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Narrow nonlinear nonresonances in a three-level system

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We investigate the possibility of transferring a narrow line-shape singularity from one of the transitions of a three-level gas system to an adjacent transition via coupling through a common level. The density matrix is determined at arbitrary saturation with respect to both resonant fields, neglecting higher spatial harmonics. Explicit expressions are obtained for the nonlinear susceptibilities at small saturation in the Doppler limit. The contrast and width of the transferred singularity (formed by an absorbing cell) in two-frequency generation on standing waves are determined. The system He–Ne/CH₄ is considered by way of example.

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1. INTRODUCTION

It is known^{1,2} that the use of narrow nonlinear resonances which arise when a low-pressure absorbing cell is placed inside the resonator, makes it possible to form on the broad $(\sim ku_0)$ Doppler contour an amplification peak (an inverted Lamb dip) with a width determined by the homogeneous broadening γ of the absorption line. A classical system of this type is the amplifying medium He-Ne with a methane absorbing cell $(\lambda = 3.39 \ \mu m)$.² An attempt can be made to get around the difficulties of producing such a system for shorter wavelengths, for example in the optical band, which are connected with the absence of such narrow absorption lines, by using the idea³ of transferring the narrow Lamb detail to an adjacent high-frequency transition in a three-level active medium. Owing to the presence of the common level, the adjacent transitions compete, therefore a narrow resonance on one of the transitions should appear also on the adjacent one.

This possibility is theoretically investigated in the present paper. Expressions are obtained for the density matrix, describing the gas three-level system in a resonant field,^{1,4} in the case of waves with arbitrary amplitudes and standing-wave coefficient. The non-linear susceptibilities averaged over the velocities are obtained for small saturation parameters. In the Doppler limit $ku_0 \gg \gamma$, expressions are obtained for the line shape, and they are analytically investigated for small detunings from resonance. It is shown that the appearance of a narrow (less than the homogeneous width) singularity on one of the transitions can lead to the appearance of a singularity on an adjacent transition. It is useful to employ two-frequency lasing for

this purpose.⁵⁻¹⁰ The contrast and width of the "transferred" singularity in two-frequency lasing on the standing waves are determined. It is shown that for the system He-Ne/CH₄ the transferred singularity is a dip rather than a peak, in contrast to the assumption made in Ref. 3.

2. DENSITY MATRIX IN RESONANT FIELDS

The equation of motion for the single-particle density matrix, which is an operator in the internal variables and a classical distribution function in the coordinates of the center of gravity of the particles, takes the form^{1,11}

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla\right) = -\frac{i}{\hbar} [\hat{H}, \hat{\rho}] - \hat{\gamma}(\hat{\rho} - \hat{\rho}_{o}), \qquad (2.1)$$

where $\hat{H} = \hat{H}_0 + \hat{V}(z, t)$ is the Hamiltonian of the particle; $\hat{\gamma}$ is the matrix of the relaxation constants,

$$(\gamma \rho)_{nm} = \gamma_{nm} \rho_{nm},$$

 $\hat{\rho}_0$ is the stationary density matrix in the absence of a field. In the presence of resonant fields, the matrix elements of the perturbation are equal to

$$V_{nm}(z,t) = \sum_{\alpha = \pm 1} V_{nm}^{\alpha} \exp\{i(\alpha k_{nm} z - \Omega_{nm} t)\} + \text{c.c.}, \qquad (2.2)$$

where $V_{nm}^{\alpha} = -\mathbf{d}_{nm}\mathbf{E}_{nm}^{\alpha}$, \mathbf{d}_{nm} is the dipole moment, \mathbf{E}_{nm}^{α} is the amplitude of the resonant field on the n-m transition, the superior index $\alpha = \pm 1$ distinguishes between waves traveling in opposite directions; Ω_{nm} are their frequencies, and k_{nm} are the wave numbers, with Ω_{nm} $= -\Omega_{mn}$, $k_{nm} = -k_{mn}$. As follows from the structure of (2.1), stationary solutions in the resonance approximation with allowance for the lower spatial harmonics¹ can be sought in the form

$$\rho_{nm}(z,t) = \sum_{\alpha=\pm i} \sigma_{nm}^{\alpha} \exp\{i(\alpha k_{nm}z - \Omega_{nm}t)\}, \quad (\sigma_{nm}^{\alpha})^* = \sigma_{mn}^{\alpha}, \quad (2.3)$$

where σ_{nm}^{α} depend slowly on the coordinates and on the time. The quantities k_{nm} and Ω_{nm} are defined such that the following equations hold:

$$\Omega_{nl} + \Omega_{lm} = \Omega_{nm}, \quad k_{nl} + k_{lm} = k_{nm}, \tag{2.4}$$

