Soft excitation of stimulated two-photon emission

G. M. Krochik and Yu. G. Khronopulo

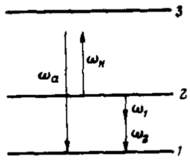
Institute of Radio Engineering and Electronics, USSR Academy of Sciences
(Submitted April 11, 1975)
ZhETF Pis. Red. **21**, No. 10, 585–589 (May 20, 1975)

It is proposed to realize soft excitation of a two-phonon laser with the aid of a four-wave parametric process. The working medium can be vapor. Estimates of the laser parameters in different regimes are presented.

PACS numbers: 42.60.C

1. The problem of developing a laser on the basis of stimulated two-photon emission (STE), i.e., a two-photon laser (TPL), was discussed about ten years ago.[1-4] The STE cross section at the frequencies ω_1 and ω_2 ($\omega_1+\omega_2=\omega_{21}$), where ω_{21} is the frequency of the transition between the levels with inverted populations), is proportional to the wave intensities $\mathcal{E}_2(\omega_2)$ and $\mathcal{E}_1(\omega_1)$. To excite a TPL it is therefore necessary to have a certain threshold intensity of the external source at the frequency ω_1, or ω_2, after the action of which the system can emit both frequencies ω_1 and ω_2 (hard triggering of the TPL [3]).

We discuss here self-excitation of a TPL with the aid of a four-wave process, the scheme of which is shown in the figure.[1)] As is well known, a parametric interaction (that depends on the phases of the fields) is produced in this case between the waves $\mathcal{E}_p(\omega_p)$, $\mathcal{E}_a(\omega_a)$, and $\mathcal{E}_{1,2}$. This interaction is the consequence of the interference of the amplitudes of the probabilities of the two-photon processes—the anti-Stokes SRS and the STE. The STE cross section contains in this case a component proportional to $\mathcal{E}_p\mathcal{E}_a$. Obviously, in this case $\mathcal{E}_p\mathcal{E}_a$ plays the role of the intensity of the triggering field, and thus the STE can be excited by realizing anti-Stokes SRS from



the excited state. We note that the last process was realized experimentally in atomic-iodine vapor.[5]

The proposed method of self-excitation of STE makes it possible to realize the following: 1) pure parametric transformation of the pump energy into the energy of the fields $\mathcal{E}_1\mathcal{E}_2$, when the generation at the frequencies $\omega_{1,2}$ stops after the pump action stops; 2) soft triggering of the TPL, when the STE continues also after the action of the pulse of the triggering field $\mathcal{E}_p$; 3) in the last regime, at $\omega_1=\omega_2$, it is possible to obtain in rare gaseous media generation in an ultranarrow line, because of the compensation of the Doppler shift in opposing waves.[6] The present paper is devoted to a discussion of these possibilities.

2. We confine ourselves to the following very simple model: a) We assume that at the frequencies ω_1 and ω_2 there exist single-mode traveling-wave resonators with identical values Q. b) The wave amplitudes $\mathcal{E}_a(\omega_a)$ and $\mathcal{E}_p(\omega_p)$ are assumed specified. A situation close to that considered here can be realized by obtaining the field $\mathcal{E}_p$ in the remaining volume via SRS field $\mathcal{E}_a$ in a noninverted medium consisting of the same working medium. In this case the equations for the squares of the real amplitudes of the field $A_j=\mathcal{E}_j(t)\exp[i\phi_j(t)]$, of the phase differences $\theta(t)=c\delta t+\phi_a-\phi_p-\phi_1-\phi_2$ (where the difference of the wave vectors is $\delta=k_a-k_p-k_1-k_2$), and the population differences of the excited ground states of one particle n take the following forms[7]:

$$A_j^2+\frac{1}{\tau}A_j^2=\frac{4\pi dF\omega_j N}{\hbar^3\epsilon_j}\,n(r_2^2A_2^2A_1^2+r_1r_2A_pA_aA_1A_2\cos\theta), \quad (1a,b)$$

$$\dot n+\frac{n-n_0}{T}=-nd\hbar^{-4}(r_2^2A_1^2A_2^2+r_1^2A_p^2A_a^2+2r_1r_2A_pA_aA_1A_2\cos\theta), \quad (1c)$$

$$\dot\theta=c\delta-\frac{2\pi dFNr_1r_2n}{\hbar^3}\left[A_pA_a\left(\frac{A_2}{A_1}\frac{\omega_1}{\epsilon_1}+\frac{A_1}{A_2}\frac{\omega_2}{\epsilon_2}\right)+A_1A_2\times\left(\frac{A_p}{A_a}\frac{\omega_a}{\epsilon_a}-\frac{A_a}{A_p}\frac{\omega_p}{\epsilon_p}\right)\right]\sin\theta. \quad (1d)$$

