The mutual mode quenching effect in optical second harmonic generation

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The nonlinear interaction of two modes during optical second harmonic generation in an optical resonator is considered. The effect of mutual mode quenching which leads to the bistability of the radiation and to hysteresis phenomena is predicted. The form of the hysteresis in the system is investigated. The possibility of using this effect to produce optical memory elements and optical logical devices is discussed. It is pointed out that the switching time in the hysteresis cycle may be less than 10^{-10} sec, and the luminous energy expendable during this time may be less than one picojoule.

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1. INTRODUCTION

There are at present a large number of papers devoted to the investigation of the nonlinear interaction (competition) between the modes in two-level lasers. The competition between the two modes during stimulated emission by two-level active particles was first considered by the present author[1] (see also his recent work published in Ref. 2). In this work it is assumed that the active medium fills the laser resonator and that the saturation of the luminescence line in the generated-radiation field can be arbitrary (including being strong). Lamb[3] has developed quite a general theory of gas lasers, and has also considered the competition between two modes in the case of weak saturation of the luminescence line. In two-level lasers the nonlinear interaction of the modes arises in the case when the frequency interval between the modes is less than the homogeneous width of the luminescence line, and is due to the saturation of this line in the field of the generated radiation. We have also investigated the nonlinear interaction of modes during stimulated Raman emission in optical resonators,[4-7] when several modes can be excited in the generated first Stokes component (or likewise in higher-order components). In this case various effects resulting from the interaction in question are possible, depending on the conditions. Recently, Kawasaki *et al.*[8] also carried out an experimental investigation of the interaction of the modes inside the first Stokes component of stimulated Mandel'shtam-Brillouin emission in an optical resonator.

However, the problem of the nonlinear interaction of the modes of an optical resonator as a result of the generation of optical harmonics has thus far not been posed in the literature. In the present paper we consider the nonlinear interaction of two modes in an optical resonator when the autoexcitation of these modes occurs as a result of negative absorption produced by one of the known processes (we shall, for definiteness, consider the case when the negative absorption is effected at the Stokes frequency, ω_S, in a Raman-active medium), while the nonlinear losses limiting the growth of the intensity of these oscillations are due to the generation of the optical second harmonic (at the frequency $2\omega_S$) under conditions when it is phase-matched with the fundamental harmonic (the case of the Cerenkov mode of emission of this harmonic is considered in Ref. 9). In the process, we predict the effect of mutual mode quenching, which leads to the bistability of the Stokes radiation, and consider the hysteresis phenomena arising from this bistability. We also discuss below the possible use of the optical resonator under consideration as an optical memory element and an element of optical logical devices.

2. DERIVATION OF THE BASIC EQUATIONS

We shall assume that the Stokes radiation is enclosed in an optical waveguide of length L, bounded by mirrors (forming a Fabry-Perot resonator), and that a section (of length l) of this waveguide possesses an appreciable second-order susceptibility tensor ($\hat{x}$), so that the optical second harmonic is generated at the frequency $2\omega_S$ as a result of the polarization induced in this medium by the Stokes radiation. We shall also assume that there are at the lengthwise boundries of the layer of medium with square-law nonlinearity elements reflecting at the frequency $2\omega_S$, and forming (for the optical second harmonic) an auxiliary Fabry-Perot resonator of length l. Thus, the polarization induced at the frequency $2\omega_S$ will excite the modes of the auxiliary resonator. For definiteness, we shall assume that the pumping wave (of frequency ω_p) propagates in a waveguide lying in the same plane as the resonators, and incident in a direction perpendicular to their axis, and that it occupies the region with the Raman-active medium (see Fig. 1). Let us note that Fig. 1 is only a schematic drawing. In fact, the mirrors of the resonator (the hatched surfaces) can be surface gratings, the cross section of the resonant-cavity waveguide need not be rectangular, etc.

