Atomic coherence and interference phenomena in resonant nonlinear optical
interactions

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1 Abstract

Interference effects in quantum transitions, giving rise to amplification without inversion, optical transparency
and to enhancements in nonlinear optical frequency conversions are considered. Review of the relevant early
theoretical and experimental results is given. The role of relaxation processes, spontaneous cascade of polarizations,
local field effects, Doppler-broadening, as well as specific features of the interference in the spectral continuum
are discussed.

Keywords: atomic coherence and interference, resonant nonlinear interactions, bound-free transitions, amplification
without inversion, relaxation-induced processes, local field effects, inhomogeneous broadening, frequency-conversion,
VUV generation

2 Introduction

There has been considerable interest recently in the study of laser-induced quantum coherence and interference,
which leads to fundamental effects in high resolution nonlinear spectroscopy, to amplification of radiation
without the requirement of population inversion (AWI) and to resonantly enhanced refraction at vanishing (without)
absorption (ERWA), to coherent population trapping and constructive contributions in resonantly enhanced
nonlinear-optical frequency conversions and, at the same time, to distractive contributions in absorption of the fundamental
and generated radiations. Wide range of applications are expected.

Resonant nonlinear optical interference effects have been subject of the extensive both theoretical
and experimental studies since the discovery of masers and lasers (see for example and ref. therein).
In this paper we briefly review some early and recent results of Russian research groups on this topic.

3 Resonant nonlinear optical interference processes

3.1 Destructive and constructive interference in classical and quantum optical physics

Interference is one of the fundamental physical phenomena. Two oscillations at one and the same, or close,
frequencies may interfere both in constructive and distractive ways. One can manipulate

1Invited review paper
by the resulting oscillations with variation of the relative phase and the amplitudes of the interfering oscillators in order to enhance or, on the contrary, to eliminate the oscillations of any nature. Interference is widely used in optical physics, including quantum optics. The concept of interference is more general, then the notions of elementary quantum-optical processes, such as one-photon, multistep and multiphoton transitions. These notions were introduced and classify at their frequency-correlation properties in the framework of the perturbation theory. Indeed, in resonant interactions, these properties may be drastically changed with growth of the intensity of the coupled fields. The latter may give rise to such effects in nonlinear spectroscopy of Doppler broadened transition, as compensation of the residual inhomogeneous broadening in Raman-like and cascade configurations.

Quantum interference may occur when coherent superposition of real states is involved in a process. Alternatively, interfering frequency-degenerate intraatomic oscillations may originate from different correlated quantum pathways, contributing in one and the same frequency. For example, in the weak-field approximation, these can be one- and two-photon contributions to an optical process, associated with the radiation at a given frequency. Such process may be thought as that started from the coherent superposition of closely spaced real energy-level and quasi-level (virtual state), created by the auxiliary strong field. Such a coherent superposition can be produced even more easily than in the case of real doublet states. In general, even in the cases, when many elementary processes contribute to an optical process and their classification is troublesome, one can explain and predict experimental results with the aid of the notion of interfering frequency-degenerated components of nonlinear polarization. The amplitudes of the components can be varied with the intensities and phases – with the frequency-determinations of the driving fields.

3.2 Effect of energy levels population and relaxation, density matrix approach

In general case of open energy-level configuration with all the levels being populated and various relaxation processes involved, density-matrix method is the most convenient for the analysis of a resonant nonlinear-optical response. Explicit formulæ, describing spectral properties of a weak probe field in the presence of an auxiliary strong one, in cascade, $V$ and $\Lambda$ configurations can be easily derived in the similar way. We shall show that on the example of the energy-level schematic, given on Fig.1.

![Atomic energy-level configuration](image)

Figure 1: Atomic energy-levels configuration.

Fields $E_1$ at frequency $\omega_1 \approx \omega_{gl}$ and $E_3$ at frequency $\omega_3 \approx \omega_{mn}$ are strong. Fields $E_2$ at frequency $\omega_2$ and $E_4$ at frequency $\omega_4$ are probe ones. We shall derive the conditions to achieve $AWI$ at the transition $gn$, as well as at transition $ml$, so that both $V$ and $\Lambda$ configurations are embedded. Frequency of the probe field may be both higher and lower compared to the driving field.

Consider energy-level configuration, shown in the Fig.1. Density matrix equations in the interaction representation, relevant to the problem under consideration, can be written in the form:

