizability, i.e., that part which depends on the field and vanishes in its absence:

$$\chi_{nl} = \frac{|p|^2 T}{\hbar}\eta_0 m \frac{k[\vartheta(1+\Delta^2) - (1-\Delta^2)] + 2\vartheta\Delta + \Delta k^2 m}{(1+\Delta^2)[1 + (\Delta + km)^2 + 2\vartheta m]}. \quad (16)$$

This part of the polarizability, as a function of m for positive k and η_0, is shown qualitatively in Fig. 2 at three characteristic values of the detuning Δ. Plots for the cases when k or η_0 or k and η_0 are negative can be obtained from Fig. 2 with the aid of relations (13). If

$$k^2 > \vartheta^2/(\vartheta^2 - 1), \quad (17)$$

the curves of Fig. 2 have extrema for all values of the detuning. These extrema are due to the Stark effect. When the inverse inequality is satisfied, there is no extremum for detuning satisfying the condition

$$(\vartheta + 1)\Delta^2 + 2\vartheta\Delta/k + \vartheta - 1 < 0$$

(see curve 3 of Fig. 2).

If $\vartheta \gg 1$ and $m_{sat} < m_{St}$, then the crests of the curves of Fig. 2 for all $|\Delta| \lesssim 1$ become flat and the value of the extremum differs little from the limiting value ($m \to \infty$) of the nonlinear polarizability without allowance for the Stark effect ($\tilde{\chi}_{nl}$). For example, in the case under consideration, at $\Delta = 0$

$$\lim_{m\to\infty} \tilde{\chi}_{nl}/\chi_{nl}^{max} = (\vartheta + |k|)/(\vartheta - 1) \approx 1.$$

The difference in the behavior of $\chi_{nl}(m)$ from that of the nonlinear polarizability of a two-level system is then due to the presence in the numerator of (16) of the

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1) See, for example, [5], where the polarizability of a two-level system was used in the discussion of the possibility of self-focusing of radiation within the working medium of a laser.

2) Such a relation between the lifetime and the reciprocal width of a homogeneous absorption line is possible, for example, in rarefied gases.

3) Incidentally, Δ_1 is never exactly at the line center, although it is sufficiently close to it when $k \gg 1$.

4) Here, as before, the results are valid for fields whose energy is smaller than the ionization energy ($m < m_{ioniz}$). Therefore, for example, the behavior of the nonlinear polarizability, described here by the asymptotic formulas, can be realized only if $k^2\vartheta m_{ioniz} \gg 1$.

term $k\vartheta(1+\Delta^2)$, as a result of which, in particular, $\lim_{m\to\infty}\tilde{\chi}_{nl}$ is larger by a factor $(\kappa_2-\kappa_1)\eta_0/2$ than the corresponding value in the two-level approximation, which equals $|p|^2 T\eta_0\Delta/\hbar(1+\Delta^2)$.

Thus, when $k \neq 0$ and $\vartheta \neq 1$, the dependence of the polarizability of the medium on the frequency and intensity of the field can differ significantly from the behavior of the polarizability of the two-level system. Accordingly, the self-action of electromagnetic waves in such media should have a different character compared with self-action in a medium consisting of two-level particles. Thus, in an absorbing medium $k > 0$, and in an amplifying medium at $k < 0$, self-focusing of the radiation of any frequency lying within the limits of the line width of the working transition is possible if (17) is satisfied. In fact, the necessary condition for self-focusing is $\partial\chi_{nl}(\Delta)/\partial m|_{m=1} > 0$; it is seen from (16) that in the indicated cases this condition is satisfied independently of the value of Δ. In the two-level approximation on the other hand, only waves with $\Delta > 0$ are self-focused in an absorbing medium and with $\Delta < 0$ in an amplifying medium[5].

