UNIVERSITÉ PARIS-SUD

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Министерство высшего и среднего специального образования РСФСР

Красноярский государственный университет

РАБОЧИЕ ПРОГРАММЫ

кафедры оптики и спектроскопии для студентов 4 курса физического факультета

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Unidirectional Doppler-Free Gain and Generation in Optically Pumped Lasers

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Abstract. The feasibility of obtaining anisotropic Doppler-free gain and unidirectional generation in optically pumped lasers is studied. The Doppler broadening is compensated due to a pump-induced velocity-dependent light shift of an atomic transition. The gain and spectral characteristics are investigated.

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Monochromatically pumped lasers are characterized by a number of new features and are especially attractive for coherent light generation in the long-wave range [1]. If the energy levels of an amplifying transition are populated in the absence of an optical pump, a relatively weak resonant monochromatic pump causes the appearance of nonlinear resonances on the Doppler profile of the gain and the oscillation line [2-5]. The gain resonances have different shapes and positions for the forward and the backward amplified waves with respect to the pump wave. It is accounted for the fact that only "resonant" velocities give contribution to the nonlinear interaction of pump and amplified waves and for the fact that resonant stepwise and two-photon radiative processes as well as their interference manifest themselves differently after velocity averaging [2–6].

When the gain exceeds the threshold value only in the maxima of the discussed resonances, it becomes possible to achieve a light source with a narrow-gain bandwidth [7] and unidirectional propagation inside a ring cavity [8]. However, it should be emphasized that only a small part of the total number of atoms is involved in the resonant nonlinear interaction with the pump and laser waves.

As it was shown in [6,9–14], there is a possibility to completely compensate for the Doppler broadening of

the gain line for an amplified wave, propagating in a certain direction. The effect is due to the light shifts compensating Doppler-frequency shifts in an intense pump field. All the atoms, irrespective of their velocities, will contribute to the gain only in a narrow frequency interval. As a result, the gain for one of the directions of the amplified wave exhibits a sharp increase compared to the others. The interest to Doppler compensation in a strong field, quasiresonant with an adjacent transition, has recently been stimulated by successful experiments [15, 16].

The present paper develops the theory of a laser based on the described effect. The gain conditions are analyzed at Doppler-free transitions, the Doppler broadening being compensated by pump-induced light shifts. The spectral characteristics and generation features at the given transitions are studied. The advantages of using a Doppler compensating pump and feasibilities to observe a unidirectional emission in ring cavities are considered.

1. Basic Expressions

Consider a Raman-like transition scheme (Fig. 1). Frequency ω of the travelling pump wave is close to the transition frequency ω_{mn} . The gain and generation characteristic in the spectral range near the frequency

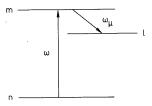


Fig. 1

 ω_{ml} of the ml transition are under consideration. Radiation at frequency ω_{μ} can be represented as the sum of two counter propagating travelling waves:

$$\begin{split} \mathbf{E}_{\mu}(z,t) = & \frac{\mathbf{E}_{\mu}^{+}}{2} \exp(\mathrm{i}\omega_{\mu}t + \mathrm{i}k_{\mu}z) \\ & + \frac{\mathbf{E}_{\mu}^{-}}{2} \exp(\mathrm{i}\omega_{\mu}t - \mathrm{i}k_{\mu}z) + \mathrm{c.c.} \,, \end{split}$$

where \mathbf{E}_{μ}^{+} and \mathbf{E}_{μ}^{-} are the waves amplitudes. The emission is provided by a nonlinear polarization at frequency ω_{μ} which can be written as follows:

$$\begin{split} \mathbf{P}_{\mu}(z,t) &= \frac{\mathbf{P}_{\mu}^{+}}{2} \exp(\mathrm{i}\omega_{\mu}t + \mathrm{i}k_{\mu}z) \\ &+ \frac{\mathbf{P}_{\mu}^{-}}{2} \exp(\mathrm{i}\omega_{\mu}t - \mathrm{i}k_{\mu}z) + \mathrm{c.c.} \\ &= \chi_{+} \frac{\mathbf{E}_{\mu}^{+}}{2} \exp(\mathrm{i}\omega_{\mu}t + \mathrm{i}k_{\mu}z) \\ &+ \chi_{-} \frac{\mathbf{E}_{\mu}^{-}}{2} \exp(\mathrm{i}\omega_{\mu}t - \mathrm{i}k_{\mu}z) + \mathrm{c.c.} \,, \end{split}$$

where χ_+ and χ_- denote the corresponding susceptibilities.

Maxwell's equations, regardless of the losses on the mirrors, provide:

$$\frac{n_{\mu}^{2}}{c^{2}}\frac{\partial^{2}\mathbf{E}_{\mu}(z,t)}{\partial t^{2}} - \frac{\partial^{2}\mathbf{E}_{\mu}(z,t)}{\partial z^{2}} = -\frac{4\pi}{c^{2}}\frac{\partial^{2}\mathbf{P}_{\mu}(z,t)}{\partial t^{2}},\tag{1}$$

where n_{μ} is the refraction index of the medium. In the case of slowly varying amplitudes $\left(k_{\mu}\frac{\partial \mathbf{E}_{\mu}}{\partial z} \gg \frac{\partial^{2} \mathbf{E}_{\mu}}{\partial z^{2}}\right)$, under a steady-state approximation, one can readily obtain for the radiation intensity $\left(I_{\mu}^{\pm} = \frac{c}{8\pi n_{\mu}^{\pm}}|E_{\mu}^{\pm}|^{2}\right)$

$$\frac{\partial I_{\mu}^{+}}{\partial z} = g_{+} I_{\mu}^{+} \quad \text{and} \quad \frac{\partial I_{\mu}^{-}}{\partial z} = g_{-} I_{\mu}^{-}, \tag{2}$$

where

$$g_{\pm} = \frac{4\pi\omega_{\mu}}{cn_{\mu}^{2}}\chi_{\pm}^{"} \qquad (\chi = \chi' + i\chi").$$
 (3)

The output radiation intensity is assumed to depend weakly on z, which is valid for small cavity losses. Then the solution of (2) has the form: $I^{\pm} = I_0^{\pm}(1 + g_{\pm}z)$, where $g_{+}z \ll 1$.

