Theory of lasers without inversion

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We here present the theory of lasing without inversion for systems with a split upper laser level. We show that the effect of amplification without inversion may be realized for at least two different situations. In the first situation, the effect is due to the asymmetry of radiative characteristics of quasidegenerate transitions. In the second situation, the effect can be provided without any asymmetry and is achievable for the definite phase of the transfer of coherence between the quasidegenerate levels (quasiparametric amplification). We also show that for laser amplification without inversion, the rate (frequency) of mixing of the quasidegenerate levels should be significantly higher than the linewidths of these levels. This mixing of quasidegenerate levels can be realized by low-frequency electromagnetic radiation or by interaction with an auxiliary third level. In addition, we derive the conditions for providing laser action for such systems rigorously, and we give some specific examples for the realization of this effect in gaseous and solid-state media. One of the most promising configurations may be realized in solid-state media when two narrow quasidegenerate levels are embedded in quasifree states (low-density state conduction band). We also discuss the use of a parametric mechanism for the creation of spatial gratings and for the construction of new types of electro-optical devices.

I. INTRODUCTION

Recently Harris[1] and Kocharovskaya and Khanin[2] predicted the possibility of amplification of light in a three-level medium without inversion of population. Harris considered the case of continuous-wave or long pulse laser modes with two upper autoionizing levels as being purely lifetime broadened and the difference between their energies as being small (quasidegenerate levels). Kocharovskaya and Khanin analyzed the amplification of ultrashort laser pulses in the three-level system, with the two lower levels being quasidegenerate ones.

The possibility of light amplification without population inversion may be based on the nonreciprocal interference of absorption and stimulated emission between a pair of upper or lower split levels and a single level; a similar effect based on Doppler recoil splitting of the emission and absorption spectra was suggested by Elton.[3] Arkhipkin and Heller[4] considered amplification without inversion with an upper single level as a field-induced autoionizinglike state.

The feasibility of laser action without population inversion has been shown by Harris[1] for the case of strong asymmetry in the line strengths and linewidth of quasidegenerate transitions. In this case, the effect is anticipated for the exact resonance of light with one component of the quasidegenerate transition which has lower frequency, which also has lower oscillator strength.

In Ref. 2 laser amplification without inversion is attributed to the population capture of two quasidegenerate lower levels and to the excitation of low-frequency coherence. Since the consideration in Ref. 2 is based on the neglect of relaxation times, it can be shown that this consideration is valid only for ultrashort laser pulses.

The feasibility of laser action in the media is usually estimated from the analyses of rate equations for population of the levels;[5] these equations for multilevel systems are derived from the kinetic equation for the density matrix and from Maxwell's equations for the radiation field in quasisteady approximation (valid when the field envelope and population difference are changing very slowly in comparison with cross-relaxation time[6]). It is known that for three- or four-level laser systems which have a single resonance of laser radiation with one of the transitions the laser action cannot be provided without population inversion.[5] The effect is possible only for the case of double resonance of laser radiation with two transitions of closed frequencies (the quasidegenerate transitions). These two transitions may be transitions from the lower single level to two discrete levels embedded in continuum,[1] to one level embedded in continuum,[4] or to two weakly interacting quasicontinuums.

This effect cannot be predicted by means of analysis of rate equations,[5,6] because interference of two resonant noncoherent processes must be taken into account and we have to consider the equations for nondiagonal components of the density matrix. The conception of inverted population was introduced for two-level systems and then was used in the analyses of multilevel systems for predictions of new laser schemes.[5] Derivation of the conditions of maser and laser oscillation was made by means of analysis of the equations describing the change in level populations due to the combined effect of pumping, spontaneous and induced radiative transitions and relaxation processes. All these processes were characterized by rates or probabilities of transitions (and relaxation), which has been introduced in the absence of a laser field by independent treatment and then were used in the equations of level populations. This theoretical formalism known as rate equations predicted all known laser systems. That is why the terminology of lasers without

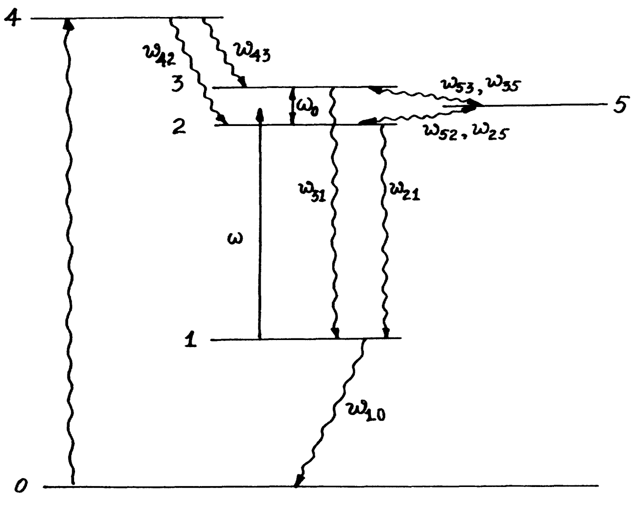

FIG. 1. The four-level laser scheme for providing amplification without inversion. ω is the laser frequency. The upper laser level is split and consists of two quasidegenerate levels 2 and 3. w_{ij} are rates of spontaneous or radiationless transitions, ω_0 is the frequency of transition between the levels 2 and 3, and 5 is the auxiliary level through which the fast coherent mixing of levels 2 and 3 can be provided.

inversion is an echo of consideration of a multilevel laser system in the framework of rate equations.

When considering the systems with split laser levels it is particularly important to go beyond rate-equation approximation, and it is one of the topics of this paper. We state the theoretical treatment of the problem of lasers without inversion in the formalism of standard kinetic equations for a density matrix.

Consider the case of a split upper laser level (Fig. 1). (The cases of the split lower level and of the transitions from the discrete level to two weakly interacting quasicontinuums will be considered in our next publications.) For the case of an upper split level, we show that, besides the ordinary probabilities of one-photon absorption and stimulated emission, there are the probabilities of transitions that arise from the interference of two noncoherent processes. Two different situations were considered.

(a) Laser gain may be provided due to the asymmetry in radiative characteristics of quasidegenerate transitions and can be attributed to the interference of absorption ($\omega \simeq \omega_{ij}$) and "hyper-Raman-scattering" ($\omega+\omega_0-\omega_0 \simeq \omega_{ij}$), where ω_0 is the frequency of the field that coherently mixes the quasidegenerate levels. This laser action is achievable if the rate of the mixing of two quasidegenerate levels is much greater than the linewidth of transition. There are not any requirements for strong asymmetry in radiative characteristics of quasidegenerate transitions as was stated in Ref. 1.