Let us discuss now the possible existence of conditions under which (17) is satisfied; in solutions and liquids, the characteristic values are $T \sim 10^{-12}-10^{-13}$ sec and $\tau \sim 10^{-7}-10^{-9}$ sec; in laser working media $T \sim 10^{-11}$ sec and $\tau \sim 10^{-3}-10^{-4}$ sec. In both cases $\vartheta \gg 1$ and (17) reduces to the condition $k > 1$, which is satisfied if $(\kappa_2-\kappa_1) > (10^{16}-10^{14})\,p^2$. In ruby, for example, $|p| \approx 3\times10^{-21}$ cgs esu and the difference of the polarizability should be larger than 10^{-25} cm^3. It should be noted that by virtue of (8) the quantity $(\kappa_2-\kappa_1)$ can be estimated from the constant of the quadratic Stark shift of the line of the corresponding transition. Unfortunately, we were unable to find in the literature any data on the shifts of weakly allowed transitions; on the other hand, measurements of the shifts of allowed transitions, for example of the transition 2S—2P in lithium[9], yields $\kappa_2-\kappa_1 \approx 4\times10^{-21}$ cm^3. The same value is obtained from data on the shifts of the lines of argon with λ = 4475 Å and λ = 4538 Å[10]. It is obvious that weakly allowed transitions can have $\kappa_2-\kappa_1$ of the same order; then $k \approx 10^4 \gg 1$. Of course, the value of $\kappa_2-\kappa_1$ can be also much smaller; however, even if we assume that $\kappa_2-\kappa_1 \approx 0.1\kappa_1$, then, starting from the data on the refractive index ($\kappa_1 \sim 10^{-23}-10^{-24}$ cm^3), we obtain $k \geq 1$.

In concluding this section let us estimate the value of the nonlinear addition to the dielectric constant $\epsilon_{nl} = 4\pi N\chi_{nl}$, which can be expected in the case of resonant interaction between the electromagnetic field and the medium. In accordance with the reasoning presented above, we can have, for example in laser media ($N \approx 10^{19}$ cm^{-3}) in the case of a saturating field, $\epsilon_{nl} \gtrsim 10^{-6}$ (at $\kappa_2-\kappa_1 = 10^{-25}$ cm^3). In the case of substances with a short lifetime in the excited state ($\tau \approx 10^{-8}$ sec, $T \approx 10^{-12}$ sec, $N \approx 10^{22}$ cm^{-3}, $|p| \sim 10^{-20}-10^{-21}$ cgs esu) the saturating fields are difficult to produce (they correspond to fluxes of several GW/cm^2), and at $E \ll E_{sat}$ we have $\epsilon_{nl} \approx 2(10^{-10}-10^{-12})\,|E|^2$, i.e., the coefficient of $|E|^2$ can be larger than or equal to the corresponding coefficient connected with the Kerr effect. Thus, ϵ_{nl} is quite readily measurable, say by interference methods; we note that such a measurement would make it possible to determine from the value of ϵ_{nl} the Stark-shift constant (see (8)) for even weakly resolved and strongly broadened transitions.

We now proceed to the polarizability in two-photon absorption and stimulated Raman scattering.

3. POLARIZABILITY IN TPA AND SRS

From (1), (6), and (7) we can obtain the stationary polarizability of one particle at a frequency of any of the fields participating in the TPA or SRS

$$\chi(\omega_i) = \chi_0^{(i)} + \chi_{nl}^{(i)} = \frac{1}{2}[\varkappa_1^{(i)} + \varkappa_2^{(i)} - (\varkappa_2^{(i)} - \varkappa_1^{(i)})\eta_0] + \frac{|r|}{\hbar}\eta_0 m_j \frac{m_i q_i\vartheta - (\Delta + m_i q_i + m_j q_j)}{1 + (\Delta + m_i q_i + m_j q_j)^2 + 2\vartheta m_i m_j}, \tag{18}$$

where i = 1, 2; j = 2, 1, and we have introduced the dimensionless quantities:

$$m_i = \hbar^{-2}T|r|\,|E_i|^2,\quad q_i = \hbar|r|^{-1}(\varkappa_2^{(i)} - \varkappa_1^{(i)}),\quad \vartheta = 2\tau/T,\qquad \Delta = \delta T. \tag{19}$$

The value of r for TPA and SRS is determined from (5), and the polarizability $\kappa_{1,2}^{(i)}$ from (3).

