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Three-photon interference spectra in four-level atomic systems*

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Abstract. We have considered the interference spectra that occur at the three-photon generated frequency arising from the interaction of three laser fields with a four-level atom, where two of the laser fields are on two-photon resonance with the three levels forming a "Λ" scheme while the third laser operates between the second ground and the second excited state of the atom. At low intensities of all three laser fields, the overall intensity of the peak at the three-photon generated frequency, describing the spectrum of an electron in the second excited state, depends on the strength of the combined field of the two laser fields that are on two-photon resonance and it takes negative values. This indicates that light amplification without population inversion is likely to occur at the three-photon generated frequency. The combined field of the three laser fields induces multiphoton excitations near the three-photon generated frequency, whose peaks are characterized by linewidths which are much less than the natural linewidths of the atoms. These excitations describe absorption or stimulating emission processes depending on the values of the detunings of the laser fields. The derived results are graphically presented and discussed.

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Considerable attention has been recently shown in achieving electromagnetic induced transparency (EIT) and light amplification without population inversion (LAWI) in three- and four-level atomic systems. The attractive applications of EIT and LAWI include the possiblity of enhancing the efficiencies of nonlinear processes and the possibility of producing laser action without population inversion [1–9]. Several mechanisms giving rise to EIT and LAWI have been proposed, which are based on simple models indicating that the foundations EIT and LAWI are the atomic coherence and the destructive quantum interference among different transitions. There have been reviews on the subject by Kocharovskaya [10], by Scully [11], by Arimondo [12], by Harris [13] and by Scully and Zubairy [14]. Experimental observation of EIT has been made in the "Λ" type atomic systems [15], while several interesting experiments have demonstrated the existence of LAWI in a number of four-level atomic systems [16–21].

Schmidt and Imamoglu [22] have shown that the four-level atomic system shown in Fig. 1 exhibits EIT and yields giant resonantly enhanced nonlinearities, while the linear susceptibilities, namely, the one-photon losses, are identically zero for all participating fields. These giant Kerr optical nonlinearities are obtained in the low-intensity limit for all laser fields provided that the two-photon resonance condition is applicable [22]. The four-level atomic system depicted in Fig. 1 consists of the levels $|1\rangle$, $|2\rangle$ and $|3\rangle$, which form a "Λ" configuration and an excited level $|4\rangle$. The two lower states $|1\rangle$ and $|2\rangle$ of the atom have very long lifetimes in comparision with the lifetimes of the two excited levels $|3\rangle$ and $|4\rangle$. The atom is pumped by the laser fields a, b and c with frequencies ω_a, ω_b and ω_c and a generated frequency $\omega = \omega_a + \omega_b - \omega_c$ and operating in the $|1\rangle \leftrightarrow |3\rangle$, $|2\rangle \leftrightarrow |4\rangle$, $|2\rangle \leftrightarrow |3\rangle$ and $|1\rangle \leftrightarrow |4\rangle$ transitions, respectively. The two-photon resonance condition occurs when the frequency of the two laser fields a and c becomes equal to the frequency splitting of the two lower levels of the atom, namely, when $\omega_{21} = \omega_a - \omega_c$, where $\omega_{21} = \omega_2 - \omega_1$. In the absence of the excited level $|4\rangle$ and, of course, in the absence of the laser field b, at two-photon resonance, the excited state $|3\rangle$ is decoupled, the population is trapped in a linear superposition of the two laser levels $|1\rangle$ and $|2\rangle$ and no absorption to the excited state $|3\rangle$ will take place [12, 23–26]. As a result, the fluorescence from the excited state $|3\rangle$ decreases sharply and a narrow dip appears in the absorption spectrum known as a black resonance [12, 23–26]. The presence of the excited level $|4\rangle$ and the laser field b operating in the $|2\rangle \leftrightarrow |4\rangle$ transition changes the dynamics of the absorption spectrum drastically.

Harris and Yamamoto [27] have shown that the four-level atomic system depicted in Fig. 1 under certain conditions and at the frequency $\omega = \omega_a$ will absorb two photons but will not absorb one photon. This is due to the fact that the presence of the laser b destroys the single-photon quantum interfer-

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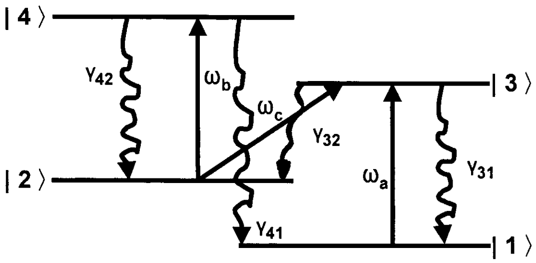

Fig. 1. Energy level diagram of a four-level atom or ion. Full lines represent laser fields operating between the levels $|1\rangle \leftrightarrow |3\rangle$, $|2\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |4\rangle$, respectively. Wavy lines represent radiative decays where $\gamma_{31}, \gamma_{32}, \gamma_{42}$ and γ_{41} denote the spontaneous decay rates for the $|3\rangle \rightarrow |1\rangle$, $|3\rangle \rightarrow |2\rangle$, $|4\rangle \rightarrow |2\rangle$ and $|4\rangle \rightarrow |1\rangle$, transitions, respectively

ence that occurs at $\omega = \omega_a$. It has been suggested [27] that such a system may function as an optical switch where a light source of one frequency will cause the absorption of light at a second frequency. Ultra-slow light pulse propagation has been recently observed in both cold and hot atoms [28–30]. Hau et al. [28] have reported the experimental observation that the speed of light pulses through a Bose–Einstein condensate is slowed to a mere 17 m/s, down by a factor of nearly 20 million from light's speed in a vacuum. The problem is based on combining EIT and cold atom technology to create a sharp excitation having a transmission linewidth, which is much less than the natural linewidth of the atoms [28]. The consequences of nonlinear optics using subnatural linewidths and ultraslow group velocities have been recently considered by Harris and Hau [31]. Experimental observations comparable to those of Hau et al. [28] have been also reported by Budker et al. [29] and by Kash et al. [30] to occur at room temperature.

In our previous work [32], the excitation spectra for an electron in the excited state $|3\rangle$ of the four-level atom depicted in Fig. 1 has been calculated. It is shown that in the low-intensity limit of all three laser fields and when the two laser fields a and c are on two-photon resonance, the laser field b operating in the $|2\rangle \leftrightarrow |4\rangle$ transition destroys the destructive interference that occurs between the spontaneous radiative decay and the process arising from the combined field of all three laser fields. The result [32] is that the overall intensity of the peak at the center of the line depends on the strength of the laser field b and it takes negative values indicating that LAWI can occur at the one-photon frequency $\omega = \omega_a$. The spectra of several induced excitations that occur at the one-photon frequency $\omega = \omega_a$ have been also considered and discussed in [32]; the reader is referred to [32] for details.

The purpose of the present study is to calculate the interference spectra of a number of excitations that occur near the generated three-photon frequency $\omega = \omega_a + \omega_b - \omega_c$ for the four-level atom depicted in Fig. 1. It is shown that at two-photon resonance of the two laser fields a and c and in the low-intensity limit of all three laser fields, the interference spectra for an electron is the excited $|4\rangle$ arising from the competition of the following processes: a spontaneous one described by the weak signal (vacuum) field that has a short lifetime, induced excitations by the combined field of all three laser fields whose lifetimes are long and determined by the strength of the laser fields in question and induced excitations arising from the combined field of the two laser fields a and c, which have short lifetimes. The induced excitations by the combined field of the three laser fields form a superposition of two asymmetric Lorentzian lines, which compete with the spontaneous radiative emissions $|4\rangle \rightarrow |1\rangle$ and $|4\rangle \rightarrow |2\rangle$ by the excited state $|4\rangle$ of the atom, and the frequency profiles of their peaks cancel each other out completely at the center of the line, namely, at the three-photon generated frequency $\omega = \omega_a + \omega_b - \omega_c$. The remaining intensity of the peak at $\omega = \omega_a + \omega_b - \omega_c$ depends on the strength of the combined field of the two laser fields a and c and takes negative values and, therefore, LAWI is likely to occur at the generated frequency $\omega = \omega_a + \omega_b - \omega_c$. Therefore, the presence of the combined field of the two laser fields a and c destroys the destructive interference, which occurs between the processes of spontaneous emissions and that arising from the combined field of the three laser fields and creating the appearance of LAWI at the generated frequency $\omega = \omega_a + \omega_b - \omega_c$. This case is analogous to that predicted in [32], where the laser field b creates the appearance of LAWI in the spectrum of an electron in the excited state $|3\rangle$ at the one-photon frequency $\omega = \omega_a$.

The excitation spectra for the state described by the operator $\Phi_{bc}(t)$ defined by (2), which represents the physical process arising from the symmetric linear superposition of the excited state $|4\rangle$ and the two-photon Raman process, where the excited state $|3\rangle$ of the atom absorbs a laser photon b and emits simultaneously a laser photon c, have been found to be similar to those describing the excitation spectra for the electron in the excited state $|4\rangle$.

The combined field of the three laser fields is found to induce the following physical processes at the generated frequency $\omega = \omega_a + \omega_b - \omega_c$: (i) the absorption of a laser photon b by the second ground state $|2\rangle$ of the atom; (ii) the three-photon process where two laser photons a and b are absorbed while a laser photon c is emitted simultaneously by the ground state $|1\rangle$ of the atom; and (iii) the antisymmetric linear superposition of the processes (i) and (ii), the state of which is defined by the operator $\Psi_{abc}(t)$ given by (41). It is shown that the spectral functions for the three processes in question describe induced absorption processes when all the laser fields are at resonance with the corresponding transitions, while at finite detunings they represent absorbing states or states exhibiting LAWI near the frequency $\omega = \omega_a + \omega_b - \omega_c$, depending on the values of the frequency detunings involved. The characteristic property of the induced excitations by the combined field of all three laser fields is that their linewidths are much less than the natural linewidths of the excited states of the atom.

The problem is formulated in Sect. 1, where the model Hamiltonian for the system under investigation is developed. The equations of motion for the required Green functions are derived in Sect. 2, where all the three laser fields have been treated as classical entities while the radiation (vacuum) field has been quantized. In Sects. 3 and 4, the expressions for the relevant Green functions have been obtained when the two laser fields a and c are on two-photon resonance and when the low-intensity limit for all three laser fields is applicable. The spectral functions describing the interference spectra under investigation have been derived in Sect. 5 while the spectra for the antisymmetric three-photon state have been considered in Sect. 6. The computed spectra are graphically presented and discussed. A brief summary of the derived results is given in Sect. 7.

1 The model Hamiltonian

The model Hamiltonian for the atomic system depicted in Fig. 1 in the electric dipole and rotating wave approximation may be taken as

$$\begin{aligned}H &= \omega_{21}\alpha_2{}^{\dagger}\alpha_2 + \omega_{31}\alpha_3{}^{\dagger}\alpha_3 + \omega_{41}\alpha_4{}^{\dagger}\alpha_4 \\ &+ \frac{ig_{\mathrm{bc}}}{2}\left[\alpha_2{}^{\dagger}\exp\left(-i\omega_{\mathrm{b}}t\right)\Phi_{\mathrm{bc}}(t) - \Phi_{\mathrm{bc}}{}^{\dagger}(t)\alpha_2\exp\left(i\omega_{\mathrm{b}}t\right)\right] \\ &+ \frac{ig_{\mathrm{a}}}{2}\left[\alpha_1{}^{\dagger}\alpha_3\exp\left(-i\omega_{\mathrm{a}}t\right) - \alpha_3{}^{\dagger}\alpha_1\exp\left(i\omega_{\mathrm{a}}t\right)\right] \\ &+ \sum_{\boldsymbol{k},\lambda} ck\beta_{k\lambda}{}^{\dagger}\beta_{k\lambda} + \frac{1}{2}i\omega_{\mathrm{p}} \\ &\times \sum_{\substack{\boldsymbol{k},\lambda \\ i,j}}\left[f_{ij}(k,\lambda)\frac{\omega_{ji}}{ck}\right]^{1/2}\left(\alpha_i{}^{\dagger}\alpha_j\beta_{k\lambda}{}^{\dagger} - \alpha_j{}^{\dagger}\alpha_i\beta_{k\lambda}\right), \end{aligned} \tag{1}$$

where

$$\Phi_{\mathrm{bc}}(t) = \left[g_{\mathrm{b}}\alpha_4 + g_{\mathrm{c}}\alpha_3\exp\left(i\omega_{\mathrm{bc}}t\right)\right]/g_{\mathrm{bc}}, \quad \omega_{\mathrm{bc}} = \omega_{\mathrm{b}} - \omega_{\mathrm{c}} \tag{2}$$