(b) The second situation is not connected with asymmetry of quasidegenerate transitions. In this case the effect may be due to the interference of absorption ($\omega \simeq \omega_{ij}$) and "Raman scattering" ($\omega-\omega_0 \simeq \omega_{ij}$). The feasibility of laser action now depends on the sign or, in a more general case, on the phase of the complex dipole or multipole moment of transition between upper quasidegenerate levels. Since the laser gain without population inversion does not connect with the intensity of the laser field or with the pulse duration (as in Ref. 2), the new type of laser may be constructed. The effect of laser amplification without inversion is based on the interference of two noncoherent multiphoton transitions, which includes the transfer of coherence between quasidegenerate levels and is similar to the interference of two noncoherent processes in four-wave mixing in two-photon resonance.[7]

II. EQUATIONS OF MOTION FOR THE DENSITY MATRIX

Density-matrix formalism is probably most convenient for the calculation of microscopic expression of optical susceptibilities and for dealing with relaxations of excitations. As in the regular theory of lasers,[5] we start from the equation for motion for density matrix ρ (Ref. 8)

$$\left[\frac{\partial}{\partial t}+i\omega_{mn}+T_{mn}^{-1}\right]\rho_{mn}=-i\hbar^{-1}[V,\rho]_{mn} , \tag{1a}$$

$$\frac{\partial \rho_{mm}}{\partial t}+\Sigma(w_{mk}\rho_{mm}-w_{km}\rho_{kk})=-i\hbar^{-1}[V,\rho]_{mm} , \tag{1b}$$

where ω_{mn} is the frequency of transition, T_{mn} is the characteristic relaxation time between the states $|m\rangle$ and $|n\rangle$ (cross relaxation), w_{mk} is the probability of radiationless transition between the states $|m\rangle$ and $|k\rangle$, and V is the Hamiltonian of interaction of a field with matter.

By way of example, we consider the standard four-level laser system[5] when the upper laser level is split (Fig. 1). The laser field interacts with transitions $1\rightarrow 2$ and $1\rightarrow 3$ simultaneously. The levels 0 and 4 serve as auxiliary levels for the pumping process. For the coherent mixing of levels 2 and 3, we consider two opportunities: (a) levels 2 and 3 are coherently mixed by an electromagnetic field with frequency $\omega_0 \simeq \omega_{32}$, and (b) they interact with each other through level 5.

For the first situation the equations for density matrix may be written

$$\frac{d\rho_{12}}{dt}+(i\omega_{12}+T_{12}^{-1})\rho_{12}=i\hbar^{-1}V_{12}(\rho_{11}-\rho_{22})-i\hbar^{-1}V_{13}\rho_{32}+i\hbar^{-1}\rho_{13}V_{32} , \tag{2a}$$

$$\frac{d\rho_{13}}{dt}+(i\omega_{13}+T_{13}^{-1})\rho_{13}=i\hbar^{-1}V_{13}(\rho_{11}-\rho_{33})-i\hbar^{-1}V_{12}\rho_{23}+i\hbar^{-1}\rho_{12}V_{23} , \tag{2b}$$

$$\frac{d\rho_{23}}{dt}+(i\omega_{23}+T_{23}^{-1})\rho_{23}=i\hbar^{-1}V_{23}(\rho_{22}-\rho_{33})-i\hbar^{-1}V_{21}\rho_{13}+i\hbar^{-1}\rho_{21}V_{13} , \tag{2c}$$

$$\frac{d\rho_{11}}{dt}+w_{10}\rho_{11}-w_{21}\rho_{22}=-i\hbar^{-1}(V_{12}\rho_{21}-\rho_{12}V_{21})-i\hbar^{-1}(V_{13}\rho_{31}-\rho_{13}V_{31})$$
$$=-2\hbar^{-1}\mathrm{Im}(\rho_{12}V_{21})-2\hbar^{-1}\mathrm{Im}(\rho_{13}V_{31}) , \tag{2d}$$

$$\frac{d\rho_{22}}{dt}-w_{42}\rho_{44}+w_{21}\rho_{22}=i\hbar^{-1}(V_{12}\rho_{21}-\rho_{12}V_{21})-i\hbar^{-1}(V_{23}\rho_{32}-\rho_{23}V_{32})$$
$$=+2\hbar^{-1}\mathrm{Im}(\rho_{12}V_{21})-2\hbar^{-1}\mathrm{Im}(\rho_{23}V_{32}) , \tag{2e}$$

$$\frac{d\rho_{33}}{dt}-w_{43}\rho_{44}+w_{31}\rho_{33}=+i\hbar^{-1}(V_{13}\rho_{31}-\rho_{13}V_{31})+i\hbar^{-1}(V_{23}\rho_{32}-\rho_{23}V_{32})$$
$$=+2\hbar^{-1}\mathrm{Im}(\rho_{13}V_{31})+2\hbar^{-1}\mathrm{Im}(\rho_{23}V_{32}) , \tag{2f}$$

$$\frac{d\rho_{44}}{dt}-w_{04}\rho_{00}+w_{43}\rho_{44}+w_{42}\rho_{44}=0 , \tag{2g}$$

where V_{23} is the Hamiltonian of interaction of the electromagnetic field with transition $2\rightarrow3$.

For the situation (b), when levels 2 and 3 interact with each other through level 5, let us analyze the equations only for the population of levels 2 and 3 [see (2e) and (2f)]. The terms V_{23}, V_{32} may be expressed in the rates $\Omega_{i5}=\hbar^{-1}V_{i5}$ and $\Omega_{5i}=\hbar^{-1}V_{5i}$ which describe the energy exchange between levels 2, 3, and 5. These terms can represent different processes of energy transfer, for example, dipole-dipole transfer, resonance-exchange transfer, and the transfer between narrow quasidegenerate levels embedded in quasifree states in solids. They serve as the perturbations of a stochastic nature.[8] The rates of such processes of the transfer can reach the order of $10^{13}-10^{14}$ sec^{-1}.[8,9] We can use in this situation the steady-state approximation to express density-matrix elements ρ_{22},ρ_{33} through ρ_{55}, and thus density-matrix element ρ_{22} through ρ_{33}. This procedure is described in Appendix A. As is shown there we can estimate the effective Rabi frequencies $\Omega_{23}=\hbar^{-1}V_{23}$ and $\Omega_{32}=\hbar^{-1}V_{32}$ through rate constants w_{i5} and w_{5i}, the experimental data for which are often known:

$$|\Omega_{23}|\propto w_{23}=(w_{25}w_{53})^{1/2} ,$$
$$|\Omega_{32}|\propto w_{32}=(w_{35}w_{52})^{1/2} . \tag{3}$$

This way we reduce the problem in situation (b) to the problem in situation (a).