\[
\rho_{gl} = r_1 \cdot \exp(i\Omega_1 t), \quad \rho_{nm} = r_3 \cdot \exp(i\Omega_3 t), \quad \rho_{ng} = r_2 \cdot \exp(i\Omega_1 t) + \tilde{r}_2 \cdot \exp[i(\Omega_1 + \Omega_3 - \Omega_4)t], \\
\rho_{mn} = r_4 \cdot \exp(i\Omega_4 t) + \tilde{r}_4 \cdot \exp[i(\Omega_1 - \Omega_2 + \Omega_3)t], \quad \rho_{ln} = r_{12} \cdot \exp[i(\Omega_1 - \Omega_2 + \Omega_3)t] + r_{43} \cdot \exp[i(\Omega_4 - \Omega_3)t], \\
\rho_{ii} = r_i, \\
P_{2r2} = iG_2 \Delta r_2 - iG_3 r_{32} + ir_{12} G_1, \quad d_2 \tilde{r}_2 = -iG_3 r_{41} + ir_{43} G_1, \\
P_{3r4} = i[G_4 \Delta r_4 - G_1 r_{41} + r_{43} G_3], \quad d_4 \tilde{r}_4 = -iG_1 r_{32} + ir_{12} G_3, \\
P_{4r1} = -iG_4 r_{14} + ir_{4} G_4, \quad P_{3r43} = -iG_4 r_{34} + ir_{43} G_3, \\
P_{32r32} = -iG_3 r_{32} + ir_{2} G_3, \quad P_{12r12} = -iG_1 r_{12} + ir_{12} G_2, \\
\Gamma_{mrm} = -2Re\{iG_3 r_3\} + \gamma_m, \quad \Gamma_{ggn} = -2Re\{iG_3 r_3\} + \gamma_{gn} + \gamma_{mn} r_m + q_n, \\
\Gamma_{gr} = -2Re\{iG_1 r_1\} + q_g, \quad \Gamma_{rl} = -2Re\{iG_1 r_1\} + \gamma_{gl} r_g + \gamma_{ml} r_m + q_l.
\]
\[ \Delta r_1 = r_l - r_g, \quad \Delta r_2 = r_n - r_g, \quad \Delta r_3 = r_n - r_m, \quad \Delta r_4 = r_l - r_m. \]

Where \( \Omega_1 = \omega_1 - \omega_{ig}, \quad \Omega_2 = \omega_3 - \omega_{inm}, \quad \Omega_2 = \omega_2 - \omega_{gm}, \quad \Omega_4 = \omega_4 - \omega_{ml}, \quad G_1 = -E_1 d_{ig}/2\hbar, \quad G_2 = -E_2 d_{gm}/2\hbar, \quad G_3 = -E_3 d_{nm}/2\hbar, \quad G_4 = -E_4 d_{ml}/2\hbar, \quad P_1 = \Gamma_{ig} + i\Omega_1, \quad P_2 = \Gamma_{ng} + i\Omega_2, \quad P_3 = \Gamma_{nm} + i\Omega_3, \quad P_4 = \Gamma_{lm} + i\Omega_4, \quad P_{12} = \Gamma_{in} + i(\Omega_1 - \Omega_2), \quad P_{34} = \Gamma_{gm} + i(\Omega_3 - \Omega_2), \quad P_{41} = \Gamma_{gm} + i(\Omega_4 - \Omega_1), \quad d_2 = \Gamma_{ng} + i(\Omega_1 + \Omega_3 - \Omega_4), \quad d_4 = \Gamma_{lm} + i(\Omega_1 - \Omega_2 + \Omega_3). \]

Here \( \Omega_i \) are frequency detunings from the resonances, \( G_i \) — Rabi frequencies, \( \Delta r_i \) — power—depending population differences, \( \Gamma_{ij} \) — homogeneous half linewidths, \( \Gamma_i^{-1} \) — lifetimes, \( \gamma_{ij} \) — relaxation rates from \( i \) to \( j \) states, \( g_i \) — population rate by an incoherent source. Density matrix amplitudes \( r_i \) determine absorption/gain and refraction indexes, \( \tilde{r}_i \) — determine four—wave mixing driving nonlinear polarizations.

The equations and their solution for the cascade atomic configurations can be derived by the simple change of the detunings signs \(^{26}\).

### 3.3 Laser—induced atomic coherence and classification of resonant nonlinear effects

Solution of the coupled density—matrix equations may be represented in the form:

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

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

\[ R_4 = \frac{\Delta r_4(1 + g_5 + v_9) - g_4(1 + g_5 - v_6)\Delta r_1 - v_1(1 + v_5 - g_6)\Delta r_3}{(1 + g_4 + v_4) + [v_5 + v_4(v_5 - g_6) + g_4(g_5 - v_6)]}, \]

\[ \Delta r_1 = \frac{(1 + a_3)\Delta n_1 + b_1 a_3 \Delta n_3}{(1 + a_3)(1 + a_3) - a_1 a_1 b_1 a_3}, \quad \Delta r_3 = \frac{(1 + a_1)\Delta n_3 + a_1 a_1 \Delta n_1}{(1 + a_1)(1 + a_3) - a_1 a_1 b_1 a_3}, \]

\[ \Delta r_2 = \Delta n_2 - b_2 a_3 \Delta r_3 - a_2 a_2 \Delta r_1, \quad \Delta r_4 = \Delta n_4 - a_3 a_3 \Delta r_1 - b_3 a_3 \Delta r_3; \]