with $g_{\mathrm{bc}}^2 = g_{\mathrm{b}}^2 + g_{\mathrm{c}}^2$ and α_i, ${}^{\dagger}\alpha_i$ are the Fermi–Dirac creation and annihilation operators describing the electronic states $i = 1, 2, 3$ and 4 and $\eta_i = \alpha_i{}^{\dagger}\alpha_i$ is the number operator of the state $|i\rangle$. The functions $f_{ij}(k,\lambda)$ are the oscillator strengths for the atomic transitions $|i\rangle \leftrightarrow |j\rangle$, ω_{p} is the atomic plasma frequency defined as $\omega_{\mathrm{p}}^2 = 4\pi\mathrm{e}^2/mV$, where $-\mathrm{e}$ and m are the charge and the mass of the electron, and V is the volume of the sample container. The functions g_{a}, g_{b}, and g_{c} are the classical Rabi frequencies of the laser fields a, b and c, defined as $g_{\mathrm{a}} = \boldsymbol{P}_{31}\cdot\boldsymbol{E}_{\mathrm{a}}$, $g_{\mathrm{b}} = \boldsymbol{P}_{24}\cdot\boldsymbol{E}_{\mathrm{b}}$ and $g_{\mathrm{c}} = \boldsymbol{P}_{32}\cdot\boldsymbol{E}_{\mathrm{c}}$, respectively, where $\boldsymbol{E}_{\mathrm{a}}$, $\boldsymbol{E}_{\mathrm{b}}$ and $\boldsymbol{E}_{\mathrm{c}}$ are the strengths of the laser fields a, b and c, while $\boldsymbol{P}_{31}$, $\boldsymbol{P}_{24}$ and $\boldsymbol{P}_{32}$ designate the transition dipole moments; units with $\hbar = 1$ are used throughout. The frequency modes ω_{a}, ω_{b} and ω_{c} are defined as $\omega_{\mathrm{a}} = \omega_{31} + \Delta_{\mathrm{a}}$, $\omega_{\mathrm{b}} = \omega_{42} + \Delta_{\mathrm{b}}$ and $\omega_{\mathrm{c}} = \omega_{32} + \Delta_{\mathrm{c}}$ where $\omega_{31} = \omega_3 - \omega_1$, $\omega_{42} = \omega_4 - \omega_2$ and $\omega_{32} = \omega_3 - \omega_2$ are the transition frequencies while Δ_{a}, Δ_{b} and Δ_{c} are the detunings of the laser fields a, b and c, which are coupled to the electronic allowed transitions $|3\rangle \leftrightarrow |1\rangle$, $|4\rangle \leftrightarrow |2\rangle$ and $|2\rangle \leftrightarrow |3\rangle$, respectively. The excited state $|2\rangle$ is a metastable one having a very long lifetime. The creation and annihilation operators $\beta_{k\lambda}{}^{\dagger}$ and $\beta_{k\lambda}$, respectively, describe the vacuum electromagnetic field, which is quantized with wavevector $\boldsymbol{k}$, frequency ck and transverse polarization $\lambda = 1, 2$. Since the state $|2\rangle$ is a metastable one, the transition $|1\rangle \leftrightarrow |2\rangle$ is electric dipole forbidden and is excluded from the last term on the right-hand side (rhs) of (1).

In writing (1) we have taken into consideration the relation $\eta_1 + \eta_2 + \eta_3 + \eta_4 = 1$.

The first three terms on the rhs of (1) describe the free atomic fields, while the fourth and fifth terms designate the interaction of the atom with the three laser fields, respectively, where the three laser fields have been treated classically. The last two terms denote the free vacuum (signal) field and its interaction with the atomic levels, respectively. The two-photon operator $\Phi_{\mathrm{bc}}(t)$ defined by (2) satisfies Fermi–Dirac statistics.

The spectral function describing the excitation spectra for an electron in the excited state $|4\rangle$ is determined by [33] the imaginary part of the Fourier transform of the single-electron Green function $G_4(\omega) = \langle\langle\alpha_4;\alpha_4{}^{\dagger}\rangle\rangle$. Our task is to calculate the Green function $G_4(\omega)$ by means of the Hamiltonian (1), and then we shall take the imaginary part of the expression $G_4(\omega)$ to derive the corresponding spectral function, which describes the required excitation spectra. In deriving the expression for $G_4(\omega)$ we shall treat the three laser fields in the Hamiltonian (1) on the same footing and then the appropriate limits will be taken to obtain the required specific solutions from the derived general expression. We shall make use of the Green function formalism, which has been described in detail by many authors [33].

2 Equations of motion for the Green functions

Using the Hamiltonian (1) we derive the following equations of motion [33] for the Green function $G_4(\omega)$

$$\begin{aligned}G_4(\omega) &= \frac{1}{2\pi d_4} - \frac{ig_{\mathrm{b}}}{2d_4}\left\langle\!\left\langle\alpha_2\exp\left(i\omega_{\mathrm{b}}t\right);\alpha_4{}^{\dagger}\right\rangle\!\right\rangle \\ &= \frac{1}{2\pi d_4} + \frac{g_{\mathrm{b}}^2}{4d_4^2}G_{2\mathrm{b}}(\omega),\end{aligned} \tag{3}$$

where

$$\begin{aligned}&d_4 = \omega - \omega_{41} + \frac{i\gamma_4}{2} = x + \Delta_{\mathrm{b}} + \Delta_{\mathrm{c}} - \Delta_{\mathrm{a}} + \frac{i\gamma_4}{2}, \\ &x = \omega - \omega_{\mathrm{ac}}{}^{\dagger}{}_{\mathrm{b}} = \omega - \omega_{\mathrm{a}} - \omega_{\mathrm{b}} + \omega_{\mathrm{c}}, \qquad \gamma_4 = \gamma_{41} + \gamma_{42}, \\ &G_{2\mathrm{b}}(\omega) = \left\langle\!\left\langle\alpha_2\exp\left(i\omega_{\mathrm{b}}t\right);\alpha_2{}^{\dagger}\left(-i\omega_{\mathrm{b}}t'\right)\right\rangle\!\right\rangle, \\ &\gamma_{41} = \frac{4}{3}\left(\omega_{41}/c\right)^3|\boldsymbol{P}_{41}|^2, \qquad \gamma_{42} = \frac{4}{3}\left(\omega_{42}/c\right)^3|\boldsymbol{P}_{42}|^2.\end{aligned}$$

The functions γ_{41} and γ_{42} denote the spontaneous transition probabilities for the radiative decays $|4\rangle \to |1\rangle$ and $|4\rangle \to |2\rangle$, respectively while $2/\gamma_4 = 2/(\gamma_{41} + \gamma_{42})$ defines the radiative lifetime of the excited state $|4\rangle$. The Green function $G_{2\mathrm{b}}(\omega)$ describes the physical process where one photon of the laser field b is absorbed by the ground state $|2\rangle$ of the atom. The equation of motion for the Green function $G_{2\mathrm{b}}(\omega)$ turns out to be

$$\begin{aligned}G_{2\mathrm{b}}(\omega) &= \frac{1}{2d_{2\mathrm{b}}} + \frac{ig_{\mathrm{bc}}}{2d_{2\mathrm{b}}}\left\langle\!\left\langle\Phi_{\mathrm{bc}}(t);\alpha_2{}^{\dagger}\exp\left(-i\omega_{\mathrm{b}}t'\right)\right\rangle\!\right\rangle \\ &= \frac{1}{2d_{2\mathrm{b}}} + \frac{g_{\mathrm{bc}}^2}{4d_{2\mathrm{b}}^2}\Phi_{\mathrm{bc}}(\omega),\end{aligned} \tag{4}$$

where the operator $\Phi_{\mathrm{bc}}(t)$ is defined by (2), the propagator $d_{2\mathrm{b}} = \omega - \omega_{21} - \omega_{\mathrm{b}} = x + \Delta_{\mathrm{a}} - \Delta_{\mathrm{c}}$ and $\Phi_{\mathrm{bc}}(\omega)$ denotes the Green function $\Phi_{\mathrm{bc}}(\omega) = \langle\langle\Phi_{\mathrm{bc}}(t);\Phi^{\dagger}{}_{\mathrm{bc}}(t')\rangle\rangle$. The Green function $\Phi_{\mathrm{bc}}(\omega)$ represents the excitation arising from the linear superposition of the excited state $|4\rangle$ and the state describing the two-photon Raman process where a laser photon b is absorbed while a laser photon c is emitted simultaneously by the excited state $|3\rangle$ of the atom.

The equation of motion for the Green function $\Phi_{\mathrm{bc}}(\omega) = \langle\langle\Phi_{\mathrm{bc}}(t);\Phi^{\dagger}{}_{\mathrm{bc}}(t')\rangle\rangle$ turns out to be

$$\begin{aligned}\Phi_{\mathrm{bc}}(\omega) &= \frac{R_{\mathrm{bc}}}{2\pi g_{\mathrm{bc}}^2} - \frac{iR_{\mathrm{bc}}}{2g_{\mathrm{bc}}}\left\langle\!\left\langle\alpha_2\exp\left(i\omega_{\mathrm{b}}t\right);\Phi^{\dagger}{}_{\mathrm{bc}}\left(t'\right)\right\rangle\!\right\rangle \\ &- \frac{g_{\mathrm{a}}g_{\mathrm{c}}}{2g_{\mathrm{bc}}d_{3\mathrm{c}}{}^{\dagger}{}_{\mathrm{b}}}\left\langle\!\left\langle\alpha_1\exp\left(i\omega_{\mathrm{ac}}{}^{\dagger}{}_{\mathrm{b}}t\right);\Phi^{\dagger}{}_{\mathrm{bc}}\left(t'\right)\right\rangle\!\right\rangle\end{aligned} \tag{5}$$

where

$$R_{bc} = g_b^2/d_4 + g_c^2/d_{3c^\dagger b},$$
$$\omega_{ac^\dagger b} = \omega_a + \omega_b - \omega_c,$$
$$d_{3c^\dagger b} = x + \Delta_a + i\gamma_3/2,$$
$$\gamma_3 = \gamma_{31} + \gamma_{32},$$
$$\gamma_{31} = \frac{4}{3}(\omega_{31}/c)^3 |\boldsymbol{P}_{31}|^2,$$
$$\gamma_{32} = \frac{4}{3}(\omega_{32}/c)^3 |\boldsymbol{P}_{32}|^2 . \tag{6}$$

The functions γ_{31} and γ_{32} denote the spontaneous transition probabilities for the radiative decays $|3\rangle \to |1\rangle$ and $|3\rangle \to |2\rangle$, respectively, while $2/\gamma_3 = 2/(\gamma_{31}+\gamma_{32})$ defines the radiative lifetime of the excited state $|3\rangle$. Using the equation of motion

$$\langle\langle \alpha_2 \exp(i\omega_b t) ; \Phi^\dagger{}_{bc}(t') \rangle\rangle = \frac{ig_{bc}}{2d_{2b}} \Phi_{bc}(\omega), \tag{7}$$

we may rewrite (5) as

$$\begin{aligned}\Phi_{bc}(\omega) &= \frac{R_{bc}/2\pi g_{bc}^2}{(1-R_{bc}/4d_{2b})} - \frac{ig_a g_c/2d_{3c^\dagger b}}{g_{bc}(1-R_{bc}/4d_{2b})} \\ &\quad \times \langle\langle \alpha_1 \exp\left(i\omega_{ac^\dagger b} t\right) ; \Phi^\dagger{}_{bc}(t') \rangle\rangle \\ &= \frac{R_{bc}/2\pi g_{bc}^2}{(1-R_{bc}/4d_{2b})} + \frac{g_a^2 g_c^2/4d_{3c^\dagger b}^2}{g_{bc}^2(1-R_{bc}/4d_{2b})} G_{1ac^\dagger b}(\omega),\end{aligned} \tag{8}$$

where the Green function $G_{1ac^\dagger b}(\omega)$ is defined as

$$G_{1ac^\dagger b}(\omega) = \langle\langle \alpha_1 \exp\left(i\omega_{ac^\dagger b} t\right) ; \alpha_1^\dagger \exp\left(-i\omega_{ac^\dagger b} t'\right) \rangle\rangle .$$

The first term on the rhs of (8) represents the excitation spectra arising from the linear superposition of the excited state $|4\rangle$ and the two-photon Raman process where the excited state $|3\rangle$ of the atom absorbs a laser photon b and emits simultaneously a laser photon c. The last term designates the interference arising from the presence of the ground level $|1\rangle$ and the laser field a operating in the $|1\rangle \leftrightarrow |3\rangle$ transition, into the three-level system in the "V" configuration formed by the levels $|2\rangle$, $|4\rangle$ and $|3\rangle$ of the atom. The Green function $G_{1ac^\dagger b}(\omega)$ describes a three-photon process where two laser photons a and b are absorbed while a laser photon c is emitted simultaneously by the ground state $|1\rangle$ of the atom.