Here $r_{1,2}$ is the second-order polarizability and determines the cross section of the Raman conversion $\mathcal{E}_a\to\mathcal{E}_p$ and two-photon absorption $\mathcal{E}_1$, $\mathcal{E}_2$; T is the inversion lifetime, N is the density of the number of particles, and d is the reciprocal line width of the 2–1 transition. In the case of a rarefied gaseous medium, d is the Doppler width, namely $d=\sqrt{\pi}/(|k_a-k_p|\bar v)\approx\sqrt{\pi}/(|k_1+k_2|\bar v)$, τ is the damping time of the field in the resonator, and F is the coefficient of filling of the resonator with the working medium.

3. If the condition

$$4\pi dNnFr_1r_2A_aA_p\omega_1/\hbar^3\epsilon_1c>\delta, \quad (2)$$

is satisfied, then $\theta\approx0$. In the case when (2) is satisfied for $n=n_{stat}$—the stationary solution of Eqs. (1)—the equality $\theta=0$ is always valid. Even if (2) is not satisfied at $t=0$, it can be assumed that $\theta\approx0$ over times $t_c\gtrsim(\delta c)^{-1}$ in which the linear synchronism conditions are satisfied. In all these situations, Eqs. (1) become greatly simplified. From (1a,b,c) we can obtain equations for the quantity $I=\approx(\omega_2\epsilon_1/\omega_1\epsilon_2)^{1/2}A_1^2=(\omega_1\epsilon_2/\omega_2\epsilon_1)^{1/2}A_2^2$ and for the population difference. Introducing the dimensionless quantities

$$x=I/I_{sat},\quad y=qr_2^2\alpha I_{sat}n,\quad I_{sat}=\hbar^2(dT)^{-1/2}r_1^{-1},$$

$$\alpha=\tau/T,\quad q=\frac{4\pi dF(\omega_1\omega_2)^{1/2}N}{\hbar^3(\epsilon_1\epsilon_2)^{1/2}};\quad S=r_1/r_2,\quad \beta=\frac{SA_pA_a}{I_{sat}}, \quad (3)$$

$$t_p=tT^{-1},$$

where I_{sat} is the intensity of the saturating field, we obtain

$$\alpha\dot x=-x+yx(x+\beta),\qquad \dot y=y_0-y-y(x+\beta)^2. \quad (4)$$

It is easy to show from (4) that generation at the frequencies ω_1 and ω_2 can be excited if the intensity of the triggering fields satisfies the relation

$$\frac{y_0}{2}-\sqrt{\frac{y_0^2}{4}-1}<\beta<\frac{y_0}{2}+\sqrt{\frac{y_0^2}{4}-1}. \quad (5)$$

The presence of an upper limit in (5) is due to the intense destruction of the population of the upper working level by the strong starting fields. When this condition is satisfied, there exists a unique physically-real equilibrium position:

$$\bar x=\frac{1}{\bar y}-\beta,\qquad \bar y=\frac{y_0}{2}-\sqrt{\frac{y_0^2}{4}-1}. \quad (6)$$

a) If $\alpha<\bar y^2(1-\bar y^2)(1+\bar y^2)^{-1}$, then in the region $\beta<\bar y^{-1}+\alpha(1+\bar y^2)\bar y^{-3}$ this state is unstable, and there exists a stable limit cycle, while the region $\beta>\bar y^{-1}+\alpha(1+\bar y^2)\bar y^{-3}$ it is stable. Thus, as a result of four-wave parametric interaction in weak starting fields, amplitude-modulated radiation is produced at the frequencies ω_1 and ω_2; with increasing β, the modulation tends to zero. It can be shown that when the starting fields are turned off, radiation at the frequencies ω_1 and ω_2 also stops.

If $\alpha>\bar y^2(1-\bar y^2)(1+\bar y^2)^{-1}$, then the equilibrium state (6) is stable. In this case, at a population difference satisfying the relation

$$y_0^2=\frac{(4\pi Nn_0rr_2)^2\omega_1\omega_2d}{\hbar^2\epsilon_1\epsilon_2T}>3+\alpha^{-1}+3+\frac{T}{\tau} \quad (7)$$

the two-photon generation in the system continues, after the triggering fields are turned off, during the entire time of action of the pump that inverts the population. Consequently, in this regime the four-wave interaction leads to soft triggering of the TPL.