Let us assume that only two modes can be excited in the primary resonator (i.e., in the resonator for the Stokes radiation). Then the electric field of the Stokes radiation in this resonator can be written in the form

$$\mathbf{E}=\sum_{s=0}^{1} a_s(t)\mathbf{E}_s(\mathbf{r}). \tag{1}$$

Here $\mathbf{E}_s(\mathbf{r})$ is the coordinate part of the s-th mode. We assume that the mirrors of the primary resonator possess sufficient reflectivity, so that the reflections from the interface between the media at the frequency ω_S do not form an auxiliary resonator at this frequency. We

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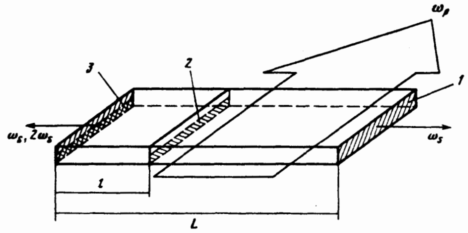

FIG. 1. The hatched surfaces (1, 2, 3) are mirrors reflecting at the following frequencies: 1) at the frequency ω_S; 2) at the frequency $2\omega_S$; 3) at the frequencies ω_S, $2\omega_S$. A material possessing appreciable square-law nonlinearity occupies the interval l along the axis of the optical waveguide and the remaining part is occupied by a Raman-active material. The pumping wave (of frequency ω_p) propagates in a direction perpendicular to the axis and occupies the space with the Raman-active material.

shall also assume that the frequency interval between these modes ($\Delta\omega = \omega_1 - \omega_0$) is substantially less than the frequency halfwidth, $\mu^{(0)}$, of the corresponding $\mathbf{E}^{(0)}(\mathbf{r})$ mode, which can be excited (on account of the phase-matching condition) at the frequency of the optical second harmonic in the auxiliary resonator ($\Delta\omega \ll \mu^{(0)}$). This condition is easily satisfied for those modes whose natural frequencies can undergo degeneration (e.g., for the modes in the double waveguide layer). In this case any of the modes (1) can excite only the $\mathbf{E}^{(0)}(\mathbf{r})$ mode, i.e., the electric field at the optical second harmonic frequency in the auxiliary resonator can be written in the form

$$\mathbf{E} = a^{(0)}(t)\mathbf{E}^{(0)}(\mathbf{r}). \tag{2}$$

For neighboring modes of the primary resonator, the spatial structures of these modes in the volume, V_l, of the auxiliary resonator may be essentially the same: $\mathbf{E}_0(r) \approx \mathbf{E}_1(r)$ (such a situation obtains if, for example, the mirrors for the second harmonic are formed by one-sided surface gratings on the double waveguide layer). In this case the resultant field in this volume can be written in the form

$$\mathbf{E} \approx [a_0(t) + a_1(t)]\mathbf{E}_0(\mathbf{r}) + a^{(0)}(t)\mathbf{E}^{(0)}(\mathbf{r}). \tag{3}$$

Making the substitution $a_0(t) + a_1(t) = \frac{1}{2}Y(t)\exp(-i\omega t) + \text{c.c.}$, $a^{(0)}(t) = \frac{1}{2}Z(t)\exp(-2i\omega t) + \text{c.c.}$ $(Y = Y_0 + Y_1$, $a_s = \frac{1}{2}Y_s\exp(-i\omega t) + \text{c.c.})$, using a procedure similar to the one used in Refs. 4, 5, and 7 to derive the equations describing stimulated Raman emission in a multimode optical resonator, and introducing into these equations the nonlinear polarization arising from the generation of the optical second harmonic, we arrive at the following closed system of equations for the complex amplitudes Z, Y_0, and Y_1:

$$\begin{aligned} \frac{dZ}{dt} &= -(\mu^{(0)} + i\Delta^{(0)})Z + \frac{i\omega^{(0)}}{2N^{(0)}}\frac{\alpha}{2}Y^2, \\ \frac{dY_s}{dt} &= -(\tilde{\mu}_s + i\Delta_s)Y_s + \frac{i\omega_0}{2N_s}\alpha Z Y^*, \end{aligned} \tag{4}$$

where

$$\alpha = \int_{V_l} \hat{\chi}(\omega) : \mathbf{E}_0\mathbf{E}^{(0)}\mathbf{E}_0 d\mathbf{r},$$

$$N_s = \frac{1}{4\pi}\int \varepsilon(\omega)\mathbf{E}_s^2 d\mathbf{r}, \quad N^{(0)} = \frac{1}{4\pi}\int \varepsilon(2\omega)\mathbf{E}^{(0)2}d\mathbf{r}, \tag{5}$$

$$\Delta_s = \omega_s - \omega, \quad \Delta^{(0)} = \omega^{(0)} - 2\omega, \quad \tilde{\mu}_s = \mu_s - \delta_s,$$

