INVERSIONLESS AMPLIFICATION AND LASER-INDUCED TRANSPARENCY AT THE DISCRETE TRANSITIONS AND THE TRANSITIONS TO CONTINUUM

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The effects of coherence of quantum transitions and the interference of resonant nonlinear optical processes on the spectra of absorption, amplification, and nonlinear-optical generation are considered. The most favorable conditions are discussed for the inversionless amplification, resonant refraction in the absence of absorption and for resonant enhancement of nonlinear-optical generation at the discrete transitions and the transitions to continuum.

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1 Introduction

Coherency and interference are the basic physical phenomena, which lead to new effects in quantum optics. Interference can be destructive or, vice versa, constructive, causing mutual suppression or amplification of simultaneous processes. In quantum optics the interference effects can reduce interaction of radiation with absorbing atoms at lower energy levels without substantial variations in the interaction with emitting atoms at overlying levels. In turn, this leads to a difference in absorption and emission spectra. The intratomic coherence conditions many fundamental effects in high-resolution nonlinear spectroscopy, the light amplification without population inversion and resonant refraction increase in the absence of absorption, coherent population trapping, increase of the resonant nonlinear-optical radiation at a simultaneous primary radiation absorption decrease, and the laser induction of structures of the autoionization type in spectral continua. The resonant nonlinear interference effects, theoretically and experimentally studied since the time of first masers and lasers creation [1-6], currently again attracted a great attention [7-10]. They are promising for new laser sources in the VUV and X-ray bands, laser accelerators of atomic particles, microscopes with increased resolution, supersensitive magnetometers, etc. (see, e.g. [11]. A lot of scientific meetings are dedicated to this problem (see, e.g. [9, 10]).

A publication flux are devoted to the related effect of electromagnetically induced transparency as applied to improve characteristics of laser light conversion to short-wave bands (see, e.g. [7]). (It appears that in many respects this is the phenomenon comprehensively studied in Russia in 1960-70 s [12-36, 40]). A quite complete survey of results acquired by western authors can be found in [7-10] and references therein. Therefore, below a basic attention would be given to certain less-known results of Russian authors.

A basic contribution into studies of the nonlinear interference processes in absorption (emission) spectra at the interaction of atom-molecular systems with electromagnetic radiation and the effect of dynamic splitting of spectral lines in strong electromagnetic fields was made by the Yerevan, Moscow,
Nizhny Novgorod, Novosibirsk, St. Petersburg, and Minsk schools. These results are generalized in monographs [4, 15-18, 22, 23, 30, 33, 36]. In [25, 28, 29, 30] the possible inversionless amplification was analyzed for three-level systems at the discrete optical transitions. Corresponding experimental studies were carried out in [21, 33, 34]. (For two-level optical systems this effect was predicted in [5] and experimentally studied in the radio band in [6, 13, 14]). The effect of self-transparency induced by a strong field at an adjacent transition was theoretically and experimentally studied in detail, e.g. in [12, 19, 20]. The coherent population trapping was first observed in [35]. Later the study in nonlinear interference phenomena at the discrete transitions was extended to the transitions to a continuous spectrum [31, 32, 36], autoionization-type resonances were predicted in [31] and experimentally revealed [32] at the atomic transitions to continuum. (Later this effect was called as the laser-induced continuum structure (LICS)). They also predicted the possible nonlinear response increase at the laser short-wave generation with a simultaneous decrease in its absorption and improvement in phase matching [31, 36]. In [37] it was shown that, similar to discrete transitions, the inversionless amplification is possible also at the transitions to autoionization and antoionization-type states. The cited papers initiated the coherence effect study, first at the transitions to continuum, and then at the discrete optical ones.

The coherence and interference phenomena are the basis for inversionless amplification, coherent population trapping, and electromagnetically induced transparency both at the discrete transitions and those to continuous spectrum. As it was already indicated, these effects offer unconventional solution for actual problems of quantum electronics. However, the peculiarities of optical transitions and real experimental designs can qualitatively change an expected manifestation of these processes. These problems remain to be a subject of great attention. Hence, it is required to develop theoretical approaches considering the most important accompanying processes and involving numerical analysis, if necessary. Now the least understood phenomena are the effects of nonuniform broadening and level degeneration, relaxation and motion of population on the coherence degradation. It appears that sometimes, vice versa, the relaxation promotes the intratomic coherence. A fairly small number of papers is dedicated to the effect of above processes on the resonant nonlinear-optical frequency mixing.

2 Resonant nonlinear-optical interference

2.1 Destructive and constructive interference in classical and quantum optics

The interference is one of fundamental physical phenomena. Oscillations of various nature depending on a phase relationship can interfere constructively or destructively. Varying oscillation phases and amplitudes, the resulting process can be amplified or suppressed. The quantum interference can proceed, when there is coherent superposition of real states. Moreover, the degenerate (in frequency) interfering intra-atomic oscillations can be conditioned by different correlating quantum transitions contributing into the same process. These are, e.g., one-and two-photon contributions into the optical process related to emission or absorption at a specified frequency. The process can result from the coherent superposition of a neighboring real energy level and a quasi-level (virtual state) created by a strong auxiliary field [26]. Such a superposition is realized even more simply than in the case of real doublet state.

The interference is more general concept, than notions of one-, two-, and multi-step and multi-photon processes. The latter were introduced and classified by their frequency-correlation properties in the framework of perturbation theory. However, these properties are significantly varied as the field intensity rises, in particular at resonant interaction [26]. As a result, the qualitative effects become possible in nonlinear spectroscopy of the Doppler-broadened transitions, such as the induced compensation of residual Doppler broadening in two-photon absorption or Raman scattering under conditions of a difference in photon frequencies [38, 39]. Even when many elementary processes contribute into the optical one at a given frequency and their classification into stepped and multi-
photon processes is difficult, experimental data can be often explained and predicted using the concept on interfering components of nonlinear polarization. Amplitudes and phases of these components are varied by controlling the corresponding field intensities and the detunings from one- and two-photon resonances.

2.2 Equation for the density matrix: the effects of intermediate level population and relaxation

In the general case of an open configuration of energy levels, when a lower level is not ground, various relaxation processes’ rates are different, and all the levels can be populated, the density matrix approach is most convenient to analyze resonant nonlinear-optical processes. Simple formulas for spectral properties of responses at a weak probing field frequency in the presence of strong one at an adjacent transition are uniformly deduced for $V$, $\Lambda$, and cascade schemes [22, 23, 27, 30, 36]. Let us show this by the example of transition diagram displayed in Fig. 1.

The scheme is assumed open, i.e. level $l$ is not ground one. For simplicity, we consider the fields $E_2$ and $E_3$ at frequencies $\omega_2$ and $\omega_4$, are probing, i.e., they do not disturb the level population. The fields $E_1$ and $E_3$ at frequencies $\omega_1 \approx \omega_{gl}$ and $\omega_3 \approx \omega_{mn}$, are strong. Below, in Sec. 3, we eliminate this limitation while considering interaction of two strong fields. Let us acquire the conditions of inversionless amplification at the transitions $gn$ and $ml$ so that to consider both the configurations $V$ and $\Lambda$. The probing field frequencies can be either higher or lower than those of strong fields. In the interaction representation the density matrix components and corresponding equations take the form

$$\rho_{1g} = r_1 \exp(i\Omega_1 t), \quad \rho_{nm} = r_3 \exp(i\Omega_3 t), \quad \rho_{ng} = r_2 \exp(i\Omega_1 t + \tilde{r}_2 \exp[i(\Omega_1 + \Omega_3 - \Omega_4)t]),$$

$$\rho_{lm} = r_4 \exp(i\Omega_4 t) + \tilde{r}_4 \exp[i(\Omega_1 - \Omega_2 + \Omega_3)t], \quad \rho_{ln} = r_{12} \exp[i(\Omega_1 - \Omega_2)t] + r_{34} \exp[i(\Omega_4 - \Omega_3)t], \quad \rho_{ii} = r_i,$$

$$P_2r_2 = iG_2\Delta r_2 - iG_3r_{32}^* + ir_2^*G_1, \quad d_2\tilde{r}_2 = -iG_3r_{41}^* + ir_4^*G_1, \quad P_4r_4 = i[G_4\Delta r_4 - G_1r_{41} + r_4G_3],$$

$$d_4\tilde{r}_4 = -iG_1r_{32}^* + ir_2G_3, \quad P_{14}r_{41} = -iG_1r_4^* + ir_4^*G_4, \quad P_34r_{43} = -iG_4r_4^* + ir_4^*G_3,$$