Let us consider the system (2) in the case when coherent mixing of quasidegenerate levels 2 and 3 occurs by mixing the electromagnetic (low-frequency) field or by energy exchange through auxiliary level 5. In both these cases we will leave the matrix elements of the Hamiltonian of interaction V_{32} and V_{23} in Eqs. (2e) and (2f) keeping in mind expressions (3) for the second situation. We analyze the first three equations in (2). We introduce slowly varying envelopes for the density matrix

$$\rho_{12}=\rho'_{12}\exp(i\omega t) \quad \rho'\equiv\rho \tag{4a}$$

and for the Hamiltonian of interaction

$$V_{12,13}=V'_{12,13}\exp(i\omega t), \quad V_{12,13}\equiv V'_{12,13} . \tag{4b}$$

We also use the notations

$$\gamma_{ij}=T_{ij}^{-1} ,$$
$$\Delta_{ij}=i\omega_{ij}+i\omega+\gamma_{ij}=i\delta_{ij}+\gamma_{ij} , \tag{5}$$
$$\eta_{ij}=\rho_{ii}-\rho_{jj} .$$

In the first approximation on laser field intensity we can neglect those terms in (2c) which are proportional to $V_{21}V_{13}$, which describe the processes of the second order. By this a way we obtain

$$\rho_{23}=i\Omega_{23}\eta_{23}\Delta_{23}^{-1} , \tag{6}$$

and

$$\frac{d\rho_{12}}{dt}+\Delta_{12}\rho_{12}=i\Omega_{12}\eta_{12}-i\Omega_{13}\rho_{32}+i\rho_{13}\Omega_{32} , \tag{7a}$$

$$\frac{d\rho_{13}}{dt}+\Delta_{13}\rho_{13}=i\Omega_{13}\eta_{13}-i\Omega_{12}\rho_{23}+i\Omega_{23}\rho_{12} . \tag{7b}$$

The steady-state solutions of Eqs. (7) have a form

$$\rho_{12}=(\Delta_{12}\Delta_{13}+|\Omega_{23}|^2)^{-1}[(i\Omega_{12}\eta_{12}-i\Omega_{13}\rho_{32})\Delta_{13}-\Omega_{32}(\Omega_{13}\eta_{13}-\Omega_{12}\rho_{23})] , \tag{8a}$$
$$\rho_{13}=(\Delta_{12}\Delta_{13}+|\Omega_{23}|^2)^{-1}[(i\Omega_{13}\eta_{13}-i\Omega_{12}\rho_{23})\Delta_{12}-\Omega_{23}(\Omega_{12}\eta_{12}-\Omega_{13}\rho_{32})] . \tag{8b}$$

III. DERIVATION OF THE CONDITIONS OF LASER AMPLIFICATION WITHOUT INVERSION

We will now derive the conditions of laser amplification without population inversion. This condition may be obtained, for example, by analyzing the equation for the population of the lower single laser level 1 [see (2d)]

$$\frac{d\rho_{11}}{dt}+w_{10}\rho_{11}-w_{21}\rho_{22}=-2\hbar^{-1}\mathrm{Im}(\rho_{12}V_{21}+\rho_{13}V_{31}) . \tag{9}$$

If the right-hand side of (7) is positive, the absorption of the laser field takes place; on the contrary, when the sign of the right-hand side is negative then one can reach the amplification of the laser field. In the case when the upper laser level is not split, this latter condition may be satisfied only for population inversion between the levels 1 and 2 and/or 3,

$$\eta_{12}=\rho_{11}-\rho_{22}<0, \quad \eta_{13}=\rho_{11}-\rho_{33}<0 . \tag{10}$$

We will show now that for the case of a split upper laser level (when laser radiation can efficiently interact simultaneously with two transitions $1\rightarrow2$ and $1\rightarrow3$) the negative sign of the right-hand side of Eq. (9) may be provided without population inversion at any of these transitions. In other words, there are not any requirements like (10).

We note that since the resonant polarization at frequency ω can be expressed as[8]

$$P_R(\omega)=d_{21}\rho_{12}(\omega)+d_{31}\rho_{13}(\omega)=\chi_R(\omega)E(\omega) , \tag{11}$$

where $\chi_R(\omega)$ is the resonant susceptibility of the medium, Eq. (9) can be rewritten in a form

$$\begin{aligned}\frac{d\rho_{11}}{dt}+w_{10}\rho_{11}-w_{21}\rho_{22}&=-2\hbar^{-1}\mathrm{Im}(\rho_{12}V_{21}+\rho_{13}V_{31})\\&=+2\hbar^{-1}\mathrm{Im}[P_R(\omega)E^*(\omega)]\\&=+2\hbar^{-1}|E(\omega)|^2\mathrm{Im}[\chi_R(\omega)] .\end{aligned} \tag{12}$$

As follows from (11) and (12) the negative sign of the right-hand side in (16) corresponds to the negative sign of the imaginary part of the resonant susceptibility of the medium at frequency ω (emission of energy). Thus the criterion of amplification without inversion can be written in the form

$$-2\hbar^{-1}\mathrm{Im}(\rho_{12}V_{21}+\rho_{13}V_{31})=+2\hbar^{-1}|E(\omega)|^2\mathrm{Im}[\chi_R(\omega)]<0 . \tag{13}$$

Now we introduce the notations

$$\begin{aligned}a&=\gamma_{12}\gamma_{13}+|\Omega_{23}|^2-\delta_{12}\delta_{13} ,\\ b&=(\delta_{12}\gamma_{13}+\delta_{13}\gamma_{12}) ,\end{aligned} \tag{14}$$

and substitute expressions (8) into Eq. (12). We obtain

$$\begin{aligned}-[P(\omega)E^*(\omega)]&=\Omega_{21}\rho_{12}+\Omega_{31}\rho_{13}\\&=(a^2+b^2)^{-1}(a-ib)\{\Omega_{21}[(i\Omega_{12}\eta_{12}+\Omega_{13}\Omega_{32}\eta_{32}\gamma_{23}^{-1})(\gamma_{13}+i\delta_{13})-\Omega_{32}(\Omega_{13}\eta_{13}-i\Omega_{12}\Omega_{23}\eta_{23}\gamma_{23}^{-1})]\\&\quad+\Omega_{31}[(i\Omega_{13}\eta_{13}+\Omega_{12}\Omega_{23}\eta_{23}\gamma_{23}^{-1})(\gamma_{12}+i\delta_{12})\\&\quad-\Omega_{23}(\Omega_{12}\eta_{12}-i\Omega_{13}\Omega_{32}\eta_{32}\gamma_{23}^{-1})]\} .\end{aligned} \tag{15}$$

Let us consider the different terms of this expression. The expression in the first set of square brackets on the right-hand side describes the processes on the transition $1\rightarrow2$. The first term inside these brackets is due to ordinary one-photon absorption at the transition $1\rightarrow2$. The second and the third terms here may be interpreted as resonant parametric or coherent (depending on the phases of the fields) three-frequency interaction of the fields with a frequency condition

$$\omega=\omega-\omega_0+\delta, \quad \omega+\omega_{12}=\delta_{12} . \tag{16}$$

