Possibility of four-photon parametric generation of light in gases

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The possiblity of four-photon parametric generation of uv and far-ir photons in gases under quasiresonance conditions is discussed. Estimates of nonlinear susceptibilities are used to show that the threshold for the four-photon generation of light in gases may be lower than the three-photon threshold in solids. The density matrix approach is used to derive expressions for the nonlinear susceptibilities and threshold excitation intensities for the interaction between fields and two- and four-level systems. It is shown that quasiresonance generation in gases is essentially determined by pressure effects, self-absorption of spontaneous emission, and saturation effects. The optimum gas pressure and pump difference from resonances in the medium, the synchronism conditions, and possible tuning of generation frequencies are discussed. The excitation thresholds are estimated and tuning curves are obtained.

1. INTRODUCTION

One of the topical problems in modern nonlinear optics is the production of coherent and frequency-tunable radiation in the far uv and ir. In these spectral regions, solids exhibit broad absorption bands and this restricts the range of application of nonlinear crystals for the generation of radiation. Possible ways of overcoming these difficulties depend on the utilization of nonlinear phenomena in gases and metal vapors. However, the use of gases as nonlinear media gives rise to a number of problems. Most crystals exhibit quadratic nonlinearity, and phenomena based on this type of nonlinearity are the most extensively investigated in optics at the present time. They include three-photon parametric processes which can be used to achieve continuous frequency tuning in the downward direction.[1] Atoms and centrally-symmetric molecules of gases form media with cubic nonlinearity in the electric dipole approximation, Four-photon parametric processes which can be used to achieve continuous frequency tuning, both in the upward and downward directions, may be associated with this nonlinearity. Among the advantages of gases are the presence of narrow resonances and the possibility of continuous variation of particle density, spectral linewidths, length of the medium, and refractive indices of the frequencies of the interacting waves (when a buffer gas is employed).

Nonlinear susceptibilities of gases may be substantially increased by using resonance phenomena. However, this is also associated with an increase in the linear absorption. Since the Lorentz linewidths and relaxation times are functions of pressure, the dependence of absorption, refraction, and nonlinear susceptibility on pressure and the form of the resonances will be different for different pressures. Moreover, in some cases, there are important saturation processes which may be looked upon as the dependence of these parameters on the field. Knowledge of these functions is essential for the use of gases as nonlinear media. Experimental studies of nonlinear resonance phenomena in potassium vapor[2-4] have shown the presence of strong violet and infrared radiation connected with four-photon parametric processes. The effects of parametric processes in the emission spectra of gases at distances less than the Doppler linewidth from the resonances were considered in[5] in the case of two-level systems and in[6] for four-level systems.

The threshold for stationary parametric generation in gases in the case of excited levels was estimated in[7] and in the case of transitions associated with the ground state in[8]. Experiments on third-harmonic generation in alkali-metal vapors and noble gases were recently reported in[9, 10].

In this paper we analyze the optimum conditions for the excitation of parametric generation of light in gases, and consider their relation to saturation phenomena and elementary processes. Particular attention is given to the possibility of producing frequency-tunable radiation in the far uv and ir.

2. THRESHOLD FOR PARAMETRIC GENERATION

Let us consider the conditions for the excitation of parametric generation when for each of the generated frequencies ω_1 and ω_2 there is a separate resonator and the conditions of phase-matching, i.e.,

$$\omega_1+\omega_2=2\omega_p, \quad \mathbf{k}_1+\mathbf{k}_2=2\mathbf{k}_p \tag{2.1}$$

are satisfied.

Sufficiently high pump intensities can be reached either by placing the nonlinear medium in the pump-laser cavity or by focusing the pump into the nonlinear medium. In both cases, the equations for slow amplitudes[11] can be used to show that the excitation conditions and the pump intensity threshold I_{th} are given by[12]

$$I=\frac{c}{8\pi}|E_p|^2>I_{th}=\frac{c}{8\pi}\left[\frac{(D_1+T_1)(D_2+T_2)}{\sigma_1\sigma_2 l^2}\right]^{1/2}. \tag{2.2}$$

Here, D_j are the absorption coefficients in the nonlinear medium of length l, T_j are the transmission losses of the mirrors at frequency ω_j per pass, $\sigma_j = \pi\omega_j|\tilde{\chi}^{(n)}|/c2^{n-4}$, and $\tilde{\chi}^{(n)}$ is the component of the parametric susceptibility tensor for the process of the corresponding order.