$$P_{32}r_{32} = -iG_2^*r_3 + ir_2^*G_3, \quad P_{12}r_{12} = -iG_1^*r_2 + ir_1^*G_2, \quad \Gamma_m r_m = -2Re\{iG_3^*r_3\} + q_m,$$

$$\Gamma_n r_n = -2Re\{iG_3^*r_3\} + \gamma_{gn} r_g + \gamma_{mn} r_m + q_n, \quad \Gamma_g r_g = -2Re\{iG_1^*r_1\} + q_g,$$

$$\Gamma \Gamma_l = -2Re\{iG_4^*r_4\} + \gamma_{g1} r_1 + \gamma_{ml} r_m + q_l,$$

where $\Delta r_1 = r_1 - r_g$, $\Delta r_2 = r_n - r_g$, $\Delta r_3 = r_n - r_m$, $\Delta r_4 = r_l - r_m$; $\Omega_1 = \omega_1 - \omega_{gl}$; $\Omega_2 = \omega_2 - \omega_{gn}$; $\Omega_4 = \omega_4 - \omega_{ml}$; $G_1 = -E_1d_{1g}/2h$, $G_2 = -E_2d_{gm}/2h$, $G_3 = -E_3d_{mn}/2h$, $G_4 = -E_4d_{ml}/2h$; $P_l = \Gamma_{lg} + i\Omega_1$, $P_2 = \Gamma_{ng} + i\Omega_2$, $P_3 = \Gamma_{nm} + i\Omega_3$, $P_4 = \Gamma_{lm} + i\Omega_4$, $P_{12} = \Gamma_{ln} + i(\Omega_1 - \Omega_2)$, $P_{34} = \Gamma_{mn} + i(\Omega_3 - \Omega_4)$, $P_{32} = \Gamma_{gm} + i(\Omega_3 - \Omega_2)$, $P_{14} = \Gamma_{gm} + i(\Omega_1 - \Omega_4)$, $d_2 = \Gamma_{ng} + i(\Omega_1 + \Omega_3 - \Omega_4)$, $d_4 = \Gamma_{lg} + i(\Omega_1 - \Omega_2 + \Omega_3)$. Here $\Omega_i$ are the frequency detunings from resonances, $G_i$ are the Rabi frequencies, $\Delta r_i$, are the population differences depending on intensity, $\Gamma_{ij}$ are the uniform halfwidths of transitions, $\Gamma_{ij}^{-1}$ are the lifetimes, $\gamma_{ij}$ is the rate of relaxation from the $i$th to $j$th levels, and $q_i$ are the rates of excitation by additional noncoherent pumping. The off-diagonal density matrix amplitudes $r_i$, define the coefficients of absorption (amplification) and refraction indices and $\tilde{r}_i$, define the nonlinear polarization of four-wave mixing. For the cascade configurations, the equations and their solutions are deduced by a simple substitution of detuning signs or by a complex conjugation of corresponding co-factors.
2.3 Laser-induced intra-atomic coherence and classification of resonant nonlinear-optical effects

Solution to the set of coupled equations for the density matrix components is given by

\[ r_{1,3} = iG_{1,3} \Delta r_1 / P_1, \quad r_{2,4} = iG_{2,4} R_{2,4} / P_{2,4}, \]

\[ R_2 = \frac{\Delta r_2 (1 + g + v_2) - v_3 (1 + v_7 - g_8) \Delta r_3 - g_3 (1 + g + v_8) \Delta r_1}{(1 + g + v_2) + [g_7 + g_2 (g_7 - g_8) + v_7 + v_2 (v_7 - g_8)]}, \]

\[ R_4 = \frac{\Delta r_4 (1 + v_5 + g_5) - g_1 (1 + g_5 - v_6) \Delta r_1 - v_1 (1 + v_5 - g_6) \Delta r_3}{(1 + g_4 + v_3) + [v_5 + v_4 (v_5 - g_6) + g_4 (g_5 - v_6)]}, \]

(1)

\[ \Delta r_1 = (1 + a_3) \Delta n_1 + b_1 a_3 n_3, \quad \Delta r_2 = \Delta n_2 - b_2 a_3 \Delta r_3 - a_2 a_1 \Delta r_1, \quad \Delta r_3 = (1 + a_1) \Delta n_3 + a_1 a_1 \Delta n_1, \]

\[ \Delta r_4 = \Delta n_4 - a_3 a_1 \Delta r_1 - b_3 a_3 \Delta r_3; \]

(2)

\[ r_m = n_m + (1 - b_2) a_3 \Delta r_3, \quad r_g = n_g + (1 - a_3) a_1 \Delta r_1, \quad r_n = n_n - b_2 a_3 \Delta r_3 + a_1 a_1 \Delta r_1, \]

\[ r_i = n_i - b_1 a_3 \Delta r_3 + a_3 a_1 \Delta r_1, \quad \Delta r_i (E_1 = 0, E_3 = 0) = \Delta n_i; \]

(3)

\[ g_1 = \frac{|G_1|^2}{P_{11} P_1}, \quad g_2 = \frac{|G_1|^2}{P_{12} P_2}, \quad g_3 = \frac{|G_1|^2}{P_{13} P_3}, \quad g_4 = \frac{|G_1|^2}{P_{12} P_3}, \quad g_5 = \frac{|G_1|^2}{P_{13} P_4}, \quad g_6 = \frac{|G_1|^2}{P_{14} P_4}, \quad g_7 = \frac{|G_1|^2}{P_{12} P_4}, \]

\[ v_1 = \frac{|G_3|^2}{P_{33} P_3}, \quad v_2 = \frac{|G_3|^2}{P_{32} P_2}, \quad v_3 = \frac{|G_3|^2}{P_{33} P_3}, \quad v_4 = \frac{|G_3|^2}{P_{32} P_3}, \quad v_5 = \frac{|G_3|^2}{P_{33} P_4}, \quad v_6 = \frac{|G_3|^2}{P_{32} P_4}, \]

\[ a_1 = \frac{\gamma_{gn} a_2}{\Gamma_{n} - \gamma_{mn}} = \frac{\gamma_{gn} \Gamma g}{\Gamma_{n} (\Gamma_{g} - \gamma_{gL})} = \frac{\gamma_{gn} \Gamma_{g}}{\Gamma_{n} (\Gamma_{g} - \gamma_{gL})}, \]

\[ b_1 = \frac{\gamma_{ml} b_2}{\Gamma_{m} - \gamma_{mn}} = \frac{\gamma_{ml} b_3}{\Gamma_{l} (\Gamma_{l} - \gamma_{ml})} = \frac{\gamma_{ml} \Gamma_{l}}{\Gamma_{l} (\Gamma_{l} - \gamma_{ml})}. \]

At \( G_3 = 0 \) equations (1) and (2) convert in solutions for \( \Lambda \) and \( V \) schemes

\[ r_2 = i \frac{G_2}{P_2} \cdot \frac{\Delta r_2 - g_3 \Delta r_1}{1 + g_2}, \quad r_4 = i \frac{G_4}{P_4} \cdot \frac{\Delta r_4 - g_1 \Delta r_1}{1 + g_4}. \]

(4)

According to [22, 23, 27, 30], it is convenient to classify the effects of the strong radiation resonant to an adjacent transition, as (i) population saturation (formulas (3)), (ii) dynamic splitting of the resonance for a probing field (or splitting of a common level, i.e., the ac Stark effect, denominators in formulas (4)), and (iii) nonlinear interference effects (NIEF) (the terms in the numerators of (4)). The two last effects are conditioned by quantum coherence.