This interaction represents an interference of two resonant processes: one-photon absorption on the transition $1\rightarrow2$, and resonant stimulated Raman scattering $1\rightarrow3\rightarrow2$ on the same transition. Although the frequency mismatch δ is not equal to zero (which is a necessary condition for parametric interactions), this process can be realized at restricted time intervals or due to the small value of energy differences between the quasidegenerate levels (which can be much less than the linewidths γ_{12} and γ_{13}). The last term in the first set of square brackets may be presented as a noncoherent resonant four-frequency process (which does not depend on the phases of the fields) with frequency condition

$$\omega=\omega+\omega_0-\omega_0, \quad \omega+\omega_{12}=\delta_{12} . \tag{17}$$

This interaction may be interpreted as an interference of two processes: the one-photon absorption on the transition $1\rightarrow2$ and the stimulated hyper-Raman-scattering $(1\rightarrow2\rightarrow3\rightarrow2)$ on the same transition. Similar interpretations concerning transition $1\rightarrow3$ can be given for the second set of square brackets in (15).

Before deriving the conditions for amplification without inversion let us make some remarks. Within dipole approximation we can represent $V_{12}=-d_{12}E(\omega)$, $V_{13}=-d_{13}E(\omega)$. In situation (a), where levels 2 and 3 are coupled by an applied electromagnetic field $E(\omega_0)$ we can also assume that $V_{23}=-d_{23}E(\omega_0)$ only for solids, since in the media with a center of inversion $d_{12}d_{23}d_{31}=0$. Let us note that in the media with a center of inversion (for example, in gaseous media) the situation when levels 2 and 3 have strong enough coupling to a common level 1 and yet also have an efficient coupling to each other is also possible, and not only through the trivial case of weak quadrupole or magnetodipole interaction (for example, the case when $1\rightarrow2,3$ are dipole transitions and $2\rightarrow3$ are magnetodipole or quadrupole ones). The coupling of all three levels is allowed, for example, for the case when levels 2 and 3 are randomly degenerative ones. Let levels 2 and 3 be close and transitions $1\rightarrow2$ and $2\rightarrow3$ are dipole. Then the transition $1\rightarrow3$ is forbidden. The strong electromagnetic field mixes levels 2 and 3 and, thus, forms an effective dipole moment d_{31} at forbidden transition $1\rightarrow3$, which is proportional to the intensity of the electromagnetic field. We do not consider this theory in detail here, however, we are going to consider it in our future publications.

Thus we can write for the terms described a three-frequency interaction:

$$\Omega_{12}\Omega_{23}\Omega_{31}=-\hbar^{-3}d_{12}d_{23}d_{31}E(\omega)E(\omega_0)[E(\omega)]^*$$
$$=-\hbar^{-3}|E(\omega)|^2 d_{12}d_{31}d_{23}E(\omega_0) . \quad (18)$$

For representation of a low-frequency electromagnetic field in a form

$$E(\omega_0)=2A(\omega_0)\cos(\omega_0 t)=A(\omega_0)f(t) \quad (19)$$

we obtain

$$\Omega_{12}\Omega_{23}\Omega_{31}=-|\Omega_{12}\Omega_{23}\Omega_{31}|f(t)\exp(i\varphi) , \quad (20)$$

where we have introduced a phase φ of the dipole moment of the transition $2\to3$; while introducing it we took into account that

$$d_{12}d_{31}E(\omega)[E(\omega)]^*=|d_{12}d_{31}|\,|E(\omega)|^2 .$$

For the coherent mixing of levels 2 and 3 through auxiliary level 5 [situation (b)] instead of

$$\Omega_{23}=-d_{23}E(\omega_0)/\hbar=-|d_{23}|A(\omega_0)f(t)\exp(i\varphi) \quad (21)$$

we have to introduce an appropriate expression for the Hamiltonian of interaction Ω_{23}. However, we still can represent the term, describing three-frequency interaction in the form (20), in which $f(t)$ and Ω_{23} depend on the actual mechanism of energy transfer and on energetic difference between levels 2, 3, and 5.

Transforming (15) we obtain the following expression for the right-hand side of Eq. (12):

$$\begin{aligned}
-2\hbar^{-1}\mathrm{Im}(\rho_{12}V_{21}+\rho_{13}V_{31})=-2(a^2+b^2)^{-1}&\\
\times[(a|\Omega_{12}|^2\eta_{12}\gamma_{13}+a|\Omega_{13}|^2\eta_{13}\gamma_{12}+b\delta_{13}|\Omega_{12}|^2\eta_{12}+b\delta_{12}|\Omega_{13}|^2\eta_{13})&\\
+a|\Omega_{12}|^2|\Omega_{23}|^2\eta_{23}\gamma_{23}^{-1}+a|\Omega_{13}|^2|\Omega_{23}|^2\eta_{32}\gamma_{23}^{-1}&\\
+a\,\mathrm{Re}(\Omega_{13}\Omega_{32}\Omega_{21})\eta_{32}\gamma_{23}^{-1}\delta_{13}+a\,\mathrm{Re}(\Omega_{12}\Omega_{23}\Omega_{31})\eta_{23}\gamma_{23}^{-1}\delta_{12}&\\
-b\gamma_{13}\mathrm{Re}(\Omega_{13}\Omega_{32}\Omega_{21})\eta_{32}\gamma_{23}^{-1}-b\gamma_{12}\mathrm{Re}(\Omega_{12}\Omega_{23}\Omega_{31})\eta_{23}\gamma_{32}^{-1}+b\,\mathrm{Re}(\Omega_{13}\Omega_{32}\Omega_{21})\eta_{13}&\\
+b\,\mathrm{Re}(\Omega_{12}\Omega_{23}\Omega_{31})\eta_{12}+a\,\mathrm{Im}(\Omega_{13}\Omega_{32}\Omega_{21})\eta_{32}\gamma_{23}^{-1}\gamma_{13}+a\,\mathrm{Im}(\Omega_{12}\Omega_{23}\Omega_{31})\eta_{23}\gamma_{23}^{-1}\gamma_{12}&\\
+b\delta_{13}\mathrm{Im}(\Omega_{13}\Omega_{32}\Omega_{21})\eta_{32}\gamma_{23}^{-1}+b\delta_{12}\mathrm{Im}(\Omega_{12}\Omega_{23}\Omega_{31})\eta_{23}\gamma_{32}^{-1}&\\
-a\,\mathrm{Im}(\Omega_{13}\Omega_{32}\Omega_{21})\eta_{13}-a\,\mathrm{Im}(\Omega_{12}\Omega_{23}\Omega_{31})\eta_{12}] .&\quad (22)
\end{aligned}$$