with corresponding analogous equations for the detunings

$$\Delta \omega_{nm} = \omega_{nm} - \Omega_{nm} \tag{2.5}$$

from the Bohr frequencies ω_{nm} of the transitions. The use of this notation is quite convenient in the resonance approximation. Substituting (2.3) in (2.1) and taking (2.4) and (2.5) into account, we obtain

$$\left(\frac{\partial}{\partial t}+i\Delta\omega_{nm}+i\alpha k_{nm}\upsilon+\gamma_{nm}\right)\sigma_{nm}^{\alpha}+\frac{i}{\hbar}\sum_{k}\left(V_{nk}^{\alpha}\sigma_{km}^{\alpha}-\sigma_{nk}^{\alpha}V_{km}^{\alpha}\right)=0.$$
(2.6)

and

$$\left(\frac{\partial}{\partial t}+\gamma_{nn}\right)\sigma_{nn}+\frac{i}{\hbar}\sum_{\mathbf{k},\mathbf{\beta}}\left(V_{n\mathbf{k}}{}^{\mathbf{\beta}}\sigma_{\mathbf{k}n}{}^{\mathbf{\beta}}-V_{\mathbf{k}n}{}^{\mathbf{\beta}}\sigma_{n\mathbf{k}}{}^{\mathbf{\beta}}\right)=\gamma_{nn}\rho_{nn}{}^{\mathbf{0}}, \quad n=m.$$
(2.7)

Regarding (2.6) as a system of equations for the offdiagonal elements of the matrix, we express them in terms of the diagonal elements that include the unperturbed values $\rho_{nn}^0 \equiv \sigma_n^0$, $\sigma_{nn} \equiv \sigma_n$. It is important here that (2.6) splits into two subsystems, each of which describes (at given σ_n) the response of the system to waves traveling in the same direction.

We confine ourselves to the case when the 2-1 transition is forbidden (Fig. 1), i.e., $V_{21}=0$ (this situation is realized, for example, in neon for the transition $2p_4 - 3p_4$ in the system $2p_4 - 3p_4 - 3s_2$). At $V_{21}=0$, these subsystems (of sixth order) split into two subsystems (of third order) for $\sigma_{31}^{\alpha}, \sigma_{21}^{\alpha}, \sigma_{23}^{\alpha}$ and $\sigma_{13}^{\alpha}, \sigma_{12}^{\alpha}, \sigma_{32}^{\alpha}$, where $\alpha = \pm 1$. Waves traveling in opposite directions already turn out to be coupled in Eqs. (2.7). The solution for arbitrary fields is given in Appendix I. In the case of weak fields we can expand σ_{nm}^{α} in the saturation parameters G_{nm}^{α} :

$$G_{nm}^{\alpha} = |V_{nm}^{\alpha}|^{2}/\hbar^{2}\tilde{\gamma}_{nm}^{2} \ll 1, \quad \tilde{\gamma}_{nm}^{2} = 2\gamma_{nm}\gamma_{n}\gamma_{m}/(\gamma_{n}+\gamma_{m}).$$
(2.8)

In accordance with the results of Refs. 4, 6, and 11 we have

$$\sigma_{s1}{}^{\alpha} = -\frac{d_{s1}E_{s1}{}^{\alpha}}{\hbar\delta_{s1}{}^{\alpha}} \left[\left(1 - 4\sum_{\rho} G_{s1}{}^{\rho} \frac{\gamma_{s1}{}^{2}}{|\delta_{s1}{}^{\rho}|^{2}} \right) n_{s1}{}^{\rho} - 2\sum_{\rho} G_{s2}{}^{\rho} \frac{\gamma_{s2}{}^{2}}{\gamma_{s2}{}^{+}} \frac{\gamma_{s2}{}^{2}}{|\delta_{s2}{}^{\rho}|^{2}} n_{s2}{}^{-} G_{s2}{}^{\alpha} \overline{\gamma_{s2}{}^{2}} \frac{n_{s1}{}^{0} \delta_{23}{}^{\alpha} - n_{23}{}^{0} \delta_{31}{}^{\alpha}}{\delta_{23}{}^{\alpha} \delta_{23}{}^{\alpha}} \right],$$
(2.9)

$$\sigma_{32}^{\alpha} = -\frac{\mathbf{d}_{32}\mathbf{E}_{32}^{\alpha}}{\hbar\delta_{32}^{\alpha}} \Big[\left(1 - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac{1}{2} \left(\frac{1}{2} - 4\sum_{\rho} G_{32}^{\rho} \frac{\gamma_{32}^{2}}{|\delta_{32}^{\rho}|^{2}} \right) n_{32}^{\alpha} + \frac$$