For a three-photon process

$$\omega_1+\omega_2=\omega_p, \quad \mathbf{k}_1+\mathbf{k}_2=\mathbf{k}_p$$

and the formula for I_{th} can be written in the form[11]

$$I_{th}=\frac{c}{8\pi}\frac{(D_1+T_1)(D_2+T_2)}{\sigma_1\sigma_2 l^2}. \tag{2.3}$$

We shall show that there exist conditions under which the threshold for four-photon generation in gases does not exceed the threshold for three-photon parametric generation in solids. Denoting the corresponding parameters by the subscripts g and s, and assuming that all the wavelengths and mirror transmission coefficients T of the mirrors are equal and that $D \ll T$, we find that the threshold given by (2.2) does not exceed that given by (2.3) when

$$\tilde{\chi}_g^{(4)} \geqslant 8\pi^2 \frac{l_s^2}{\lambda l_g T} (\chi_s^{(3)})^2. \quad (2.4)$$

For $l_s = 1$ cm, $l_g = 10$ cm, $T = 0.05$, $\lambda = 10^{-4}$ cm, and $\tilde{\chi}_s^{(3)} = 3 \times 10^{-9}$ esu,[1] the condition given by (2.4) corresponds to $\tilde{\chi}_g^{(4)} \geq 1.4 \times 10^{-11}$ esu. An order-of-magnitude estimate (see Secs. 3 and 4) is $\tilde{\chi}_g^{(4)} \approx Nd^4/\hbar^3|\Omega|^3$ where N is the density of atoms in the nonlinear medium, d is the modulus of the matrix element of the dipole moment of the transition, and $\Omega = \omega - \omega_0$ is the characteristic frequency difference from the atomic resonances for the interacting fields. It will be shown below that, in many cases, it is convenient to take $N \sim 10^{14}$ cm^{-3}, for which $D \ll T$ if $|\Omega| \gtrsim 10^{12}$ sec^{-1}. Assuming that $d = 5 \times 10^{-18}$ esu, we obtain $\tilde{\chi}_g^{(4)} \approx 6 \times 10^{-11}$ esu and, consequently, the threshold is lower than for the three-photon generator.

We must now consider particular expressions for $\tilde{\chi}$ and D; including saturation effects, and establish the connection between the threshold pump intensity and microscopic parameters of the medium and elementary processes in the gas.

3. PARAMETRIC FREQUENCY TRANSFORMATION IN THE CASE OF A TWO-LEVEL SYSTEM

One of the nonlinear effects which arise during the interaction between a two-level system and a strong field of frequency ω_p and weak radiation of another frequency ω_1 is the appearance of polarization at the frequency $\omega_2 = 2\omega_p - \omega_1$. Similarly, the field at the frequency ω_2 facilitates the appearance of polarization at frequency ω_1. When the self-excitation conditions are satisfied, the generation of fields at frequencies ω_1 and ω_2 in a strong field of frequency ω_p becomes possible. The threshold value of the pump intensity, which is necessary for the excitation of this generation, can be determined from the material constants of the medium. This means that we must calculate the components of the density matrix $\hat{\rho}(\omega_j)$ at the corresponding frequencies. If we use the density matrix, we can find the polarization at any frequency ω_j

$$\mathscr{P}(\omega_j) = \mathrm{Sp}\langle \hat{\rho}(\omega_j) \hat{d} \rangle_v. \quad (3.1)$$

In this expression, $\hat{d}$ is the dipole-moment operator for the transition. The angular brackets indicate averaging with respect to the velocities of the atoms.