Substituting (20) in (22) and then in (13) we find the following condition of amplification:

$$\begin{aligned}
&a|\Omega_{12}|^2\eta_{12}\gamma_{13}+a|\Omega_{13}|^2\eta_{13}\gamma_{12}+b\delta_{13}|\Omega_{12}|^2\eta_{12}+b\delta_{12}|\Omega_{13}|^2\eta_{13}+a|\Omega_{12}|^2|\Omega_{23}|^2\eta_{23}\gamma_{23}^{-1}+a|\Omega_{13}|^2|\Omega_{23}|^2\eta_{32}\gamma_{23}^{-1}\\
&-a|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)\eta_{32}\gamma_{23}^{-1}\delta_{13}-a|\Omega_{12}\Omega_{23}\Omega_{31}|f(t)(\cos\varphi)\eta_{23}\gamma_{23}^{-1}\delta_{12}+b\gamma_{13}|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)\eta_{32}\gamma_{23}^{-1}\\
&+b\gamma_{12}|\Omega_{12}\Omega_{23}\Omega_{31}|f(t)(\cos\varphi)\eta_{23}\gamma_{32}^{-1}-b|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)\eta_{13}-b|\Omega_{12}\Omega_{23}\Omega_{31}|f(t)(\cos\varphi)\eta_{12}<0 . \quad (23)
\end{aligned}$$

A. Amplification based on the asymmetry of radiative characteristics of quasidegenerate transitions

Let us now consider the case when $\delta_{12}>0$, $\delta_{13}<0$ (the laser frequency is located between the frequencies of quasidegenerate transitions) and let us introduce notations $\delta_{12}=|\delta_{12}|$, $\delta_{13}=|\delta_{13}|$. For $b=(\delta_{12}\gamma_{13}-\delta_{13}\gamma_{12})=0$, we rewrite (23) in the form

$$\begin{aligned}
&a|\Omega_{12}|^2\eta_{12}\gamma_{13}+a|\Omega_{13}|^2\eta_{13}\gamma_{12}-b\delta_{13}|\Omega_{12}|^2\eta_{12}+b\delta_{12}|\Omega_{13}|^2\eta_{13}\\
&+a|\Omega_{23}|^2\eta_{23}\gamma_{23}^{-1}(|\Omega_{12}|^2-|\Omega_{13}|^2)+a|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)\eta_{23}\gamma_{23}^{-1}(\delta_{13}+\delta_{12})\\
&-b|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)\eta_{23}\gamma_{23}^{-1}(\gamma_{13}-\gamma_{12})-b|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)(\eta_{13}+\eta_{12})<0 . \quad (24)
\end{aligned}$$

Let us first consider the case when the three-frequency interaction is negligible, and the four-frequency interaction dominates (for example, it is possible when the average over the time value of $\cos\varphi=0$, or $\cos\varphi<0$, or φ is arbitrary but $|\Omega_{23}|\gg\delta_{12}+\delta_{13}$). As is shown in Appendix B in this case for the assumptions

$$|\Omega_{23}|^2\gg\gamma_{12}\gamma_{13},\quad |\Omega_{23}|^2\gg\delta_{12}\delta_{13} ,$$
$$\gamma_{23}\propto\gamma_{12},\gamma_{13},\quad \eta_{23}\propto\eta_{12},\eta_{13} , \quad (25)$$

the inequality (24) can be reduced to the following one:

$$|\Omega_{23}|^2 c>\gamma_{13}\gamma_{12} ,$$

$$\text{where } c=\frac{\eta_{23}(|\Omega_{13}|^2-|\Omega_{12}|^2)}{(|\Omega_{12}|^2\eta_{12}+|\Omega_{13}|^2\eta_{13})} . \quad (26)$$

Conditions (25) and (26) are satisfied for a wide range of parameters. For example, they can be satisfied for $|\Omega_{13}|^2\gtrsim2|\Omega_{12}|^2$ and for the rate of coupling of levels 2

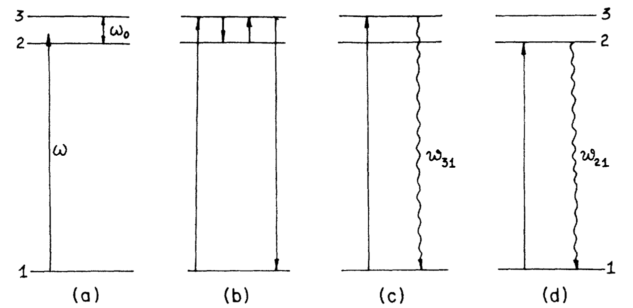

FIG. 2. The schematic diagram of the four-frequency process for producing laser action without inversion. Four-frequency interaction (b) induced by laser field ω and coherent-mixing field ω_0 is more efficient than absorption on the transitions $1\rightarrow 3$ (c) and $1\rightarrow 2$ (d).

and 3 significantly higher than the linewidths of transitions $1\rightarrow 2$ and $1\rightarrow 3$. In the case when the linewidths of quasidegenerate transitions are approximately equal to each other, the condition $\eta_{23}\propto\eta_{12},\eta_{13}$ may be provided if the laser pumping is used, which makes the population of the single lower level and the populations of the upper laser levels equal to each other. For the case of different linewidths of quasidegenerate levels, the population difference η_{23} can be of the order of the population differences η_{12} and η_{13} without any laser pumping. Thus four-frequency interaction can provide the amplification without inversion only due to asymmetry of radiative characteristics of quasidegenerate transitions.

Figure 2 illustrates the effect of laser action without inversion based on the asymmetry of radiative characteristics. In this case the four-frequency interaction [see Fig. 2(b)] induced by a laser field ω and coherent-mixing field ω_0, is more efficient than the process of absorption on the transitions $1\rightarrow 3$ [see Fig. 2(c)] and $1\rightarrow 2$ [see Fig. 2(d)]. This is due to the higher rate of coupling of levels 2 and 3, compared to the rate of relaxation of levels 2 and 3 to level 1. The probability of parametric reradiation on the frequency ω is, thus, more efficient than the ordinary absorption on the transitions.

The case considered in particular includes the situation which was treated in Ref. 1 ($\delta_{12}=0$, $|\Omega_{13}|^2\gg|\Omega_{12}|^2$, $\gamma_{13}\gg\gamma_{12}$). This follows from (23) by letting $b\rightarrow 0$ through $\delta_{12}\rightarrow 0$, and $\gamma_{13}\rightarrow\infty$. The more rigorous derivation is given in Appendix B.

We note that as it follows from (25) and (26), the stronger inequality $|\Omega_{23}|^2\gg\gamma_{12}\gamma_{13}$, the weaker asymmetry of radiative characteristics ($|\Omega_{12}|^2\ll|\Omega_{13}|^2$, $\gamma_{12}\ll\gamma_{13}$, $\rho_{22}>\rho_{33}$) of quasidegenerate transitions is required.