Substitution of (8) into (4) and then the derived result into (3) we obtain

$$G_{2b}(\omega) = \frac{2d_4 d_{3c^\dagger b}}{2\pi D_{bc}} + \frac{g_a^2 g_c^2 d_4^2}{16 D_{bc}^2} G_{1ac^\dagger b}(\omega), \tag{9}$$

$$G_4(\omega) = \frac{(d_{2b} d_{3c^\dagger b} - g_c^2/4)}{2\pi D_{bc}} + \frac{g_a^2 g_b^2 g_c^2}{64 D_{bc}^2} G_{1ac^\dagger b}(\omega), \tag{10}$$

where

$$D_{cb} = D_{cb}(\omega) = d_{2b} d_4 d_{3c^\dagger b} - \frac{g_b^2}{4} d_{3c^\dagger b} - \frac{g_c^2}{4} d_4. \tag{11}$$

The last terms on the rhs of (8)–(10) indicate that the three-photon process described by the Green function $G_{1ac^\dagger b}(\omega)$ creates the interference into the spectra of those represented by the Green functions $\Phi_{bc}(\omega)$, $G_{2b}(\omega)$ and $G_4(\omega)$, respectively.

Using the Hamiltonian (1) we derive now the equation of motion for the Green function $G_{1ac^\dagger b}(\omega)$, which turns out to be

$$G_{1ac^\dagger b}(\omega) = \frac{1}{2\pi d_{c^\dagger b}} + \frac{ig_a}{2d_{c^\dagger b}} G_{3c^\dagger b, 1ac^\dagger b}(\omega), \tag{12}$$

$$G_{3c^\dagger b, 1ac^\dagger b}(\omega) = \frac{ig_a}{2d_{3c^\dagger b}} G_{1ac^\dagger b}(\omega) - \frac{ig_c}{2d_{3c^\dagger b}} G_{2b, 1ac^\dagger b}(\omega), \tag{13}$$

where use has been made of the following notation:

$$G_{3c^\dagger b, 1ac^\dagger b}(\omega) = \langle\langle \alpha_3 \exp(i\omega_{bc} t) ; \alpha_1^\dagger \exp\left(-i\omega_{ac^\dagger b} t'\right) \rangle\rangle,$$

$$G_{2b, 1ac^\dagger b}(\omega) = \langle\langle \alpha_2 \exp(i\omega_b t) ; \alpha_1^\dagger \exp\left(-i\omega_{ac^\dagger b} t'\right) \rangle\rangle,$$

$$d_{c^\dagger ab} = \omega - \omega_a - \omega_b + \omega_c = x.$$

Substitution of (13) into (12) yields

$$\begin{aligned}G_{1ac^\dagger b}(\omega) &= \left[\frac{d_{3c^\dagger b}}{2\pi} + \frac{g_a g_c}{4} G_{2b, 1ac^\dagger b}(\omega)\right] \Big/ D_a \\ &= \frac{d_{3c^\dagger b}}{2\pi D_a} + \frac{g_a^2 g_c^2}{16 D_a^2} G_{2b}(\omega),\end{aligned} \tag{14}$$

where

$$D_a = d_{3c^\dagger b} d_{c^\dagger ab} - g_a^2/4.$$

The solution of (8)–(10) and (14) yields

$$G_4(\omega) = \left(d_{2b} D_a - g_c^2 d_{c^\dagger ab}/4\right)/2\pi D_4 \tag{15}$$

$$G_{2b}(\omega) = d_4 D_a / 2\pi D_4, \tag{16}$$

$$G_{1ac^\dagger b}(\omega) = D_{bc}/2\pi D_4, \tag{17}$$

$$\begin{aligned}\Phi_{bc}(\omega) &= d_{2b}\left(g_b^2 D_a + g_c^2 d_4 d_{c^\dagger ab}\right)/ \\ &\quad 2\pi g_{bc}^2 D_4,\end{aligned} \tag{18}$$

where

$$\begin{aligned}D_4 = D_4(\omega) &= \left(d_{c^\dagger ab} d_{3c^\dagger ab} - g_a^2/4\right)\left(d_4 d_{2b} - g_b^2/4\right) \\ &\quad - g_c^2 d_4 d_{c^\dagger ab}/4.\end{aligned} \tag{19}$$

The expression (15) for the Green function $G_4(\omega)$ describes the excitation spectrum of an electron in the excited state $|4\rangle$, while the expression (16) represents the excitation spectrum for the physical process where a laser photon b is absorbed by the ground state $|2\rangle$ of the atom. The Green function $G_{1ac^\dagger b}(\omega)$ given by (17) designates the excitation spectrum of a three-photon process where two laser photons a and b are absorbed while a laser photon c is emitted simultaneously by the ground state $|1\rangle$ of the atom. The Green function $\Phi_{\rm bc}(\omega)$ given by the expression (18) denotes the excitation spectrum arising from the linear superposition of the excited state $|4\rangle$ and the two-photon Raman-type process where a laser photon b is absorbed while a laser photon c is emitted simultaneously by the excited state $|3\rangle$ of the atom. No approximations have been made in the derivation of expressions (15)–(18), where the three laser fields have been treated classically and on the same footing; hence, (15)–(18) are exact within the limits of the model Hamiltonian (1).

3 Two-photon resonance

When the two lasers a and c are tuned to the two-photon resonance, the detunings $\Delta_{\rm a}$ and $\Delta_{\rm c}$ become equal and the frequency separation between the two laser modes is equal to the splitting between the two lower states of the atom, i.e., $\omega_{21}=\omega_{\rm a}-\omega_{\rm c}$. At this limit, the propagators $d_{2\rm b}$ and $d_{\rm c}{}^{\dagger}{}_{\rm ab}$ become equal to $d_{2\rm b}=d_{\rm c}{}^{\dagger}{}_{\rm ab}=x$ while $d_4=x+\Delta_{\rm b}+i\gamma_4/2$ and $d_{3\rm c}{}^{\dagger}{}_{\rm ab}=x+\Delta_{\rm a}+i\gamma_3/2$ in the expressions (15)–(19).

4 The low-intensity limit

The low-intensity limit for all the three laser fields occurs when $\gamma_{31}^2\gg g_{\rm a}^2$, $\gamma_{32}^2\gg g_{\rm c}^2$, $\gamma_{42}^2\gg g_{\rm b}^2$ and $\gamma_{41}^2\gg g_{\rm b}^2$. At the low-intensity limit and at two-photon resonance, the function $D_4(\omega)$ defined by (19) for $\Delta_{\rm a}=\Delta_{\rm c}$ factorizes into

$$D_4(\omega)=D_4=(x+\Delta_{\rm b}+i\gamma_4/2)\,(x+\Delta_{\rm a}+i\gamma_3/2)\times(x+i\gamma_+/4)\,(x+i\gamma_-/4)\,, \tag{20}$$

where

$$\gamma_\pm=\gamma_{\rm abc}\mp\left(\gamma_{\rm abc}^2-4\gamma_{\rm a}\gamma_{\rm b}/\lambda_{\rm a}\lambda_{\rm b}\right)^{1/2},\ \gamma_++\gamma_-=2\gamma_{\rm abc},\quad \gamma_+\gamma_-=4\gamma_{\rm a}\gamma_{\rm b}/\lambda_{\rm a}\lambda_{\rm b}, \tag{21}$$

and the following definitions have been used: $\gamma_{\rm abc}=\gamma_{\rm ac}/\lambda_{\rm a}+\gamma_{\rm b}/\lambda_{\rm b}$, $\gamma_{\rm ac}=\gamma_{\rm a}+\gamma_{\rm c}$, $\gamma_{\rm a}=g_{\rm a}^2/\gamma_3$, $\gamma_{\rm b}=g_{\rm b}^2/\gamma_4$, $\gamma_{\rm c}=g_{\rm c}^2/\gamma_3$, $\lambda_{\rm a}=1+4n_{\rm a}^2$, $\lambda_{\rm b}=1+4n_{\rm b}^2$, $n_{\rm a}=\Delta_{\rm a}/\gamma_3$ and $n_{\rm b}=\Delta_{\rm b}/\gamma_4$.

Substituting (20) into (15)–(18), and after expanding the numerators in power series at the roots of the corresponding denominators, and retaining only the first nonvanishing terms of the order $g_{\rm ac}^2/\gamma_3^2$ and $g_{\rm b}^2/\gamma_4^2$, we obtain

$$G_4(\omega)=\frac{1}{2\pi}\times\left[\frac{1-\mu_{\rm ac}\,(v_{\rm ba}+2im_{\rm ba})}{x+\Delta_{\rm b}+i\gamma_4/2}+\left(\frac{\gamma_{\rm ac}}{\gamma_{34}\lambda_{\rm a}\lambda_{\rm ab}}\right)\frac{(v_{\rm ba}+2im_{\rm ab})}{x+\Delta_{\rm a}+i\gamma_3/2}\right]-\frac{1}{4\pi\gamma_4\lambda_{\rm b}(\gamma_+-\gamma_-)}\times\left[\gamma_+\left(\frac{\mu_++2iv_+}{x+i\gamma_+/4}\right)-\gamma_-\left(\frac{\mu_-+2iv_-}{x+i\gamma_-/4}\right)\right], \tag{22}$$

$$G_{2\rm b}(\omega)=\frac{1}{2\pi(\gamma_+-\gamma_-)}\times\left[\frac{\gamma_+-2\gamma_{\rm a}(1+2in_{\rm a})/\lambda_{\rm a}}{x+i\gamma_+/4}-\frac{\gamma_--2\gamma_{\rm a}(1+2in_{\rm a})/\lambda_{\rm a}}{x+i\gamma_-/4}\right], \tag{23}$$

$$G_{1\rm ac}{}^{\dagger}{}_{\rm b}(\omega)=\frac{1}{2\pi(\gamma_+-\gamma_-)}\left[\frac{\gamma_+-2\mu-4is}{x+i\gamma_+/4}-\frac{\gamma_--2\mu-4is}{x+i\gamma_-/4}\right], \tag{24}$$

$$\Phi_{\rm bc}(\omega)=\frac{g_{\rm b}^2}{2\pi g_{\rm bc}^2}\left[\frac{1-\mu_{\rm a}\,(v_{\rm ba}+2im_{\rm ba})}{x+\Delta_{\rm b}+i\gamma_4/2}+\frac{g_{\rm c}^2/g_{\rm b}^2+\gamma_{\rm a}(v_{\rm ab}+2im_{\rm ab})/\gamma_{34}\lambda_{\rm a}\lambda_{\rm ab}}{x+\Delta_{\rm a}+i\gamma_3/2}\right]-\frac{1}{8\pi g_{\rm bc}^2(\gamma_+-\gamma_-)}\left[\gamma_+^2\left(\frac{2\mu-v\gamma_-+4i(s-m\gamma_-/2)}{x+i\gamma_+/4}\right)-\gamma_-^2\left(\frac{2\mu-v\gamma_++4i(s-m\gamma_-/2)}{x+i\gamma_-/4}\right)\right], \tag{25}$$

where use has been made of the following notation: $\mu_{\rm ac}=\gamma_3\gamma_{\rm ac}/\gamma_4\gamma_{34}\lambda_{\rm b}\lambda_{\rm ab}$, $v_{\rm ab}=1-4n_{\rm a}n_{\rm ab}$, $m_{\rm ab}=n_{\rm a}+n_{\rm ab}$, $v_{\rm ba}=1-4n_{\rm b}n_{\rm ab}$, $m_{\rm ba}=n_{\rm b}+n_{\rm ab}$, $n_{\rm ab}=\Delta_{\rm ab}/\gamma_{34}$, $\Delta_{\rm ab}=\Delta_{\rm a}-\Delta_{\rm b}$, $\gamma_{34}=\gamma_3-\gamma_4$, $\lambda_{\rm ab}=1+4n_{\rm ab}^2$, $v=1-4n_{\rm a}n_{\rm b}$, $m=n_{\rm a}+n_{\rm b}$, $\mu=\gamma_{\rm b}/\lambda_{\rm b}+\gamma_{\rm c}/\lambda_{\rm a}$, $s=n_{\rm b}\gamma_{\rm b}/\lambda_{\rm b}+n_{\rm a}\gamma_{\rm c}/\lambda_{\rm a}$, $\mu_\pm=\gamma_\pm-2m\gamma_{\rm ac}/\lambda_{\rm a}$, $v_\pm=\gamma_\pm n_{\rm b}-2m\gamma_{\rm ac}/\lambda_{\rm a}$ and $\mu_{\rm a}=\gamma_{\rm a}\gamma_3/\gamma_4\gamma_{34}\lambda_{\rm b}\lambda_{\rm ab}$. The expressions (22)–(25) are valid in the low-intensity limit for the three laser fields and when the two-photon resonance condition $\omega_{21}=\omega_{\rm a}-\omega_{\rm b}$ is applicable, namely when $\Delta_{\rm a}=\Delta_{\rm c}$.