ε is the linear part of the permittivity of the medium; ω_s is the natural field-oscillation frequency for the $\mathbf{E}_s(\mathbf{r})$ mode; $\omega^{(0)}$ is the natural frequency for the $\mathbf{E}^{(0)}(\mathbf{r})$ mode; the term δ_s takes into account the negative absorption in the medium for the $\mathbf{E}_s$ modes; $s = 0, 1$ (normally, it is convenient to set $N_0 = N_1$). The Eqs. (4) were derived under the assumption that the frequency halfwidth, $\mu^{(0)}$, for the $\mathbf{E}^{(0)}(\mathbf{r})$ mode is substantially less than the halfwidth, μ, of the spontaneous Raman scattering line ($\mu^{(0)} \ll \mu$). At frequencies, ω_0 and ω_1, lying, for example, near the center of this line, the quantities δ_s are real, and are determined by the expression

$$\delta_s = \frac{2\pi\omega_s}{\varepsilon(\omega_s)}\int \Gamma(\mathbf{E}_s\mathbf{E}_p)^2 d\mathbf{r} \Big/ \int \mathbf{E}_s^2 d\mathbf{r}, \tag{6}$$

where the quantity Γ for, for example, the transitions in which the fully symmetric oscillations of the molecules participate is given by the relation $\Gamma = 3\lambda_S^4 N Q_0 / 2^9\pi^5\hbar\mu$ (λ_S is the wavelength of the Stokes radiation in a vacuum, Q_0 is the total cross section for spontaneous Raman scattering per molecule, and N is the density of the molecules in the material). Normally, $\delta_0 \approx \delta_1 = \delta$. According to (6), the estimate $\delta_s \sim \gamma E_p^2$, where the coefficient γ for such materials as crystalline quartz and calcite has the value $\gamma \sim 5\times10^2 - 3\times10^4$ cgs esu, is correct.

3. THE STEADY-STATE SOLUTIONS AND THEIR STABILITY

Under the condition that $\Delta\omega \ll \mu^{(0)}$, the phase space of the system (4) splits into regions of "fast" and "slow" motions, it following from the first equation that the entire slow-motion region is stable against the fast motions. In order to obtain the equations describing the slow motions, it is necessary, as is usual in systems of this type (see Ref. 10), to formally set $dZ/dt = 0$, as a result of which we find

$$dY_s/dt = -(\tilde{\mu}_s + i\Delta_s)Y_s - \beta|Y|^2 Y, \tag{7}$$

where

$$\beta = \omega_0^2\alpha^2/4N_0N^{(0)}(\mu^{(0)} + i\Delta^{(0)}). \tag{8}$$

Let us write out here the conditions of applicability of the system of equations (7):

$$\Delta\omega \ll \mu^{(0)}, \ \mu. \tag{9}$$

The conditions (9) are broader than the region, $\Delta\omega \ll \mu^{(0)} \ll \mu$, of applicability of Eqs. (4), which is due to the broadening of the corresponding region after the double application of the method of splitting the phase space of the system into fast- and slow-motion regions [the partitioning procedure was first used in the derivation of Eqs. (4), in much the same way as is done in Ref. 4].

Let us now consider the single-frequency emission regimes, i.e., let us consider solutions of the form $Y_s \equiv \tilde{Y}_s = \text{const}$ under, for simplicity, the condition $|\Delta^{(0)}| \ll \mu^{(0)}$, which implies that the detunings of the optical second harmonic frequency and the corresponding natural frequency $\omega^{(0)}$ are substantially smaller than the frequency width of the latter frequency (under this condition, the quantity β is real, $\beta > 0$). When $\mu_s/\Delta\omega$, $\delta/\Delta\omega < 1$ (which are easy to realize for, among others, the neighboring modes in the double wave-guide layer), Eqs. (7) have two different solutions, $\tilde{Y}_s^{(0)}$ and $\tilde{Y}_s^{(1)}$, which describe field oscillations with different frequencies $\omega = \omega_S^{(0)}$, $\omega_S^{(1)}$ given by the relations

$$\omega_S^{(0)} \approx \omega_0 + \frac{(\delta-\mu_0)^2}{\Delta\omega}, \quad \omega_S^{(1)} \approx \omega_1 - \frac{(\delta-\mu_1)^2}{\Delta\omega}. \tag{10}$$

The corresponding values of the amplitudes $\tilde{Y}^{(m)} = \tilde{Y}_0^{(m)} + \tilde{Y}_1^{(m)}$ are given by the equalities

$$|\tilde{Y}^{(m)}|^2 \approx (\delta-\mu_m)/\beta, \tag{11}$$

the quantities $|\tilde{Y}_s^{(m)}|$ for $s \neq m$ having values of the order of $|\tilde{Y}^{(m)}|\mu_m/\Delta\omega$. Thus, the emission occurs essentially at the frequency of one or the other mode, and this mode makes the dominant contribution to the amplitude of the light oscillations. According to (11), the condition for the self-excitation of these oscillations in the m-th mode has the form $\delta > \mu_m$.