$$2\sum_{\rho}G_{31}^{\rho}\frac{\gamma_{1}}{\gamma_{31}^{+}}\frac{\gamma_{31}^{2}}{|\delta_{31}^{\rho}|^{2}}n_{31}^{\rho}-G_{31}^{\alpha}\tilde{\gamma}_{31}^{2}\frac{n_{32}^{0}\delta_{13}^{\alpha}-n_{13}^{0}\delta_{32}^{\alpha}}{\delta_{13}^{\alpha}\delta_{12}^{\alpha}\delta_{32}^{\alpha}}\Big], \qquad (2.10)$$

$$\sigma_{21}^{\alpha} = -\frac{(\mathbf{d}_{31}\mathbf{E}_{31}^{\alpha})(\mathbf{d}_{32}\mathbf{E}_{32}^{\alpha})}{\hbar^{2}} \cdot \frac{n_{31}^{0}\delta_{23}^{\alpha} - n_{23}^{0}\delta_{31}^{\alpha}}{\delta_{21}^{\alpha}\delta_{23}^{\alpha}}, \qquad (2.11)$$

where $\delta_{nm}^{\alpha} = \Delta \omega_{nm} + \alpha k_{nm}v - i\gamma_{nm}$ are the complex detunings with allowance for the Doppler effect and the damping, $\gamma_{nm}^{\star} = (\gamma_n + \gamma_m)/2; n_{nm}^0 \equiv \rho_{nn}^0 - \rho_{mm}^0$ are the stationary differences of the populations in the absence of the field.



FIG. 1.

The first terms in (2.9) and (2.10) describe the linear response, the second terms describe the saturation due to the transitions under the influence of the intrinsic field. The third and fourth terms are due respectively to a steplike and two-photon (coherent) transitions.¹ Owing to the absence of an external field for the 2 - 1 transition, a nonzero matrix element σ_{21}^{α} appears in second order perturbation theory in the field on account of two-photon processes.

Equations (2.9) and (2.10) contain the denominators δ_{31}^{α} and δ_{32}^{α} , which are responsible for the nonlinear resonances that occur when the detunings $\Delta \omega_{31} \pm k_{31}v$ and $\Delta \omega_{32} \pm k_{32}v$ vanish simultaneously, i.e., in the case when both fields interact with one and the same group of particles from the Maxwellian distribution:

$$\frac{\Delta \omega_{31}}{k_{31}} = \pm \frac{\Delta \omega_{32}}{k_{32}},$$

$$|v| = \left| \frac{\Delta \omega_{31}}{k_{31}} \right| = \left| \frac{\Delta \omega_{32}}{k_{32}} \right|.$$
(2.12)

The plus (minus) sign in (2.12) corresponds in this case to resonance with the co-moving (opposing) waves.

3. NONLINEAR SUSCEPTIBILITIES IN RESONANT FIELDS

To determine the nonlinear susceptibilities χ , we must find the polarization of the medium p(z, t)

 $\mathbf{P}(z,t) = \operatorname{Sp} \langle \hat{\mathbf{d}\rho} \rangle,$

where the angle brackets denote averaging over the velocities of the particles with Maxwellian distribution

$$\langle A(z,t) \rangle = \int_{-\infty}^{\infty} A(z,v,t) W(v) dv, \quad W(v) = \frac{1}{\sqrt{\pi} u_0} e^{-v^{t/u_0}t}.$$
 (3.1)

Taking (2.3) into account, we obtain

$$\mathbf{P}(z,t) = \sum_{\substack{n,m \\ \alpha = \pm 1}} \mathbf{P}_{nm}^{\alpha} \exp\{i(\alpha k_{nm} z - \Omega_{nm} t)\}, \qquad (3.2)$$

with

$$(P_{nm}^{\alpha})_{j} = (\chi_{nm}^{\alpha})_{ji} (E_{nm}^{\alpha})_{ii}.$$

$$(3.3)$$

Assuming for simplicity that all waves are plane-polarized, we shall omit the tensor indices from now on .

In the general case of strong fields (see Appendix I), averaging of the density matrix over the velocities calls for numerical integration. For weak fields, however, this averaging can be carried out analytically. At an arbitrary ratio of the homogeneous and Doppler widths, the result of the averaging is expressed in terms of the tabulated probability integral $w((\Delta\omega + i\gamma)/ku_0)$. In the Doppler limit $|k_{nm}|u_0 \gg \gamma_{nm}$ the result of the integration,

as is well known, is expressed in terms of elementary functions.¹ We note, however, that in neon this relation between the thermal and homogeneous widths is not always valid. For example, it is satisfied for the $3s_2 - 2p_4$ transition, but not for $3s_2 - 3p_4$.