It is of interest to consider the χ_{nl} connected with the TPA in the case of degenerate frequencies $\omega_1 = \omega_2 = \omega$. In experiments on TPA one encounters most frequently precisely this situation. It can be shown that in this case the nonlinear part of the polarizability is given by

$$\chi_{nl} = \frac{|r|}{\hbar}\eta_0 m \frac{mq\vartheta - (\Delta + mq)}{1 + (\Delta + mq)^2 + 2\vartheta m^2}. \tag{20}$$

The $\chi_{nl}(m)$ dependence is shown in Fig. 3 for several values of the detuning and for $q > 0$ and $\eta_0 > 0$. The case of negative q or η_0 can be obtained with the aid of the relations

$$\chi_{nl}(\eta_0, -q, -\Delta) = -\chi_{nl}(\eta_0, q, \Delta),\quad \chi_{nl}(-\eta_0, q, \Delta) = -\chi_{nl}(\eta_0, q, \Delta).$$

It is seen from Fig. 3 that

$$\lim_{m\to\infty}\chi_{nl}(m,\Delta) = \frac{q|r|(\vartheta-1)}{\hbar(2\vartheta+q^2)}\eta_0$$

is independent of the detuning, in contrast to the polarizability in single-photon resonance. Just as in Sec. 2, it is convenient to introduce the characteristic fields with the aid of the relations $\tilde{m} = (q\vartheta)^{-1}$, $m_{sat} = (2\vartheta)^{-1/2}$, $m_{St} = q^{-1}$. If $\vartheta \gg 1$ and $m_{St} \gg m_{sat}$, i.e.,

$$\hbar T^{1/2}|\varkappa_2 - \varkappa_1| \gg 2|r|\tau^{1/2},$$

then the limiting value of χ_{nl} is close to $(\kappa_2-\kappa_1)\eta_0/2$, just as in the case of single-photon resonance. If an inequality inverse to (21) is satisfied, then the quantity

$$\lim_{m\to\infty}\chi_{nl} = \frac{|r|^2(2\tau - T)\eta_0}{\hbar^2(\varkappa_2-\varkappa_1)T}$$

is always smaller than $(\kappa_2-\kappa_1)\eta_0/2$. The curves of Fig. 3 corresponding to $\Delta > 0$ have minima, and those corresponding to $\Delta < 0$ have maxima. The values of the latter likewise do not exceed $(\kappa_2-\kappa_1)\eta_0/2$. The positions of the extrema are determined by the positive roots of the equation

$$m^2\Delta[2\vartheta(1+q^2) - q^2] + 2mq(\vartheta-1)(1+\Delta^2) - \Delta(1+\Delta^2) = 0.$$

It is easy to show that if m_{sat} and M_{St} are essentially

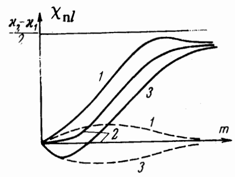

FIG. 3. Dependence of the nonlinear part of the polarizability of the intensity of the field in two-photon absorption (see (20)) at $q > 0$ and $1-\Delta = \tilde{\Delta} < 0$; $2-\Delta = 0$, $3-\Delta = -\tilde{\Delta} > 0$. The dashed lines represent plots of $\chi_{nl}(m)$ for $\kappa_1 = \kappa_2 (q = 0)$ at the same values of the detuning.

different, then χ_{nl}^{max} is reached at $m \sim \min\{m_{sat}, m_{St}\}$. When $m \ll \min\{m_{sat}, m_{St}\}$, then we can use for the nonlinear polarizability (20) the approximate expression

$$\chi_{nl} = \frac{|r|\eta_0}{\hbar(1+\Delta^2)} m\left[-\Delta + mq\left(\vartheta - \frac{1-\Delta^2}{1+\Delta^2}\right)\right]. \quad (20a)$$