3 Difference in pure emission and absorption spectra due to the nonlinear interference effects: inversionless amplification, resonantly amplified refraction in the absence of absorption, and laser-induced transparency

A light emitted of absorbed, e.g., at the frequency \( \omega_2 \) whose power is proportional to \( \text{Re}(iG_2 r_2) \), can be considered as a difference between pure emission (a term proportional to \( r_g \)) and pure absorption (other terms in formulas (1) and (4)). The two constituents are positive, but differently depend on detuning due to the NIE. Thus, a sign alternation arises in spectral line contour, resulting in the inversionless
amplification. This was emphasized in [24, 27] (see also [22, 23, 30, 36]). Optimum conditions for 
the inversionless amplification in a uniformly broadened three-level system were analyzed in [28-30] in 
detail. The refraction index at frequency \( \omega_2 \) is defined as \( Iml(-iG^2_nr_2) \) and, generally, the laser-induced 
minimum (including zero) absorption can coincide with the resonant refraction index maximum [11, 
40]. As is emphasized in [22-30, 36], the splitting effect and the NIE as a whole, varying the spectral 
line shape and causing the difference in pure emission (spontaneous or induced) and absorption spectra, 
does not vary its integral intensity, which is defined only by saturation effects,

\[
\int d\Omega_2 \text{Re}(-ir_2/G_2) = \Delta r_2, \quad \int d\Omega_4 \text{Re}(-ir_4/G_4) = \Delta r_4. \tag{5}
\]

Thus, namely NIE lead to the coherent population trapping, electromagnetically induced transparency 
and the inversionless amplification, e.g., at the transition \( gm \) (or \( ml \)), when the second terms 
in nominators of (4) become equal or begin to exceed \( \Delta r_2 \) (or \( \Delta r_4 \)). It is seen from the density matrix 
equation that the considered effects are finally defined by the coherence at transitions \( gm \) and \( ln \) (\( r_{32} \) 
and \( r_{12} \)), induced jointly by probing and strong fields.

### 3.1 Inversionless amplification of the probing wave

Now we enlarge on the problem, what are the elementary processes usually defined by the perturbation 
theory, which contribute into absorption (amplification) in the analyzed cases. For instance, let us 
consider the absorption index \( \alpha(\Omega_4) \) at frequency \( \omega_4 > \omega_1 \) (Fig. 1) and \( E_3 = 0 \) normalized to its 
maximum \( \alpha^0(0) \), in the absence of all strong fields. From formulas (4) we find

\[
\frac{\alpha(\Omega_4)}{\alpha^0(0)} = \text{Re} \left\{ \frac{\Gamma_4[\Delta r_4 - g_1 \Delta r_1]}{P_2 n_4(1 + g_4)} \right\}. \tag{6}
\]

Further we consider the two following cases. (i) Great yields from one-photon resonances

\[
|\Omega_4| \approx |\Omega_4| \gg \Gamma_1, \Gamma_4, \quad |g_4| \ll 1, |g_1| \ll 1, \quad P_4 \approx i\Omega_4, \quad P_1 \approx i\Omega_1 \approx i\Omega_4.
\]

Formula (6) takes on the form

\[
\frac{\alpha(\Omega_4)}{\alpha^0(0)} \approx \frac{\Gamma_4^2 \Delta r_4}{\Omega_4^2 \Delta n_4} - \text{Re} \left\{ \frac{\Gamma_4(\Delta r_4 g_4 + \Delta r_1 g_1)}{i\Omega_4^2 \Delta n_4} \right\} \approx \frac{\Gamma_4^2 \Delta r_4}{\Omega_4^2 \Delta n_4} - \frac{\Gamma_4 \Gamma_1}{\Omega_4^2 + (\Omega_4 - \Omega_1)^2} \frac{|G_1|^2(\Delta r_1 - \Delta r_4)}{\Omega_4^2 \Delta n_4}.
\]

\[
= \frac{\Gamma_{lm}(\Gamma_{lm})}{(n_l - n_m)\Omega_4^2} - \frac{\Gamma_{gm} \Gamma_{lm}}{\Omega_4^2 + (\Omega_4 - \Omega_1)^2} \frac{|G_1|^2(r_m - r_g)}{\Omega_4^2(n_l - n_m)}. \tag{7}
\]

The two last co-factors in (7) describe the Raman scattering and arise from the nominator (NIE) and 
denominator in (6). It ensues from (7) that the population inversion of initial and final unperturbed 
states (\( r_m = n_m > r_g \)) is required for the probing field amplification.

(ii) Resonance \( \Omega_1 = \Omega_4 = 0 \).

The amplification and transparency conditions have the form

\[
g_1 \Delta r_1 \geq \Delta r_4, \quad \frac{|G_1|^2}{\Gamma_{lg}\Gamma_{gm}} (r_l - r_g) \geq r_l - r_m. \tag{8}
\]

As it follows from (8), the amplification, due to NIE, does not require population inversion between 
initial and final states. The lower is the relaxation rate at a two-photon transition as compared to 
the coherence relaxation at coupled one-photon transitions, the more favorable are conditions for 
the inversionless amplification. An optimum strong field intensity is defined by the common level 
splitting into two quasilevels, which reduces the interference and, hence, the amplification at the \( ml \) 
transition center. The population difference saturation at the strong field transition also reduces the 
system coherence. There is an optimum relationship between the initial population differences \( \Delta n_4 \) 
and \( \Delta n_3 \), created by an additional noncoherent excitation. Optimum conditions for the inversionless 
amplification and transparency for opened and closed systems are analyzed in [28-30] in more detail.
3.2 Three-level system in strong fields: inversionless amplification for the strong wave

Above expressions can be easily generalized to the case of inversionless amplification of the strong fields which can drive a quantum system. This case is of interest in connection with creation of "laser without population inversion". For certainty, let us consider the interaction of two strong fields $E_3$ and $E_4$ (Fig. 1). Taking the strong field $E_4$ effects in density matrix equations into account, the set of equations can be reduced to an algebraic. The solution has the form

$$r_4 = i \frac{G_4 (1 + u_2^* \Delta r_4 - v_1 \Delta r_3)}{1 + v_4 + u_2}, \quad r_3 = i \frac{G_3 (1 + v_4^* \Delta r_2 - u_3 \Delta r_4)}{1 + v_4^* + u_2},$$

where

$$u_2 = |G_4|^2 / P_3 P_{43}^*, \quad u_3 = |G_4|^2 / P_4 P_{43}^*,$$

other notations are the same.

It is seen comparing (4) and (9) that, apart from the population difference saturation, a growth in the amplified wave intensity makes more difficult to achieve the conditions for inversionless amplification and self-transparency at the line center (factors $(1 + u_2^2)$ and $(1 + v_4^2)$ in the nominators), as well as reduces the gain due to an additional resonance splitting (the additional term in denominators).

An extended analysis of this problem, accounting for the saturated populations, will be published elsewhere.

3.3 Inversionless amplification and resonantly amplified refraction in the absence of absorption in sodium vapor: a simple experiment

Currently a small number of experiments contrasts to a flux of theoretical publications. The most experiments concern with coherent excitation of a doublet or a set of neighboring sublevels in the short-pulse mode, as well as with accompanying interference effects. In [41-43] there was proposed a design, comprehensive theoretical grounds, and estimations for a possible experiment on simultaneous observation of inversionless amplification and absorptionless resonant refraction. This was a scheme of interfering two-quantum transitions induced by an auxiliary field in the uniformly broadened three-level system with collisions. Such an experiment is of interest due to minimized accompanying processes. Meanwhile, this simplest model enters more complex experimental schemes.

Let us consider again the energy level diagram displayed in Fig. 1. We assume the level $n$ to be a ground one and the field $E_1$ and $E_4$ to be absent. Thus, we separate the V-shaped three-level configuration $g - n - m$. The strong field $E_3$ at frequency $\Omega_3$ couples the levels $m$ and $n$. The weak field at frequency $\omega_2$ probes the transition $gn$. Using (3) and (4) we derive the absorption $\alpha_2$ and refraction $n_2$ indices at frequency $\omega_2$, (see also [28-30]),

$$\alpha_2(\Omega_2) = \alpha_2^0(0) \text{Im} f(\Omega_2, |E_3|^2),$$

$$\Delta n(\Omega_2) = n(\Omega_2) - n(\Omega_2)^{nr} = \delta n_2 \text{Re} f(\Omega_2, |E_3|^2),$$

$$f(\Omega_2, |E|^2) = -i \frac{\Gamma_{gm} \Gamma_{g} G_{3}}{\Delta n_2 \Gamma_{gn} + i(\Omega_2 - \Omega_3)(r_n - r_g) - i r_{mn} G_3},$$

where $\alpha_2^0(0)$ is the absorption (or amplification, depending on the population difference $\Delta n_2$ sign) at the spectral line center if the strong field $E_3$ is turned off, $\delta n_2^0$ is the maximum contribution of transition $ng$ to the refraction index at $E_3 = 0$, and $n(\Omega_2)^{nr}$ is the linear contribution of all other nonresonant levels.