For a ring cavity the generation requirements are applied to each wave separately:

$$g_{+} = T_{+} ; \quad g_{-} = T_{-} ,$$
 (4)

where T_{\pm} denotes the cavity losses per unit length of the amplifying medium. Frequencies of the output waves are determined by the expressions:

$$\omega_{\mu}^{+} = \omega_{\mu}^{+q} \left[1 - \frac{2\pi}{(n_{\mu}^{0})^{2}} \chi_{+}' \right];$$

$$\omega_{\mu}^{-} = \omega_{\mu}^{-q} \left[1 - \frac{2\pi}{(n_{\mu}^{0})^{2}} \chi_{-}' \right],$$
(5)

where n_{μ}^{0} is the nonresonant part of the linear refraction index, $\omega_{\mu}^{\pm q}$ is the intrinsic cavity frequency in the absence of a medium $(\chi = 0)$.

The pump radiation is assumed to have the form of a travelling wave:

$$\mathbf{E}(z,t) = \frac{\mathbf{E}}{2} \exp[i(\omega t - kz)] + \text{c.c.},$$

and only level n is supposed to be populated in the absence of a pump. Below it will be shown that the properties of the optical medium at frequency ω_{μ} are highly anisotropic. The most interesting is the case when the gain index for the wave \mathbf{E}_{μ}^{-} considerably exceeds that for \mathbf{E}_{μ}^{+} so that only the wave \mathbf{E}_{μ}^{-} is generated in a ring cavity. Consider a three-level system (Fig. 1), showing interaction only with the two co-propagating waves \mathbf{E} and \mathbf{E}_{μ}^{-} .

Neglecting spontaneous transitions from m to l level, the steady-state density matrix elements are defined by the following set of equations:

$$\begin{split} P_{nm}r_{nm} &= iG\Delta_{nm} + iG_{\mu}r_{nl} \\ P_{ml}r_{ml} &= -iG_{\mu}^{*}\Delta_{em} - iG^{*}r_{ne} \\ P_{nl}r_{nl} &= -iGr_{ml} + iG_{\mu}^{*}r_{nm} \\ \Gamma_{m}r_{m} &= -2\operatorname{Re}\left\{iG^{*}r_{nm} + iG_{\mu}^{*}r_{lm}\right\} \\ \Gamma_{l}r_{l} &= 2\operatorname{Re}\left\{iG_{\mu}^{*}r_{lm}\right\}, \end{split} \tag{6}$$

where r_{ij} denotes the Fourier amplitudes of the density matrix elements in interaction representation (e.g. $\varrho_{nm} = r_{nm} \exp\{i\omega - \omega_{nm}t - ikz\}$);

$$\begin{split} & \varDelta_{ij} = \varrho_{ii} - \varrho_{jj} = r_i - r_j \;; \\ & G = - \operatorname{Ed}_{nm} / 2\hbar \;; \\ & G_{\mu} = - \operatorname{E}_{\mu}^{-} \operatorname{d}_{em} / 2\hbar \;; \quad r_{ii} \equiv r_i \\ & P_{nm} = \Gamma_{nm} + \mathrm{i} \varOmega' = \Gamma_{nm} + \mathrm{i} (\varOmega - k V) \\ & = \Gamma_{nm} + \mathrm{i} (\omega - \omega_{mn} - k V) \;; \\ & P_{ml} = \Gamma_{ml} - \mathrm{i} \varOmega'_{\mu} = \Gamma_{ml} - \mathrm{i} (\varOmega_{\mu} - k_{\mu} V) \\ & = \Gamma_{ml} - \mathrm{i} (\omega_{\mu} - \omega_{ml} - k_{\mu} V) \;; \\ & P_{nl} = \Gamma_{nl} + \mathrm{i} \varOmega'_{nl} = \Gamma_{nl} + \mathrm{i} (\varOmega' - \varOmega'_{\mu}) \\ & = \Gamma_{nl} + \mathrm{i} (\varOmega - \varOmega_{\mu} - k V + k_{\mu} V) \;. \end{split}$$

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The susceptibility χ is related to r_{lm} by the ratioes:

Doppler-Free Gain in Lasers

$$\chi' = -2N\hbar |E_{\mu}^{-}|^{-2} \operatorname{Im} \{\langle iG_{\mu}^{*}r_{lm} \rangle \} ;$$

$$\chi'' = 2N\hbar |E_{\mu}^{-}|^{-2} \operatorname{Re} \{\langle iG_{\mu}^{*}r_{lm} \rangle \}$$

$$= N\hbar |E_{\mu}^{-}|^{-2} \langle \Gamma_{l}r_{l} \rangle .$$
(7)

The angular brackets $\langle ... \rangle$ denote velocity averaging with a Maxwell distribution.