The first term of (20a) coincides with the customarily employed expression for the polarizability in two-photon processes (for example,[11]). Obviously, the remaining term may in the general case not be smaller than the first. For example, when $m > \tilde{m}$ and $\vartheta > 2$ the contribution of the second term is larger than that of the first in the entire band $|\delta| < T^{-1}$. When $\vartheta \gg 1$, the influence of the Stark shift on the polarizability is small, and it is necessary to retain from among the terms that depend on the field in (20a) only the term $mq\vartheta$. Thus, in the range $\tilde{m} \ll m \ll m_{sat}$ it is possible in this case to use the following expression for the polarizability:

$$\chi_{nl} = \frac{|r|q\vartheta\eta_0}{\hbar(1+\Delta^2)} m^2 = 2\hbar^{-4}|r|^2\tau T(\varkappa_2 - \varkappa_1)|E|^4. \quad (20b)$$

The region of applicability of (20b) can be sufficiently broad, for in many cases

$$\frac{m_{sat}}{\tilde{m}} = \frac{\hbar(\varkappa_2 - \varkappa_1)}{|r|}\left(\frac{\tau}{T}\right)^{1/2} \gg 1.$$

For example, in TPA in vapor and in gas ($\tau \approx 10^{-7}$ sec, $T \approx 10^{-9}$ sec, $r = 10^{-51}$ cgs esu) at $\kappa_2 - \kappa_1 \sim 10^{-24}-10^{-21}$ cm^3 we have $m_{sat}/\tilde{m} \approx 10-10^4$; the value of m in these cases corresponds to a flux $5 \times 10^3-5$ kW/cm^2.

Let us estimate the value of χ_{nl} from (20b). At a pressure of 1 atm ($N \approx 10^{19}$ cm^{-3}) in gases, at the given values of $\kappa_2 - \kappa_1$, the change of the dielectric constant is $\epsilon_{nl} = 4\pi N\chi_{nl} \approx 10^{-6}-10^{-3}$ in a flux of 5 MW/cm^2. In condensed media, the TPA on the electronic transitions ($\tau \approx 10^{-9}$ sec, $T \sim 10^{-11}$, $N = 10^{22}$ cm^{-3}) can lead to $\epsilon_{nl} \approx 10^{-6}$ in a flux of 50 MW/cm^2 even if $(\kappa_2 - \kappa_1) \approx 0.1\kappa_1 \approx 10^{-25}$ cm^3. We note that the formulas obtained in this case are suitable for the description of the polarizability under the influence of radiation pulses from Q-switched lasers ($t_{pulse} \approx 10^{-8}$ sec). Thus, the ϵ_{nl} due to the TPA is comparable with the nonlinear dielectric constant connected with the Kerr effect in liquids. We can therefore expect the presence of TPA to have a strong influence on the interaction of light both in gases and in condensed media.

Let us discuss now the nonlinear polarizability connected with SRS. For fields that are small compared with saturating fields ($m_1 m_2 \ll 1/2\vartheta$) and with Stark fields, we easily obtain from (18)

$$\chi_{nl}(\omega_i) = \frac{|r|}{\hbar}\eta_0 \frac{m_j}{1+\Delta^2}\left[-\Delta + m_i q_i\left(\vartheta - \frac{1-\Delta^2}{1+\Delta^2}\right) - m_j q_j \frac{1-\Delta^2}{1+\Delta^2}\right]. \quad (18a)$$

The generation of the Stokes component occurs most effectively near the line center (with allowance for its Stark shift), i.e., $\Delta_{optim} = -\Sigma m_i q_i$. When $m_i \ll (m_i)_{St}$, we have $|\Delta_{optim}| \ll 1$. By substituting Δ_{optim} in (18a), we can easily verify that in the case when $\vartheta \gg 1$ the term that determines χ_{nl} is the one proportional to the product $m_1 m_2$, and

$$\chi_{nl}(\omega_i) = \frac{|r|}{\hbar}\eta_0\vartheta q_i m_i m_j. \quad (18b)$$

The threshold and certain features of self-focusing due to the polarizability (18b) were considered by us in[6]. When $\vartheta \approx 1$ the contribution made to χ_{nl} by terms connected with the Stark effect is significant and it is necessary to use expression (18a).