B. Quasiparametric amplification

The second opportunity to achieve laser action without inversion does not connect with asymmetry of radiative characteristics of split transition and is based on three-frequency interaction. To show this we assume in (24)

$$b=0,\quad |\Omega_{12}|^2\simeq|\Omega_{13}|^2,\quad \gamma_{12}\simeq\gamma_{13}\ . \tag{27}$$

The assumptions (27) may lead to the effect of amplification without inversion which is not based on the asymmetry of quasidegenerate transitions. The analysis of inequality (24) (see Appendix B) shows that in this case if we assume in (24) $\rho_{22}>\rho_{33}$ then for $\varphi=\pi$, the conditions of laser action without inversion are

$$\begin{aligned}&|\omega_{23}|\propto\gamma_{12},\gamma_{13},\quad f(t)\propto 1\ ,\\&|\Omega_{23}|\gg\gamma_{12},\gamma_{13},\quad \eta_{23}\propto\eta_{12},\eta_{13}\ .\end{aligned} \tag{28}$$

Conditions (28) are similar to those obtained for four-frequency interaction. To reach the effect one has to provide the phase $\varphi=\pi$ and the rate of coupling of the levels 2 and 3 much greater than the linewidths of quasidegenerate transitions, the difference of frequencies of which should not exceed their linewidths. For $\eta_{23}<0$ ($\rho_{22}<\rho_{33}$) the required phase is $\varphi=0$. The discussion of a way to provide conditions of amplification without inversion is considered in the next section.

IV. DISCUSSION AND CONCLUSIONS

Here we have treated the problem of lasers without inversion by using a standard nonlinear-optical approach (analysis of equations for the density matrix). Until now, several different particular cases for providing laser amplification without inversion were considered;[1-4] these can hardly be applied to calculations of the parameters of actual systems. Our theory predicts both new processes and new laser materials because it formulates some general conditions for providing amplification without inversion.

We have shown that the effect of amplification without inversion may be realized for at least two different laser systems with split upper laser levels. The first situation describes amplification based on the interference of two noncoherent processes: one-photon absorption and hyper-Raman scattering [see frequency condition (17)]. For the laser frequency located between the frequencies of quasidegenerate transitions, the effect can be achieved if the asymmetry in radiation characteristics (the oscillator strength and linewidth) of these transitions takes place. In the second situation the effect can be provided without any asymmetry in radiative characteristics and it has the parametric nature: amplification without inversion is achievable for the definite phase of the transfer of the coherence between the quasidegenerate levels. Phenomenologically, it can be represented as a resonant three-frequency interaction based on the interference of one-photon absorption and Raman scattering [see frequency condition (16)]. One of the particular cases of strong asymmetry in radiative characteristics of split transition was considered in Ref. 1; in this situation, the requirement of strong asymmetry is not necessary if the rate of coupling of quasidegenerate levels is high enough.

We derive the necessary condition of laser amplification without inversion: the rate (the frequency) of mixing of quasidegenerate levels should be significantly higher than the linewidths of these levels. This condition can be satisfied by at least two ways. One can mix quasidegenerate levels with electromagnetic, low-frequency (microwave, millimeter-wave) field; in this case the rate of mixing of quasidegenerate levels can be designated as a Rabi frequency. In the second situation the coherent mixing of quasidegenerate levels can be provided through an auxiliary level. This can be due, for example, to dipole-dipole or resonance-exchange interactions (See Fig. 1).

The parametric three-frequency mechanism (see Fig. 3) can provide only the amplification for definite phase of transfer of coherence between levels 2 and 3 and for time intervals $\Delta t \lesssim \omega_0/2\pi$, where ω_0 is the frequency between quasidegenerate levels [see conditions (24) and (B15)]. This means that such a process can be used to generate short pulses (for example, picosecond and femtosecond pulses). We have shown that the phase of transfer may be prescribed to the phase of generalized dipole or multipole moment of transition $2\rightarrow 3$; for producing this effect, the phase should be equal to π if the population of level 2 is greater than the population of level 3.

One of the conditions for producing the effect is the rough equality of the population differences η_{23}, η_{13}, and η_{12} ($\eta_{23} \propto \eta_{12}, \eta_{13}$). This can be provided by two ways: when the population of level 1 is significantly higher than the population of levels 2 and 3, and in the same time the population of level 2 is much higher than the population of level 3. The asymmetry in populations of quasidegenerate levels in this case may be due, for example, to asymmetry in radiative characteristics ($\gamma_{31} \gg \gamma_{21}$). Thus, in such a situation one does not need to have laser pumping of levels 2 and 3. The second way is to pump the levels 2 and 3 to the degree at which their populations are closed to the population of level 1, so the population differences η_{23}, η_{13}, and η_{12} are approximately equal to each other. For the latter case, there is no requirement in population asymmetry of levels 2 and 3, but one needs laser pumping (which, however, does not necessarily provide the inverted population).

To provide the splitting of the laser transition, the application of constant electric or magnetic fields may be used. In both cases, there is an opportunity to govern the frequency of transition ω_0 between the quasidegenerate levels. In such situations the rate of coherent mixing of

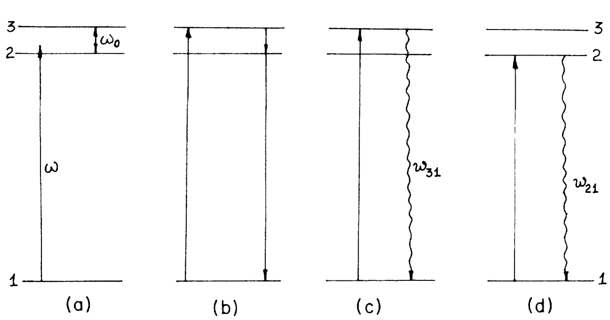

FIG. 3. The schematic diagram of the three-frequency parametric process for producing laser action without inversion. Parametric process (b) induced by laser field ω and coherent-mixing field ω_0 is more efficient than the absorption on the transitions $1\rightarrow 3$ (c) and $1\rightarrow 2$ (d).

quasidegenerate transitions depends on the value of splitting (frequency difference between the split transitions); there is an optimal field amplitude for providing the maximum effect. One has to make detailed calculations for the valid prediction of concrete laser systems working without inversion of level population. These must take into account the mechanisms of coherence transfer between two quasidegenerate levels.

We are now going to give some general examples of appropriate media based on the results of our considerations.