As was noted above, the NIE leads to inversionless amplification and is created there by the coherence at the transition $gm$. The coherence is induced by the strong field (factor $r_{mn}$) in combination
with the probing field. The greater is $|G_3 r_{nm}/\Gamma_{gm}|$ as compared to $r_n - r_g$, the more pronounced is the effect,

$$r_{mn} = -\frac{i G_3 (r_n - r_m)}{\Gamma - i \Omega_3},$$

(13) hereafter $\Gamma \equiv \Gamma_3$. At $\Omega_3 = 0$ the absorption (amplification) maximum corresponds to $\Omega_2 = 0$, hence,

$$f(0) = \frac{r_n - r_g - (r_n - r_m)|G_3|^2/\Gamma \Delta n_2}{(1 + |G_3|^2/\Gamma_{gm} \Delta n_2)^2}.$$  
(14)

Thus, even at $(r_n - r_g) > 0$ and $(r_n - r_m) > 0$, a negative absorption, i.e. amplification, could take place, if

$$|r_n - r_m||G_3|^2/\Gamma_{gm} > |r_n - r_g|.  
(15)$$

The lower is the coherence relaxation rate $\Gamma_{gm}$ at two-photon transition $gm$ as compared to the coherence relaxation at coupled one-photon transitions, the more favorable are conditions for inversionless amplification. At $|G_3|^2 \gg \Gamma_{gm} \Gamma_{gn}$ a splitting of the level $n$ into two quasi-levels significantly reduces interference and, hence, amplification at the transition $gn$ center. There is also an optimum relationship between saturated population differences at the interacting transitions. It depends on the strong field intensity and the relation between initial population differences $\Delta n_2 = n_n - n_g$ and $\Delta n_3 = n_n - n_m$ [28-30], created by an additional noncoherent radiation. To vary this relation in a wide range, we proposed in [41-43] to use alkali atoms placed into a high-pressure buffer gas. The strong field couples $P_{3/2}$ and the ground $S$-level. A fast collisional exchange furnishes population transfer from $P_{3/2}$ to the lower level $P_{1/2}$. For simplicity, it can be believed that the population distribution over the fine structure levels is Boltzmann’s one due to collisions. Thus, it becomes possible to vary the population difference at the probing transition in a wide range varying the strong field intensity and buffer gas pressure. Due to the saturation of $P_{3/2} - S$ transition, even the population inversion at the $P_{1/2} - S$ becomes possible (similar to a ruby laser). Hence, $P_{1/2} - S$ can be chosen as a probing transition. The population inversion was experimentally observed by a similar scheme in the mixture of sodium and helium vapors [44].

Collisions play a double part, i.e., on the one hand they considerably worsen coherence, on the other hand the population transfer due to collisions furnishes simple control and optimization of the population differences at the coupled transitions. Moreover, a wide collisional broadening allows one to neglect the nonuniformity of interaction with atoms due to the Doppler effect, hyperfine splitting, and some other processes. This makes an experiment be governed by the simplest theoretical model. By estimations and numerical examples for sodium atoms, now we show that inversionless amplification and absorptionless resonant refraction can be significant under proposed experimental conditions, in spite of collisions reducing the coherence.

We will discuss the concrete $D-1$ and $D_2$ transitions in sodium. According to [44], we write the kinetic equations of level populations

$$(\Gamma_m + \nu_{mg}) r_m - \nu_{gm} r_g - P = 0,$$

$$P = \frac{2|G_3|^2 \Gamma (r_n - r_m)}{\Gamma^2 + i \Omega_3^2},$$

(16)

$$(\Gamma_g + \nu_{gm}) r_g - \nu_{mg} r_m = 0, \quad (\Gamma_m r_m + \Gamma_g r_g - P = 0,$$

$$r_m + r_n + r_g = N,  
(17)$$

(18)

where $\nu_{gm}$ and $\nu_{mg}$, are the frequencies of collisions transferring populations, $\Gamma_g^{-1}$ and $\Gamma_m^{-1}$ are the lifetimes of relevant levels. From (16)-(18) we find

$$r_n - r_m = \frac{N}{1 + \alpha}, \quad r_n - r_g = \frac{N}{1 + \alpha} \left[ \frac{\nu_{mg} - (\nu_{gm} + \Gamma)}{\nu_{mg} + 2(\nu_{gm} + \Gamma)} - 1 \right],$$

(19)
where

\[ \alpha = \frac{2|G_3|^2 \Gamma}{\Gamma^2 + \Omega_3^2 \Gamma_g \nu_{mg} + \Gamma_m (\nu_{gm} + \Gamma)} \]  \tag{20} \]

Let us assume that \( \Gamma_g \approx \Gamma_m \) and a buffer gas pressure is high that \( (\nu_{mg} - \nu_{gm}) \gg \Gamma_g \). Taking under given conditions \( \nu_{gm} = \nu_{mg} \cdot \exp(-\Delta E/k_B T) \), where \( \Delta E = E_m - E_g \), is the fine splitting energy, \( k_B \) is the Boltzmann constant, and \( T \) is the temperature, we get

\[ r_n - r_g = \frac{N}{1 + \alpha} \left[ \frac{1 - \exp(-\Delta E/k_B T)}{1 + 2 \exp(-\Delta E/k_B T) - 1} \right], \]  \tag{21} \]

\[ \alpha = \frac{2|G_3|^2 \Gamma}{\Gamma_m (\Gamma^2 + \Omega_3^2)} \frac{1 + 2 \exp(-\Delta E/k_B T)}{1 + \exp(-\Delta E/k_B T)}. \]  \tag{22} \]

For sodium \( \Delta E = 17.2 \text{ cm}^{-1} \) and, at \( T = 550 \text{ K} \), estimations yield \( \Delta E/k_B T = 4.3 \cdot 10^{-2} \), \( \alpha \approx 3|G_3|^2/\Gamma \approx 9\lambda^3 I/64\pi^3 \epsilon_0 h c \),

\[ r_n - r_g = \frac{N}{1 + \alpha} \left[ 1.310^{-2} \alpha - 1 \right]. \]  \tag{23} \]

where \( \lambda \) and \( I \) are the strong field wavelength and energy flux density, \( \epsilon_0 \) is the dielectric constant of vacuum. From (13), (14), (20), and (21), it is seen that principally attainable inversionless amplification rises as \( \Delta E \) grows (e.g., in \( K \) and \( Rb \)).

The inelastic collision cross section in sodium and helium for the transition \( 3P_{3/2} - 3P_{1/2} \) is \( \sigma_{mg} \approx 4 \cdot 10^{-15} \text{ cm}^2 \). At \( T=550 \text{ K} \) and helium atmospheric pressure, estimations yields \( \nu_{mg} = N H e \bar{v}_0 \sigma_{mg} \approx 7.5 \cdot 10^9 \text{ s}^{-1} \). Since \( \Gamma_g \approx \Gamma_m \approx 6.2 \cdot 10^{-7} \text{ s}^{-1} \), the validity conditions for approximation (21) are satisfied. Using data [45] for the collisional broadening of sodium and helium lines, we estimate the collisional halfwidth as \( \Gamma \approx 5 \cdot 10^{10} \text{ s}^{-1} \), which exceeds the Doppler’s width of this transition, \( \Delta \omega_D/2 = 4.7 \cdot 10^9 \text{ s}^{-1} \). For our conditions, we have

\[ \frac{|G_3|^2}{\Gamma \Gamma_{gm}} \approx \frac{|G_3|^2}{\Gamma \Gamma_{gm}} \approx \frac{\alpha \Gamma_m}{3 \Gamma_{gm}}, \]

\( \alpha \approx 5 \cdot 10^9 I \), where \( I \) is expressed in \( \text{W} \cdot \text{cm}^{-2} \).

For the radiation power of 0.1 W focused into the footprint \( A = 10^{-5} \text{ cm}^2 \) (the confocal parameter is \( b \approx 1 \text{ cm} \)), we find \( |G_3| \approx 3.6 \text{ GHz} \), \( \alpha \approx 5 \cdot 10^9 \), and \( |G_3|^2/\Gamma \approx 0.1 \). These values are optimal to vary the population difference around zero at the probing transition \( r_n - r_g \). Above estimations for the intensity 1-10 kW-cm\(^2\) required to change noticeably the line shape agree well to the experiment with a change in the population difference ratio at the coupled transitions (44). Inversionless amplification at \( r_n - r_g = 0 \) is estimated as \( \alpha_2(0)/\alpha_2(0) \approx -\Gamma_m/3 \Gamma_{gm} \). Accepting \( \Gamma_{gm} \approx \nu_{mg} \), we find a value about 0.3% of the absorption in the absence of strong field. It is seen that this value is very sensitive to the coherence decay rate at the transition \( gm \).