The expressions for the populations of the levels r_l and r_m can be derived from (6) [13]:

$$r_{l} = \frac{\eta_{1} I^{-2\kappa}}{\tilde{\Omega}^{\prime 2} + \tilde{\Gamma}^{2}(1+\kappa)};$$

$$r_{m} = \frac{\eta_{3} \tilde{\Gamma}^{2} \kappa}{\tilde{\Omega}^{\prime 2} + \tilde{\Gamma}^{2}(1+\kappa)}$$

$$+ 4 \frac{\eta_{2}}{\gamma} \frac{|G|^{2}}{\Omega^{2}} \frac{\Gamma_{nm}}{\Gamma_{l}} \left(\Gamma_{l} + 2\Gamma_{ml} \frac{|G_{\mu}|^{2}}{\Omega_{\mu}^{2}} \right) \frac{\tilde{\Omega}^{\prime 2} + \tilde{\Gamma}^{2}}{\tilde{\Omega}^{\prime 2} + \tilde{\Gamma}^{2}(1+\kappa)},$$

$$(8)$$

wher

$$\kappa = 4 \frac{|GG_{\mu}|^2 \eta_2}{\Omega \Omega_{\mu} \tilde{\Gamma} \Gamma_l} \; ; \qquad \eta_1 = \overline{\Gamma}_m / \gamma \; ; \qquad \eta_3 = \left(\frac{\Omega_{\mu}}{\Omega} \Gamma_l - \overline{\Gamma}_l \right) / \gamma \; ;$$

$$\begin{split} \eta_2 &= \tfrac{1}{2} \gamma \bigg[\varGamma_m + 2 \frac{\varGamma_{ml}}{\varGamma_l} (\varGamma_m + \varGamma_l) \frac{|G_\mu|^2}{\Omega_\mu^2} \\ &+ 4 \frac{\varGamma_{nm}}{\varGamma_l} \bigg(\varGamma_l + 3 \varGamma_{ml} \frac{|G_\mu|^2}{\Omega_\mu^2} \bigg) \frac{|G|^2}{\Omega^2} \bigg]^{-1} \;; \end{split}$$

$$\gamma = \bar{\Gamma}_{m} - 2\Gamma_{l} + \frac{\Omega}{\Omega_{u}} \Gamma_{m} - \Gamma_{l} \left(1 - 2 \frac{\Omega_{u}}{\Omega} - \frac{\Omega}{\Omega_{u}} \right);$$

$$\tilde{\Gamma} = \Gamma_{nl} + \Gamma_{ml} \frac{|G|^2}{\Omega_u^2} + \Gamma_{nm} \frac{|G_\mu|^2}{\Omega^2}; \tag{9}$$

$$\bar{\varGamma}_{\it m} = \varGamma_{\it m} + 2(\varGamma_{\it nm}|G|^2 + \varGamma_{\it ml}|G_{\it \mu}|^2)/\Omega\Omega_{\it \mu} \ ; \label{eq:gamma}$$

$$\tilde{\Gamma}_l = \Gamma_l - 2(\Gamma_{nm}|G|^2 + \Gamma_{ml}|G_{\mu}|^2)/\Omega\Omega_{\mu}$$

$$\tilde{\Omega}' = \Omega' - \Omega'_{\mu} + \frac{|G|^2}{\Omega'_{\mu}} - \frac{|G_{\mu}|^2}{\Omega'}.$$

According to (7), the expression for r_l determines χ'' . Note that (8) has been obtained for the following approximations: $|\tilde{\Omega}| \ll |\Omega|$, $|\Omega_{\mu}|$; $|\Omega| \gg k \bar{v}$; $|\Omega_{\mu}| \gg k_{\mu} \bar{v}$ (\bar{v} being the most probable thermal velocity). Under this approximation one finds for χ' :

$$\chi' = \frac{N|d_{ml}|^2}{2\hbar} \left\langle \frac{|G|^2}{\Omega \Omega_{\mu}} \left[1 - 4 \left(2 - \frac{\Omega}{\Omega_{\mu}} \right) \frac{\eta_2}{\gamma} \frac{|G^2|}{\Omega^2} \frac{\Gamma_{nm}}{\Gamma_l} \right. \right. \\ \left. \cdot \left(\Gamma_l + 2 \frac{|G_{\mu}|^2}{\Omega_{\mu}^2} \Gamma_{ml} \right) \right] \frac{\tilde{\Omega}'}{\tilde{\Omega}'^2 + \tilde{\Gamma}^2 (1 + \kappa)} - \frac{\Delta_{ml}}{\Omega_{\mu}} \right\rangle. \tag{10}$$

2. Doppler-Free Gain

Equations (9) determine the position of a perturbed resonance $\tilde{\Omega}'$:

$$\begin{split} \Omega' &= \Omega' - \Omega'_{\mu} + \frac{|G|^2}{\Omega'_{\mu}} - \frac{|G_{\mu}|^2}{\Omega'} \simeq \left(\Omega - \Omega_{\mu} + \frac{|G|^2}{\Omega_{\mu}} - \frac{|G_{\mu}|^2}{\Omega}\right) \\ &- (k - k_{\mu})V - \frac{|G_{\mu}|^2}{\Omega^2}kV + \frac{|G|^2}{\Omega_{\mu}^2}k_{\mu}V \\ &= \tilde{\Omega} - \tilde{k}V + \tilde{k}_{\mu}V = 0 \;. \end{split} \tag{11}$$

These relations demonstrate an important fact: the appearance of additional intensity- and velocity-dependent resonance shifts. These shifts account for the dependence of the magnitude of the resonance displacement in a strong electromagnetic field on the detuning and, hence, on the atomic velocity.