Laser action without inversion may be realized in free atoms, molecules, or ions if they contain two levels that are quasidegenerate with respect to magnetic quantum number or angular moment, and if they exchange very fast their excitations with the third level that is random degenerate with these two levels. Another widely spread possibility is when these two levels are embedded in continuum and exchange energy with it. In this case, if the radiative characteristics of quasidegenerate transitions are different, the effect of laser amplification based on the four-frequency mechanism may be achieved. By applying a constant electric or magnetic field, we can increase the energetic difference between quasidegenerate transitions and asymmetry in their radiative characteristics. Systems described on hydrogenlike ions may serve as an example of a promising laser medium for vacuum-ultraviolet and soft-x-ray regions.

There is also an opportunity for constructing lasers without inversion on solid-state materials, which are in general more rich for the realization of different energetic level configurations. For example, when two narrow quasidegenerate levels are embedded in quasifree states (low-density state conduction band), the rate of coherent exchange between these levels with continuum may be very high, and provide the sufficient rate of mixing of quasidegenerate levels (Ω_{23}).

The parametric mechanism (which has never been treated before) has some principally new applications. Because the effect in this case is due to the interference of two noncoherent processes and because it depends on the phase of transfer of coherence between the split transitions, this mechanism can be used for the creation of spatial grating, the period of which is defined by frequency difference between the quasidegenerate levels. Such systems may be very promising in the design of new electro-optical devices such as beam deflectors, devices for visualization of electric field distribution, and new types of optical storage. We will discuss these opportunities in our next publications.

APPENDIX A

For the second situation (the levels 2 and 3 interact with each other through level 5) let us analyze the equations for the population of levels 2 and 3 [(2e) and (2f)]:

$$\frac{d\rho_{22}}{dt} - w_{42}\rho_{44} + w_{21}\rho_{22} = i\hbar^{-1}(V_{12}\rho_{21} - \rho_{12}V_{21}) - i(\Omega_{25}\rho_{52} - \rho_{25}\Omega_{52}) , \quad \text{(A1a)}$$

$$\frac{d\rho_{33}}{dt}-w_{43}\rho_{44}+w_{31}\rho_{33} = i\hbar^{-1}(V_{13}\rho_{31}-\rho_{13}V_{31})-i(\Omega_{35}\rho_{53}-\rho_{35}\Omega_{53}) . \quad \text{(A1b)}$$

The right-hand sides of Eqs. (A1) contain the terms proportional to $\Omega_{i5}=\hbar^{-1}V_{i5}$ and $\Omega_{5i}=\hbar^{-1}V_{5i}$ which describe energy exchange between levels 2 and 3 and 5. These terms can describe different processes of energy transfer, for example, dipole-dipole transfer, resonance-exchange transfer, and the transfer between narrow quasidegenerate levels embedded in quasifree states in solids. These processes of energy transfer between the levels 2, 3, and 5 serve as the perturbations of a stochastic nature. The standard procedure of quantum-statistical-mechanical theory of relaxation[8] allows us to obtain from (A1) the following equations for the steady-state response to such perturbations:

$$\frac{d\rho_{22}}{dt}-w_{42}\rho_{44}+w_{21}\rho_{22}+w_{25}\rho_{22}-w_{52}\rho_{55} = i\hbar^{-1}(V_{12}\rho_{21}-\rho_{12}V_{21}) , \quad \text{(A2a)}$$

$$\frac{d\rho_{33}}{dt}-w_{43}\rho_{44}+w_{31}\rho_{33}+w_{35}\rho_{22}-w_{53}\rho_{55} = i\hbar^{-1}(V_{13}\rho_{31}-\rho_{13}V_{31}) , \quad \text{(A2b)}$$

where the probabilities of transitions (the relaxation rates) w_{i5} and w_{5i} are of the order of $|\Omega_{i5}|$ and $|\Omega_{5i}|$. We note that if we continue to consider our five-level system in the terms of rate constants w_{i5} and w_{5i} we could never derive the conditions of the effect of laser action without inversion, because this effect does not exist in this approximation. We will use Eqs. (A2) only for estimation of effective Rabi frequencies Ω_{23} and Ω_{32} while solving the problem within the framework of density-matrix formalism. For such estimation let us first define w_{23} and w_{32} through w_{i5} and w_{5i}.

We assume that w_{i5} and w_{5i} are much larger than $w_{31}, w_{21}, w_{43}, w_{42}, w_{51}$. This assumption is justified for most examples of the energy-transfer mechanism which we have been given before. The rates of the dipole-dipole and resonance-exchange mechanisms can reach the order of $10^{13}-10^{14}$ $\sec^{-1}$;[9] the probabilities of nonradiative electron excitation transfer in solids are the same order.[9] Taking this into account we reduce Eqs. (A2) to the form

$$\frac{d\rho_{22}}{dt}+w_{25}\rho_{22}-w_{52}\rho_{55}=0 , \quad \text{(A3a)}$$

$$\frac{d\rho_{33}}{dt}+w_{35}\rho_{33}-w_{53}\rho_{55}=0 . \quad \text{(A3b)}$$

The equilibrium state of these equations ($d\rho_{22}/dt=0$, $d\rho_{33}/dt=0$) is

$$\rho_{22}=w_{52}\rho_{55}/w_{25}=\rho_{33}w_{35}w_{52}/w_{25}w_{53} . \quad \text{(A4)}$$

Since the rates w_{i5} and w_{5i} are much faster than all other rates w_{ij} in our five-level system we can replace Eqs. (A3) by an effective system

$$\frac{d\rho_{22}}{dt}+w_{23}\rho_{22}-w_{32}\rho_{33}=0 , \quad \text{(A5a)}$$

$$\frac{d\rho_{33}}{dt}+w_{32}\rho_{33}-w_{23}\rho_{22}=0 . \quad \text{(A5b)}$$

The steady-state solution of this system can be easily found:

$$\rho_{22}=w_{32}\rho_{33}/w_{23} . \quad \text{(A6)}$$

The comparison of (A4) and (A6) gives

$$w_{32}=(w_{35}w_{52})^{1/2}, \quad w_{23}=(w_{25}w_{53})^{1/2} . \quad \text{(A7)}$$

Thus we can estimate the rate of mixing of levels 2 and 3 through level 5 by introducing effective rate constants w_{32} and w_{23} defined in (A7).

Now for the estimation of effective Rabi frequencies $\Omega_{23}=\hbar^{-1}V_{23}$, $\Omega_{32}=\hbar^{-1}V_{32}$ [see (A1)] we can use (A7) and experimental data for the values of rate constants w_{i5}, w_{5i}:

$$\begin{aligned} |\Omega_{23}| &\propto w_{23}=(w_{25}w_{53})^{1/2} , \\ |\Omega_{32}| &\propto w_{32}=(w_{35}w_{52})^{1/2} . \end{aligned} \quad \text{(A8)}$$

By this way we have reduced the problem in situation (b) to the problem in situation (a).