Absorption-amplification spectral line shapes, frequency interval positions and halfwidths can be controlled, as analyzed in [29,30]. The line shape is very sensitive to the strong field intensity and frequency detunings from resonance. The amplification halfwidth increases and maximum decrease as the strong field intensity grows above a certain value. The population difference saturation at the strong field transition and the common energy level splitting reduce the inversionless amplification, so that it should be optimized by an appropriate choice of the strong field intensity and detuning. In our case the inelastic collision frequency is the important optimization parameter. The refraction index (dispersion) is described by \( \text{Re} f(\Omega_3, |E_3|^2) \). In the framework of proposed experiment the absorption-amplification coefficient and refraction index shapes can be controlled so that the refraction maximum falls within the spectral interval of vanishing absorption.

Thus, the considered model of three-level system with the interference controlled by collisions makes it possible the amplification without population inversion and the resonantly amplified refraction at
vanishing absorption. Exact formulas are presented to analyze optimum experimental conditions. Collisions destroying the coherence reduce the effects, as compared to atomic beams. However, this decrease is comparable to the effect of Doppler broadening in metal vapors. Advantages of the proposed experimental design are a simplicity and the possibility to control populations at the coupled transitions and to avoid interfering effects. This makes experiment adequate to a simple theoretical model. An experiment on inversionless amplification in the continuous mode in potassium vapor using collisional population of the upper level of probing transition was carried out in [46], however the four-level configuration contribution was significant there. The latter was conditioned by the transitions between hyperfine splitting sublevels and the incomplete overlapping of optical transitions due to insufficient collisional broadening.

4 Coherence and frequency mixing: multiple resonances at the condition of induced transparency

A nonlinear-optical response sharply increases as interacting wave frequencies approach one- and multiphoton resonances. This reduces required intensities of initial fields down to the values corresponding to cw lasers [47-51]. However, due to resonant absorption of primary and generated waves, there arise limitations from above onto the atomic density. Quantum coherence alternatively manifests itself in various optical processes. In particular, as was shown by an example of bound-free transitions [31, 32, 36], the absorption decrease can be not accompanied by an effective nonlinear susceptibility decrease on frequency mixing and varies the refraction index in another way.

Recently, an interest is growing to control matter-optical properties via the quantum coherence effects, especially promising for shortwave generation [7, 52, 53]. Therewith, an accent is on the wave conversion at frequency mixing under the condition of resonance with an absorbing transition between discrete levels and only by the generated field. In [54] a scheme of totally resonant multiphoton interaction was proposed, in which the quantum transition coherence and interference suppress absorption of both primary and generated field. Therewith the atomic nonlinear susceptibility is not subject to a significant destructive interference and rises by many orders of magnitude due to simultaneous multiphoton and one-photon resonances. Now available great atomic concentration rises additionally the nonlinear-optical response of medium and yields new spectral dependencies conditioned by local field effects (to be considered further).
In this section we consider several qualitative effects conditioned by intratomic coherence and possible totally resonant four-wave interaction under low absorption of both generated and initial fields. We show also the possibility of significant efficiency of generation. The results are easily extended to nonlinear-optical processes of higher order. Now we turn to the energy level diagram shown in Fig.2a [54].

The strong fields $E_3$ and $E_2$ at frequencies $\omega_3$ and $\omega_2$ couple nonpopunited levels 3, 2 and 2, 1, respectively. The fields $E_1$ and $E_s$, generated at frequencies $\omega_1 \approx \omega_0$ and $\omega_s = \omega_1 + \omega_2 + \omega_3$ are assumed to be weak and not changing the level populations. The latter fields are considered only in the lowest order of perturbation theory. The absorption coefficient and refraction index at frequencies $\omega_1$ and $\omega_s$, as well as the nonlinear polarization generating the wave at frequency $\omega_s$ are defined by real and imaginary parts of effective linear

$$\chi(-\omega_1; \omega_1) = (\chi_1^0/P_{01})f_1, \quad \chi_s(-\omega_s; \omega_s) = (\chi_s^0/P_{03})f_s, \quad (24)$$

and nonlinear

$$\chi^{NL}(-\omega_s; \omega_s + \omega_2 + \omega_3) = (\chi_0^{NL}/P_{01}P_{02}P_{03})f \quad (25)$$

susceptibilities, which, in turn, are proportional to the pre-exponential factors $r_i$ and $\tilde{r}_i$ of the corresponding components of nondiagonal density matrix elements (see the similar equations of Sec. 2). Here $\chi_1^0, \chi_s^0$ and $\chi_s^{NL}$ – are the resonant susceptibilities at negligibl $G_2$ and $G_3$. The factors $f_1$, $f_2$, and $f$ describe the strong field effects. Simple calculations by the density matrix procedure similar to [30,36] yield

$$f_1 = \{1 + g_2/P_{01}P_{02}[1 + (g_3/P_{02}D_{03})]\}^{-1}, \quad (26)$$

$$f_s = \{1 + g_3/P_{03}D_{02}[1 + (g_2/D_{02}D_{01})]\}^{-1}, \quad (27)$$

$$f = f_1[1 + g_3/D_{03}P_{02}]^{-1} = [1 + (g_2/D_{02}D_{01}) + (g_3/D_{03}P_{02})]^{-1}, \quad (28)$$

where

$$P_{01} = 1 + ix_1, \quad P_{02} = 1 + ix_2, \quad P_{03} = 1 + ix_s,$$

$$D_{01} = 1 + iy_{1}, \quad D_{02} = 1 + iy_{2}, \quad D_{03} = 1 + iy_s,$$

$$x_1 = \omega_1 - \omega_0 \Gamma_{10} = 0, \quad x_2 = \omega_1 + \omega_2 - \omega_2 \Gamma_{20} = 0, \quad x_s = \omega_s - \omega_3 \Gamma_{30} = 0,$$

$$y_1 = \omega_s - \omega_3 - \omega_2 - \omega_0 \Gamma_{10} = 0, \quad y_2 = \omega_s - \omega_3 - \omega_2 \Gamma_{20} = 0, \quad y_s = \omega_1 + \omega_2 + \omega_3 - \omega_0 \Gamma_{30} = 0,$$

$$g_2 = G_2^2/\Gamma_{10}\Gamma_{20}, \quad g_3 = G_3^2/\Gamma_{30}\Gamma_{20},$$

and $\Gamma_{ij}$ are the uniform halfwidths of corresponding transitions. To analyze the cases. when $E_s$, is not an independent probing field, we should put $\omega_s = \omega_1 + \omega_2 + \omega_3$ and $D_{0i} = P_{0i}$.

Differences of the factors $f_1$, $f_2$, and $f$ from unity and between each other, as well as their frequency dependency, are conditioned by two different coherence initiation channels $\rho_{02}$ (two combinations of strong and weak fields, $E_1, E_2$ and $E_s, E_3$) and their evolution as the field intensities rise. The absorption coefficients are defined by imaginary parts of the relevant susceptibilities, relative to which the considered processes manifest themselves as resonance splitting and absorption minima. The generated wave power $P \propto g_2g_3|\chi^{NL}|^2$ is defined by not only an imaginary part, but also by the real part of $\chi^{NL}$. (A relative phase of the generated wave depends on their ratio.) Therefore, the quantum coherence effects can be used to match the most important generation conditions, i.e., significant decrease of absorption for all the interacting fields without a noticeable decrease in atomic nonlinear-optical response. Furthermore, these effects can be used to increase additionally the generation efficiency improving the wave phase velocity matching. The laser-induced spectral structures in the susceptibility real parts (additional dispersion conditioned by the coherence) enable such a matching by a slight detuning from the resonance for $\omega_1$ or $\omega_s$.  

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An approach to the two resonances increases $|\chi^{NL}|^2$ by the factor $x_i^{-2}$, equal to $10^6$ and more. The triple resonance gives a gain of the order of $10^{18}$. Due to the induced transparency, the atomic density $N$ and, hence, the power $P \propto N^2$ can be additionally increased by a few orders of magnitude.

These results are easily generalized to the case of higher order mixing. For instance, if the 3-2 and (or) 2-1 transitions are multiphoton, the generalization is achieved by substituting the one-photon Rabi frequencies $G_2$ and $G_3$ with the corresponding matrix elements for multiphoton transitions.