5 Spectral functions

Taking the imaginary part of (22)–(25) we obtain

$$I_4(\omega)=\frac{2}{\gamma_4}\frac{(1-\mu_{\rm ac}v_{\rm ba})\gamma_4^2/4+(x+\Delta_{\rm b})\mu_{\rm ac}m_{\rm ba}\gamma_4}{(x+\Delta_{\rm b})^2+\gamma_4^2/4}+\frac{2\gamma_{\rm ac}}{\gamma_3\gamma_{34}\lambda_{\rm a}\lambda_{\rm ab}}\frac{v_{\rm ab}\gamma_3^2/4-(x+\Delta_{\rm a})m_{\rm ab}\gamma_3}{(x+\Delta_{\rm a})^2+\gamma_3^2/4}-\frac{2}{\gamma_4(\gamma_+-\gamma_-)\lambda_{\rm b}}\left[\frac{\mu_+\gamma_+^2/16+xv_+\gamma_+/2}{x^2+\gamma_+^2/16}-\frac{\mu_-\gamma_-^2/16-xv_-\gamma_-/2}{x^2+\gamma_-^2/16}\right], \tag{26}$$

$$I_{2\rm b}(\omega)=\frac{4}{\gamma_+-\gamma_-}\left[\frac{(1-2\gamma_{\rm a}/\lambda_{\rm a}\gamma_+)\gamma_+^2/16+x\gamma_{\rm a}n_{\rm a}/\lambda_{\rm a}}{x^2+\gamma_+^2/16}-\frac{(1-2\gamma_{\rm a}/\lambda_{\rm a}\gamma_-)\gamma_-^2/16+x\gamma_{\rm a}n_{\rm a}/\lambda_{\rm a}}{x^2+\gamma_-^2/16}\right], \tag{27}$$

$$I_{1\rm ac}{}^{\dagger}{}_{\rm b}(\omega)=\frac{4}{\gamma_+-\gamma_-}\left[\frac{(1-2\mu/\gamma_+)\gamma_+^2/16+xs}{x^2+\gamma_+^2/16}-\frac{(1-2\mu/\gamma_-)\gamma_-^2/16+xs}{x^2+\gamma_-^2/16}\right], \tag{28}$$

$$I\Phi_{\rm bc}(\omega)=\frac{2}{g_{\rm bc}^2}\left\{\gamma_{\rm b}\left(\frac{(1-\mu_{\rm ac}v_{\rm ba})\gamma_4^2/4+(x+\Delta_{\rm b})\mu_{\rm a}m_{\rm ba}\gamma_4}{(x+\Delta_{\rm b})^2+\gamma_4^2/4}\right)\right.$$
$$+\frac{(\gamma_{\rm c}+\gamma_{\rm a}\gamma_{\rm b}\gamma_4 v_{\rm ab}/\gamma_3\gamma_{34}\lambda_{\rm a}\lambda_{\rm ab})\gamma_3^2/4-(x+\Delta_{\rm a})\gamma_{\rm a}\gamma_{\rm b}m_{\rm ab}\gamma_4/\gamma_3\gamma_{34}\lambda_{\rm a}\lambda_{\rm ab}}{(x+\Delta_{\rm a})^2+\gamma_3^2/4}$$
$$-\frac{1}{2(\gamma_+-\gamma)}\left[\gamma_+\left(\frac{(2\mu+v\gamma_-)\,\gamma_+^2/16+x\,(s-m\gamma_-/2)\,\gamma_+}{x^2+\gamma_+^2/16}\right)\right.$$
$$\left.\left.-\gamma_-\left(\frac{(2\mu-v\gamma_+)\,\gamma_-^2/16-x\,(s-m\gamma_+/2)\,\gamma_-}{x^2+\gamma_-^2/16}\right)\right]\right\}, \qquad (29)$$

where $I_4(\omega)=-2\pi\,\mathrm{Im}\,G_4(\omega)$, $I_{2\rm b}(\omega)=-2\pi\,\mathrm{Im}\,G_{2\rm b}(\omega)$, $I_{1\rm ac}{}^{\dagger}{}_{\rm b}(\omega)=-2\pi\,\mathrm{Im}\,G_{1\rm ac}{}^{\dagger}{}_{\rm b}(\omega)$, $I\Phi_{\rm bc}(\omega)=-2\pi\,\mathrm{Im}\,\Phi_{\rm bc}(\omega)$ and Im denotes the imaginary part of the expression in question.

The spectral function (26) describes the excitation spectrum of an electron in the excited state $|4\rangle$ when the two laser fields a and c are tuned to the two-photon resonance, $\omega_{21}=\omega_{\rm a}-\omega_{\rm c}$ and when the low-intensity limit for the three laser fields is applicable. It represents the interference spectra arising from the competition of four excitations which have different lifetimes. The first term on the rhs of (26) describes an asymmetric Lorentzian line which is peaked at the frequency $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}-\Delta_{\rm b}$ and has a spectral width equal to $\gamma_4/2$. The maximum intensity of this peak at the frequency $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}-\Delta_{\rm b}$ is equal to $2(1-\mu_{\rm ac}v_{\rm ba})/\gamma_4$, where the first term $\gamma_4/2$ denotes the radiative lifetime due to the spontaneous radiative decays $|4\rangle\to|2\rangle$ and $|4\rangle\to|1\rangle$ while the second term $-2\mu_{\rm ac}v_{\rm ba}/\gamma_4$, takes negative values, and has been induced by the combined field of the two lasers a and c that operate in the $|1\rangle\leftrightarrow|3\rangle$ and $|2\rangle\leftrightarrow|3\rangle$ transitions, respectively. The asymmetry of this peak at the frequency $\omega\neq\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}-\Delta_{\rm b}$ is also induced by the combined field of the two laser fields a and c, it is equal to $\mu_{\rm ac}v_{\rm ba}\gamma_4$ and its importance depends on the numerical values of the detuning $\Delta_{\rm a}$ and $\Delta_{\rm b}$. The second term on the rhs of (26) denotes an asymmetric Lorentzian line that peaks at the frequency $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}-\Delta_{\rm a}$ and has a spectral width equal to $\gamma_3/2$. This peak is induced by the combined field of the two lasers a and c and its maximum intensity at $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}-\Delta_{\rm a}$ is equal to $2\gamma_{\rm ac}v_{\rm ab}/\gamma_3\gamma_{34}\lambda_{\rm a}\lambda_{\rm ab}$. The asymmetry of this peak at $\omega\neq\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}-\Delta_{\rm a}$ is equal to $m_{\rm ab}\gamma_3=(n_{\rm a}+n_{\rm ab})\gamma_3$ that depends on the numerical values of the detunings $\Delta_{\rm a}$ and $\Delta_{\rm b}$.

The last two terms on the rhs of (26) designate the superposition of the two asymmetric Lorentzian lines that are peaked at the generated frequency $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}$ but have different spectral widths of the order $\gamma_+/2$ and $\gamma_-/2$, respectively. These two superposed excitations are induced by the combined field of the three lasers a, b and c when the two lasers a and c are on a two-photon resonance, $\omega_{21}=\omega_{\rm a}-\omega_{\rm c}$. At the frequency $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}$, the maximum intensity arising from the superposition of these two induced peaks is equal to $-2(\mu_+-\mu_-)/\lambda_{\rm b}\gamma_4(\gamma_+-\gamma_-)=-2/\lambda_{\rm b}\gamma_4$, which takes negative values. The asymmetry of these two peaks at $\omega\neq\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}$ depends on the numerical values of the expressions $\nu_+\gamma_+/2$ and $\nu_-\gamma_-/2$, respectively. Peaks having positive and negative maximum intensities (heights) indicate that the physical processes of absorption (attenuation) and stimulated emission (amplification), respectively, are likely to occur at the corresponding frequencies.

The total maximum intensity $I_4(\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c})$ at the generated frequency $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}$ may be determined from (26) and is equal to

$$I_4(\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c})=\frac{2}{\gamma_4}\left(\frac{1}{\lambda_{\rm b}}-\frac{1}{\lambda_{\rm b}}-\frac{\gamma_{\rm ac}}{\gamma_{34}\lambda_{\rm ab}}S_{\rm ab}\right)$$
$$=\frac{-2\gamma_{\rm ac}}{\gamma_4\gamma_{34}\lambda_{\rm ab}}S_{\rm ab}\,, \qquad (30)$$

where

$$S_{\rm ab}=\frac{\gamma_3}{\gamma_4\lambda_{\rm b}^2}\left(1-4n_{\rm b}^2-8n_{\rm b}n_{\rm ab}\right)-\frac{\gamma_4}{\gamma_3\lambda_{\rm a}^2}\left(1-4n_{\rm a}^2-8n_{\rm a}n_{\rm ab}\right)\,. \qquad (31)$$

The first two terms on the rhs of (30) imply that at $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}$, the maximum intensities of the peaks describing the spontaneous radiative decay and the excitations induced by the combined field of the three laser fields cancel each other out completely, while the remaining last term denotes the height of the peak which is induced by the combined field of the lasers a and c. In this case the combined field of the two lasers a and c destroys the coherence, which is achieved by the cancellation of the spontaneous radiative decay processes $|4\rangle\to|1\rangle$ and $|4\rangle\to|2\rangle$ and by the processes arising from the combined field of the three laser fields. The remaining net intensity depends on $\gamma_{\rm ac}$, which is the strength of the combined field of the lasers a and c.

When the three lasers are tuned at resonance with the corresponding transitions, i.e. when $\Delta_{\rm a}=\Delta_{\rm b}=\Delta_{\rm c}=0$, which implies that $\omega_{21}=\omega_{\rm a}-\omega_{\rm c}$ and $\omega_{43}=\omega_{\rm b}-\omega_{\rm c}$, where the laser fields a and c as well as those of b and c are at two-photon resonance, respectively, then (30) becomes equal to

$$I_4(\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c})=\frac{-2\gamma_{\rm ac}}{\gamma_4}\left(\frac{1}{\gamma_4}+\frac{1}{\gamma_3}\right). \qquad (32)$$

In this case the height of the peak at the frequency $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}$ takes negative values indicating that the process of stimulating emission (amplification) is likely to occur at the corresponding frequency. This behaviour is illustrated in Fig. 2, where the relative intensity $\gamma_4 I_4(\omega)$ in units of $1/\gamma_4$ is plotted versus the relative frequency $(\omega-\omega_{\rm a}-\omega_{\rm b}+\omega_{\rm c})/\gamma_4$ for $\Delta_{\rm a}=\Delta_{\rm b}=\Delta_{\rm c}=0$, $\gamma_3=\gamma_4$ and for values of $\gamma_{\rm a}=\gamma_{\rm c}=\gamma_{\rm b}=0.125\gamma_4$, $0.25\gamma_4$ and $0.5\gamma_4$, respectively. Figure 2 indicates that for values of $\gamma_{\rm ac}=0.125\gamma_4$, $0.25\gamma_4$ and $0.5\gamma_4$, the height $I_4(\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c})$ of the peak at the frequency $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}$ takes the numerical values of $-1/\gamma_4$, $-2/\gamma_4$ and $-4/\gamma_4$, respectively.

We may rewrite (32) as

$$\gamma_4 I_4(\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c})/\left(\gamma_{\rm ac}/\gamma_4\right)=-2\left(1+\gamma_4/\gamma_3\right), \qquad (33)$$

which implies that for given values of $\gamma_{\rm ac}/\gamma_4$, the maximum height of the peak at $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}$ takes always negative values and its magnitude depends on the value of the ratio γ_4/γ_3. The behaviour of the spectral function (26) as a function of the ratio γ_4/γ_3 is illustrated in Fig. 3, where the relative intensity $\gamma_4 I_4(\omega)$ in units of $1/\gamma_4$ is plotted versus the relative frequency $(\omega-\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c})/\gamma_4$ for $\Delta_{\rm a}=\Delta_{\rm b}=\Delta_{\rm c}=0$, $\gamma_{\rm a}=\gamma_{\rm b}=\gamma_{\rm c}$, $\gamma_{\rm a}/\gamma_4=0.5$ and for different values of $\gamma_4/\gamma_3=0.6$, 1, 1.5, 3, 4 and 5, respectively. It is shown in Fig. 3 that the higher the value of the ratio γ_4/γ_3, the higher is the negative value (in absolute units) of the relative intensity at the frequency $\omega=\omega_{\rm a}+\omega_{\rm b}-\omega_{\rm c}$.