For the transition scheme under consideration, the pump fields being weak, compensation of Doppler broadening is impossible. As the intensity increases, there appears such a possibility due to the induced Doppler shifts, and the compensation condition is satisfied for all the velocities simultaneously. The condition has the following form:

$$\tilde{k} = \tilde{k}_{\mu}$$
, i.e. $k \left(1 + \frac{|G_{\mu}|^2}{\Omega^2} \right) = k_{\mu} \left(1 + \frac{|G|^2}{\Omega_{\mu}^2} \right)$. (12)

The position of the resonance Ω_{μ} can be readily found from (11):

$$(\Omega_{\mu})_{\text{res}} \equiv \Omega_{1, 2} = \frac{1}{2} \left(\Omega - \frac{|G_{\mu}|^2}{\Omega} \right) \pm \frac{1}{2} \sqrt{\left(\Omega - \frac{|G_{\mu}|^2}{\Omega} \right)^2 + 4|G|^2} \,. \tag{13}$$

The condition for the resonance at one of the detunings $\Omega_{\mu} = \Omega_{1,2}$ to be Doppler-free can be derived by substituting the detuning value given by (13) into (12):

$$k_{u} = M_{1,2}k$$
;

$$M_{1,2} = \frac{1}{2} \left(1 + \frac{|G_{\mu}|^2}{\Omega^2} \right) \left[1 \pm \frac{\Omega - \frac{|G_{\mu}|^2}{\Omega}}{\sqrt{\left(\Omega - \frac{|G_{\mu}|^2}{\Omega}\right)^2 + 4|G|^2}} \right]. \quad (14)$$

It should be noted that, according to (8), the resonance experiences power-broadening which becomes significant when $\kappa \lesssim 1$. However, the Doppler compensating condition, on the one hand, and the condition for the appearance of a narrow resonance with the width of the order of $\tilde{\Gamma}$ (i.e. $\kappa \gtrsim 1$), on the other hand, can be satisfied simultaneously at $|G|^2/\Omega^2 \lesssim 1$, and $|G_{\mu}|^2 \gtrsim \tilde{\Gamma} \Gamma_e$.

Assuming $|G_{\mu}|^2 \gtrsim \tilde{\Gamma} \Gamma_e \ll \Omega^2$ the following expression can be obtained for the resonance location Ω_{μ} and for

the condition of its Doppler broadening cancellation:

$$\Omega_{1,2} = \frac{1}{2} (\Omega \pm \sqrt{\Omega^2 + 4|G|^2}),$$
 (15)

$$k_{\mu} = M_{1,2} k; \qquad M_{1,2} = \frac{1}{2} \left(1 \pm \frac{\Omega}{\sqrt{\Omega^2 + 4|G|^2}} \right).$$
 (16)

A joint solution of (15) and (16) allows one to determine the optical pump intensity I^f required for a complete cancellation of Doppler broadening of the gain line, and to find the frequency ω_{μ}^f of the maximum gain:

$$I^{f} = \frac{c\hbar^{2}\Omega^{2}}{8\pi|d_{nm}|^{2}n} \left[\left(\frac{\omega_{mn}}{2\omega_{me} - \omega_{mn}} \right)^{2} - 1 \right], \tag{17}$$

$$\omega_{\mu}^{f} = \omega_{me} + \Omega \frac{\omega_{me}}{2\omega_{me} - \omega_{mn}},\tag{18}$$

where *n* is the refraction index for the pump wave. At $|\Omega| \sim 1 \, \mathrm{cm}^{-1}$, $|d_{mn}| \sim 10$ Debye, the estimation for I_f yields $\sim 1 \, \mathrm{kW/cm^2}$.

As it follows from (15), in weak fields the resonance $\Omega_{\mu} = \Omega_1 \rightarrow \Omega$ and, hence, corresponds to a double-quantum transition, and the resonance $\Omega_{\mu} = \Omega_2 \rightarrow 0$ and is associated with a cascade one. As G is increased, both resonances are shifted. However, resonance shifts cancel at $\Omega_{\mu}^{(\alpha)} = -|d_{m_{\alpha}n}/d_{m_{\beta}n}|^2 \Omega_{\mu}^{(\beta)}$, when m level consists of two sublevels m_{α} and m_{β} . In this case an unshifted "double-quantum" resonance can be observed at the frequency of $\omega_{\mu} = \omega - \omega_{en}$. The pump intensity being such that

$$\frac{k_{\mu}}{k} = \frac{\Omega_{\alpha}^{2}}{\Omega_{\alpha}^{2} + \frac{|\mathbf{Ed}_{m_{\alpha}n}|^{2}}{4\hbar^{2}} (1 + |d_{m_{\alpha}n}/d_{m_{\beta}n}|^{2})}$$
(19)

the unshifted resonance is Doppler-free. In case, level n consists of two sublevels n_{α} and n_{β} , it is the frequency $\omega_{\mu} = \omega_{ml}$ that is resonant, i.e. an unshifted "cascade" resonance can be observed, if $\Omega_{\alpha} = -|d_{n_{\alpha}m}/d_{n_{\beta}m}|^2 \Omega_{\beta}$. When

$$\frac{k_{\mu}}{k} = \frac{\frac{|\mathbf{Ed}_{n_{\alpha}m}|^{2}}{4\hbar^{2}} (1 + |d_{n_{\alpha}m}/d_{n_{\beta}m}|^{2})}{\Omega_{\alpha}^{2} + \frac{|\mathbf{Ed}_{n_{\alpha}m}|^{2}}{4\hbar^{2}} (1 + |d_{n_{\alpha}m}/d_{n_{\beta}m}|^{2})}$$
(20)

the resonance broadening is homogeneous. Levels n_{α} and n_{β} can be either Zeeman sublevels, splitted by a magnetic field, or the fine structure levels. The corresponding cases have experimentally been treated in [15, 16].