In a particular case, when the transparency is induced only at the frequency $\omega_s$, by the wave $E_4$ not directly participating in conversion (Fig. 2c), the relevant expressions are also deduced by a simple subscript substitution. It is necessary to put $g_2 = 0$ and change subscripts in the formulas for $g_3$ and the corresponding resonance denominator. This case is similar to considered in [31, 32, 36] for the transition to continuum (Fig. 26) (see also [52, 53]).

Thus, the intratomic coherence effects in strong electromagnetic fields enable one to control absorption, refraction, and nonlinear-optical generation spectral properties. In particular, the above choice of transition diagram and interacting wave intensities make it possible to gain a medium nonlinear-optical response increased by many orders, meanwhile eliminating the initial and generated field absorption.

5 Atomic coherence: effects of a local field and the spectral line nonuniform broadening

5.1 Local field

The local field acting on an atom substantially differs from the external field both in value and phase, as the atomic density grows. This varies shapes of the spectral lines conditioned by the quantum transitions’ interference [55]. The effect is revealed at particle concentration of the order of $10^{17}$ cm$^3$.

Let us show by a simple example that qualitatively new spectral dependencies can arise in above problems due to the local field formation and drastic variation in the field of additional strong laser radiation at an adjacent transition.

According to a conventional (but approximate) concept (see, e.g. [56, 57]), the local $E_L$ and external $E$ fields in isotropic media are related by the simple formula

$$E_L = E + \frac{P}{3\epsilon_0}. \quad (29)$$

The medium polarization $P$ to a linear approximation can be presented by $P = \epsilon_0 N \alpha E_L$, where $N$ is the atomic concentration, $\alpha$ is the microscopic (atomic) polarizability, and $\epsilon_0$ is the dielectric constant of vacuum.

One of the consequences from (29) is the Clausius-Mossotti equation [56, 57] relating polarizability $\alpha$ to the dielectric constant $\epsilon_0$ of material

$$\epsilon = 1 + LN\alpha, \quad (30)$$

where the local field factor $L = (\epsilon + 2)/3 = (1 - \alpha N/3)^{-1}$ shows how the local field differs from the external one. The former plays an important part in linear and nonlinear optical phenomena (see, e.g. [56-59]). In spite of an approximate nature of formula (29), the authors of [58] showed that it well describes linear and nonlinear responses of the dense atomic gas.

Let us consider the interaction of two optical fields with three-level systems of the cascade or $\Lambda$-schemes. Therewith, one field is strong and (for simplicity) interacts with the transition between nonpopulated or equipopulated levels. According to the classification of resonant nonlinear processes (see Sec. 2), only the resonance splitting effect would be observed for the probing field in this case. For certainty we consider the $n-m-l$ $\Lambda$-scheme (Fig. 1), where the levels $m$ and $n$ are nonpopulated, but the state $l$ is ground. The strong field of frequency $\omega_3$ and amplitude $E_3$ acts at the $nm$ transition,
the weak (probing), wave of frequency \( \omega_4 \) and amplitude \( E_4 \) acts at the \( lm \) transition. The set of equations for this problem differs used in Sec. 3 only by the substitution of \( E_4 \) by \( E_{4L} \). Such an approach is widely used in the local field theory [56-59].

For the macroscopic complex polarization \( P_4(\omega_4) = d_{ml} \rho_{lm} N \) using formulas of Sec. 3 we find

\[
P_4(\omega_4) = \epsilon_0 \chi_4(\omega_4) E_4,
\]

\[
\chi_4(\omega_4) = \chi_0^4 f(\omega_4), \quad (31)
\]

\[
\chi_0^4 = i N |d_{ml}|^2 / \epsilon_0 \Gamma_{lm}, \quad f(\omega_4) = \Gamma_{lm} P_{43} / (P_4 - i \delta_{4L}) P_{35} + |G_4|^2, \quad (32)
\]

where \( \chi_4 \) is the macroscopic susceptibility at the probing field frequency \( \omega_4 \) in the presence of strong one at the frequency \( \omega_3 \), \( \chi_0^4 \) is the susceptibility in the absence of the latter, and \( f(\omega_4) \) is the form-factor. The parameter

\[
\delta_{4L} = |d_{ml}|^2 N / 3 \epsilon_0 \hbar
\]

appears as the transition \( lm \) frequency shift conditioned by the concentration rise (local field). It is substantial that in so doing the two-photon and strong field transition frequencies are not varied. As a result, the local field effect is not reduced to redefining the detuning of the weak field resonance, but qualitatively changes the whole spectral line shape, if this factor becomes comparable to the resonance width. At \( d_{ml}=1 \) Db and \( N = 10^{23} \) m\(^{-3} \) we estimate this shift as \( \delta_{4L} = 8 \cdot 10^{11} \) s\(^{-1} \), which can be comparable to characteristic shock widths of resonances. For instance, when the shock width is defined by resonance exchange (self-broadening) and significantly exceeds a natural one, \( \Gamma_{ml} \) has the form (see, e.g. (58,59] and references therein)

\[
\Gamma_{ml} \approx |d_{ml}|^2 N / 6 \epsilon_0 \hbar,
\]

whence it follows that the ratio \( C_4 = \delta_{4L} / \Gamma_{ml} \) can approach two in this case.

Peculiarities of the local field, exhibited in spectral nonlinear interference dependencies of absorption, are analyzed in [55]. The red shift, entering all the resonant denominators, and hence, not leading to a simple redefinition of the detuning from one-photon resonance, changes qualitatively the spectral dependencies as the absorbing particle concentration rises. Thus, the local field strongly varies spectral dependencies of the probing field absorption in the presence of a strong field at the adjacent transition.

The factor \( L_4 = E_{4L} / E_4 \) characterizing the difference between local and external fields by value and phase can be presented in the form

\[
L_4 = 1 + i G_4 f(\omega_4). \quad (34)
\]

Whence it follows that this difference also increases as the parameter \( C_4 \) rises. Varying the strong field intensity and frequency, the local field spectral dependence can be also significantly varied.

The resonant exchange also shifts the frequency of the ground-to-excited state transition. This shift is proportional to atomic concentration, but is usually two- or threefold shorter than the broadening and can be often neglected.

The acquired results are genual and applicable to other interaction schemes. In the case of cascade transitions under the conditions of zero or equal level populations at the strong field transition, the results are found by a simple redefinition of corresponding quantities.

Since the local field acting on an atom can substantially differ by value and phase from the external field as atomic density rises, this drastically changes spectral dependencies not only for absorption and refraction, but also for generating nonlinear polarizations [54]. Similar, it can be shown using the Lorentz-Lorentz approximation that the local field induces red shifts in the resonant denominators at the allowed transitions for effective nonlinear susceptibilities (Sec. 4). Thus, a supplementary
opportunity appears to increase efficiency sharply by controlling concentration, initial field intensities, and detunings from resonances at the conditions of multiple resonances, induced transparency, and phase matching. The considered local field effects should be taken into account when designing and interpreting experiments.

5.2 Constructive and destructive interference as a consequence of atomic velocity distribution: nonuniform broadening of spectral lines

As it was already mentioned, the laser-induced coherence contribution into spectra can be constructive or destructive depending on a detuning sign. Therefore, inversionless amplification and resonance splitting in gases can significantly differ from those for stationary atoms at the Doppler broadening prevailing above uniform one. Nevertheless, as was shown in [22, 23, 27, 30, 60, 61], spectral profiles with alternating signs are induced sometimes even in this case. In the relatively weak laser fields the NIE manifest itself in narrow spectral structure variations within the wide Doppler’s contour. This structure is anisotropic, i.e., depends on the angle between the strong and probing field wave vectors, as well as on collisions changing atom velocities. Destructivity or constructive effect of the Maxwell velocity distribution depends on the position of probing wave frequency compared to the strong-field frequency. The probing field transition line is being deformed as a whole as the strong-field transition structure is anisotropic, i.e., depends on the angle between the strong and probing field wave vectors, as well as on collisions changing atom velocities. Destructivity or constructivity effect of the Maxwell velocity distribution depends on the position of probing wave frequency compared to the strong-field frequency. The probing field transition line is being deformed as a whole as the strong-field transition uniform broadening or the wave intensity rise. Formulas of averaging over velocities for a number of cases are presented in [27, 30, 60, 61].