Although formulas (18)—(18b) describe the stationary polarizability, they can be used to calculate χ_{nl} in the case of SRS on vibrational levels in liquids for nanosecond pulses, and in a number of cases also picosecond pulses, if $t_{pulse} \gg \tau$ ($\tau \sim 10^{-10}-10^{-12}$ sec in liquids). χ_{nl} can assume appreciable values in thin filaments produced during self-focusing (power flux ~ 2 GW/cm^2 [12]). Assuming $\kappa_2 - \kappa_1 \sim 10^{-25}$ cm^3, $\tau \sim 10^{-10}$ sec, $T \sim 10^{-11}$ sec, and $m_2/m_1 \sim 0.5$ we obtain $\epsilon_{nl} \approx 5 \times 10^{-5}$. In the case of SRS of a picosecond pulse (power flux $\sim 10^3$ GW/cm^2, $t_{pulse} \approx 3 \times 10^{-12}$ sec[13]) we have $\epsilon_{nl} \approx 5 \times 10^{-4}$ even for very broad lines ($T \approx 10^{-13}$ sec) with a short time $\tau \approx 10^{-12}$ sec.

Thus, SRS can exert a noticeable influence on the self-action of light[6]. We note that mechanisms for the change of polarizability under the action of fields, which have been considered here, were not taken into account in[14], where the influence of TPA and SRS on self-focusing of light was investigated.

4. POLARIZABILITY OF MATTER UNDER THE INFLUENCE OF SHORT LIGHT PULSES

As already mentioned in the introduction, observation of self-focusing of nanosecond[15] and picosecond[13] pulses in air was reported in the literature recently. Apparently, this self-focusing can be connected with only one of the phenomena considered here, since the Kerr effect and striction in air make a negligibly small contribution to the nonlinear polarizability ($\chi_{nl}^{Kerr} \approx (10^{-15}-10^{-16})|E|^2$; $\epsilon_{nl}^{strict} \approx 5 \times 10^{-15}|E|^2$).

The relaxation times in gases at atmospheric pressure are $\tau \sim 10^{-3}$ sec for vibrational transitions[16] and $\tau \sim 10^{-7}-10^{-8}$ for electronic transition, the line width being of the same order in both cases ($T \sim 10^{-10}$ sec). It is therefore of interest to investigate the nonlinear polarizability in single- and two-photon absorption and SRS in the nonstationary case, i.e., under the influence of pulses whose duration[5)] does not exceed either of the relaxation times τ and T.

1) Let $T \ll t_{pulse} \ll \tau$. Then we can assume σ_{12} in (6) to be stationary; substituting σ_{12} from (6) in the corresponding equation (7), we can obtain the population-balance equation. Integrating the latter under the initial condition $\eta|_{t=0} = (\sigma_{11} - \sigma_{22})_{t=0} = \eta_0$ and substituting the solution in (1), we can easily verify that, for example in the case of single-photon processes, $\chi(t_{pulse})$ differs

5)The pulse is assumed to be rectangular.

from the stationary value (9) of the polarizability by the amount

$$-\frac{|p|^2 T}{\hbar}\eta_0 \frac{mk\vartheta + 2m\vartheta(\Delta + mk)/[1+(\Delta+mk)^2]}{1+(\Delta+mk)^2+2\vartheta m} \times \exp\left\{-\frac{t_{\text{pulse}}}{\tau}\left[1+\frac{2m\vartheta}{1+(\Delta+mk)^2}\right]\right\}; \quad (22)$$