To investigate the stability of, for example, the steady-state solution $\tilde{Y}^{(0)}$, we linearize Eq. (7) in the vicinity of this solution. As a result, we find the following expression for the corresponding characteristic polynomial:

$$D(\lambda) = \lambda[\lambda^3 + 2\lambda^2(2\delta+\mu_1-3\mu_0) + (\lambda+2\delta-2\mu_0)\Delta\omega^2] \tag{12}$$

(the trivial root $\lambda = 0$ is due to the fact that the solution has an arbitrary constant factor, $e^{i\psi}$). Arising from (12) is the following condition for the stability of the $\tilde{Y}^{(0)}$ regime:

$$\mu_1 > 2\mu_0 - \delta. \tag{13}$$

As can be seen, for $\delta > 2\mu_0$ (i.e., when the threshold for generation in the mode with the index 0 is exceeded by a factor of two), the regime in question is stable regardless of the magnitude of the linear losses, μ_1, for the mode with the index 1. In the interval $\mu_0 < \delta < 2\mu_0$ this regime is stable only when the linear losses for the mode with the index 1 are not too small as compared to the losses for the mode 0. Similarly, the regime of emission into the mode with the index 1 is stable when $\mu_0 > 2\mu_1 - \delta$. Thus, mutual mode quenching is realized in the Stokes radiation, and hysteresis phenomena turn out to be possible in the system when the quantities μ_0 and μ_1 are varied in the interval $\mu_m < \delta < 2\mu_m$.

The μ_1 dependences of the Stokes-radiation intensity $|\tilde{Y}|^2$ for both regimes (with allowance for the results of the investigation of their stability) for fixed values of μ_0 and δ ($\mu_0 < \delta < 2\mu_0$) are shown in Fig. 2, where the solid curves correspond to the stable values of $|\tilde{Y}|^2$, while the dashed lines correspond to the unstable values. The arrows indicate the hysteresis cycle when μ_1 is varied. As can be seen, there is realized in the interval $2\mu_0 - \delta < \mu_1 < (\mu_0 + \delta)/2$ depending on the initial conditions, one of the two considered stable regimes of emission (respectively into one or the other mode), i.e., bistability is realized in the system. In this case, with allowance for the equality

$$Z = \tilde{Z} = \frac{i\omega^{(0)}\alpha}{4N^{(0)}\mu^{(0)}}\tilde{Y}^2 \tag{14}$$

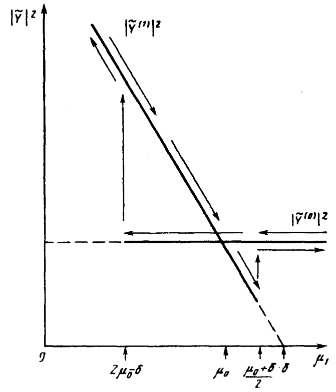

FIG. 2. Dependence of the intensity, $|\tilde{Y}|^2$, of the Stokes radiation (of frequency ω_S) on the magnitude of the passive losses, μ_1, of the mode with the index 1 for a fixed value of the passive losses, μ_0, of the mode with the index 0. The solid curves correspond to the stable values of $|\tilde{Y}|^2$; the dashed lines, to the unstable values. The arrows indicate the hysteresis occurring when μ_1 is varied.

we find the bistability and the hysteresis will also be simultaneously observed in the optical second harmonic radiation. Let us also note that a change in the ratio of the passive mode losses (i.e., in the ratio of the quantities μ_0 and μ_1) can be effected by, for example, an electro-optical method on account of the deviations that occur in the natural frequencies ω_0, ω_1 during the selective frequency reflection by the mirrors (which is realizable, for example, from surface gratings), or on account of the deviations of the location of the frequency peak of the light reflected by the same mirrors.