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

\[ r_n = n_n - b_2 a_3 \Delta r_3 + a_1 a_1 \Delta r_1, \quad \Delta r_i (E_1 = 0, E_3 = 0) = \Delta n_i; \]

\[ g_1 = \frac{[G_1]^2}{P_{12}P_2}, \quad g_2 = \frac{[G_1]^2}{P_{12}P_1}, \quad g_3 = \frac{[G_1]^2}{P_{12}P_4}, \quad g_4 = \frac{[G_1]^2}{P_{12}P_4}, \quad g_5 = \frac{[G_1]^2}{P_{32}P_3}, \quad g_6 = \frac{[G_1]^2}{P_{32}P_4}, \quad g_7 = \frac{[G_1]^2}{P_{32}P_4}, \quad g_8 = \frac{[G_1]^2}{P_{32}P_4}, \]

\[ v_1 = \frac{[G_3]^2}{P_{32}P_3}, \quad v_2 = \frac{[G_3]^2}{P_{32}P_3}, \quad v_3 = \frac{[G_3]^2}{P_{32}P_3}, \quad v_4 = \frac{[G_3]^2}{P_{32}P_3}, \quad v_5 = \frac{[G_3]^2}{P_{32}P_3}, \quad v_6 = \frac{[G_3]^2}{P_{32}P_3}, \quad v_7 = \frac{[G_3]^2}{P_{32}P_3}, \quad v_8 = \frac{[G_3]^2}{P_{32}P_3}; \]

\[ a_1 = \frac{\gamma_{gm} a_2}{\Gamma_l \Gamma_g \Gamma_{ig}}, \quad a_2 = \frac{2(\Gamma_l + \Gamma_g - \gamma_{gl})}{\Gamma_l \Gamma_g \Gamma_{ig}}, \quad a_3 = \frac{a_1^2 \Gamma_3}{\Gamma_3} \Gamma_m \Gamma_N \gamma_{nm}, \quad a_6 = \frac{2(\Gamma_m + \Gamma_g - \gamma_{gm})}{\Gamma_m \Gamma_N \gamma_{nm}}; \]

By substituting frequency deviations \( \Omega_i \) for that Doppler-shifted \( \Omega_i - k_i v \) (\( v \) is atomic velocity) we can take into account the effect of atomic motion. Imaginary part of density-matrix amplitudes \( r_2 \) and \( r_4 \) represent absorption or gain at the corresponding probe-field frequencies. At \( G_3 = 0 \) equations (2) and (3) convert in solutions for \( \Lambda \) and \( V \) schemes

\[ r_2 = i \frac{G_2}{P_2} \frac{\Delta r_2 - g_3 \Delta r_1}{1 + g_2}, \quad r_4 = i \frac{G_4}{P_4} \frac{\Delta r_4 - g_1 \Delta r_1}{1 + g_4}. \]
Following\textsuperscript{2b,c} we can classify resonant nonlinear effects as 1) power saturation of the populations (eq. (4)); 2) strong-field induced splitting of the probe-field resonances, or ac Stark effect (denominators in eqs. (4)); and 3) nonlinear interference effect (NIEF) (second and third terms in the denominators of eqs. (2)).

4 Difference in absorption and emission spectra due to the nonlinear interference effects, amplification without inversion, resonance–enhanced refraction without absorption

Power of emitted or absorbed radiation, for example at the frequency $\omega_2$, which is proportional to $Re(-iG_2^2r_2)$, can be considered as a difference between pure emission (the term, proportional to $r_g$) and pure absorption (the rest terms in eqs.(2)). The difference in frequency-dependence of these terms, induced by the auxiliary driving field, is the origin of AWI\textsuperscript{2b}. Refractive index at $\omega_2$ is determined by $Im(-iG_2^2r_2)$ and, in general, laser-induced minimum in absorption may coincide with the resonance-enhanced maximum in refraction\textsuperscript{1,3}. Thus, laser-induced resonance splitting and NIEF transform only spectral shape of absorption/gain and refractive indices, give rise to difference in the line shapes of spontaneous (or pure induced) emission and absorption, but do not affect the integral intensity of the spectral lines\textsuperscript{2b,c}:

$$\int d\Omega_2 Re(-ir_2/G_2) = \Delta r_2, \quad \int d\Omega_4 Re(-ir_4/G_4) = \Delta r_4. \quad (5)$$

Indeed, NIEF give rise to electromagnetically induced transparency (EIT) and to AWI at the transitions $gn$ (or $ml$), when contributions of second and third terms in the denominators of eqs.(2) are equal or dominate over $\Delta r_2$ (or over $\Delta r_4$), correspondingly. From the above presented density-matrix equations one can see that the coherence at the transitions $gn$ and $ln$ ($r_{32}$ and $r_{12}$), induced in cooperation of the strong and the probe fields, is the source of the EIT and AWI effects.

A great number of elementary processes, introduced and defined for the bare states in the framework of the perturbation theory, may give contribution to the absorption/gain index $\alpha(\Omega)$. Consider, for example, $\alpha(\Omega_4)$ at the frequency $\omega_4 > \omega_1$ (Fig.1), reduced by it’s maximum value $\alpha^0(0)$ in the absence of the all strong fields, for the case when $E_3 = 0$. From the eqs.(2) one finds:

$$\frac{\alpha(\Omega_4)}{\alpha^0(0)} = Re\left(\frac{G_4}{P_4} \frac{\Delta r_4 - g_1\Delta r_1}{\Delta n_4(1 + g_4)}\right) \quad (6)$$

Consider two subcases:

\textit{a. Off resonance:} $|\Omega_1| \approx |\Omega_4| >> \Gamma_1, \Gamma_4; |g_4| << 1; |g_1| << 1; P_4 \approx i\Omega_4; P_1 \approx i\Omega_1 \approx i\Omega_4$.