We present here the polarizability for the most interesting case of small detunings of the field relative to the centers of the Doppler contours $|\Delta\omega_{nm}|$ $\ll |k_{nm}|u_0$. The main contribution to the integrals of (3.1), which determined the nonlinear susceptibilities, is made by the region of low velocities $k_{nm}v \sim \Delta\omega_{nm}$ and $\gamma_{nm} \ll |k_{nm}|u_0$; the exponential in the Maxwellian distribution can therefore be set equal to unity. Under this assumption, the integrals can be calculated explicitly. In terms of the type

$$\int_{-\infty}^{\infty} \frac{dv}{(v-v_1)(v-v_2)(v-v_3)}$$

a nonzero contribution is made by those which have poles on opposite sides of the real axis in the complex velocity plane. After averaging, we obtain for the susceptibilities, at an arbitrary ratio of the amplitudes of the opposing waves,

$$\chi_{c}^{\alpha} = \chi_{a}^{\alpha} - \sum_{b,\beta} \chi_{ab}^{\alpha\beta} \frac{|E_{b}^{\beta}|^{2}}{4\pi}; \quad \beta, \alpha = \pm 1, \quad a, b = 1, 2,$$
(3.4)

where we have left out the index 3 corresponding to the common upper level, $\chi_a^{\alpha} \equiv \chi_{3a}^{\alpha}$, $\mathbf{E}_a^{\alpha} \equiv \mathbf{E}_{3a}^{\alpha}$. The coefficients $\chi_{alb}^{\alpha\beta} \equiv \chi_{3a3b}^{\alpha\beta}$ have the symmetry property $\chi_{alb}^{-\alpha-\beta} = \chi_{alb}^{\alpha\beta}$ and are equal to:

$$\chi_{a|a}^{a\beta} = \theta_{a} \frac{|d_{aa}|^{2}}{\hbar^{2}} \frac{\gamma_{3a}^{+}}{\gamma_{3}\gamma_{a}} \left[\frac{|\alpha - \beta|}{2} \Delta \omega_{3a} - i\gamma_{3a} \right]^{-1}$$
(3.5)

for the "opposing and co-moving" waves on one transition;

$$\chi_{a|b}^{+-} = \theta_{b} \frac{|d_{3a}|^{2}}{\hbar^{2}} \frac{1}{\gamma_{3}} \frac{1}{\Delta_{ab} - i\Gamma_{3a3b}}; \quad a \neq b$$
(3.6)

for "opposing" waves on adjacent transitions;

$$\chi_{1|2}^{++} = \theta_{2} \frac{|d_{31}|^{2}}{\hbar^{2}} \frac{1}{\gamma_{3}} \left[\frac{1}{\Delta_{12}^{+} - i\Gamma_{3133}} + \frac{-i\gamma_{3}}{(\Delta_{12}^{+} - i\Gamma_{3132})(\Delta_{13}^{+} - i\Gamma_{3132})} \right], \quad (3.7)$$

$$\chi_{3|1}^{++} = \theta_{1} \frac{|d_{32}|^{2}}{\hbar^{2}} \frac{1}{\gamma_{3}} \left[\frac{1}{\Delta_{31}^{+} - i\Gamma_{3234}} + \frac{-i\gamma_{3}}{(\Delta_{21}^{+} - i\Gamma_{3232})(\Delta_{21}^{+} - i\Gamma_{3232})} + \frac{-i\gamma_{3}}{(\Delta_{21}^{+} - i\Gamma_{3232})^{2}} \right] \quad (3.8)$$

for "co-moving" waves on adjacent transitions. In this case

$$\chi_{a}^{o} = \frac{\theta_{a}}{2\gamma_{5a}} \left(\Delta \omega_{3a} + i\gamma_{5a} \right)^{-1}, \quad \theta_{a} = -2\pi^{\nu_{i}} n_{3a}^{o} |d_{3a}|^{2} \frac{1}{\hbar k_{3a} u_{0}},$$

$$\Delta_{ab}^{\pm} = \Delta \omega_{3a} \pm \frac{k_{3a}}{k_{3b}} \Delta \omega_{3b} = \pm \frac{k_{3a}}{k_{3b}} \Delta_{ba}^{\pm}, \quad \Gamma_{iklm} = \gamma_{ik} \pm \frac{k_{ik}}{k_{im}} \gamma_{im} = \frac{k_{ik}}{k_{im}} \Gamma_{lmik}.$$
(3.9)

In the particular case of standing waves $E_a^* = E_a^- = E_a$ and, as follows from (3.4), the dependence of the susceptibility on the propagation direction of the wave vanishes:

$$\chi_{a} = \chi_{a}^{0} - \sum_{b} \chi_{a|b} \frac{|E_{b}|^{2}}{4\pi}, \quad \chi_{a|b} = \sum_{b} \chi_{a|b}^{+\beta}. \quad (3.4')$$

Additional averaging over the orientation of the molecule can be carried out directly in expressions (3.5)-(3.8). For example, at identical polarization of the

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field on both transitions it is necessary to make the substitutions $|d_{mn}|^2 - \frac{1}{3}|d_{mn}|^2$, $|d_{mn}|^2|d_{kl}|^2 - \frac{1}{5}|d_{mn}|^2|d_{kl}|^2$.