Since the dependence of responses on detuning signs often has a sign-alternating character at coherent interaction, some interfering components can vanish when averaged over velocities. We present an example showing the effect of nonuniform broadening on coherent interaction. Let us consider the four-wave mixing in the lowest order of perturbation theory (Fig. 1). From the solutions to equations for \( \tilde{r}_4 \) (Sec. 2) we deduce the nonlinear susceptibility at frequency \( \omega_4 = \omega_1 - \omega_2 + \omega_3 \)

\[
\chi(\omega_4 = \omega_1 - \omega_2 + \omega_3) = \frac{iK}{\Gamma_{ml} + i(\Omega_1 - \Omega_2 + \Omega_3)} \times \\
\left\{ \frac{1}{\Gamma_{gm} + i(\Omega_3 - \Omega_2)} \left[ \frac{n_g - n_n}{\Gamma_{ng} + i\Omega_2} + \frac{n_m - n_n}{\Gamma_{mn} + i\Omega_3} \right] + \frac{1}{\Gamma_{in} + i(\Omega_1 - \Omega_2)} \left[ \frac{n_g - n_n}{\Gamma_{ng} - i\Omega_1} + \frac{n_g - n_l}{\Gamma_{lg} + i\Omega_1'} \right] \right\}
\]

where \( K \) is the constant, \( \Omega_1' = \omega_1 - \omega_{gl} - k_1\mathbf{v} = \Omega_1 - k_1\mathbf{v} \) is the detuning from resonance tailing the Doppler’s shift into account, other \( \Omega_2' \) are the similar detunings depending on velocity, and \( n_i \) are the populations of corresponding levels, also depending on velocity.

As is seen from (35), all the terms, besides those proportional to \( n_g - n_n \) as functions of velocity, have poles at the same complex semiplane. Therefore, if the Doppler’s shifts corresponding to the heat velocity \( u \) much exceed uniform halfwidths of transitions, then only the polarization components proportional to \( n_g - n_n \) are nonzero after averaging over velocities with the Maxwell distribution. The averaging result has the form

\[
\langle \chi \rangle_v = \frac{iK \pi^{1/2} \exp \left\{ - (\Omega_2/k_2u)^2 \right\} (N_g - N_n)}{k_2u[\tilde{\Gamma}_1 + i(\Omega_1 - k_1\Omega_2/k_2)][\tilde{\Gamma}_3 + i(\Omega_3 - k_3\Omega_2/k_2)]},
\]

where \( N_g \) and \( N_n \) are the integral over velocities unperturbed level populations, \( \Omega_4 = \Omega_1 - \Omega_2 + \Omega_3 \), \( \Omega_1 = \omega_1 - \omega_{gl} \), \( \Omega_2 = \omega_2 - \omega_{gm} \), \( \Omega_3 = \omega_3 - \omega_{mn} \), \( k_i = \omega_i/c \) and

\[
\tilde{\Gamma}_1 = \Gamma_{ml} + (k_1/k_2 - 1)\Gamma_{ng}, \quad \tilde{\Gamma}_3 = \Gamma_{gm} + (k_3/k_2 - 1)\Gamma_{ng}.
\]

Substituting \( \Omega_2' \) by \( -\Omega_2' \) in the sum \( \omega_4 = \omega_1 - \omega_2 + \omega_3 \) in the resonant cascade level configuration, we find that all the poles appears at the same complex plane. This significantly decreases the averaged susceptibility as compared to the frequency subtraction scheme.
The effect of velocity distribution on the coherent four-wave mixing can be described by the following way. For stationary atoms and a totally resonant process, the nonlinear polarization is proportional to the factor $\Gamma^{-3}$. For a gas and the frequency subtraction at an available intermediate level population, this factor is substituted by $1/ku\Gamma^2$, i.e., decreased by $ku/\Gamma$. A more detailed analysis [30] shows that for frequency summation or at $N_n - N_g = 0$, the coherence and nonlinear polarization suppressed by the interference of various velocity contributions yields this factor equal to $(ku)^{-3}$. In other words, the susceptibility decreases by the factor of $(ku/\Gamma)^3$ as compared to stationary atoms and by the factor of $(ku/\Gamma)^2$ as to optimum conditions for frequency subtraction in gases. Hence, the difference scheme was chosen for continuous four-wave generation in the field of helium-neon laser in [47] as distinct from [48].

When only one resonance ($\omega_1 - \omega_2 \approx \omega_{ln}$) of the Raman-type scattering presents, we have

$$\chi^{(3)} \propto \frac{1}{\Omega_1 \Omega_4} \exp \left[ - \left( \frac{\Omega_1 - \Omega_2}{(k_1 - k_2)u} \right)^2 \right].$$

In the absence, of resonances (for both frequency summation and subtraction), we find

$$\chi^{(3)} \propto \frac{1}{\Omega_1 \Omega_4 (\Omega_1 - \Omega_2)}.$$ 

Since the NIE and mixing processes are defined by a common one source, i.e., the coherence induced at a forbidden transition, dose effects occur also at averaging over velocities.

Thus, the transition nonuniform broadening can significantly change the influence of various level population on the coherent processes, so that some level contributions prevail. Varying the level populations, strong field intensities and detunings, the amplification-absorption, refraction, and nonlinear polarization spectra can be controlled, thus considerably increasing the generation yield.

6 Nonlinear interference effects at bound-free transitions: laser-induced autoionization-type resonances in the continuum

Nonlinear interference effects similar to those occurring at the discrete transitions (including inversionless amplification and induced transparency) manifest themselves also in continuous spectra, e.g., at the transition into an ionization continuum. The corresponding theory was generalized in [30-32, 36, 62]. Similar phenomena for crystal bands were considered in [29]. The laser-induced and autoionization-type resonances theoretically analyzed in [31] were experimentally observed in [32], and then the nonlinear interference processes became a subject of intense studies in the context of laser-induced continuum structures (LICS), inversionless amplification and electromagnetically induced transparency, first, at bound-free transitions (see, e.g. [62, 64, 65]) and then also at discrete transitions (see [7-11, 40, 52, 53]).

Let us show the potentials to control simultaneously two LICs and to split discrete resonances by strong electromagnetic fields to reduce absorption, correct phase matching, and to improve nonlinear-optical short-wave generation techniques (Fig. 2b). The wave at frequency $\omega_1$ is weak, but those at frequencies $\omega_3$, and $\omega$ are strong. We also take into account possible strong nonresonant transitions to the discrete levels $k$. The detunings $|\omega_1 - \omega_{gm}|$, $|\omega_1 + \omega_2 - \omega_{gn}|$ and $|\omega_1 - \omega_3 - \omega_{ln}|$ are assumed to be much smaller than all other. Density matrix calculations similar to [31, 36] yield the following expressions for nonlinear susceptibility $\chi^{(3)}(\omega_\mu = \omega_1 + \omega_2 + \omega_3)$, the probing wave absorption coefficients $\alpha(\omega_1)$ and $\alpha(\omega_\mu)$ at the corresponding frequencies,

$$\frac{\chi^{(3)}(\omega_\mu = \omega_1 + \omega_2 + \omega_3)}{\chi^{(3)}_{0\mu}} = \frac{K}{D_{gm}X},$$