Eq.(6) takes the form:

$$\frac{\alpha(\Omega_4)}{\alpha^0(0)} \approx \frac{\Gamma_4^2\Delta r_4}{\Omega_4^2\Delta n_4} - Re\left(\frac{\Gamma_4(\Delta r_4g_4 + \Delta r_1g_1)}{i\Omega_4\Delta n_4}\right) \approx \frac{\Gamma_4^2\Delta r_4}{\Omega_4^2\Delta n_4} - \frac{\Gamma_4\Gamma_1}{\Gamma_4^2 + (\Omega_4 - \Omega_1)^2} \cdot \frac{|G_1|^2(\Delta r_1 - \Delta r_4)}{\Omega_4^2\Delta n_4} =$$

$$= \frac{\Gamma_4^2(r_l - r_m)}{(n_l - n_m)\Omega_4^2} - \frac{\Gamma_{gm}\Gamma_{lm}}{\Gamma_{gm}^2 + (\Omega_4 - \Omega_1)^2} \cdot \frac{|G_1|^2(r_m - r_g)}{\Omega_4^2(n_l - n_m)} \quad (7)$$

The last terms in eqs.(7) describe Raman-like coupling and originate both from the nominator and the denominator in eq.(6). Population inversion between initial and final bare states ($r_m = n_m > r_g$) is required for amplification of the probe field.

\textit{b. Resonance:} $\Omega_1 = \Omega_4 = 0$.

Conditions for AWI and EIT are:
\[ g_1 \Delta r_1 \geq \Delta r_4, \quad \text{or} \quad \left| \frac{G_1}{\Gamma g \Gamma g_m} \right| \cdot (r_l - r_g) \geq r_l - r_m \]  

Eq.(8) shows that due to NIEF, population inversion between initial and final bare states is not required in order to attain AWI in this case. Small relaxation rate of the coherence, induced in cooperation of the driving and probe fields, compared to the other relaxation rates is the most important.

Analysis of the condition for EIT and AWI as well as of sign-changing line shape in the more details can be found in ref.\(^{2b}\) both for open and closed (l is ground state) atomic configurations. The analysis shows strong dependence of the line shape on the ratios of both population and coherence relaxation rates as well as on the ratios of initial unsaturated population differences on the coupled transitions.

4.1 Constructive and destructive interference due to the atomic velocity distribution

Furthermore, the analysis shows that the contributions of the coherence driving fields to the spectra may be both constructive and destructive, depending on the detunings of the probe as well as of the strong fields. This indicates that in gases with inhomogeneous broadening of the coupled transitions, dominating over homogeneous one, conditions for AWI and EIT may considerably differ from that for atoms at rest. Nevertheless, it was found out that under certain conditions sign-changing spectral profiles may be produced too\(^{2b,c,7a,b}\). At weak intensities of driving field narrow structure, superimposed on the Doppler background, appears. The shape of the structure is anisotropic and depends on the angle between the wave vectors of the interacting radiations. Optically-pumped unidirectional-emitting ring laser may operate by that. The line shape is also dependent on the intensity of the driving field and velocity-changing collisions. Special features may occur, when some of the coupled transitions are homogeneously, and some of them are inhomogeneously broadened. It was found out that destructive or constructive character of the effect of Maxwell’s velocity distribution depends on the fact whether a frequency of the probe field is less or greater than that of the strong one too. Analytical results describing general behavior of the velocity-averaged functions for some limiting cases, including Rabi frequencies larger then homogeneous linewidths, can be found in ref.\(^{2a,b,c,7a,b}\).

5 Coherence and nonlinear-optical conversion, Enhancements in nonlinear-optical conversion due to multiple resonance and induced transparency, Local-field effects

Nonlinear optical response of a medium experiences a giant enhancements in one- and multiphoton resonances. This reduces required fundamental powers down to cw regime\(^8\), however imposes severe limitations on the number density of the medium due to absorption of fundamental and generated radiations. As it is discussed above, in the presence of a strong electromagnetic radiation resonances for a weak probe radiation experience splitting\(^2,9\), which exhibits itself in a different ways in real and imaginary parts of linear and nonlinear susceptibilities. Later makes possible to combine decrease in absorption with increase in squared module of nonlinear susceptibilities, responsible for optical generation, and at the same time with improvements in phase-matching and increasing density of the medium\(^2f,10\).

With the increase of the atom number density, local field, acting on an atom, may pretty much differ from the external field both in the amplitude and phase. This may drastically change shape of nonlinear spectroscopic structures, including electromagnetically induced transparency\(^{11,12}\).