Let us now consider a numerical example, bearing in mind the possible use of the system under consideration as an optical-memory element or as an element of optical logical devices. The condition for phase matching in optical second harmonic generation in plane optical waveguides has been realized for different materials (see Refs. 11–19). Under such a condition (in the volume V_l), we obtain for the quantity β on the basis of (8) the following estimate

$$\beta \sim f\pi^2\omega_S^2|\hat{\chi}(\omega_S)|^2 V_l/\mu^{(0)} n_{\text{eff}}^4 V_L,$$

where

$$n_{\text{eff}}^4 = \frac{\int\varepsilon(\omega_S)\mathbf{E}_0^2 d\mathbf{r}}{\int\mathbf{E}_0^2 d\mathbf{r}} \frac{\int\varepsilon(2\omega_S)\mathbf{E}^{(0)2} d\mathbf{r}}{\int\mathbf{E}^{(0)2} d\mathbf{r}},$$

$$f = \frac{\sin^4 x}{x^2}, \quad x = (k^{(0)} - 2k_0)l/2$$

(V_L is the volume of the primary resonator). For example, for $L \sim 0.1$ cm, $l \sim 3\times10^{-2}$ cm, $r_{2\omega_S} \sim 0.5$ ($\mu^{(0)} \sim 10^{11}$ rad/sec), $n_{\text{eff}} \sim 2$ for the GaAs crystal ($|\hat{x}| \sim 10^{-5}$ cgs esu), and $\omega_S \sim 10^{15}$ rad/sec we obtain $\beta \sim 10^8$ cgs esu. For the above-indicated Raman-active materials, and for reflection coefficients of the mirrors at the Stokes frequency $r_{\omega_S} \sim 0.8$, the threshold value of the pump power is $P \approx 10^2 - 10^4$ W; the characteristic switching time, τ_{ch}, in the hysteresis cycle at, for example, a pump intensity of $I_p \sim (1.5-2)I_{p\,\text{thr}}$ has a value of the order of $\tau_{\text{ch}} \sim 1/2\mu_m \sim 3\times10^{-11}$ sec; the output power of the generated Stokes radiation when the optical waveguide has, for example, a width $a \sim 5\times10^{-4}$ cm and an effective thickness $\Lambda_{\text{ch}} \sim 2\times10^{-4}$ cm has the value 3×10^{-4}

W. Let us point out that this value is smaller than the pump power by roughly five-seven orders of magnitude. Thus, 10^5-10^7 resonators of the type under consideration can be placed one after another in one and the same pumping beam. Let us also note that, in the considered example, the luminous energy expendable in each of such resonators during the period τ_{ch} has a value of the order of 10^{-14} J.

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Two-mode He–Ne laser in an axial magnetic field

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We consider the two-mode operation of a gas laser placed in an axial magnetic field. The dependences of the critical frequency, of the region of stable two-mode lasing, and of the frequency of the intermode beats on the magnetic field are investigated for different characters of polarizations of the fields of the interacting modes. It is observed that with increasing magnetic field the behavior of the critical frequency varies significantly with the character of the polarizations of the interacting-mode fields.

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Investigations, for example those reported in Refs. 1–3, have shown that an axial magnetic field affects strongly the emission characteristics of a single-mode gas laser. At the same time two-mode gas lasers with orthogonally polarized modes are presently offering great promise for use in a number of fundamental physical experiments and applications.[4] This explains the interest in the effect of the magnetic field on the characteristics of the emission of such lasers. In particular, even the very first investigations of a two-mode He–Ne/CH_4 laser in an axial magnetic field have shown that under certain conditions the amplitude of the output-power resonance increases substantially and the shift of the resonance peak as a result of the influence of the gain contour decreases. This improves greatly the characteristics of stabilized lasers of this type.

We report here, for the first time ever, the results of theoretical and experimental investigations of the influence of an axial magnetic field on the principal characteristics of two-mode gas lasers—the critical frequency, the range of continuous tuning of the intermode distance, the region of two-mode generation, and the frequency of the intermode beats at various polarizations of the interacting-mode fields.

1. EXPERIMENTAL RESULTS

The experiments were performed with an He–Ne laser operating on the $3s_2-3p_4$ transition of Ne ($j_a=2, j_b=1$, where j_a and j_b are the angular momenta of the lower and upper working levels, respectively). The distance ω_{12} between the axial modes was varied smoothly with a Fabry–Perot phase-anisotropy resonator in the form of two $\lambda/4$ plates.[5] The active medium was placed either between the phase plates or on the side of one of them. The magnitude and direction of the magnetic field were varied by changing the current flowing through the solenoid in which the discharge tube was placed.

By separating the emission of any mode with the aid

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