4. TWO-FREQUENCY LASING IN THE PRESENCE OF AN ABSORBING CELL, AND TRANSFER OF SINGULARITY TO AN ADJACENT TRANSITION

The transfer of a narrow singularity produced by an absorbing cell on the transition $3 \rightarrow 2$ (Ref. 1) to the adjacent transition $3 \rightarrow 1$ can be effective in the two-frequency lasing regime. Confining ourselves to the case of standing waves and small saturation, we write down the equations for the intensities $I_a(t)$

 $= (c/4\pi)|E_a(t)|^2$, assuming that a single-mode regime is realized on each transition

$$\frac{1}{c}\frac{\partial I_{1}}{\partial t} + \frac{k_{21}}{Q_{1}}I_{1} = \varkappa_{1}I_{1}, \qquad \varkappa_{a} = \varkappa_{a}^{0} - \sum_{b} \varkappa_{ab}I_{b},$$

$$\frac{1}{c}\frac{\partial I_{2}}{\partial t} + \frac{k_{22}}{Q_{2}}I_{2} = [\varkappa_{2} - \varkappa(\Delta\omega)]I_{2}, \qquad \varkappa(\Delta\omega) = \varkappa^{0} - \varkappa'(\Delta\omega)I_{2}.$$
(4.1)

Here $\kappa_a = -4\pi k_{3a} \operatorname{Im} \chi_a$ is the gain of the three-level system on the transition $3 \rightarrow a$, $\kappa_{ab} = -(4\pi k_{3a}/c) \operatorname{Im} \chi_{a|b}$ [see (3.4')], $\kappa(\Delta \omega)$ is the absorption coefficient introduced by the two-level system,¹ with

$$\varkappa'(\Delta\omega) = \frac{\varkappa'}{2} \left[1 + L\left(\frac{\Delta\omega}{\gamma}\right) \right], \quad L(x) = (1 + x^{2})^{-1}, \quad (4.2)$$

where $\Delta \omega = \Omega_2 - \omega_{abs}$ is the detuning of the lasing frequency from the transition frequency ω_{abs} , γ is the homogeneous absorption line width ($\gamma \ll \gamma_{nm}$), Q_a is the Q of the mode, and the quantities in (4.1) are assumed averaged over the resonator length.

The solution of the system (4.1), corresponding to the stationary regime of the two-frequency generation, is of the form

$$I_{1}^{\circ}(\Delta\omega) = \frac{\alpha_{1}(\varkappa_{22}-\varkappa'(\Delta\omega)) - \alpha_{2}\varkappa_{12}}{\varkappa_{11}(\varkappa_{22}-\varkappa'(\Delta\omega)) - \varkappa_{12}\varkappa_{21}} = \frac{D_{1}(\Delta\omega)}{D_{0}(\Delta\omega)} > 0,$$

$$I_{2}^{\circ}(\Delta\omega) = \frac{\alpha_{2}\varkappa_{11} - \alpha_{1}\varkappa_{21}}{\varkappa_{11}(\varkappa_{22}-\varkappa'(\Delta\omega)) - \varkappa_{12}\varkappa_{21}} = \frac{D_{2}}{D_{0}(\Delta\omega)} > 0,$$
(4.3)

where $\alpha_1 = \varkappa_1 - k_{31}/Q_1$ or $\alpha_2 = \varkappa_2 - \varkappa^0 - k_{32}/Q_2$ is the excess of gain over the loss. This solution is stable only in region III on Fig. 2, where

$$D_1(\Delta \omega) > 0, \quad D_2 > 0, \quad D_0(\Delta \omega) > 0,$$
 (4.4)

as can be easily verified by investigating, with the aid of (4.1), the roots of the characteristic equation for small perturbations $\delta I_a(t) \sim e^{-pt}$. The mutual influence of the transitions² is due to the nonlinear cross co-



FIG. 2.