Consider experimental schematics, proposed in ref.\(^{13}\), that combines the advantages of both multiple resonance enhancements and increase in atom number density of nonlinear medium due to the above
induced at the transition 02 by the two combinations of strong and weak fields (field, but generated radiation, ω\textsubscript{f}) and are weak, do not change populations of the levels and are accounted for only in the lowest order of the perturbation theory. Absorption and refraction indexes for the probe fields at ω\textsubscript{1} and ω\textsubscript{s} are represented by the imaginary and real parts of

\[ \chi_1(-\omega_1;\omega_1) = (\chi_0^L/P_01)f_1, \quad \chi_s(-\omega_s;\omega_s) = (\chi_0^L/P_03)f_s \]  

Nonlinear susceptibility is:

\[ \chi_{NL}(-\omega_s;\omega_1 + \omega_2 + \omega_3) = (\chi_0^L/P_01P_02D_03)f, \]  

where \( \chi_1^L, \chi_0^L \) and \( \chi_{NL}^L \) are resonant values of the susceptibilities at negligibly small \( G_2 \) and \( G_3 \). Factors \( f_1, f_2 \) and \( f \) describe effects of the strong fields. Simple density - matrix calculations, similar to given in\textsuperscript{2b,c,10a}. yield:

\[ f_1 = \{1 + g_2/P_01P_02[1 + (g_3/P_02D_03)]\}^{-1}, \]  

\[ f_s = \{1 + g_3/P_03D_02[1 + (g_2/D_02D_01)]\}^{-1}, \]  

\[ f = f_1[1 + g_3/D_03P_02]^{-1} = [1 + (g_2/D_02D_01) + (g_3/D_03P_02)]^{-1} \]

\[ P_{01} = 1 + ix_1, \quad P_{02} = 1 + ix_0, \quad P_{03} = 1 + ix_s; \quad D_{01} = 1 + iy_1, \quad D_{02} = 1 + iy_0, \quad D_{03} = 1 + iy_s; \]  

\[ x_1 = (\omega_1 - \omega_{10})/\Gamma_{10} = 0, \quad x_0_2 = (\omega_1 + \omega_2 - \omega_{21})/\Gamma_{20} = 0, \quad x_s = (\omega_s - \omega_{30})/\Gamma_{30} = 0; \]  

\[ y_1 = (\omega_s - \omega_3 - \omega_2 - \omega_{10})/\Gamma_{10} = 0, \quad y_0_2 = (\omega_s - \omega_3 - \omega_{21})/\Gamma_{20} = 0, \quad y_s = (\omega_1 + \omega_2 + \omega_3 - \omega_{30})/\Gamma_{30} = 0; \]  

\[ g_2 = G_2^2/\Gamma_{10}\Gamma_2, \quad g_3 = G_3^2/\Gamma_{30}\Gamma_{20}. \]

\( \Gamma_{ij} \) are homogeneous halfwidth of the corresponding transitions. In the case, when \( E_s \) is not a probe field, but generated radiation, \( \omega_s = \omega_1 + \omega_2 + \omega_3 \) and \( D_{0i} = P_{0i} \).

Factors \( f_1, f_s \) and \( f \) are different and describe splitting of the corresponding resonances. Frequency-dependence and difference from unity of the factors \( f_1, f_s \) and \( f \) is determined by the coherence, induced at the transition 02 by the two combinations of strong and weak fields (\( E_1, E_2 \) and \( E_s, E_3 \)).
Generated power $P \propto g_{2g3} |\chi^{NL}|^2$, depends not only on imaginary but on real part of $\chi^{NL}$ too, and because of that may not deplete in the spectral range of induced transparency and phase-matching. Each resonance increases $|\chi^{NL}|^2$ by the factor of $x_i^{-2}$, which may be on the order of $10^6$. Laser-induce spectral structures in real parts of $\chi_1$ and $\chi_s$(dispersion caused by the coherence at the 02 transition), provide additional means to phase-match frequency - conversion by the small detunings of the fundamental radiations from the resonances. Triple resonance may yield total enhancement in generated power on the order of $10^{18}$. Due to the induced transparency, number density of the atoms $N$ and consequently $P \propto N^2$ may be increased by several orders of the magnitude in addition.

At high number density of the atoms, local fields may significantly differ from the external electromagnetic fields both in amplitudes and in phases. As it was shown in $^{12,13}$, that may drastically change spectral properties of the induced transparency as well as of the generating nonlinear polarization. Similar to $^{11,12}$, making use Lorentz-Lorenz approximation, local field effects can be accounted for by the substituting one- photon resonances on that red-shifted (by substituting $x_1$ and $x_s$ for $x_1 + C_1$ and $x_s + C_s$, $C_1 = N |d_{10}|^2 / 3\epsilon_0\Gamma_{10}$; $C_s = N |d_{30}|^2 / 3\epsilon_0\Gamma_{30}$, $\epsilon_0$ is permittivity of free space). Due to the fact that this does not influence transition frequencies between the excited states and that of the multiphoton transitions, the introduced shifts may drastically change effects of strong electromagnetic fields at $\omega_2$ and $\omega_3$ on both dressed linear and nonlinear responses.

Equations, given above, can be easily generalized on the cases of the higher order processes. For example, when 1–0 and/or 3–2, 2–1 are multiphoton transitions, generalization can be done simply by substituting one-photon Rabi frequencies and detunings for the corresponding multiphoton ones. Manipulations by the nonlinear susceptibility, absorption and refractive indexes for the generating radiation with the auxilliary strong fields, coupled to the adjacent transitions (both bound and continuum states, Figs. 2b, c.), were considered in ref.$^{2f,10}$.