3. Generation Features at Doppler-Free Transitions

Substituting (8) into (7) and using (3), the expression for the gain coefficient $g(\tilde{\Gamma}(1+\kappa)^{1/2} \ll |\tilde{k}-k_{\mu}|\tilde{v})$ can be derived

derived
$$g = N \frac{4\pi^{3/2} \omega_{\mu} |d_{me}|^2}{cn_{\mu} \hbar} \frac{\eta_1 \eta_2 |G|^2}{\Omega \Omega_{\mu}} \frac{\exp\left[-\left(\frac{\Omega}{|\tilde{k} - \tilde{k}_{\mu}|\bar{v}}\right)^2\right]}{|\tilde{k} - \tilde{k}_{\mu}|\bar{v} \cdot \sqrt{1 + \kappa}}.$$
(21)

Provided the condition (14) satisfied, one obtains

$$g = N \frac{4\pi\omega_{\mu}|d_{me}|^2}{cn_{\mu}\hbar} \frac{\eta_1\eta_2|G|^2}{\Omega\Omega_{\mu}} \frac{\tilde{\Gamma}}{\tilde{\Omega}^2 + \tilde{\Gamma}^2(1+\kappa)}. \tag{22}$$

The comparison of (21) and (22) shows that the gain coefficients for co-propagating \mathbf{E}_{μ}^{-} and counter propagating \mathbf{E}_{μ}^{+} waves with respect to the pump wave relate, in the maxima, as $g_{-}/g_{+} \sim |\tilde{k} + \tilde{k}_{\mu}| \overline{v} \sim 10^{2}$, when $\kappa \gtrsim 1$. Thus, the threshold condition being satisfied only for the co-propagating wave, a unidirectional generation can be observed.

Assuming in further considerations the Doppler compensation condition to be satisfied and using (4), the following expression for the output radiation intensity can be found:

$$\kappa = \frac{I}{\bar{I}} \left(1 - \frac{\bar{I}}{\bar{I}} \frac{\tilde{\Omega}^2 + \tilde{\Gamma}^2}{\tilde{\Gamma}^2} \right) \tag{23}$$

or in an another form:

$$I_{\mu} = \frac{N}{T} \frac{\eta_{1} \hbar \omega_{\mu} \Gamma_{e}}{2n_{\mu}^{2}} \left(1 - \frac{\overline{I}}{I} \frac{\tilde{\Omega}^{2} + \tilde{\Gamma}^{2}}{\tilde{\Gamma}^{2}}\right), \tag{24}$$

where I is the pump intensity, and \bar{I} denotes its threshold value:

$$\bar{I} = \frac{T}{N} \frac{c^2 \hbar^3 (n_{\mu}/n) \tilde{\Gamma} \Omega \Omega_{\mu}}{8\pi^2 \eta_1 \eta_2 \omega_{\mu} |d_{nm} d_{me}|^2}.$$
 (25)

To obtain a narrow gain bandwidth, apart from the condition (17), the pump intensity should not exceed, substantially the threshold value, i.e. $I = I^f \lesssim \overline{I}$. Otherwise, when the saturation parameter $\kappa \gg 1$, the power broadening of the gain line becomes significant.

Assuming

$$\begin{split} |\Omega| \sim & |\Omega_{\mu}| \sim 10^{11} \, \mathrm{s}^{-1} \,; \\ \tilde{\Gamma} \sim & 10^9 \, \mathrm{s}^{-1} \,; \\ \Gamma_m \sim & 10^7 \, \mathrm{s}^{-1} \,, \\ \omega_{\mu} \sim & 10^{14} \, \mathrm{s}^{-1} \,; \\ |d_{nm}d_{me}| \sim & 10^{-36} \, \, \mathrm{units} \, \, \mathrm{CGSE} \,; \\ L \sim & 10 \, \mathrm{cm} \,; \\ T \sim & 10^{-3} \, \mathrm{cm}^{-1} \,; \\ \eta_1 \sim & \eta_2 \sim 1 \,; \\ N \sim & 10^{14} \, \mathrm{cm}^{-3} \,, \end{split}$$

the threshold value of the pump intensity becomes $\bar{I} \sim 1 \, \text{kW/cm}^2$.

It should be noted that for the given parameters, the pump absorption $\alpha L(\alpha)$ being the absorption index) is insignificant $(\alpha L \sim 10^{-1})$ and may be neglected.

A. K. Popov and V. M. Shalaev

the condition of its Doppler broadening cancellation:

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$$k_{\mu} = M_{1,2}k; \qquad M_{1,2} = \frac{1}{2} \left(1 \pm \frac{\Omega}{\sqrt{\Omega^2 + 4|G|^2}} \right).$$
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$$I^{f} = \frac{c\hbar^{2}\Omega^{2}}{8\pi |d_{nm}|^{2}n} \left[\left(\frac{\omega_{mn}}{2\omega_{me} - \omega_{mn}} \right)^{2} - 1 \right], \tag{17}$$

$$\omega_{\mu}^{f} = \omega_{me} + \Omega \frac{\omega_{me}}{2\omega_{me} - \omega_{me}},\tag{18}$$

where n is the refraction index for the pump wave. At $|\Omega| \sim 1 \,\mathrm{cm}^{-1}$, $|d_{mn}| \sim 10$ Debye, the estimation for I_f vields $\sim 1 \,\mathrm{kW/cm^2}$.