4. In rarefied gaseous media, the condition (7) is particularly easy to satisfy for the generation of degenerate frequencies $\omega_1=\omega_2=\omega$, in a standing-wave

resonator. In this case, the polarization at the frequency ω has a component with a natural (rather than Doppler) line width γ of the working transition,[6] as a result of which we have in condition (10) $d=\gamma \gg \sqrt{\pi}/2k\bar{v}$. Two-photon emission has in this case an extremely narrow spectral width and a stable frequency.

5. Inasmuch as at present population inversion has been produced in a number of vapors,[8] we present estimates, for example, for atomic iodine.[5,8] At $\omega_{1,2} \approx 10^{15}$ sec^{-1}, $\omega_p = 1.78\times10^{15}$ sec^{-1}, $\omega_a = 3.2\times10^{15}$ sec^{-1}, $\tau = 5\times10^{-8}$ sec, $r_2 = r_1 = 1.2\times10^{-51}$ cgs esu, and $d = 4\times10^{-9}$ sec^{-1}, if $Nn_0T^{-1} = 10^{22}$ cm^{3}sec^{-1}, $FNn_0 = 10^{15}$ cm^{-3} and $T = 5\times10^{-5}$ sec (these population lifetimes are typical of population inversion by electron impact or by photodissociation), then $y_0 = 2.5 > 2$. At a starting-field energy density 4×10^{-6} J-cm$^{-3} < A_pA_a < 1.6\times10^{-5}$ J-cm^{-3}, we obtain regime (a). The characteristic growth time of the generation pulses is $t_p = 1/\mathrm{Re}\lambda \approx 10^{-7}$ sec, where λ is the root of the characteristic equation of the system (4). The average power of the amplitude-modulated STE is ~200 W/cm^3. For degenerate frequencies $\omega_1 = \omega_2 = \omega$ at $FNn_0 = 3\times10^{17}$, the regime (b) can be realized, in which the soft triggering leads to a quasistationary generation within the limits of the natural line width.

In the case when $FNn_0T^{-1} \approx 5\times10^{22}$, the regime (b) can be realized also for the generation of nondegenerate frequencies. The value $T = 3\times10^{-6}$ sec needed for this purpose can be obtained, for example, by inversion production with laser radiation. After the starting fields are turned off, fields will be radiated at the frequencies ω_1 and ω_2, with power of several kW/cm^3.

We are grateful to R.V. Khokhlov for interest in the work and a useful evaluation, and to I.I. Sobel'man, V.S. Butylkin, and G.V. Venkin for fruitful discussions.

[1)]This excitation makes it possible to vary the frequencies $\omega_{1,2}$ by retuning the natural frequencies of the corresponding resonators.

[1]A.M. Prokhorov and A.S. Selivanenko, Inventor's Certificate 24/XII-1963 No. 872303.

[2]P. P. Sorokin and N. Braslau, IBM J. Res. Develop. 8, 182 (1964).

[3]G. L. Gurevich and Yu.G. Khronopulo, Izv. vyssh. uch. zav. ser. Radiofizika 8, 493 (1965).

[4]G. L. Gurevich and M.I. Kheifets, *ibid.* 10, 43 (1967).

[5]R. L. Karman and V. Kh. Laudermilk, Tezisy dokladov predstavlennykh na 7 Vsesoyuznuyu konf. po kogerentnoĭ i nelineĭnoĭ optike (Abstracts of Papers at 7th All-Union Conf. on Coherent and Nonlinear Optics), Moscow State Univ. (1974), p. 240.

[6]L.S. Vasilenko, V.P. Chebotaev, and A.V. Shishaev, ZhETF Pis. Red. **12**, 161 (1970) [JETP Lett. **12**, 113 (1970)].

[7]V.S. Butylkin, G.M. Krochik, and Yu.G. Khronopulo, Zh. Eksp. Teor. Fiz. **68**, 506 (1975) [Sov. Phys.-JETP **41**, No. 2 (1975)].

[8]A.V. Vinogradov and E.A. Yukov, ZhETF Pis. Red. **16**, 631 (1972) [JETP Lett. **16**, 447 (1972)].