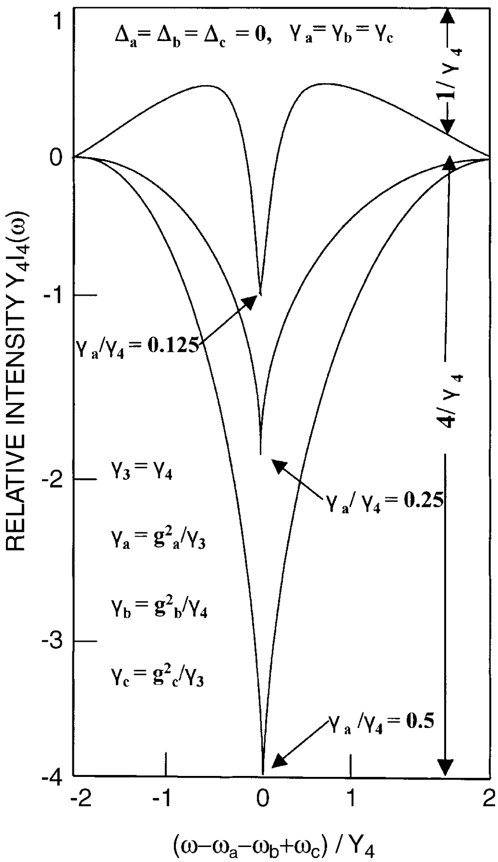

Fig. 2. Interference spectra for an electron in the excited state $|4\rangle$ in the low-intensity limit of all three laser fields and when the laser fields a and c are on two-photon resonance. The relative intensity $\gamma_4 I_4(\omega)$ computed from (26) is plotted versus the relative frequency $(\omega-\omega_a-\omega_b+\omega_c)/\gamma_4$ for $\Delta_a=\Delta_b=\Delta_c=0$, $\gamma_a=\gamma_b=\gamma_c$, $\gamma_3=\gamma_4$ and for different values of $\gamma_a=g_a^2/\gamma_4$, namely, for $\gamma_a=0.125\gamma_4$, $0.25\gamma_4$ and $0.5\gamma_4$, respectively

When $n_a=n_b$, namely when $\Delta_b/\Delta_a=\gamma_4/\gamma_3$ then (30) becomes

$$\gamma_4 I_4\left(\omega_a+\omega_b-\omega_c\right)=-2\gamma_{ac}\left(\frac{1}{\gamma_3}+\frac{1}{\gamma_4}\right)\frac{\left(1-12n_a^2\right)}{\left(1+4n_a^2\right)^3}. \quad (34)$$

The behaviour of (34) is illustrated in Fig. 4 where $\gamma_4 I_4(\omega_a+\omega_b-\omega_c)/2\gamma_{ac}(\frac{1}{\gamma_3}+\frac{1}{\gamma_4})$ is plotted versus the relative detuning $n_a=\Delta_a/\gamma_3$. It is shown in Fig. 4 that when $\Delta_b/\Delta_a=\gamma_4/\gamma_3$, the height of the peak at $\omega=\omega_a+\omega_b-\omega_c$ takes negative values for n_a in the interval $0\le n_a<0.29$, it becomes positive for n_a in the interval $0.29<n_a<2$, while it vanishes for $n_a=0.29$ and for $n_a>2$.

When the detunings Δ_a and Δ_b become equal, i.e. when $\Delta_a=\Delta$ and $\Delta_b=\Delta$, then $n_{ab}=0$, $\lambda_{ab}=1$ and (30) takes the form

$$\gamma_4 I_4\left(\omega_a+\omega_b-\omega_c\right)=\frac{2\left(\gamma_{ac}\right)}{\gamma_4}\left(1+\frac{\gamma_4}{\gamma_3}\right)\times\left[-1+\frac{48\Delta^4}{\gamma_3^2\gamma_4^2}+4\Delta^2\left(\frac{1}{\gamma_3^2}+\frac{1}{\gamma_4^2}\right)\right]\Big/\lambda_a^2\lambda_b^2. \quad (35)$$

The expression (35) is illustrated in Fig. 5, where the quantity $\gamma_4 I_4(\omega_a+\omega_b-\omega_c)/(\gamma_{ac}/\gamma_4)$ in units of (γ_{ac}/γ_4^2) is

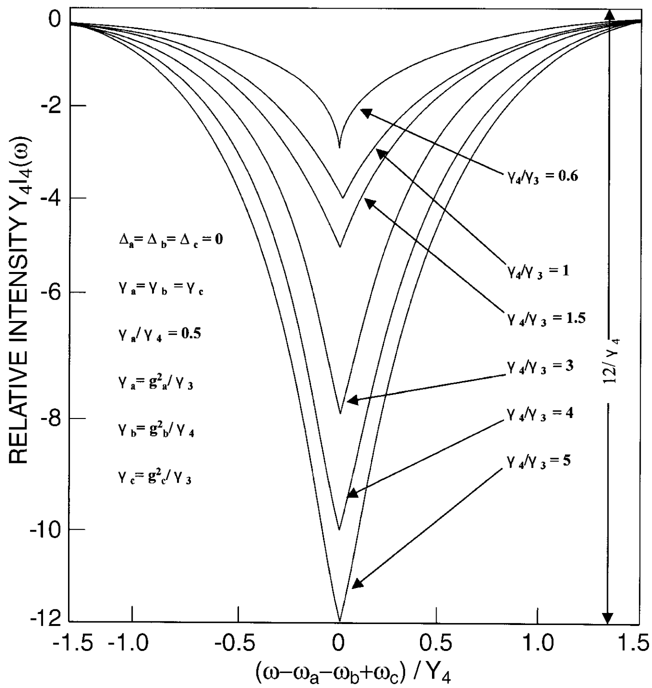

Fig. 3. As in Fig. 2 but for $\gamma_a=0.5\gamma_4$ and for different values of $\gamma_4/\gamma_3=$ 0.6, 1, 1.5, 3, 4 and 5, respectively

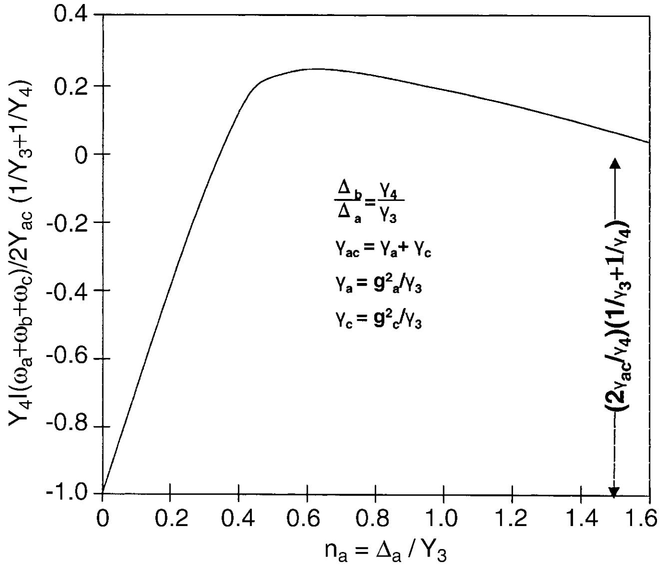

Fig. 4. Variation of the relative height $\gamma_4 I_4(\omega_a+\omega_b-\omega_c)$ of the peak at the frequency $\omega=\omega_a+\omega_b-\omega_c$. The relative intensity $\gamma_4 I_4(\omega_a+\omega_b-\omega_c)/2\gamma_{ac}(1/\gamma_3+1/\gamma_4)$ in units of $(2\gamma_{ac}/\gamma_4)(1/\gamma_3+1/\gamma_4)$ computed from (34) is plotted versus the relative detuning $n_a=\Delta_a/\gamma_3$ for $\Delta_a/\gamma_3=\Delta_b/\gamma_4$

plotted versus the relative detuning $n=\Delta/\gamma_4$ for different values of the ratio $\gamma_4/\gamma_3=0.5, 1, 6$ and 10, respectively. It is shown in Fig. 5 that the height $I_4(\omega_a+\omega_b-\omega_c)$ of the peak at the frequency $\omega=\omega_a+\omega_b-\omega_c$ takes negative values for very small values of $n=\Delta/\gamma_4$, it becomes positive only in a very narrow range of values for $n=\Delta/\gamma_4$ and then it practically vanishes for higher values than $n=\Delta/\gamma_4>0.3$.

When $\Delta_a\neq\Delta_c$ and $\Delta_{ab}^2\gg\gamma_{34}^2$, then in the limit when γ_3 takes values close to those of γ_4, i.e. when $\gamma_3\to\gamma_4$, (30) becomes equal to

$$\gamma_4 I_4(\omega_a+\omega_b-\omega_c) = \frac{4\gamma_{ac}}{\gamma_4 \lambda_a^2 \lambda_b^2} \times \left\{ \frac{8\Delta_a\Delta_b}{\gamma_4^2}\left[1+\frac{2}{\gamma_4^2}\left(\Delta_a^2+\Delta_b^2+\Delta_a\Delta_b\right)\right]-1\right\}. \quad (36)$$

In this case the height $I_4(\omega_a+\omega_b-\omega_c)$ of the peak at the frequency $\omega=\omega_a+\omega_b-\omega_c$ may take positive or negative values depending on the values of the parameters Δ_a, Δ_b and γ_4. However, when $I_4(\omega_a+\omega_b-\omega_c)$ is positive, which occurs only when the detunings Δ_a, and Δ_b become much larger than γ_4, the magnitude of the height of the peak is negligible.

The rhs of the expressions (32) and (33) for the height $I_4(\omega_a+\omega_b-\omega_c)$ are similar to those of $I_3(\omega_a)$ given by (25) and (26) in [32], respectively, and one can be obtained from the other through the interchange $g_b \rightleftharpoons g_{ac}$ and $\gamma_3 \rightleftharpoons \gamma_4$. The basic difference is that $I_4(\omega_a+\omega_b-\omega_c)$ is the height of the peak at the frequency $\omega=\omega_{41}=\omega_a+\omega_b-\omega_c$ arising from a three-photon process while $I_3(\omega_a)$ in [32] is the height of the peak at the frequency $\omega=\omega_{31}=\omega_a$, which corresponds to one-photon process.

The spectral function (27) represents the excitation spectra for the physical process where a laser photon b is absorbed by the second ground state $|2\rangle$ of the atom. It consists of the superposition of two asymmetric Lorentzian lines peaked at the same frequency $\omega=\omega_a+\omega_b-\omega_c$ but having different spectral widths of the order $\gamma_+/4$ and $\gamma_-/4$, respectively. The maximum intensity of the peak arising from the superposition of the two asymmetric lines at the generated frequency $\omega=\omega_a+\omega_b-\omega_c$ is determined from (27) and is equal to

$$I_{2b}(\omega_a+\omega_b-\omega_c) = \frac{8\gamma_a}{\gamma_+\gamma_-\lambda_a} = \frac{2\lambda_b}{\gamma_b}, \quad (37)$$

which implies that the height of the peak at frequency $\omega=\omega_a+\omega_b-\omega_c$ takes always positive values indicating that the physical process of absorption (attenuation) takes place. The behaviour of the spectral function (27) is illustrated in Fig. 6, where the relative intensity $(\gamma_b/\lambda_b)I_{2b}(\omega)$ in units of λ_b/γ_b is plotted versus the relative frequency $4(\omega-\omega_a-\omega_b+\omega_c)/\gamma_+$ for $\gamma_a/\lambda_a=\gamma_b/\lambda_b=\gamma_c/\lambda_a$ and for different values of the relative detuning $n_a=\Delta_a/\gamma_3=0, 0.5, 1, 1.5$ and 2, respectively. Inspection of Fig. 6 indicates that for $n_a\neq 0$, the relative intensity $(\gamma_b/\lambda_b)I_{2b}(\omega)$ takes negative and positive values for positive and negative values of the relative frequency $4(\omega-\omega_a-\omega_b+\omega_c)/\gamma_+$, respectively.

The spectral function (28) describes a three-photon process where two laser photons a and b are absorbed while a laser photon c is emitted simultaneously by the ground $|1\rangle$ of the atom. It consists of the superposition of two asymmetric Lorentzian lines that peak at the same frequency $\omega=\omega_a+\omega_b-\omega_c$ but they have different spectral widths equal to $\gamma_+/4$ and $\gamma_-/4$, respectively. The maximum intensity arising from the superposition of the two asymmetric lines at the generated frequency $\omega=\omega_a+\omega_b-\omega_c$ is calculated from (28) and equal to

$$I_{1ac^\dagger b}(\omega_a+\omega_b-\omega_c) = \frac{8\mu}{\gamma_+\gamma_-} = 2\left(\frac{\lambda_a}{\gamma_a}+\frac{\lambda_b\gamma_c}{\gamma_a\gamma_b}\right) \quad (38)$$

which takes always positive values indicating that the physical process of absorption takes place for the three-photon

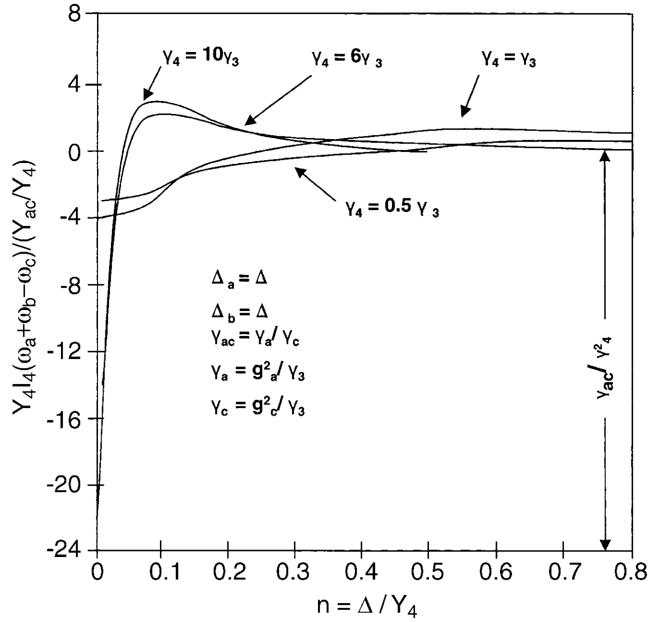

Fig. 5. As in Fig. 4. The relative intensity $\gamma_4 I_4(\omega_a+\omega_b-\omega_c)/(\gamma_{ac}/\gamma_4)$ in units of γ_{ac}/γ_4^2 computed from (35) is plotted versus the relative detuning $n=\Delta/\gamma_4$ for $\Delta_a=\Delta$, $\Delta_b=\Delta$ and for different values of the ratio $\gamma_4/\gamma_3=0.5$, 1, 6 and 10, respectively