### 6 Nonlinear interference effects at bound-free transitions, Laser-induced autoionizing-like resonances (laser induced continuum structure)

Nonlinear interference phenomena, similar to those at bound-bound transitions, including AWI and EIT, can occur at the transitions to ionization continuum. Appropriate theory was developed in ref. $^{2f,10a,14}$. Similar case, relevant to the zone bands in crystals, was considered in ref.$^{15}$. Laser induced autoionizing like resonances – laser induced continuum structure (LICS) was observed in the experiments ref.$^{16}$, and since the end of 80’s studies of the resonant interference processes in the context of LICS, AWI and EIT, first at bound–free and then at bound–bound transitions, have involved a number of research groups$^{17,18}$.

Potential feasibilities to manipulate both by LICS and by the splitting of the discrete resonances in order to enhance short - wavelengths frequency - mixing output and to decrease resonant absorption of the both fundamental and generated radiations can be shown with the example of Fig.2b., generalized for the case, when $\omega_1$ is close to $\omega_{10}$, and radiations at $\omega_2$, $\omega_3$ and $\omega$ are strong. The example combines opportunities to manipulate by two LICS and by depletion of absorption at the discrete transitions. Contribution of strong off - resonant $k$ levels are taken into account too. By that, the detunings $|\omega_1-\omega_{gm}|$, $|\omega_1 + \omega_2 - \omega_{gm}|$ and $|\omega - \omega_3 - \omega_{nl}|$ are assumed being much less than all the rest. Density - matrix calculations give the expressions for nonlinear susceptibility $\chi^{(3)}(\omega_{\mu} = \omega_1 + \omega_2 + \omega_3)$, which determines generated power at the frequency $\omega_{\mu}$, as well as for absorption indexes $\alpha(\omega_1)$ and $\alpha(\omega_{\mu})$ for probe radiations at corresponding frequencies as follows $^{19}$.

$$\chi^{(3)}(\omega_{\mu} = \omega_1 + \omega_2 + \omega_3)/\chi^{(3)}_{0\mu} = K/(D_{gm}X),$$  \hspace{1cm} (14)

$$\alpha(\omega_1)/\alpha_{01} = Re\{1 - g_{nm}/(D_{gm}X)]/D_{gm}\},$$  \hspace{1cm} (15)
\[ \alpha(\omega_{\mu})/\alpha_{0\mu} = 1 - k_3\beta_l + k_3\beta_l(y_l + q_{gl})^2/(1 + y_l^2) - \]
\[-Re\{k_{4gmn}A^2(1 - iq_{gn})^2/Y\} \]

where \(\chi^{(3)}_{0\mu}\), \(\alpha_{01}\) and \(\alpha_{0\mu}\) - are corresponding resonant values at the intensities of all the fields being negligibly weak. The rest parameters are as follows:

\[ K = 1 - k_1\beta_l[(1 - iq_{nl})(1 - iq_{lg})]/[(1 - iq_{ng})(1 + iy_l)], \]
\[ A = 1 - k_1\beta_l[(1 - iq_{ln})(1 - iq_{gl})]/[(1 - iq_{gn})(1 + iy_l)], \]
\[ X = (1 + g_{mn})[1 + ix_n + g_{mn}/D_{gm}(1 + g_{mn}) - k_2\beta_l\beta_n(1 - iq_{nl})^2/(1 + ix_l)], \]
\[ Y = (1 + g_{mn})[1 + iy_n + g_{mn}/p_{gm}(1 + g_{mn}) - k_2\beta_l\beta_n(1 - iq_{nl})^2/(1 + iy_l)], \]
\[ D_{gm} = 1 + i(\omega_1 - \omega_{gm})/\Gamma_{gm}, \quad p_{gm} = 1 + i(\omega_1 - \omega_3 - \omega_{gm} - \omega_{gn})/\Gamma_{gm}, \]
\[ x_l = (\omega_1 + \omega_2 + \omega_3 - \omega - \omega_{gl} - \delta_{ll})/\Gamma_{gl} + \gamma_{ll}, \quad x_n = (\omega_1 + \omega_2 - \omega_{gm} - \delta_{nn})/\Gamma_{gm} + \gamma_{nn}, \]
\[ y_l = (\omega_1 - \omega - \omega_{gl} - \delta_{ll})/\Gamma_{gl} + \gamma_{ll}, \quad y_n = (\omega_1 - \omega_3 - \omega_{gm} - \delta_{nn})/\Gamma_{gm} + \gamma_{nn}, \]
\[ k_1 = (\gamma_{ll}\gamma_{ln}/(\gamma_{gn}\gamma_{nn}), k_2 = (\gamma_{ll}\gamma_{ln}/(\gamma_{ll}\gamma_{nn}), k_3 = (\gamma_{ll}\gamma_{lg}/(\gamma_{gg}\gamma_{ll}), k_4 = (\gamma_{gg}\gamma_{ll}/(\gamma_{gg}\gamma_{nn}), \]
\[ g_{mn} = | G_{mn} |^2 / \Gamma_{gm} \Gamma_{gn}, \quad \beta_l = g_{ll}/(1 + g_{ll}), \quad \beta_n = g_{nn}/(1 + g_{nn}), \]
\[ g_{ii} = \gamma_{ii}/\Gamma_{gi}, \quad q_{ij} = \delta_{ij}/\gamma_{ij}, \quad \gamma_{ij} = \pi\hbar G_{i\mu}G_{ej}|\epsilon=\hbar\omega_{\mu} + Re\{\sum_k G_{ij}G_{kj}/p_{gk}\}, \]

\[ \delta_{ij} = \hbar P \int dG_{i\mu}G_{ej}/(\hbar\omega_{\mu} - \epsilon) + Im\{\sum_k G_{ik}G_{kj}/p_{gk}\} \]

Factors \(0 \geq k_i \geq 1\), depending on whether continuum states are not degenerate or degenerate (unity).