(38)
\[
\frac{\alpha(\omega_1)}{\alpha_{01}} = \text{Re} \left\{ \frac{1 - g_{nn}/(D_{gm}X)}{D_{gm}} \right\},
\]
\[
\frac{\alpha(\omega_\mu)}{\alpha_{0\mu}} = 1 - k_3 \beta_l + \frac{k_3 \beta_l (y_l + q_{gl})^2}{1 + y_l^2} - \text{Re} \left\{ k_4 g_{nn} A^2 \frac{(1 - i q_{gl})^2}{Y} \right\},
\]
where \(\chi^{(3)}_0, \alpha_{01}\) and \(\alpha_{0\mu}\) are the relevant resonant values for negligible intensities of all the fields,
\[
K = 1 - \frac{k_1 \beta_l [(1 - i q_{nl})(1 - i q_{pl})]}{(1 - i q_{nl})(1 + ix_l)},
\]
\[
A = 1 - \frac{k_1 \beta_l [(1 - i q_{nl})(1 - i q_{gl})]}{(1 - i q_{gn})(1 + iy_l)},
\]
\[
X = (1 + g_{nn}) \left[ 1 + ix_n + \frac{q_{mn}}{D_{gm}(1 + g_{mn})} - k_2 \beta_l \beta_n \frac{(1 - i q_{nl})^2}{1 + ix_l} \right],
\]
\[
Y = (1 + g_{nn}) \left[ 1 + iy_n + \frac{q_{mn}}{p_{gm}(1 + g_{mn})} - k_2 \beta_l \beta_n \frac{(1 - i q_{gl})^2}{1 + iy_l} \right],
\]
\[
D_{gm} = 1 + \frac{i(\omega_1 - \omega_{gm})}{\Gamma_{gm}}, \quad p_{gm} = 1 + \frac{i(\omega_\mu - \omega_3 - \omega_2 - \omega_{gm})}{\Gamma_{gm}},
\]
\[
x_l = \frac{\omega_1 + \omega_2 + \omega_3 - \omega - \omega_{gl} - \delta_l}{\Gamma_{gl} + \gamma_{ll}}, \quad x_n = \frac{\omega_1 + \omega_2 - \omega_{gm} - \delta_{nn}}{\Gamma_{gn} + \gamma_{nn}},
\]
\[
y_l = \frac{\omega_\mu - \omega - \omega_{gl} - \delta_l}{\Gamma_{gl} + \gamma_{ll}}, \quad y_n = \frac{\omega_\mu - \omega_3 - \omega_{gm} - \delta_{nn}}{\Gamma_{gn} + \gamma_{nn}},
\]
\[
k_1 = \frac{\gamma_{gl} \gamma_{nn}}{\gamma_{gn} \gamma_{mn}}, \quad k_2 = \frac{\gamma_{il} \gamma_{ln}}{\gamma_{ll} \gamma_{mn}}, \quad k_3 = \frac{\gamma_{gl} \gamma_{lq}}{\gamma_{gg} \gamma_{ll}}, \quad k_4 = \frac{\gamma_{gn} \gamma_{ng}}{\gamma_{gg} \gamma_{mn}}
\]
\[
g_{mn} = \frac{|G_{mn}|^2}{\Gamma_{gm} \Gamma_{gn}}, \quad \beta_l = \frac{g_{ll}}{1 + g_{ll}}, \quad \beta_n = \frac{g_{nn}}{1 + g_{nn}},
\]
\[
g_{ii} = \frac{\gamma_{ii}}{\Gamma_{ii}}, \quad g_{ij} = \frac{\delta_{ij}}{\gamma_{ij}},
\]
\[
\gamma_{ij} = \pi h G_{ie} G_{ej} |_{\epsilon = \hbar \omega_\mu} + \text{Re} \left\{ \sum_k \left( \frac{G_{ik} G_{kj}}{p_{jk}} \right) \right\}, \quad \delta_{ij} = h P \int \frac{de G_{ie} G_{ej}}{h \omega_\mu - \epsilon} + \text{Im} \left\{ \sum_k \frac{G_{ik} G_{kj}}{p_{jk}} \right\},
\]
P in (51) designates the sign of the integral principal value. The factors \(k_i\), take the values \(0 \geq k_i \geq 1\) depending on a degree of degeneracy for continuum states (unity for the nondegenerate states).

Formulas (38) and (40) generalize the expressions from [31, 36] to the case of several strong fields. Together with (39) these formulas show the possibility to reduce absorption both for the primary and the generated fields. The absorption falls exponentially as the medium length rises. Since the absorption coefficients, as functions of frequency, and the ratios of squared module of nonlinear susceptibility to these coefficients, do not coincide at certain conditions. These ratios define the generation power in absorbing media. This power quadratically rises with growing atomic concentration and the initial fields intensity under the condition of optimized medium perturbation. Comparing (38) and (40) to corresponding formulas from [31, 36] and Sec. 4, we see new interference spectral structures caused by the joint action of strong fields \(E_3\) and \(E\) (terms proportional to \(\beta_n\) and \(g_n\)), arising in nonlinear polarization and absorption (refraction). These nonlinear resonances give additional opportunities for absorption nonlinear spectroscopy and enhancement of efficiency for nonlinear-optical conversion to the short-wave spectral range.
Nonlinear interference and relaxation

Relaxation processes can extraordinarily exhibit the coherence effects. For instance, the spontaneous relaxation coupling two transitions with close frequencies [64, 65] can promote inversionless amplification even in the absence of strong fields. Let us consider an example [47], when certain relaxation mechanisms and external dc fields suppressing the destructive interference make the resonant nonlinear-optical interaction allowed. The experiment was carried out with the He–Ne laser beam ($\lambda = 1.52 \mu m$) resonant to the transition $2s_2 - 2p_4$ of Ne atoms. Upper and lower levels contain three ($J_1 = 1$) and one ($J_0 = 0$) Zeeman sublevels, respectively. The initial beam contained two linear orthogonally polarized components $E_1$ and $E_2$ with the frequency shift $\Delta = \omega_2 - \omega_1$ much lower than the natural transition width. The wave intensity at frequency $\omega_1$ was significantly higher than at $\omega_2$. The four-wave generation of $E_s$, arose at the frequency $\omega_s = 2\omega_1 - \omega_2 = \omega_2 - 2\Delta$ with the same polarization as in $E_2$. The generation power sharply rose with growing collision frequency and external dc magnetic field. This effect can be explained in the following way. It is convenient to expand each wave and nonlinear polarization $P^{(NL)}(\omega_s)$ into two circular components $P^{(NL)}_+(\omega_s)$ and $P^{(NL)}_-(\omega_s)$. These components contain two terms. One describes the four-wave mixing with the same polarization at two-level subsystems, while another does the radiation with contrary polarizations at three-level Zeeman subsystems (Fig. 3a).

At such a choice of polarizations, these two contributions interfere destructively and totally suppress one another, if relaxation rates for populations and quadrupole moments (alignment) are equal for the Zeeman sublevels in the upper electron state. The spontaneous radiation trapping, anisotropic collisions, and/or the external magnetic field violate the amplitude balance of destructively interfering components of nonlinear polarization and induce the four-wave mixing. The magnetic field effect on

Figure 3: Interfering oscillations in various resonant processes: (a) destructive interference of four-wave mixing processes in the double $V$-shaped and two-level configurations between the Zeeman sublevels of transition $2s_2 - 2p_4$ Ne [47], (b,c) interference of the doublet sublevels contributions into four-wave mixing [66, 67], (d) up-conversion of the weak infrared ($\omega_2$) wave (fields $E_1$ and $E_3$ are strong mainly destructive interference at the frequencies $\omega_s - \omega_3 = \omega_{ng} = \omega_1 + \omega_2$ limits the conversion efficiency [69], (e) interference of multiphoton and one-photon polarization components limits the upper level population, (f) interference of nonresonant nonlinear polarization of the seventh order and the resonant one of the ninth order are used to detect the seventh harmonic generation at the resonant nonlinearity of the ninth order [70], and (g,h) interference of the doublet component contributions into the two-photon and nonresonant one-photon processes (spontaneous emission from the upper levels).
the two-level configuration is compensated for the Doppler shifts. The second component of nonlinear polarization represents a double V-configuration (Fig. 3a), which is removed from resonance by the magnetic field.

Now let us consider another example (Fig. 3a). The coherence induced at the transition \( n' n \), defining four-wave mixing \( \omega_s = 2 \omega_1 - \omega_2 \), is given by

\[
\rho^{(2)}_{n'n'} V_{n'g} \rho^{(1)}_{gm} \left[ \frac{1}{\Omega_1 + i\Gamma_{ng}} - \frac{1}{\Omega_2 + i\Gamma_{n'g}} \right] \frac{1}{\Omega + i\Gamma_{n'n'}} = \frac{1}{(\Omega_2 + i\Gamma_{n'g})(\Omega_1 - i\Gamma_{ng})} \left[ 1 - i \frac{\Gamma_{nn'} - \Gamma_{n'g} - \Gamma_{ng}}{\Omega + i\Gamma_{nn'}} \right],
\]

where \( \Omega_1 = \omega_1 - \omega_{ng} \), \( \Omega_2 = \omega_2 - \omega_{n'g} \), \( \Omega = \omega_2 - \omega_1 - \omega_{n'n} \). A spontaneous relaxation we have \( \Gamma_{ij} = (\Gamma_i + \Gamma_j)/2 \) and the resonance \( \Omega = 0 \) disappears. Collisions violate the relevant frequency equality and induce this resonance, suppressing the destructive interference [66, 67].

8 Conclusion

The purpose of this paper is to show the diverse exhibition of nonlinear interference effects in optics and to survey certain earlier works in this field. Interference phenomena can play a governing part in numerous experiments on resonant nonlinear optics [68]. Some such processes are sketched at Fig. 3 and explained in the figure caption.

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