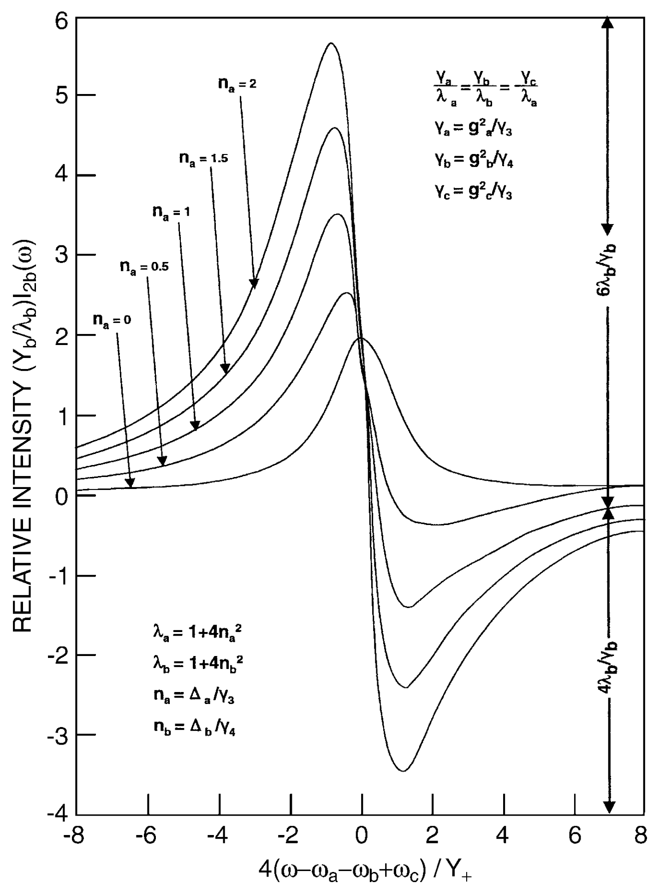

Fig. 6. Interference spectra describing the physical process where a laser photon b is absorbed by the second ground state $|2\rangle$ of the atom. The relative intensity $(\gamma_b/\lambda_b)I_{2b}(\omega)$ in units of λ_b/γ_b computed from (27) is plotted versus the relative frequency $4(\omega-\omega_a-\omega_b+\omega_c)/\gamma_+$ for $\gamma_a/\lambda_a=\gamma_b/\lambda_b=\gamma_c/\lambda_a$ for different values of the relative detuning $n_a=0, 0.5, 1, 1.5$ and 2, respectively

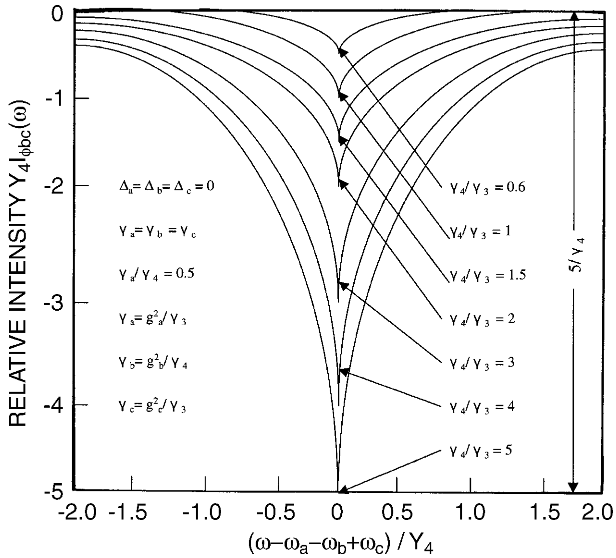

Fig. 8. Interference spectra describing the state of the operator $\Phi_{bc}(t)$, which arises from the linear superposition of the excited state $|4\rangle$ and the state describing the two-photon Raman process where a laser photon b is absorbed while a laser photon c is emitted simultaneously by the excited state $|3\rangle$ of the atom. The relative intensity $\gamma_4 I\phi_{bc}(\omega)$ in units of $1/\gamma_4$ computed from (29) is plotted versus the relative frequency $(\omega-\omega_a-\omega_b+\omega_c)/\gamma_4$ for $\gamma_a=\gamma_b=\gamma_c$, $\Delta_a=\Delta_b=\Delta_c$, $\gamma_a/\gamma_4=0.5$ and for different values of the ratio $\gamma_4/\gamma_3=0.6$, 1, 1.5, 2, 3, 4 and 5, respectively

process in question. The asymmetric behaviour of the spectral function (28) is demonstrated in Fig. 7, where the relative intensity $(\gamma_a/\lambda_a)I_{1ac}{}^{\dagger}{}_b(\omega)$ in units of λ_a/γ_a is plotted versus the relative frequency $4(\omega-\omega_a-\omega_b+\omega_c)/\gamma_+$ for $\gamma_a/\lambda_a=\gamma_b/\lambda_b=\gamma_c/\lambda_a$ and different values of the relative detuning $n_a=n_b=0$, 0.5, 1, 1.5 and 2, respectively. Figure 7 indicates that the height of the overall peak $I_{1ac}{}^{\dagger}{}_b(\omega)$ takes negative and positive values for positive and negative values of the relative frequency $4(\omega-\omega_a-\omega_b+\omega_c)/\gamma_+$, respectively.

The spectral function (29) represents the excitation spectrum arising from the linear superposition of the excited state $|4\rangle$ and the state describing the two-photon Raman process where a laser photon b is absorbed while a laser photon c is emitted simultaneously by the excited state $|3\rangle$ of the atom. The four terms on the rhs of (29) are similar in form with the corresponding ones in (26). The maximum intensity of the overall peak at the frequency $\omega=\omega_a+\omega_b-\omega_c$ is derived from (29) in the form

$$I\phi_{bc}\,(\omega_a+\omega_b-\omega_c)=\frac{2}{g_{bc}^2}\left(\frac{\gamma_b}{\lambda_b}+\frac{\gamma_c}{\lambda_a}-\frac{\gamma_b}{\lambda_b}-\frac{\gamma_c}{\lambda_a}-\frac{\gamma_a\gamma_b S_{ab}}{\gamma_{34}\lambda_{ab}}\right)$$
$$=\frac{-2\gamma_a\gamma_b}{g_{bc}^2\gamma_{34}\lambda_{ab}}S_{ab}, \tag{39}$$

where the function S_{ab} is determined by (31). The first four terms on the rhs of (39) indicate that at the frequency $\omega=\omega_a+\omega_b-\omega_c$ the contributions to the height of the peaks describing the excitations induced by the laser fields b and c are cancelled out completely by those excitations induced by the combined field of the three laser fields, while the remaining last term denotes the height of the peak arising from the cross terms corresponding to the strengths of the laser fields a and b.

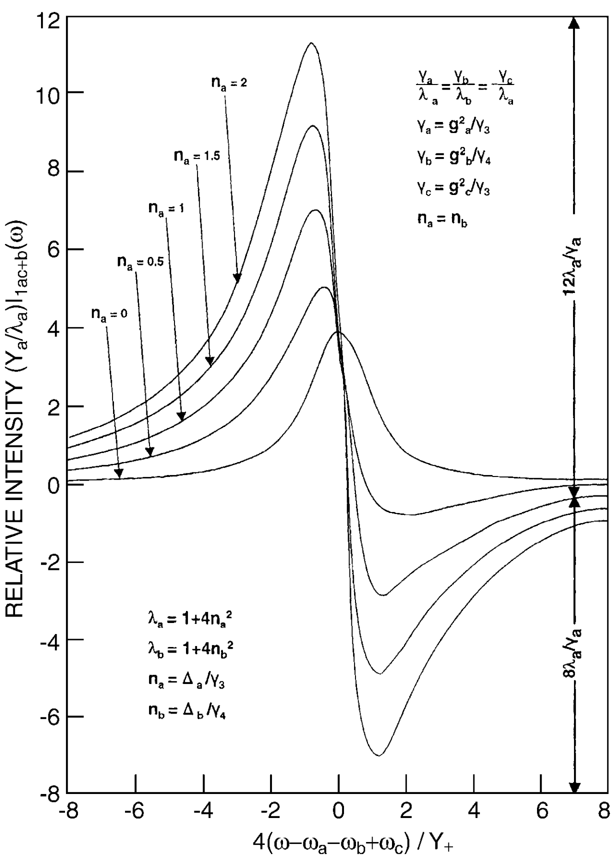

Fig. 7. Three-photon interference spectra describing the physical process where two laser photons a and b are absorbed while a laser photon c is emitted simultaneously by the ground state $|1\rangle$ of the atom. The relative intensity $(\gamma_a/\lambda_a)I_{1ac\dagger b}(\omega)$ in units of λ_a/γ_a is plotted versus the relative frequency $4(\omega-\omega_a-\omega_b+\omega_c)/\gamma_+$ for $\gamma_a/\lambda_a=\gamma_b/\lambda_b=\gamma_c/\lambda_a$, $n_a=n_b$ and for different values of the relative detuning $n_a=0$, 0.5, 1, 1.5 and 2, respectively

When the three laser fields are tuned at resonance with the corresponding transitions, namely when $\Delta_a=\Delta_b=\Delta_c=0$, then (39) is reduced to

$$I\phi_{bc}\,(\omega_a+\omega_b-\omega_c)=\frac{-2\gamma_a\gamma_b}{g_{bc}^2}\left(\frac{1}{\gamma_3}+\frac{1}{\gamma_4}\right), \tag{40}$$

which implies that the height of the peak at the frequency $\omega=\omega_a+\omega_b-\omega_c$ takes always negative values indicating that the process of stimulating emission will occur at the corresponding frequency. The behaviour of the spectral function (29) is illustrated in Fig. 8, where the relative intensity $\gamma_4 I\phi_{bc}(\omega)$ is plotted in units of $1/\gamma_4$ versus the relative frequency $(\omega-\omega_a-\omega_b+\omega_c)/\gamma_4$ for $\Delta_a=\Delta_b=\Delta_c=0$, $\gamma_a=\gamma_b=\gamma_c$, $\gamma_a/\gamma_4=0.5$ and for different values of $\gamma_4/\gamma_3=0.6, 1, 1.5, 3, 4$ and 5, respectively. Figure 8 is similar to Fig. 3 and implies that the relative intensity $I\phi_{bc}(\omega)$ at the frequency $\omega=\omega_a+\omega_b-\omega_c$ takes negative values which are higher in absolute units the higher the values of the ratio γ_4/γ_3.

6 Antisymmetric three-photon state

We introduce the antisymmetric three-photon operator $\Psi_{abc}(t)$ as

$$\Psi_{abc}(t)=[g_c\alpha_1\exp i(\omega_a+\omega_b-\omega_c)t-g_a\alpha_2\exp(i\omega_b t)]/g_{ac}, \tag{41}$$

which satisfies Fermi–Dirac statistics and $g_{ac}^2 = g_a^2 + g_c^2$. Using the Hamiltonian (1), we derive the equation of motion for the Green function $\Psi(\omega) = \langle\langle \Psi_{abc}(t); \Psi_{abc}{}^{\dagger}(t') \rangle\rangle$ in the form

$$\Psi(\omega) = \left(\frac{g_a^2}{d_{2b}} + \frac{g_c^2}{d_c{}^{\dagger}{}_{ab}} \right) / 2\pi g_{ac}^2 - \frac{i g_a g_c}{2 g_{ac}} \left(\frac{1}{d_{2b}} - \frac{1}{d_c{}^{\dagger}{}_{ab}} \right) \langle\langle \alpha_3 \exp(i\omega_{bc} t); \Psi_{abc}{}^{\dagger}(t') \rangle\rangle - \frac{i g_a g_b}{g_{ac} d_{2b}} \langle\langle \alpha_4; \Psi_{abc}{}^{\dagger}(t') \rangle\rangle \,. \tag{42}$$

At two-photon resonance, where $\omega_{21} = \omega_a - \omega_c$ and $d_{2b} = d_c{}^{\dagger}{}_{ab}$, the second term on the rhs of (42) vanishes and then considering the equation of motion of the Green function $\langle\langle \alpha_4; \Psi_{abc}{}^{\dagger}(t') \rangle\rangle$ with respect to the time argument t' we obtain

$$\Psi(\omega) = \frac{1}{2\pi d_{2b}} + \frac{g_a^2 g_b^2}{4 g_{ac}^2 d_{2b}^2} G_{4,4}(\omega) \tag{43}$$

where the expression for the Green function $G_{4,4}(\omega)$ is determined by (15) for $d_{2b} = d_c{}^{\dagger}{}_{ab} = \omega - \omega_a - \omega_b + \omega_c$. In the absence of the laser field b, i.e. when $g_b = 0$, the last term on the rhs of (43) vanishes and the result for $\Psi_{ac}(t) = [g_c \alpha_1 \exp i(\omega_a - \omega_c)t - g_a \alpha_2]/g_{ac}$ is given by

$$\langle\langle \Psi_{ac}(t); \Psi_{ac}{}^{\dagger}(t') \rangle\rangle = \frac{1}{2\pi(\omega - \omega_a + \omega_c)} \tag{44}$$

which is a stationary state. The expression (44) describes the well-known coherent nonabsorbing state for a three-level atom in the "Λ" configuration [23–25, 34].