We have used here the notation of Sec. 2. An analogous expression is obtained from (22) for TPA by substituting in all the quantities of (22)

$$|p|^2 \to (|r|\ |E|\hbar^{-1})^2. \quad (23)$$

2) Let now

$$t_{\text{pulse}} \ll T, \tau, \left(\delta + \frac{\varkappa_2 - \varkappa_1}{\hbar}|E|^2\right)^{-1}. \quad (24)$$

The last of these conditions denotes that we are considering a situation where the frequency of the field is close to the line shifted as a result of the Stark effect. Equations (6) and (7) will be solved by an iteration method. In the first approximation $\eta = \eta_0$. Substituting this value in (6), substituting the solution of this equation in (7), and then using (1) we obtain, taking (24) into account,

$$\chi = \chi_0 + \frac{|p|^2}{2\hbar}\eta_0 t_{\text{pulse}}^2\left[\frac{\varkappa_2 - \varkappa_1}{\hbar}|E|^2 - \delta\right] \quad (25)$$

for single-photon resonance. The condition for the applicability of the iteration method $|\eta - \eta_0| \ll \eta_0$ reduces in this case to

$$|p|^2|E|^2\hbar^{-2}t_{\text{pulse}}^2 \ll 1. \quad (26)$$

The polarizability in the case of TPA (when $\omega_1 = \omega_2$) is obtained from (25) by making the substitution (23); for SRS and TPA with essentially different ω_1 and ω_2, the polarizability is equal to

$$\chi(\omega_i) = \chi_0(\omega_i) + \frac{|r|^2|E_j|^2}{2\hbar^3}\eta_0 t_{\text{pulse}}^2\left[\frac{\varkappa_2^{(i)} - \varkappa_1^{(i)}}{\hbar}|E_i|^2 - \frac{\varkappa_2^{(j)} - \varkappa_1^{(j)}}{\hbar}|E_j|^2 - \delta\right]. \quad (27)$$

The condition for the applicability of the iteration method is given in this case by

$$|r|^2|E_1|^2|E_2|^2\hbar^{-4}t_{\text{pulse}}^2 \ll 1.$$

We call attention to the following circumstance. The field-dependent terms in (25) and (27) are due both to the change of the polarizability as a result of the redistribution of the populations among the levels with $\kappa_1 \neq \kappa_2$ and to the high-frequency Stark effects. It turns out here that in (25) the contribution of the Stark effect is exactly half as large as the contribution connected with the redistribution of the populations, and has the opposite sign; the ratio of the contributions due to these causes is the same in the terms proportional to $|E_i|^2$ in (27). Such simple relations are obtained because of the presence of the connection (8) between the difference $\kappa_2 - \kappa_1$ and the Stark-shift constants.

With increasing pulse duration or with increasing deviation from the line center (with allowance for the line shift under the influence of the fields), the polarizability begins to oscillate with a frequency $(\delta + (\kappa_2 - \kappa_1)|E|^2/\hbar$.

In concluding this section, let us estimate the value that ϵ_{nl} assumes at the end of a pulse with $t_{\text{pulse}} = 10^{-11}$ sec and a power flux 100 GW/cm^2 in single-photon resonance with a weakly resolved transition ($|p| \approx 10^{-21}$ cgs esu) in a gas under a pressure on the order of 1 atm ($N = 10^{19}$ cm^{-3}, $\tau \geq 10^{-9}$ sec, $T = 10^{-10}$ sec). In this case, as can be easily verified, the first two conditions from (24) and (26) are satisfied. The third condition of (24) is satisfied for $\delta = 0$ only if $\kappa_2 - \kappa_1 \lesssim 10^{-25}$ cm^3. In this case ϵ_{nl} is described by expression (25). Its value is 1.2×10^{-7}. If $\kappa_2 - \kappa_1$ is larger than 10^{-25} cm^3, it is necessary, in order to satisfy (24), to choose $\delta = -(\kappa_2 - \kappa_1)|E|^2/\hbar$. Then ϵ_{nl} can reach a value 2.4×10^{-3} (if $\kappa_2 - \kappa_1 = 10^{-21}$ cm^3). This means that even for ultrashort pulses self-focusing due to resonant mechanisms is perfectly feasible.