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efficients \varkappa_{12} and \varkappa_{21} , which in accordance with (3.6)– (3.8) are different from zero only near the nonlinear resonances (2.12). Thus, the transfer of the singularity to an adjacent transition is possible in two "interaction bands"

$$\left|\frac{\Delta\omega_{3_1}}{k_{3_1}}\pm\frac{\Delta\omega_{3_2}}{k_{3_2}}\right|\leq\frac{\gamma_{3_1}}{k_{3_1}},\quad\frac{\gamma_{3_2}}{k_{3_2}}$$

(see Fig. 3). The presence of a nonlinearly absorbing substance leads, according to (4.3) and (4.2), to formation of a narrow intensity peak $I_2^0(\Delta\omega)$ at $\Delta\omega = 0$ with width $\sim \gamma$, just as in the case of single-frequency lasing.^{11,1} As seen from (4.3) besides the narrow peak on the long-wave transition at $\Omega_2 = \omega_{abs}$, a narrow singularity is formed also on the short-wave transition, if the frequency Ω_1 satisfies the resonance condition. To determine the character of the transferred singularity, we introduce the contrast

$$h_a = 1 - I_a^{o}(0) / I_a^{o}(\infty), \quad a = 1, 2,$$
 (4.5)

where $I_a^0(\infty) = I_a^0(|\Delta \omega| \gg \gamma)$, and by virtue of $\gamma \ll \Gamma_{iklm}$ we can neglect the changes of $\Delta \omega_{31}$ and $\Delta \omega_{32}$ with changing $\Delta \omega$. From (4.3) we get

$$h_{1} = -\frac{\kappa_{12}}{\kappa_{11}} \frac{I_{2}^{\circ}(\infty)}{I_{2}^{\circ}(0)} h_{2}, \quad h_{2} = 1 - \frac{D_{0}(\infty)}{D_{0}(0)}, \quad (4.6)$$

or, using (4.2),

$$h_1 = \frac{\kappa'(0)\kappa_{12}D_2}{2D_0(0)D_1(\infty)}, \quad h_2 = -\frac{\kappa'(0)\kappa_{12}}{2D_0(0)}.$$
(4.7)

Taking (4.4) into account, we find from this that at \varkappa_{12} >0 ("normal competition") the singularities on the coupled transitions are of opposite sign, while at \varkappa_{12} <0 they have the same sign, i.e., a dip is transferred to the adjacent transition at $\kappa_{12} > 0$ and a spike at κ_{12} <0. Incoherent processes make a positive contribution to \varkappa_{12} [see (3.6) and the first term of (3.7)], while coherent ones make a negative contribution only at $(\Delta_{12}^*)^2 > \Gamma_{31,32}\Gamma_{21,32}$ [second term of (3.7)]. Therefore at small detunings compared with the homogeneous widths of the transition, the transferred singularity corresponds to a minimum of the intensity. For the transitions $3s_2 - 2p_4$ and $3s_2 - 3p_4$ in neon, for example, the coherent processes make a small contribution to κ_{12} (and κ_{21}) at all detunings, by virtue of $\gamma_3/\Gamma_{31,32}$ $\ll 1, \gamma_3/\Gamma_{21,32} \ll 1$, since $\gamma_3 \ll \gamma_{31}, \gamma_{32}$ and therefore in this case $\varkappa_{12} > 0$, and a dip is formed on the short-wave transition, rather than a peak as proposed in Ref. 3.



FIG. 3. I, II—bands of interaction of co-moving and opposing waves, III—absorption band, IV—resonator retuning line.

The width of the transferred singularities at half-maximum

$$\delta\omega = [D_0(0)/D_0(\infty)]^{\gamma_1}\gamma \qquad (4.8)$$

is of the order of γ , with $D_0(0)/D_0(\infty) \leq 1$.

We consider now the line shape when the lasing frequencies are changed by changing the resonator length l, disregarding both the linear and nonlinear frequency shifts, which in the case of the $He-Ne/CH_4$ system at a neon pressure 1 Torr do not exceed 5×10^4 Hz, which is less than the width of the singularity $\gamma/2\pi \approx 5 \cdot 10^5$ Hz.¹ Since the wavelengths λ_a satisfy, at integer Z_a , the condition $Z_a \lambda_a = 2l, a = 1, 2$, it follows that the lasing frequencies change in such a way that $\Omega_1/Z_1 = \Omega_2/Z_2$ (i.e., the retuning line goes parallel to the interaction band of the co-moving waves). On the shortwave transition there will be observed a narrow intensity dip at the frequency $\Omega_1 = (Z_1/Z_2)_{abs}$, with a width $\sim (k_{31}/k_{32})\delta\omega \sim (k_{31}/k_{32})\gamma$. Thus, under the conditions of single-mode lasing, the position of the transferred singularity can be calculated accurate to a value of the order of its width. On the other hand, if the linear and nonlinear frequency shifts are taken into account, the position of the singularity can be established with a relative error of the same order as on the long-wave transition.