Comparing eqs. (14) and (16) with corresponding equations from ref. 10a,2f, one can see additional interference LICS structures in generating nonlinear polarization, absorption and refraction indexes, produced in cooperation by the \(E_3\) and \(E\) fields (terms, proportional to \(\beta_n\) and \(q_n\)), which provide with the supplementary means in absorption spectroscopy and for enhancements of generated short-wavelength radiation.

### 7 Relaxation-induced coherence processes

As it was outlined above, relaxation may influence interference processes both in negative and positive ways. Consider examples, when role of relaxation is positive.

#### 7.1 AWI due to interference in spontaneous cascade of polarizations

The features in absorption and emission spectra, discussed above, are caused by interference of contributions of probe field and combination of probe and auxiliary strong field in atomic polarization. As it was outlined above, there may be other sources of interfering intraatomic oscillations. One of the means to obtain AWI without making use of auxiliary strong fields has been suggested recently in ref. 20. The origin is interference through the correlations in spontaneous decay.

Consider four-level atomic configuration shown in Fig 3a. All four transitions are allowed. Suppose, that the transition frequency \(\omega_{mn}\) is close to \(\omega_{m_{1n}},\) and \(\omega_{m_{1m}}\) is close to \(\omega_{n_{1n}},\) that is difference \(\Delta\)

\[ \Delta = \omega_{m_{1n}} - \omega_{mn} = \omega_{m_{1m}} - \omega_{n_{1n}} \]

is small. In this case interference between considered four radiating channels is possible. It is caused by the coherence transfer due to interaction with the vacuum oscillations, besides the populations
Figure 3: Energy-level schematics for relaxation-induced coherent processes. 

**a. AWI through spontaneous cascade of polarizations.**

**b. Relaxation-induced FWM.**

decay and spontaneous emissions of photons. For the absorption index in the frequency range around $\omega_{mn}$ calculations give:

$$\alpha(\Omega) = \frac{\lambda^2}{4\pi} \left\{ N_{nm}A_{mn} \frac{\Gamma}{\Gamma^2 + \Omega^2} + N_{n1m1}A_{m1n1}[\frac{\Gamma_1}{\Gamma_1^2 + (\Omega - \Delta)^2} + \frac{KC}{\Gamma_1} f(\Omega)] \right\},$$

(29)

$$C = \sqrt{A_{m1m}A_{n1n}A_{mn}/A_{m1n1}}, \quad K = (-1)^{J_m + J_{n1}} \sqrt{2J_m + 1} \sqrt{2J_{n1} + 1} \left\{ \frac{J_m}{J_{n1}} \frac{J_n}{J_{m1}} \frac{1}{1} \right\},$$

(30)

$$f(\Omega) = \text{Re} \frac{\Gamma \Gamma_1}{(\Gamma - i\Omega)[\Gamma_1 - i(\Omega - \Delta)]} = \frac{\Gamma \Gamma_1 [\Gamma_1 - \Omega(\Omega - \Delta)]}{(\Gamma^2 + \Omega^2)[\Gamma_1^2 + (\Omega - \Delta)^2]},$$

(31)

$$N_{nm} = (2J_m + 1)(\rho_n - \rho_m), \quad N_{n1m1} = (2J_{m1} + 1)(\rho_{n1} - \rho_{m1}).$$

(32)

Here $\Omega = \omega - \omega_{mn}$, $A_{ij}$ – Einstein coefficients, $\Gamma$, $\Gamma_1$ – are line halfwidths for the interfering transitions, $J_i$ – energy level momenta, $N_{ij}$ – population differences.

The interference term is described by the function $f(\Omega)$, $\int f(\Omega) d\Omega = 0$. Coefficient $K$ is determined by the moments of four levels under consideration and may vary in the interval $-1 \leq K \leq 1$. The case $K \geq 0$ corresponds to constructive interference (enhancements in the oscillations), the case $K \leq 0$ — to destructive interference. The analysis of the lineshape eq.29 shows it sign-changing behavior. For example, at $|\Omega| \gg \Delta$

$$\alpha(\Omega) = \frac{\lambda^2}{4\pi \Omega^2} \left\{ N_{nm}A_{mn}\Gamma + N_{n1m1}A_{m1n1}(\Gamma_1 - KC) \right\},$$

(33)

According to eq.33, absorption index may occur negative (AWI), if the requirements

$$K > O, \quad (KC/\Gamma_1 - 1)N_{n1m1}A_{m1n1}\Gamma_1 > N_{nm}A_{mn}\Gamma$$

(34)

are met. When $K \leq 0$, $\Delta = 0$, the condition

$$(|K|C/\Gamma - 1)N_{n1m1}A_{m1n1}\Gamma > N_{nm}A_{mn}\Gamma_1$$

(35)

means appearance of AWI in the line center ($\Omega = 0$). Similar phenomena may occur in the spectral range of the doublet $\omega_{m1m}$, $\omega_{n1n}$. Thus, in the considered atomic configuration AWI may be provided by the correlations in the spontaneous decay without any external action.
7.2 Collision-induced four-wave mixing