Substituting (22) into the last term on the rhs of (43) when the low intensity limit is applicable, we obtain

$$\Psi(\omega) = \frac{1}{2\pi(\omega - \omega_a - \omega_b + \omega_c)} + \frac{2\gamma_a \gamma_b}{2\pi \lambda_b \gamma_{ac} (\gamma_+ - \gamma_-)} \times \left[\frac{(\mu_+ + 2i\nu_+)/\gamma_+}{\omega - \omega_a - \omega_b + \omega_c + 2i\gamma_+/4} - \frac{(\mu_- + 2i\nu_-)/\gamma_-}{\omega - \omega_a - \omega_b + \omega_c + i\gamma_-/4} \right]. \tag{45}$$

In deriving (45) we have retained only the last term on the rhs of (22), because the contributions arising from the first two terms of (22) to the final expressions of the intensities of the peaks in question are negligible. Taking the imaginary part of (45) we get

$$I_\Psi(\omega) = \delta(\omega - \omega_a - \omega_b + \omega_c) + \frac{8\gamma_a \gamma_b}{\lambda_b \gamma_{ac} (\gamma_+ - \gamma_-)} \times \left[\left(\frac{1}{\gamma_+^2} \right) \frac{\mu_+ \gamma_+^2/16 - (\omega - \omega_a - \omega_b + \omega_c)\nu_+ \gamma_+/2}{(\omega - \omega_a - \omega_b + \omega_c)^2 + \gamma_+^2/16} - \left(\frac{1}{\gamma_-^2} \right) \frac{\mu_- \gamma_-^2/16 - (\omega - \omega_a - \omega_b + \omega_c)\nu_- \gamma_-/2}{(\omega - \omega_a - \omega_b + \omega_c)^2 + \gamma_-^2/16} \right], \tag{46}$$

where $I_\Psi(\omega) = -2\pi \operatorname{Im} \Psi(\omega)$. The first term on the rhs of (46) describes a delta-function distribution representing the stationary part of the state at $\omega = \omega_a + \omega_b - \omega_c$ while the second term describes the spectral function of the state of question when $g_b \neq 0$ and consists of the superposition of two asymmetric Lorentzian lines which are peaked at the generated frequency $\omega = \omega_a + \omega_b - \omega_c$ and have different spectral widths of the order $\gamma_+/4$ and $\gamma_-/4$, respectively. The overall height of the peak at the generated frequency $\omega = \omega_a + \omega_b - \omega_c$ is determined from (46) and is equal to

$$I_\Psi(\omega_a + \omega_b - \omega_c) = 2\left[\nu \left(\frac{\lambda_a}{\gamma_a} + \frac{\lambda_b}{\gamma_b} + \frac{\gamma_c \lambda_b}{\gamma_a \gamma_b} \right) - \frac{\lambda_a}{\gamma_a \gamma_c} \right]. \tag{47}$$

For values of $\Delta_a = \Delta_b = \Delta_c = 0$ as well as for $n_a n_b < 1/4$, (47) implies that the height $I_\Psi(\omega_a + \omega_b - \omega_c)$ of the peak at the frequency $\omega = \omega_a + \omega_b - \omega_c$ takes positive values indicating that the physical process of absorption takes place. For values of $\Delta_a \neq 0$, $\Delta_b \neq 0$ and for $n_a n_b \geq 1/4$, the height of the peak takes negative values indicating that stimulated emission (amplification) is likely to occur at $\omega = \omega_a + \omega_b - \omega_c$.

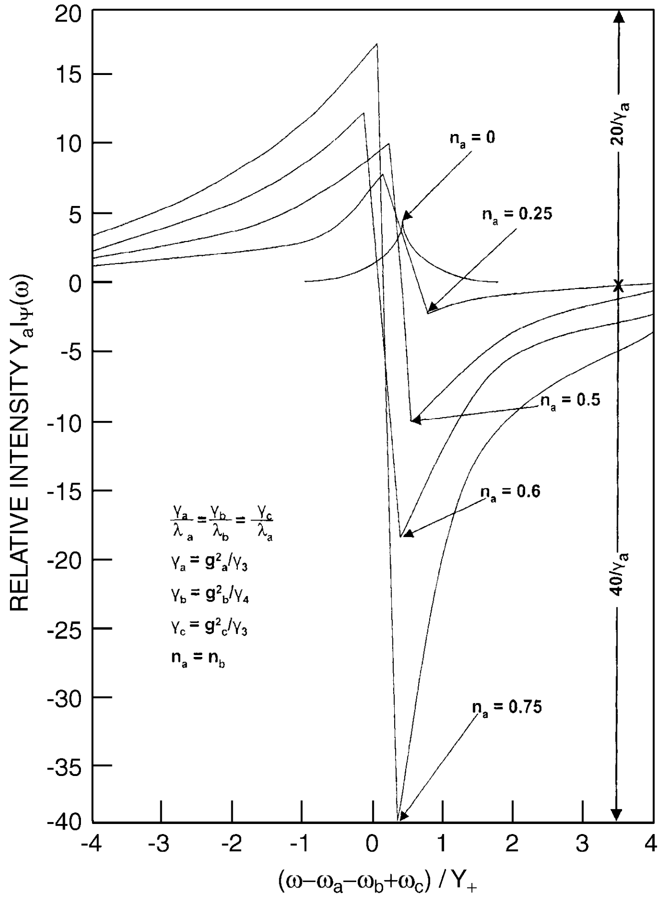

Fig. 9. Interference spectra describing the antisymmetric three-photon state $\Psi_{abc}(t)$. The relative intensity $\gamma_a I_\Psi(\omega)$ in units of $1/\gamma_a$ computed from (46) is plotted versus the relative frequency $(\omega - \omega_a - \omega_b + \omega_c)/\gamma_+$ for $\gamma_a/\lambda_a = \gamma_b/\lambda_b = \gamma_c/\lambda_a$, $n_a = n_b$ and for different values of the relative detunings $n_a = 0, 0.25, 0.5, 0.6$ and 0.75, respectively

The behaviour of the spectral function (46) is illustrated in Fig. 9 where the relative intensity $\gamma_a I_\Psi(\omega)$ in units of $1/\gamma_a$ is plotted versus the relative frequency $(\omega - \omega_a - \omega_b + \omega_c)/\gamma_+$ for $\gamma_a/\lambda_a = \gamma_b/\lambda_b = \gamma_c/\lambda_a$, $n_a = n_b$ and for different values of the detuning $n_a = 0, 0.25, 0.5, 0.6$ and 0.75, respectively. In this case the spectral widths γ_+ and γ_- are equal to $\gamma_+ = 0.763\gamma_a/\lambda_a$ and $\gamma_- = 5.24\gamma_a/\lambda_a$ and the lifetimes of the states in question are of the order of $4/\gamma_+ = 5.24\lambda_a/\gamma_a$ and $4/\gamma_- = 0.763\lambda_a/\gamma_a$ which are long in comparison to the short radiative lifetimes $2/\gamma_3$ and $2/\gamma_4$ of the atomic states $|3\rangle$ and $|4\rangle$, respectively. It is shown in Fig. 9 that for $n_a = 0$ the spectral function is a symmetric Lorenztian line with positive height while for $n_a > 0$, the spectral functions describe asymmetric Lorentzian lines where the height of the peaks takes positive and negative values for negative and positive values of the relative frequency $(\omega - \omega_a - \omega_b + \omega_c)/\gamma_+$, respectively. Figure 9 indicates that the negative values of the height of the peaks are higher in absolute units than the corresponding positive ones, the higher the values of the relative detunings $n_a = n_b$. Considering that $\gamma_a \ll \gamma_3$ and $\gamma_a \ll \gamma_4$ and that the heights of the peaks in Fig. 9 are measured in units of $1/\gamma_a$ where $1/\gamma_a \gg \gamma_3$ and $1/\gamma_a \gg \gamma_4$ are applicable, the physical process of induced absorption and stimulated emission described in Fig. 9 for different values of n_a are of fundamental importance since they are substantial in magnitude.

7 Summary and discussion

We have calculated the three-photon interference spectra for a single electron in the excited state $|4\rangle$ of the four-level atom depicted in Fig. 1. Using the Green function formalism [33] we have derived the following expressions describing the three-photon interference spectra: (i) for a single electron in the excited state $|4\rangle$ determined by (15); (ii) for the physical process where a laser photon b is absorbed by the second ground state $|2\rangle$ of the atom given by (16); (iii) for the physical process where two laser photons a and b are absorbed while a laser photon c is emitted simultaneously by the ground state $|1\rangle$ of the atom described by (17); and (iv) for the electron state $\Phi_{abc}(t)$ arising from the symmetric linear superposition of the electron state $|4\rangle$ and the two-photon Raman process where a laser photon b is absorbed while a laser photon c is emitted simultaneously by the excited state $|3\rangle$ of the atom determined by (18). An observation of the rhs of (8)–(10) indicates that the three-photon Green function defined by $G_{1ac^\dagger b}(\omega)$ causes the interference into the spectra for the three-level atom in the "V" configuration and is due to the presence of both the ground state $|1\rangle$ and of the laser field a. In the low-intensity limit of all three laser fields and when the two laser fields a and c are at two-photon resonance, then (15)–(18) are reduced to (22)–(25), respectively. At this limit, the Green function defined by (24) is due to the combined field of the three laser fields and, therefore, the interference into the spectra previously discussed is entirely induced by the presence of the combined field of the three laser fields. The spectral functions determined by (26)–(29) describe the physical processes in question and they have been derived from (22)–(25), respectively.

When the three laser fields are at resonance with the corresponding transitions and when $\gamma_a = \gamma_b = \gamma_c$, the spectral function (26) is illustrated in Figs. 2 and 3. The height of the peak at the generated frequency $\omega = \omega_a + \omega_b - \omega_c$ takes negative values as shown in Fig. 2 for $\gamma_3 = \gamma_4$ and for $\gamma_a/\gamma_4 = 0.125, 0.2$ and 0.5, as well as in Fig. 3 for $\gamma_a/\gamma_4 = 0.5$ and different values of $\gamma_4/\gamma_3 = 0.6, 1, 1.5, 3, 4$ and 5. It is indicated in Fig. 3 that for a given value of γ_a/γ_4 the higher the value of the ratio γ_4/γ_3 the higher is the negative value in absolute units of the relative intensity indicating that LAWI occurs at the generated frequency $\omega = \omega_a + \omega_b - \omega_c$. This behaviour is analogous to that predicted in [32] for the spectra of an electron in the excited state $|3\rangle$ at the one-photon frequency $\omega = \omega_a$. The height of the peak at $\omega = \omega_a + \omega_b - \omega_c$ as a function of $n_a = \Delta_a/\gamma_3$ is shown in Fig. 4, while Fig. 5 indicates the variation of the height of the peak as a function of $n = \Delta/\gamma_4$ for $\Delta_a = \Delta$, $\Delta_b = \Delta$ and for different values of the ratio γ_4/γ_3. It is shown that at finite values of the detunings and for special values of γ_4/γ_3, the height of the peak $I_4(\omega_a + \omega_b - \omega_c)$ may take zero or even positive values, which are very small and, hence, without any practical use.

The spectral functions (27) and (28) are induced entirely by the combined field of the three laser fields and they differ only in the expressions for their numerators arising from contributions of the laser field a and from the laser fields b and c, respectively. They are illustrated in Figs. 6 and 7 for $\gamma_a/\lambda_a = \gamma_b/\lambda_b = \gamma_c/\lambda_a$ and for different values of n_a and for $n_a = n_b$, respectively. It is shown in Figs. 6 and 7 that the spectral functions (27) and (28) for $n_a = 0$ and for $n_a = n_b = 0$, respectively, are represented by symmetric Lorentzian lines where the height of the peaks takes positive values describing induced absorption processes. When $n_a \neq 0$ and $n_b \neq 0$, the height of the peaks takes negative and positive values for positive and negative values of the relative frequency $(\omega - \omega_a - \omega_b + \omega_c)/\gamma_+$, respectively, implying that induced absorption and stimulated emission takes place for negative and positive values of the relative frequency $(\omega - \omega_a - \omega_b + \omega_c)/\gamma_+$, respectively. The positive and negative values in absolute units of the height of the peaks are higher, the higher the values of the detuning n_a and $n_a = n_b$. Since $\gamma_a/\lambda_a = \gamma_b/\lambda_b = \gamma_c/\lambda_a$ and $n_a = n_b$, the heights of the peaks in Fig. 7, which arise from contributions of the laser fields b and c, are larger by a factor of two than those in Fig. 6 that arise from contributions only from the laser field a.