APPENDIX B

We derive now the conditions of amplification without inversion from inequality (23). We allow $\delta_{12}>0$, $\delta_{13}<0$ and introduce the notations $\delta_{12}=|\delta_{12}|$, $\delta_{13}=|\delta_{13}|$ for which we obtain from (14)

$$\begin{aligned} a &=\gamma_{12}\gamma_{13}+|\Omega_{23}|^2+\delta_{12}\delta_{13} , \\ b &=(\delta_{12}\gamma_{13}-\delta_{13}\gamma_{12}) . \end{aligned} \quad \text{(B1)}$$

In the notations (B1) we obtain the following condition instead of condition (23):

$$\begin{aligned} & a|\Omega_{12}|^2\eta_{12}\gamma_{13}+a|\Omega_{13}|^2\eta_{13}\gamma_{12}-b\delta_{13}|\Omega_{12}|^2\eta_{12}+b\delta_{12}|\Omega_{13}|^2\eta_{13} \\ & +a|\Omega_{23}|^2\eta_{23}\gamma_{23}^{-1}(|\Omega_{12}|^2-|\Omega_{13}|^2)+a|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)\eta_{23}\gamma_{23}^{-1}(\delta_{13}+\delta_{12}) \\ & -b|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)\eta_{23}\gamma_{23}^{-1}(\gamma_{13}-\gamma_{12})-b|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)(\eta_{13}+\eta_{12})<0 . \end{aligned} \quad \text{(B2)}$$

We assume now that

$$|\Omega_{23}|^2 \gg \gamma_{12}\gamma_{13} ,$$
$$|\Omega_{23}|^2 \gg \delta_{12}\delta_{13} , \quad \text{(B3)}$$
$$b=(\delta_{12}\gamma_{13}-\delta_{13}\gamma_{12})=0 ,$$

then $a \simeq |\Omega_{23}|^2$, and we obtain from (B2)

$$-|\Omega_{23}|^2\eta_{23}\gamma_{23}^{-1}(|\Omega_{12}|^2-|\Omega_{13}|^2) - |\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)\eta_{23}\gamma_{23}^{-1}(\delta_{13}+\delta_{12}) > |\Omega_{12}|^2\eta_{12}\gamma_{13}+|\Omega_{13}|^2\eta_{13}\gamma_{12} . \quad \text{(B4)}$$

(a) For the case when four-frequency interaction dominates, we let γ_{23} be of the order of γ_{12} and γ_{13}, and η_{23} be of the order of η_{12},η_{13}. We also assume the average over the time value of $\cos\varphi=0$, or $\cos\varphi<0$, or φ is arbitrary but

$$|\Omega_{23}| \gg \delta_{12}+\delta_{13} . \quad \text{(B5)}$$

Then the condition (B4) is reduced to the following one:

$$|\Omega_{23}|^2 c > \gamma_{13}\gamma_{12} ,$$

where

$$c=\frac{\eta_{23}(|\Omega_{13}|^2-|\Omega_{12}|^2)}{(|\Omega_{12}|^2\eta_{12}+|\Omega_{13}|^2\eta_{13})} . \quad \text{(B6)}$$

The inequality (B6) is satisfied for

$$c \propto 1, \quad \eta_{23} \propto \eta_{12},\eta_{13} \quad (\text{for } \rho_{22}>\rho_{33}) . \quad \text{(B7)}$$

In the particular case $\delta_{12}=0$ ($\delta_{13}=-|\delta_{13}|$), which was considered in Ref. 1, we can also assume

$$|\Omega_{23}|^2 \gg \gamma_{12}\gamma_{13} \quad \text{(B8)}$$

and rewrite (B2) in the form

$$|\Omega_{23}|^2(|\Omega_{12}|^2\eta_{12}\gamma_{13}+|\Omega_{13}|^2\eta_{13}\gamma_{12})+|\delta_{13}|^2\gamma_{12}|\Omega_{12}|^2\eta_{12}-|\Omega_{23}|^2\gamma_{23}^{-1}|\Omega_{23}|^2\eta_{23}|\Omega_{13}|^2 < |\Omega_{23}|^2\gamma_{23}^{-1}\eta_{23}|\delta_{13}|\,|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)-|\delta_{13}|\,|\Omega_{23}|^2\gamma_{13}\gamma_{23}^{-1}|\Omega_{12}\Omega_{23}\Omega_{31}|f(t)(\cos\varphi)\eta_{23} - |\delta_{13}|\gamma_{12}|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)(\eta_{13}+\eta_{12}) . \quad \text{(B9)}$$

It is easy to show that this inequality can be satisfied for

$$|\Omega_{23}|^2 \gg |\delta_{13}|^2, \quad \gamma_{12} \ll \gamma_{13}, \quad |\Omega_{12}|^2 \ll |\Omega_{13}|^2, \quad \eta_{23} \propto \eta_{12},\eta_{13} . \quad \text{(B10)}$$

(b) Now we consider the case when three-frequency interaction dominates. Let $b=0$, $|\Omega_{12}|^2 \simeq |\Omega_{13}|^2$, and $\gamma_{12} \simeq \gamma_{13}$. Then we reduce (B4) to the form

$$-|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)(\cos\varphi)\eta_{23}\gamma_{23}^{-1}(\delta_{13}+\delta_{12}) > |\Omega_{12}|^2\eta_{12}\gamma_{13}+|\Omega_{13}|^2\eta_{13}\gamma_{12} . \quad \text{(B11)}$$

For $\varphi=\pi$ we obtain

$$|\Omega_{13}\Omega_{32}\Omega_{21}|f(t)\eta_{23}\gamma_{23}^{-1}\omega_{32} > |\Omega_{12}|^2\eta_{12}\gamma_{13}+|\Omega_{13}|^2\eta_{13}\gamma_{12} \quad \text{(B12)}$$

or

$$|\Omega_{23}|f(t)\omega_{32}\{\eta_{23}|\Omega_{13}\Omega_{21}|/(|\Omega_{12}|^2\eta_{12}\gamma_{13}+|\Omega_{13}|^2\eta_{13}\gamma_{12})\} > \gamma_{12}\gamma_{13} . \quad \text{(B13)}$$

At

$$|\Omega_{23}|f(t)\omega_{32} \gg \gamma_{12}\gamma_{13} \quad \text{(B14)}$$

the inequality (B14) is satisfied for

$$|\omega_{23}| \propto \gamma_{12},\gamma_{13}, \quad f(t) \propto 1, \quad |\Omega_{23}| \gg \gamma_{12},\gamma_{13}$$

and

$$\eta_{23} \propto \eta_{12},\eta_{13} \quad (\rho_{22}>\rho_{33}) . \quad \text{(B15)}$$

For $\eta_{23}<0$ ($\rho_{22}<\rho_{33}$) the required phase is $\varphi=0$.

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