As it follows from (15), in weak fields the resonance $\Omega_{n} = \Omega_{1} \rightarrow \Omega$ and, hence, corresponds to a doublequantum transition, and the resonance $\Omega_{11} = \Omega_{22} \rightarrow 0$ and is associated with a cascade one. As G is increased, both resonances are shifted. However, resonance shifts cancel at $\Omega_{\mu}^{(\alpha)} = -|d_{m,n}/d_{m,n}|^2 \Omega_{\mu}^{(\beta)}$, when m level consists of two sublevels m_{π} and m_{θ} . In this case an unshifted "double-quantum" resonance can be observed at the frequency of $\omega_{\mu} = \omega - \omega_{en}$. The pump intensity being such that

$$\frac{k_{\mu}}{k} = \frac{\Omega_{\alpha}^{2}}{\Omega_{\alpha}^{2} + \frac{|\mathbf{Ed}_{m_{\alpha}n}|^{2}}{4\hbar^{2}} (1 + |d_{m_{\alpha}n}/d_{m_{\beta}n}|^{2})}$$
(19)

the unshifted resonance is Doppler-free. In case, level nconsists of two sublevels n_{α} and n_{β} , it is the frequency $\omega_n = \omega_{ml}$ that is resonant, i.e. an unshifted "cascade" resonance can be observed, if $\Omega_{\alpha} = -|d_{n,m}/d_{n,m}|^2 \Omega_{\beta}$.

$$\frac{k_{\mu}}{k} = \frac{\frac{|\mathbf{Ed}_{n_{\alpha}m}|^{2}}{4\hbar^{2}} (1 + |d_{n_{\alpha}m}/d_{n_{\beta}m}|^{2})}{\Omega_{\alpha}^{2} + \frac{|\mathbf{Ed}_{n_{\alpha}m}|^{2}}{4\hbar^{2}} (1 + |d_{n_{\alpha}m}/d_{n_{\beta}m}|^{2})}$$
(20)

the resonance broadening is homogeneous. Levels n_{α} and $n_{\rm g}$ can be either Zeeman sublevels, splitted by a magnetic field, or the fine structure levels. The corresponding cases have experimentally been treated in Г15, 167.

3. Generation Features at Doppler-Free Transitions

Substituting (8) into (7) and using (3), the expression for the gain coefficient $q(\tilde{\Gamma}(1+\kappa)^{1/2} \ll |\tilde{k}-k_n|\bar{v})$ can be

derived
$$g = N \frac{4\pi^{3/2} \omega_{\mu} |d_{me}|^2}{cn_{\mu} \hbar} \frac{\eta_1 \eta_2 |G|^2}{\Omega \Omega_{\mu}} \frac{\exp\left[-\left(\frac{\tilde{\Omega}}{|\tilde{k} - \tilde{k}_{\mu}|\bar{v}}\right)^2\right]}{|\tilde{k} - \tilde{k}_{\mu}|\bar{v} \cdot \sqrt{1 + \kappa}}.$$
(21)

Provided the condition (14) satisfied, one obtains

$$g = N \frac{4\pi\omega_{\mu}|d_{me}|^2}{cn_{\mu}\hbar} \frac{\eta_1 \eta_2 |G|^2}{\Omega\Omega_{\mu}} \frac{\tilde{\Gamma}}{\tilde{\Omega}^2 + \tilde{\Gamma}^2 (1+\kappa)}.$$
 (22)

The comparison of (21) and (22) shows that the gain coefficients for co-propagating \mathbf{E}_{u}^{-} and counter propagating E⁺ waves with respect to the pump wave relate, in the maxima, as $g_-/g_+ \sim |\tilde{k} + \tilde{k}_u|\bar{v} \sim 10^2$, when $\kappa \approx 1$. Thus, the threshold condition being satisfied only for the co-propagating wave, a unidirectional generation can be observed.

Assuming in further considerations the Doppler compensation condition to be satisfied and using (4), the following expression for the output radiation intensity can be found:

$$\kappa = \frac{I}{\bar{I}} \left(1 - \frac{\bar{I}}{\bar{I}} \frac{\tilde{\Omega}^2 + \tilde{\Gamma}^2}{\tilde{\Gamma}^2} \right) \tag{23}$$

or in an another form:

$$I_{\mu} = \frac{N}{T} \frac{\eta_1 \hbar \omega_{\mu} \Gamma_e}{2n_{\mu}^2} \left(1 - \frac{\bar{I}}{I} \frac{\tilde{\Omega}^2 + \tilde{\Gamma}^2}{\tilde{\Gamma}^2} \right), \tag{24}$$

where I is the pump intensity, and \bar{I} denotes its threshold value:

$$\bar{I} = \frac{T}{N} \frac{c^2 \hbar^3 (n_{\mu}/n) \tilde{\Gamma} \Omega \Omega_{\mu}}{8\pi^2 \eta_1 \eta_2 \omega_{\mu} |d_{nm} d_{me}|^2}.$$
 (25)

To obtain a narrow gain bandwidth, apart from the condition (17), the pump intensity should not exceed, substantially the threshold value, i.e. $I = I^f \lesssim \overline{I}$. Otherwise, when the saturation parameter $\kappa \gg 1$, the power broadening of the gain line becomes significant.

Assuming

$$\begin{split} |\Omega| \sim & |\Omega_{\mu}| \sim 10^{11} \, \mathrm{s}^{-1} \,; \\ \tilde{\Gamma} \sim & 10^9 \, \mathrm{s}^{-1} \,; \\ & I_m \sim 10^7 \, \mathrm{s}^{-1} \,, \\ & \omega_{\mu} \sim 10^{14} \, \mathrm{s}^{-1} \,; \\ |d_{nm} d_{me}| \sim & 10^{-36} \, \mathrm{units \ CGSE} \,; \\ & L \sim & 10 \, \mathrm{cm} \,; \\ & T \sim & 10^{-3} \, \mathrm{cm}^{-1} \,; \\ & \eta_1 \sim & \eta_2 \sim & 1 \,; \\ & N \sim & 10^{14} \, \mathrm{cm}^{-3} \,, \end{split}$$

the threshold value of the pump intensity becomes $\bar{I} \sim 1 \text{ kW/cm}^2$.