The spectral function (29) is illustrated in Fig. 8 for $\Delta_a = \Delta_b = \Delta_c = 0$, $\gamma_a = \gamma_b = \gamma_c$, $\gamma_a/\gamma_4 = 0.5$ and for different values of γ_4/γ_3. It is shown in Fig. 8 that the height of the peaks takes negative values which are higher in absolute units the higher the values of the ratio γ_4/γ_3. Figure 8 is similar to Fig. 3 but the negative heights of the peaks in Fig. 8 are smaller in absolute units than those in Fig. 3 since from (34) and (40), we have $\gamma_4 I_4(\omega - \omega_a - \omega_b + \omega_c) = -2(1 + \gamma_4/\gamma_3)$ and $\gamma_4 I\Phi_{bc}(\omega_a + \omega_b - \omega_c) = -\gamma_4/\gamma_3$, respectively. The negative values of the intensities in Fig. 8 imply that LAWI is likely to occur at the generated frequency $\omega = \omega_a + \omega_b - \omega_c$.

The excitation spectra for the antisymmetric three-photon state $\Psi_{abc}(t)$ defined by (41) have been calculated in Sect. 6. In the absence of the laser field b and the excited state $|4\rangle$, the two-photon state $\Psi_{ac}(t)$ is a stationary state given by (44) and is the well-known coherent nonabsorbing state for a three-level atom in the "Λ" configuration [23–25, 34]. The spectral function for the three-photon state $\Psi_{abc}(t)$ is determined by (46), the first term of which represents the coherent part that is a delta-function distribution, while the last term consists of two asymmetric Lorentzian lines that are induced by the combined field of the three laser fields and peak at the same frequency $\omega = \omega_a + \omega_b - \omega_c$ but have different spectral widths of the order $\gamma_+/4$ and $\gamma_-/4$, respectively. The overall height of the peak at the generated frequency $\omega = \omega_a + \omega_b - \omega_c$ is given by (47) and is a function of the expressions γ_a, γ_b and γ_c as well as of the detunings n_a and n_b. The behaviour of the spectral function (46) is illustrated in Fig. 9 for $n_a = n_b = 0$ and for $n_a \neq 0$ and $n_b \neq 0$, respectively. It is shown in Fig. 9 that for $n_a = n_b = 0$ the spectral line is a symmetric one while for $n_a \neq 0$ and $n_b \neq 0$ but $n_a = n_b$, the asymmetry of the spectral lines favours LAWI at the three-photon generated frequency $\omega = \omega_a + \omega_b - \omega_c$.

In conclusion, it has been demonstrated that the combined field of the three laser fields, which occurs at low intensities of all three laser fields, generates three-photon excitations having characteristic spectral widths that are much less than the natural linewidths of the excited states of the atom. The linewidths γ_+ and γ_- defined by (21) are induced by the combined field of the three laser fields and they are functions of the expressions γ_a, γ_b and γ_c as well as of the detunings n_a and n_b. The lifetime of these excitations are much longer than those of the excited states of the atoms, namely $1/\gamma_\pm \gg 1/\gamma_3$ and $1/\gamma_\pm \gg 1/\gamma_4$. We hope that these results will motivate and prove useful in experimental endeavours, especially in nonlinear optics and cold atom technology by using excitations which have linewidths much less than the natural linewidths of the excited states of the atoms [28–30].

References

1. S.E. Harris: Phys. Rev. Lett. **62**, 1033 (1989)
2. V.G. Arkhipkin, Y.I. Heller: Phys. Lett. **A98**, 12 (1983); A. Imamoglu, S.E. Harris: Opt. Lett. **14**, 1344 (1989); M.O. Scully, S.-Y. Zhu, A. Gavrielides: Phys. Rev. Lett. **62**, 2183 (1989)
3. A. Imamoglu: Phys. Rev. A **40**, 2835 (1989); A. Lyras, Z. Tang, P. Lambropoulos, J. Zhang: Phys. Rev. A **40**, 4131 (1989); S.E. Harris, J.J. Macklin: Phys. Rev. A **40**, 4135 (1989)
4. G.S. Agarwal, S. Ravi, J. Cooper: Phys. Rev. A **41**, 4721, 4727 (1990); E.E. Fill, M.O. Scully, S.-Y. Zhu: Opt. Commun. **77**, 36 (1990); S. Basile, P. Lambropoulos: Opt. Commun. **78**, 163 (1990)
5. O.A. Kocharovskaya, Y.I. Khanin: Pis'ma Zh. Eksp. Teor. Fiz. **48**, 581 (1988) [JETP Lett. **48**, 630 (1988)]; O. Kocharovskaya, P. Mandel: Phys. Rev. A **42**, 523 (1990); O. Kocharovskaya, R.-D. Li, P. Mandel: Opt. Commun. **77**, 215 (1990); V.R. Blok, G.M. Krochik: Phys. Rev. A **41**, 1517 (1990); A **44**, 2036 (1991); A. Imamoglu, J.E. Field, S.E. Harris: Phys. Rev. Lett. **66**, 1154 (1991)
6. L.M. Narducci, M.O. Scully, G.-L. Oppo, P. Ru, J.R. Tredicce: Phys. Rev. A **42**, 1630 (1990); L.M. Narducci, H.M. Doss, P. Ru, M.O. Scully, S.Y. Zhu, C. Keitel: Opt. Commun. **81**, 379 (1991); L.M. Narducci, M.O. Scully, C.H. Keitel, S.-Y. Zhu, H.M. Doss,: Opt. Commun. **86**, 324 (1991)
7. R.M. Whitley, C.R. Stroud, Jr.: Phys. Rev. A **14**, 1498 (1976); M. Lewenstein, Y. Zhu, T.W. Mossberg: Phys. Rev. Lett. **64**, 3131 (1990); Y. Zhu, Q. Wu, S. Morin, T.W. Mossberg: Phys. Rev. Lett. **65**, 1200 (1990); D.Z. Gauthier, Q. Wu, S.E. Morin, T.W. Mossberg: Phys. Rev. Lett. **68**, 464 (1992)
8. A. Karawajczyk, J. Zakrzewski, W. Gawlik: Phys. Rev. A **45**, 420 (1992); Y. Zhu: Phys. Rev. A **45**, R6149 (1992); Z.-R. Luo, Z.-Z. Xu: Phys. Rev. A **45**, 8292 (1992); P. Mandel, O. Kocharovskaya: Phys. Rev. A **46**, 2700 (1992); A **47**, 5003 (1993); W. Tan, W. Lu, R.G. Harrison: Phys. Rev. A **46**, R3613 (1992); H.Y. Ling: Phys. Rev. A **49**, 2827 (1994); H.Y. Ling, Y.-Q. Li, M. Xiao: Phys. Rev. A **53**, 1014 (1996)
9. C. Mavroyannis: Phys. Rev. A **46**, R6785 (1992); Quantum Opt. **4**, L145 (1992); **5**, L141 (1993); **5**, 43 (1993); **5**, L317 (1993); Opt. Soc. Am. B **10**, 2406 (1993); Z. Phys. B **99**, 429 (1996); Appl. Phys. B **62**, 547 (1996); Z. Phys. D **38**, 295 (1996); J. Phys. B **29**, 571 (1996): Appl. Phys. B **64**, 465 (1997)
10. O. Kocharovskaya: Phys. Rep. **219**, 175 (1992)
11. M.O. Scully: Phys. Rep. **219**, 191 (1992); Quantum Opt. **6**, 203 (1994)
12. E. Arimondo: In Progress in Optics XXXV, ed. by E. Wolf (Elsevier Science, Amsterdam 1996) p. 257
13. S.E. Harris: Phys. Today **50** (7), 36 (1997)
14. M.O. Scully, M.S. Zubairy: *Quantum Optics* (Cambridge University Press 1997) and references therein
15. K. Hakuta, L. Marmet, B.P. Stoicheff: Phys. Rev. Lett. **66**, 596 (1991); K.-J. Boller, A. Imamoglu, S.E. Harris: Phys. Rev. Lett. **66**, 2593 (1991); J.E. Field, K.H. Hahn, S.E. Harris: Phys. Rev. Lett. **67**, 3062 (1991); A. Kasapi, M. Jain, G.Y. Yin, S.E. Harris: Phys. Rev. Lett. **74**, 2447 (1995); Y. Zhu, T.N. Wasserlauf: Phys. Rev. A **54**, 3653 (1996); Y.-g. Li, M. Xiao: Phys. Rev. A **51**, R2703 (1995); S.A. Hopkins, E. Usadi, H.X. Chen, A.V. Durrant: Opt. Commun. **138**, 185 (1997); A.V. Durrant, H.X. Chen, S.A. Hopkins, J.A. Vaccaro: Opt. Commun. **151**, 138 (1998)
16. J. Gao, C. Guo, X. Guo, G. Jin, P. Wang, J. Zhao, H. Zhang, Y. Jiang, D. Want, D. Jiang: Opt. Commun. **93**, 323 (1992)
17. A. Nottelman, C. Peters, W. Lange: Phys. Rev. Lett. **70**, 1783 (1993)
18. E.S. Fry, X. Li, D. Nikonov, G.G. Padmabandu, M.O. Scully, A.V. Smith, F.K. Tittel, C. Wang, S.R. Wilkinson, S.Y. Zhu: Phys. Rev. **70**, 3235 (1993)
19. W.E. van der Veer, R.J.J. van Diest, A. Donszelmann, H.B. van Linden van den Heuvell: Phys. Rev. Lett. **70**, 3243 (1993)
20. W. Lange, A. Nottelman, C. Peters: Quantum Opt. **6**, 273 (1994)
21. A.S. Zibrou, M.D. Lukin, L. Hollberg, D.E. Nikonov, M.O. Scully, H.G. Robinson, V.L. Velichansky: Phys. Rev. Lett. **76**, 3935 (1996)
22. E. Schmidt, A. Imamoglu: Opt. Lett. **21**, 1936 (1996)
23. G. Alzetta, A. Gozzini, L. Moi, G. Orriols: Nuovo Cimento B **36**, 5 (1976); E. Arimondo, G. Orriols: Lett. Nuovo Cimento **17**, 333 (1976)
24. H.R. Gray, R.M. Whitley, C.R. Stroud, Jr.: Opt. Lett. **3**, 218 (1978)
25. G. Alzetta, L. Moi, G. Orriols: Nuovo Cimento B **52**, 209 (1979); G. Orriols: Nuovo Cimento B **53**, 1 (1979); S. Swain: J. Phys. B: At. Mol. Phys. **12**, 3201 (1979); J.D. Stettler, C.M. Bowden, N.M. Witriol, J.H. Eberly: Phys. Rev. Lett. **73A**, 171 (1979); G.S. Agarwal, S.S. Jha: J. Phys. B: At. Mol. Phys. **12**, 2655 (1979); M.S. Feld, M.M. Burns, P.G. Pappas, D.E. Murnick: Opt. Lett. **5**, 79 (1980)
26. C. Mavroyannis: Can. J. Phys. **70**, 1 (1992)
27. S.E. Harris, Y. Yamamoto: Phys. Rev. Lett. **81**, 3611 (1998)
28. L.V. Hau, S.E. Harris, Z. Dutton, C.H. Behroozi: Nature **397**, 594 (1999)
29. D. Budker, D.F. Kimball, S.M. Rochester, V.V. Yashchuk: Phys. Rev. Lett. **83**, 1167 (1999)
30. M.M. Kash, V.A. Sautenkou, A.S. Zibrou, L. Holliberg, G.R. Welch, M.D. Lukin, Y. Rostovtsev, E.S. Fry, M.O. Scully: Phys. Rev. Lett. **82**, 5229 (1999)
31. S.E. Harris, L.V. Hau: Phys. Rev. Lett. **82**, 4611 (1999)
32. C. Mavroyannis: Quantum Semiclass. **2**, 735 (2000)
33. D.N. Zubarev: Usp. Fiz. Nauk **71**, 71 (1960) [Sov. Phys Usp. **3**, 320 (1960)]; *Nonequilibrium Statistical Thermodynamics* (Plenum, New York 1976) Chapt. 16; V.L. Bonch-Bruevich, S.V. Tyablikov: *The Green Function in Statistical Mechanics* (North-Holland, Amsterdam 1963); Adv. Phys. **11**, 317 (1962); S.V. Tyablikov: *Methods in Quantum Theory of Magnetism* (Plenum, New York 1967); C. Mavroyannis: *The Green Function Method*. In *Physical Chemistry*, ed. by H. Eyring, W. Jost, D. Henderson (Academic, New York 1975) Vol. 11A
34. O.A. Kocharovskaya, F. Mauri, E. Arimondo: Opt. Commun. **84**, 393 (1991)