In conclusion, we consider it our pleasant duty to thank V.S. Solov'ev, who has called our attention to the considered problem.

APPENDIX I

The solution of Eqs. (2.6) and (2.7) for the offdiagonal matrix elements is of the form

$$\sigma_{31}{}^{\alpha} = \frac{\left(\delta_{21}{}^{\alpha}\delta_{23}{}^{\alpha} - \hbar^{-2}|V_{31}{}^{\alpha}|^{2}\right)\left(\sigma_{3} - \sigma_{1}\right) + \hbar^{-2}|V_{32}{}^{\alpha}|^{2}\left(\sigma_{3} - \sigma_{2}\right)}{D^{\alpha}} \frac{V_{31}{}^{\alpha}}{\hbar}, \quad (I.1)$$

$$\sigma_{32}^{\alpha} = \frac{\left(\delta_{12}^{\alpha}\delta_{13}^{\alpha} - \hbar^{-2} | V_{32}^{\alpha} |^{2}\right) \left(\sigma_{3} - \sigma_{2}\right) + \hbar^{-2} | V_{31}^{\alpha} |^{2} \left(\sigma_{3} - \sigma_{1}\right)}{-(D^{\alpha})^{*}} \frac{V_{32}^{\alpha}}{\hbar}, \quad (I.2)$$

$$\sigma_{21}{}^{\alpha} = -\frac{\delta_{23}{}^{\alpha}(\sigma_{3}-\sigma_{1})+\delta_{31}{}^{\alpha}(\sigma_{3}-\sigma_{2})}{D^{\alpha}}\frac{V_{31}{}^{\alpha}(V_{32}{}^{\alpha})}{\hbar^{2}}.$$
 (I.3)

The differences of the diagonal elements, which enter in (I.1)-(I.3) are equal to

$$\sigma_{3} - \sigma_{2} = \mathcal{D}^{-1}[(1 + A_{1})(\sigma_{3}^{\circ} - \sigma_{2}^{\circ}) - B_{2}(\sigma_{3}^{\circ} - \sigma_{1}^{\circ})], \qquad (I.4)$$

$$\sigma_{3}-\sigma_{4}=\mathscr{D}^{-1}[(1+A_{2})(\sigma_{3}^{\circ}-\sigma_{1}^{\circ})-B_{4}(\sigma_{3}^{\circ}-\sigma_{2}^{\circ})], \qquad (I.5)$$

where

$$\begin{split} A_{i} &= 2 \sum_{\alpha} \frac{|V_{s1}^{\alpha}|^{2}}{\hbar^{2}} \operatorname{Im} \left[\frac{1}{D^{\alpha}} \frac{2\gamma_{s1}^{+}}{\gamma_{s}\gamma_{1}} \left(\delta_{21}^{\alpha} \delta_{23}^{\alpha} - \frac{|V_{s1}^{\alpha}|^{2}}{\hbar^{2}} + \frac{\gamma_{1}}{2\gamma_{31}^{+}} \frac{|V_{s2}^{\alpha}|^{2}}{\hbar^{2}} \right) \right], \\ A_{z} &= 2 \sum_{\alpha} \frac{|V_{s2}^{\alpha}|^{2}}{\hbar^{2}} \operatorname{Im} \left[\frac{1}{D^{\alpha}} \frac{2\gamma_{s2}^{+}}{\gamma_{s}\gamma_{2}} \left(\delta_{21}^{\alpha} \delta_{31}^{\alpha} - \frac{|V_{s2}^{\alpha}|^{2}}{\hbar^{2}} + \frac{\gamma_{2}}{2\gamma_{32}^{+}} \frac{|V_{31}^{\alpha}|^{2}}{\hbar^{2}} \right) \right], \\ B_{i} &= 2 \sum_{\alpha} \frac{|V_{s2}^{\alpha}|^{2}}{\hbar^{2}} \operatorname{Im} \left[\frac{1}{D^{\alpha}} \frac{1}{\gamma_{s}} \left(\delta_{21}^{\alpha} \delta_{31}^{\alpha} - \frac{|V_{s2}^{\alpha}|^{2}}{\hbar^{2}} + \frac{2\gamma_{s1}^{+}}{\gamma_{1}} \frac{|V_{s1}^{\alpha}|^{2}}{\hbar^{2}} \right) \right], \\ B_{z} &= 2 \sum_{\alpha} \frac{|V_{s1}^{\alpha}|^{2}}{\hbar^{2}} \operatorname{Im} \left[\frac{1}{D^{\alpha}} \frac{1}{\gamma_{s}} \left(\delta_{21}^{\alpha} \delta_{23}^{\alpha} - \frac{|V_{s1}^{\alpha}|^{2}}{\hbar^{2}} + \frac{2\gamma_{s2}^{+}}{\gamma_{2}} \frac{|V_{s2}^{\alpha}|^{2}}{\hbar^{2}} \right) \right], \\ \mathcal{D} &= (1+A_{1}) (1+A_{2}) - B_{i} B_{2}. \end{split}$$

Expanding (I.1)-(I.5) in terms of the small saturation parameters, we obtain the expressions (2.9)-(2.11).