It should be noted that for the given parameters, the pump absorption $\alpha L(\alpha)$ being the absorption index) is insignificant ($\alpha L \sim 10^{-1}$) and may be neglected.

Doppler-Free Gain in Lasers

Using (5) and (10), one easily obtains the expression for the output-radiation frequency $(|\Omega| \gg k\bar{v}; |\Omega_u| \gg k_u\bar{v})$:

$$\Omega_{q} = \Omega_{\mu} + \delta \omega_{\mu}^{q} \theta \frac{\tilde{\Omega}}{\tilde{\Gamma}} - N \frac{\omega_{\mu} |d_{me}|^{2} \pi}{\hbar (n_{\mu}^{0})^{2}} \frac{\Delta_{me}(|G|^{2}, I_{\mu})}{\Omega_{\mu}}$$
 (26) where

$$\begin{split} &\Omega_{q} = \omega_{\mu}^{q} - \omega_{me} \; ; \quad \delta \omega_{\mu}^{q} = c \, T \; ; \\ & \theta = (8\eta_{1}\eta_{2})^{-1} \bigg[1 - \bigg(2 - \frac{\Omega}{\Omega_{u}} \bigg) 2 \frac{|G|^{2}}{\Omega^{2}} \frac{\Gamma_{nm}}{\Gamma_{m}} \bigg(1 + 4 \frac{|G|^{2}}{\Omega^{2}} \frac{\Gamma_{nm}}{\Gamma_{m}} \bigg)^{-1} \bigg] \end{split}$$

The analysis of (26) shows that depending on a concrete correlation between the parameters G, Ω , Ω ... both output frequency pulling and pushing relative to the cavity frequency can be observed. The dependence of the radiation frequency on the intrinsic frequency of the cavity at the line center $\tilde{\Omega} \simeq 0$ (i.e. $\Omega_n \simeq \Omega_{1/2}$) exhibits a strong resonant behavior and is increased when $\theta > 0$ and decreased when $\theta < 0$:

$$\left(\frac{d\Omega_{\mu}}{d\Omega_{q}}\right)_{\widetilde{\Omega}=0} \simeq \left[1 - \theta \frac{\delta \omega_{\mu}^{q}}{\widetilde{\Gamma}} \left(1 + \frac{|G|^{2}}{\Omega_{1,2}^{2}}\right)\right]^{-1}.$$
(27)

In the latter case the output frequency weakly depends on the fluctuations of the cavity frequency. The effect can be employed for passive stabilization of the output frequency.

The difference in the refraction indices for copropagating and counter-propagating waves results in the difference of their frequencies ω_{u}^{+} and ω_{u}^{-} , the threshold conditions being satisfied for the both waves. Lets evaluate this difference. According to (5), (10),

$$\begin{split} \frac{\Delta\omega_{\mu}}{\omega_{\mu}^{q}} &= \frac{\omega_{\mu}^{+} - \omega_{\mu}^{-}}{\omega_{\mu}^{q}} \\ &= n_{\mu}^{-} - n_{\mu}^{+} = \frac{2\pi}{(n_{\nu}^{0})^{2}} (\chi_{-}^{\prime} - \chi_{+}^{\prime}) \simeq \frac{2\pi}{(n_{\nu}^{0})^{2}} \chi_{-}^{\prime} \,. \end{split}$$

When

$$N \sim 10^{14} \, \mathrm{cm}^{-3}$$
;
 $|d_{ml}| \sim 1 \, \mathrm{Debye}$;
 $|\tilde{\Omega}| \sim \tilde{\Gamma} \sim 10^9 \, \mathrm{s}^{-1}$;
 $|G| \sim |\Omega| \sim |\Omega_{\mu}|$;
 $g_{\pm} = T_{\pm}$,
we find $\Delta \omega_{\nu}/\omega_{\nu}^q \sim$

we find $\Delta\omega_u/\omega_u^q \sim 10^{-4}$. Then, at $v_u \sim 10^3 \, \mathrm{cm}^{-1}$ one obtains

$$\Delta v_{\mu} = (\omega_{\mu}^{+} - \omega_{\mu}^{-})/2\pi \sim 0.1 \text{ cm}^{-1}$$
.

4. Conclusion

In conclusion we summarize the basic features of light generation at Doppler-free transitions. In lasers with the gain observed at a Doppler broadened transition only a small part of the total number of excited atoms (of the order of $\Gamma/k\overline{v} \ll 1$) is involved into the process of

monochromatic light generation. In particular, it results in the appearance of a Lamb dip, being significantly narrower than the gain line. The total number of atoms, contributing to the generation at the transition center is proportional to the Lamb dip area.

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The considered possibility to increase the part of the atoms contributing to the generation is due to the following. The pump intensity can be chosen so that Doppler-free gain components appear at Raman-like transitions. In other words, all the atoms, irrespective of their velocities, occur to be involved in such transitions. The total gain spectrum of these optically pumped lasers appears to be concentrated in the narrow spectral interval of the order of the natural transition width. The given gain properties are attributed only to the forward emission. The gain spectrum for a counter propagating wave is Doppler broadened with the width of the order of $(\tilde{k} + \tilde{k}_u)\bar{v}$ and, hence, is $[(k+k_u)\overline{v}/\Gamma]$ times weaker. It can result in a unidirectional generation. The dependence of the output radiation frequency also undergoes a significant modification. In particular, passive stabilization of the output frequency becomes possible at the line of the Dopplerfree transition. It is important to emphasize that for the cavity-less superfluorescent lasers the usage of the discussed Doppler-free gain makes the realization of the threshold-like condition significantly easier, and a narrow-band unidirectional emission can be obtained.