Consider examples, when collisions and spontaneous relaxation, as well as external magnetic field, break destructive interference. This removes elimination of four-wave mixing process and provides with the test, selectively sensitive to the specific modes of relaxation. The experiment was carried out with He–Ne laser, \( \lambda = 1.52 \mu m \), which is resonant to \( 2s_2–2p_4 \) transition of Ne. The upper level consists of three Zeeman’s sublevel (\( J_1 = 1 \)), the lower one is singlet (\( J_0 = 0 \)). Fundamental beam consisted of two linear and orthogonal polarized components \( E_1 \) and \( E_2 \), frequency-shift \( \Delta = \omega_2 - \omega_1 \) being much less than natural transition linewidth. Intensity of the radiation at \( \omega_1 \) was much greater than that at \( \omega_2 \). Collision and magnetic field sensitive four-wave mixing output \( E_s \) at \( \omega_s = 2\omega_1 - \omega_2 = \omega_2 - 2\Delta \) and with the same polarization as \( E_2 \) was detected. Growth of the FWM signal with the increase of collision rate and strength of magnetic field was observed, that can be explained as follows.

Each field and emitting nonlinear polarization \( P_{NL}(\omega_s) \) may be represented as combination of two circular polarized components \( P_{NL}^+((\omega_s)) \) and \( P_{NL}^-(\omega_s) \). Formulae for each of these components of nonlinear polarization consist of two terms. One of them describes FWM of the radiations with one and the same polarizations in two-level subsystem, another one – FWM of the waves with opposite polarizations in three-level Zeeman’s subsystem (Fig.3b). In the schematic under consideration, it turned out, that the two contributions interfere in the distractive way and completely eliminate each other, provided by the relaxation rates of population and quadruple moment (alignment) in the upper level are equal. It is obvious that trapping of the spontaneous radiation from the upper level, anisotropic collisions, as well as external magnetic field break the counterbalance and, therefore, induce FWM output. Such dependence was observed in the experiments. External magnetic field turns the second channel into fully resonant double-V schematics.

8 Review of early theory and experiments on NIEF, AWI and related phenomena

Coherence phenomena in three-level systems were studied since discovery of masers. Feasibility to attain AWI in these systems was discussed in some of publications of that period both for microwave\(^ {21} \) and optical transitions\(^ {22} \). AWI in optical two-level systems was predicted in ref.\(^ {23} \) and first was observed in radio-frequency transitions\(^ {24,2d} \). In optical range AWI and corresponding features in refractive index were observed in ref.\(^ {2c,25} \). Studies of coherence and interference phenomena in quantum transitions is growing research area, since they are embedded in many optical processes of basic and practical importance.

9 Concluding remarks

As it was outlined, interference is basic and very general phenomenon of optical physics, which may play a crucial role in many experimental schematics of resonant nonlinear optics. Some of such schematics are shown in the Fig.4.

Fig.4a. shows upconversion of weak infrared radiation at the frequency \( \omega_2 \). Fields \( E_1 \) and \( E_3 \) are strong. Destructive interference of oscillations at the frequency \( \omega_s - \omega_3 = \omega_{ng} = \omega_1 + \omega_2 \) was shown to be one of the main process, limiting the conversion efficiency\(^ {26} \). Fig.4b. – interference of multiphoton transition and one-photon, induced by the generating radiation eliminates population of the upper level. Fig.4c. – off-resonant 7th-order seventh-harmonic generation interfere with resonant 9th-order seventh-harmonic generation, that was used for detection of the processes\(^ {26} \). Figs.4d, e. – interference of contributions of the doublet sublevels in two-photon and off-resonant one-photon transitions. Figs.4f, g. – interference of doublet sublevels in FWM.
Pressure-induced resonance was first proposed and experimentally proved in\textsuperscript{8} and later in\textsuperscript{27}. The entire analogy between the schemes 3b and 4f is seen from the formula for the driving coherence (scheme 4f)

\[
\rho_{n'n}^{(2)} \propto V_{n'g}\rho_{gn}^{(1)} + \rho_{n'g}^{(1)}V_{gn} \propto \frac{1}{\Omega_2 + i\Gamma_{n'g}} - \frac{1}{\Omega_1 - i\Gamma_{ng}} \frac{1}{\Omega + i\Gamma_{n'n}} = \\
= \frac{1}{(\Omega_2 + i\Gamma_{n'g})(\Omega_1 - i\Gamma_{ng})}[1 - i\frac{\Gamma_{nn'} - \Gamma_{n'g} - \Gamma_{ng}}{\Omega + i\Gamma_{n'n}}].
\]

(36)

Here \(\Omega_1 = \omega_1 - \omega_{ng}\), \(\Omega_2 = \omega_2 - \omega_{n'g}\), \(\Omega = \omega_2 - \omega_1 - \omega_{n'n}\). At spontaneous relaxation, \(\Gamma_{ij} = (\Gamma_i + \Gamma_j)/2\), and resonance \(\Omega = 0\) disappears. Collisions induce this resonance.

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