CONCLUSION

Let us formulate the main conclusions of our investigation.

1. The nonlinear polarizability of matter (χ_{nl}) accompanying the resonant interaction of the matter with the field, such as single-photon absorption and emission, TPA, and SRS, is formed under the influence of three factors: (a) redistribution of the populations of the excited and ground states with unequal κ_2 and κ_1 under the influence of the fields (see (2) and (3)); (b) saturation due to the fact that the pouplation difference tends to zero when the field energy increases; (c) the Stark shift of the anomalous-dispersion curve together with the center of the absorption (emission) line.

Both causes (a) and (b) are connected with changes of the population, which depend also on the value of the level Stark shift. The latter in turn is directly connected, as shown here (see (8)), with the difference $(\kappa_2 - \kappa_1)$. Therefore there are no situations (with the exception of the case $\kappa_2 = \kappa_1$) in which the indicated causes would appear separately. The expressions for the nonlinear polarizabilities with allowance for the simultaneous action of these causes can be obtained with the aid of the density-matrix formalism. χ_{nl} is described by formula (9) for single-photon absorption and emission, by (18) and (20) for TPA, and by (18) for SRS; these expressions are valid up to fields exceeding the saturating and Stark values.

2. Allowance for the difference between κ_1 and κ_2 means, in essence, allowance for the influence exerted on χ_{nl} by all the remaining levels of the system; this influence inevitably drops out from consideration if one confines himself to the two-level model. As shown in Sec. 2, this model is suitable for the calculation of χ_{nl} only in the case $\kappa_1 = \kappa_2$; the difference between χ_{nl} and the polarizability of a two-level system can also be negligibly small up to Stark fields, if $2\tau = T$.

3. With decreasing pulse duration, the relative role of the Stark effect in the formation of χ_{nl} increases, since it is not subject to time delay whereas a definite time is required to change the populations (this time depends on the magnitudes of the fields interacting with the medium). However, the Stark shift takes part in the formation of χ_{nl} via the off-diagonal density-matrix element, which also is subject to time delay. In this connection, for very short pulses the contribution of the Stark effect does not exceed half the contribution connected with the redistribution of the populations.

4. With the aid of measurements of χ_{nl} it is possible to determine the polarizability of the excited state, and

also, by virtue of the relation (8), the constant of the high-frequency Stark shift of the working transition. For this measurement, it suffices to have a field $|E|^2 \sim \hbar\tau^{-1}|\kappa_2 - \kappa_1|^{-1}$, which is usually much weaker than the field necessary for the measurement of the Stark shift by the known method[9]. The value of χ_{nl} reached in these processes is sufficient to be able to measure it, for example, by interference methods. We note that $(\kappa_2 - \kappa_1)$, and consequently the constant of the Stark shift, can also be determined by measuring (using a nonresonant test field) the increment of ϵ of the medium due to the resonant processes; in this case χ for the test field is χ_0 (see (9)), where η_0 should be replaced by the value of η produced as a result of resonant absorption, TPA, or SRS, respectively[6)].

5. Estimates of the value of χ_{nl} connected with the effects considered here allow us to state that the resonant mechanisms of formation of χ_{nl} can greatly influence the character of self-action of light (even in the presence of the Kerr effect and striction). In a number of cases these mechanisms are the main cause of self-focusing. By way of an example, we can point to the self-action of picosecond pulses in gases, since in gases the χ_{nl} connected with the Kerr effect and with the striction is small, while thermal self-focusing cannot develop within the duration of such a pulse.

In such cases, the self-action of the light should have a number of distinctive features, connected with the fact that the formation of χ_{nl} is accompanied by a decrease (increase) of the total beam energy, and in the case of SRS it is accompanied by transfer of energy from one frequency to another. An investigation of the self-action processes produced by resonant mechanisms, with allowance for the changes in the energy of the waves taking part in the process, will be reported by us in a future paper.

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6)Measurement of the population difference η entails no difficulty (see, for example, [17]).

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