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Department of Chemistry

University of Toronto Toronto ONTARIO M5S 1A1

May 13, 1992.

Professor V.M.Shalaev L.V.Kirensky Institute of Physics Siberian Branch of the Academy of Science 660036 Krasnoyarsk Russia

Dear Professor Shalaev:

It is a pleasure to be able to offer you a position as a Visiting Associate Professor at the University of Toronto and Ontario Laser and Lightwave Research Center for one year up to June 30, 1993. I expect that you will continue our collaborative research program on optics of clusters.

Kindly take this letter to your nearest Canadian Consular office and they will issue you and your family with the appropriate visa. Please do not hesitate to let me know if there is anything further I can do to assist you.

Yours sincerely,

Martin Moskovits

Professor of Chemistry.

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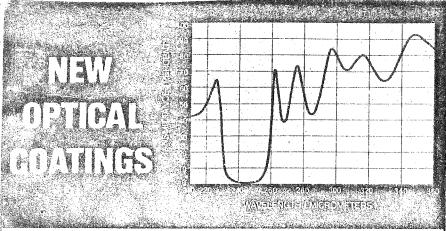
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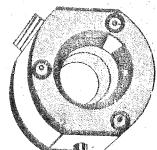


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be reduced to match the from the first to the second state the process would be repeated as many times as needed.

In addition to keeping the time required to excite the molecule much shorter than the relaxation time, the technique would hold photon waste to a minimum. For laser-induced decomposition of boron trichloride, Stevering calculates that chirping would increase efficiency tenfold. Such a reduction in the photon efficiency af other photochemical reactions—typically about 150 photons per reaction—"would bring photochemistry into a more competitive position" with thermochemistry.

Laser projection television

B. M. Lavrushin and E. S. Shemchuk, Television Sci Res Inst, Moscow Kvant Elekt 3 12, 2605 (Dec in Russian)

Evaluating the application of electronbeam-pumped semiconductor lasers to projection television, the authors conclude that the system has potential resolution of a few thousand television lines and high brightness on a medium-size screen. Commercial prototypes of e-beam-pumped semiconductor lasers [LF Dec '74 p22] have been tested for applications including high-speed nonimpact printing of computer output in the United States, but have found no major applications.

SF, studies in USSR

V. T. Plamonenko Moscow State U Pisma JETP 25 1, 52 (Jan 5 in Russian)

Studies of the mechanisms underlyisotopically photodissociation of polyatomie molecules are proliferating in the Soviet Union as in the United States. This paper, "On a mechanism of collisionless dissociation of molecules in a strong infrared laser field," by an author at Moscow State University, describes calculations indicating that the transfer of "considerable" vibrational energy into rotational energy compensates for the anharmonicity of energy levels.

Resonant upconversion

A. K. Popov and V. P. Timofeev, Kirensky Inst of Physics, Krasnoyarsk, USSR Opt Comm 201, 94 (Jan)

Two methods of resonant two-photon upconversion are proposed by Popov and Timofeev at a Siberian institute. In one, a "broad range of infrared radiation" would be upconverted into the green via double-quantum transitions in cesium vapor; the major advantage, they write, is the possibility

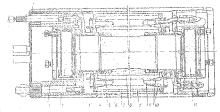
of controlling the phase-matching conditions for a wide range of infrared frequencies by tuning one of the pumping frequencies within the doublet."

Upconversion it mercury vapor and krypton, they write, could permit generation of 42 new vacuum-ultraviolet and soft-x-ray lines with wavelengths as short as 70.8 nanometers. Basis is two-photon resonances of the fourth harmonic of neodymium-glass with Hg I and of the fifth harmonic of Nd-glass with Kr I.

Pumping glass slabs

E. F. Zholobov et al Kvant Elekt 4 1; 122 (Jan in Russian)

The six authors, of undisclosed affiliation, describe a coaxial lamp for pumping slabs of neodymium-glass



Top view of coaxial lamp for pumping glass rod shows 1 isolator, 2 totally reflecting mirror, 3 electrode, 4 copper bellows, 5 active element 6 metal casing, 7 outer wall of coaxial lamp(s), 8 inner wall, 9 reflective coating, 10 damping ring and 11 output mirror

920 millimeters long and 44 mm in diameter. Laser efficiency was 3.3% with 250-microsecond flashlamp pulses of 660 joules, and increased with pump-pulse duration.

'Competitive-beam' studies

A. N. Rubinov, S. A. Batishche and V. A. Mostovnikov, Minsk Inst Phys Kvant Elekt S 11, 1516 (Nov in Russian)

To measure weak absorptions, they propose a "competitive-beam" technique in which two laser cavities include the same active medium. The sample being studied would be placed into the cavity of one of the resonators arranged to produce narrow, tunable output. They say the method is suitable for measurement of wide absorption bands as well as of parrow lines, and can be used to measure two photon absorption spectra.

H.-laser mass spectrometer

V. K. Potapov et al, Inst of Spectroscopy, Moscow Knowt Elekt 3 12, 2610 (Dec in Russian)

Hydrogen laser output in the vacuum ultraviolet is applied, for the first time according to the five authors, to study molecular photognization in a



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