APPENDIX II

Transfer of Singularity at Arbitrary Saturation Parameters

At G_{23}^{α} , $G_{31}^{\alpha} \ge 1$, the gains in (4.1) are given by the exact expressions $\varkappa_{\alpha} = \varkappa_{\alpha}(I_1, I_2, \Omega_1, \Omega_2)$, which can be obtained, for example, after averaging the expressions obtained in Appendix I over the velocities. The absorption coefficient is $\varkappa = \varkappa(\Delta \omega, I_2)$. Assuming $\varkappa \ll \varkappa_2$, we obtain the rapidly varying increments $\delta I_a(\Delta \omega)$ to the generation intensities, due to the nonlinear absorption:

$$\delta I_1(\Delta \omega) = -\eta_1 \delta I_2(\Delta \omega), \quad \delta I_2(\Delta \omega) = \frac{\kappa(\Delta \omega, I_2)}{\xi_2(1-\eta_1\eta_2)}, \quad (\text{II.1})$$

where

$$\begin{split} \xi_{a} &= \left(\frac{\partial \varkappa_{a}}{\partial I_{a}}\right), \quad \eta_{1} &= -\left(\frac{\partial I_{1}}{\partial I_{2}}\right)_{\varkappa_{1}}, \\ \eta_{3} &= -\left(\frac{\partial I_{2}}{\partial I_{1}}\right)_{\varkappa_{3}}, \end{split}$$

 I_1 and I_2 are the stationary lasing intensities in the absence of an absorbing cell, with \varkappa_a taken to be functions of only I_a after eliminating the frequencies Ω_a with the aid of the equations for the phases. The contrasts of the singularities

$$h_a = \left[\delta I_a(\infty) - \delta I_a(0)\right] / I_a \tag{II.2}$$

have opposite signs in the case of normal competition $\eta_1 > 0$, with

$$h_{i} = -\eta_{i} \frac{I_{2}}{I_{i}}, \quad h_{2} = \frac{\kappa(\infty, I_{2}) - \kappa(0, I_{2})}{\xi_{2}(1 - \eta_{1}\eta_{2})I_{2}}.$$
 (II.3)

From the condition of the stability of the generation when account is taken of the fact that $\xi_1, \xi_2 < 0$ (saturation under the influence of the intrinsic field) it follows that $1 - \eta_1 \eta_2 > 0$, so that a peak is formed on the transition 3-2, and the dip is transferred to the transition 3-1.

- ¹⁾Neglect of the higher spatial harmonics means smallness of the parameters $(\tilde{\gamma}_{3a}^2/\gamma_{3a}^2)G_{3a} \ll 1$, a = 1, 2 (cf. Ref. 12), where the saturation parameters G and $\tilde{\gamma}_{nm}$ are given in (2.8). At $\gamma_{3a}/\gamma_{3a} \ll 1$, which is the case, e.g., for neon,^{1,12} neglect of the harmonics is permissible also for a saturation $G \ge 1$ which is not small.
- ²⁾With the aid of (4.3) it is easy to investigate also the line shape for two-frequency lasing, in analogy with the procedure used by Melekhin⁷ for the transition scheme $3 \rightarrow 1$, $2 \rightarrow 1$.
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Reflection and refraction of a plane wave by the interface of two media, with allowance for positron polarization of the medium

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Integral equations are obtained for the propagation of electromagnetic waves in an atomic system in whose spectrum negative energy states are included as intermediate states (positron polarization of the medium). Additional terms that depend on the coherence properties of the medium are obtained in the Lorentz-Lorenz and Fresnel formulas for a plane wave. It is shown that they are likely to play an important role in systems with small inhomogeneous broadening of the spectral lines (for example, in a rarefied gas).

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1. When optical radiation propagates in a medium, the resultant field acting on an arbitrary *j*-th dipole is made up of the fields \tilde{H}_{j} , \tilde{E}_{j} of the incident wave (which propagates at the speed of light in vacuum), and the

fields produced by the surrounding atom at the location of the *j*-th dipole. Usually in the calculation of the field of the surrounding atoms one confines oneself to the socalled dipole field H_{j}^{e}, E_{j}^{e-1} As shown by us in Ref. 2, the

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