Suppose that all the frequencies of the interacting fields are closest to the mn transition frequency ($E_m > E_n$). Solving the equations for the density matrix, normalized to a unit volume, and retaining only first-order terms in the weak fields, we can obtain the following expression for the nondiagonal elements of the density matrix at the frequencies ω_j of the weak fields:

$$\rho_{mn} = r_{nm} \exp\{i(\omega_j t - \mathbf{k}_j \mathbf{r})\} + \tilde{r}_{nm} \exp\{i[(2\omega_p - \omega_i)t - (2\mathbf{k}_p - \mathbf{k}_i)\mathbf{r}]\},$$

$$r_{nm} = i \frac{(N_n - N_m) G_j (\Gamma^2 + \Omega_p'^2)}{[\Gamma^2(1+\varkappa) + \Omega_p'^2](\Gamma + i\Omega_j')}$$

$$\times \left\{ 1 - \frac{(2\Gamma - i\Delta_j')[\Gamma - i(2\Omega_p' - \Omega_j')]\tau_j^* |G_p|^2}{(\Gamma - i\Omega_p')[(\Gamma + i\Omega_j')(\Gamma - i2\Omega_p' + i\Omega_j) + 2\tau_j^*(\Gamma - i\Delta_j')|G_p|^2]} \right\},$$

$$\tilde{r}_{nm} = -i \frac{1}{[\Gamma^2(1+\varkappa) + \Omega_p'^2]}$$

$$\times \frac{(N_n - N_m) G_p^2 G_i^* (\Gamma^2 + \Omega_p'^2)(2\Gamma + i\Delta_i')\tau_i}{\{(\Gamma - i\Omega_i')(\Gamma + i2\Omega_p' - i\Omega_i') + 2\tau_i(\Gamma + i\Delta_i')|G_p|^2\}(\Gamma + i\Omega_p')} \quad (3.2)$$

In these expressions, $\Omega_{j,p}' = \Omega_{j,p} - \mathbf{k}_{j,p}\mathbf{v} = \omega_{j,p} - \omega_{mn} - \mathbf{k}_{j,p}\mathbf{v}$, $\Delta_j = -\Delta_i$, ω_{mn} is the frequency of the transition under consideration, $\Delta_j' = \Omega_p' - \Omega_j'$, N_n and N_m are the level populations in the absence of the field, integrated with respect to the velocity,

$$G_{j,p} = -\frac{\mathbf{E}_{j,p}\mathbf{d}_{nm}}{2\hbar}; \quad \Gamma \equiv \Gamma_{mn}; \quad \varkappa = 2\frac{\Gamma_m + \Gamma_n - \gamma_{mn}}{\Gamma_m \Gamma_n \Gamma} |G_p|^2;$$

$$\tau_j = \frac{\Gamma_m + \Gamma_n - \gamma_{mn} + 2i\Delta_j'}{(\Gamma_m + i\Delta_j')(\Gamma_n + i\Delta_j')};$$

i, j = 1, 2, Γ is the homogeneous transition width, and $\gamma_{m,n}$ is the probability of relaxation per unit time from level m to level n. Using (3.2), we can calculate the corresponding susceptibilities at the frequencies ω_j which, in general, are functions of both $|G_p|^2$ and κ.

The component r_{nm} represents the change in the absorption and refraction at the frequency ω_j due to the different nonlinear effects, and $\tilde{r}_{nm}$ represents the change in the parametric polarization in a strong field. If we can expand in terms of G_p, the resonances at $\Delta_i = 0$ and $\Omega_j = 2\Omega_p$ can be interpreted as being due to two- and three-photon processes. The strong field mixes different elementary resonances, forming new nonlinear resonances which are the poles of (3.2). The result is that the classification into one- and two-photon processes becomes meaningless.[13] This phenomenon can be interpreted as the splitting of energy levels in a strong electromagnetic field.[14, 13] When $\Omega_j = 2\Omega_p$, $\Omega_i = 2\Omega_p - \Omega_j = 0$, which is accompanied by strong absorption of this wave. In the ensuing analysis, we shall confine our attention to the case when all the departures from resonances are much greater than the homogeneous (Γ) and Doppler ($k\bar{v}$) linewidths

$$|\Omega_p, \Omega_{1,2}, \Delta_{1,2}| \gg \Gamma, k\bar{v}, \quad (3.3)$$

and, moreover,

$$|G_p|^2/\Omega_p^2, \quad |G_p|^2/|\Omega_1\Omega_2| \ll 1. \quad (3.4)$$

Conditions (3.3) and (3.4) enable us to neglect the motion of particles, and to remove averaging over the velocities in (3.2) by substituting v = 0. When (3.4) is satisfied, the splitting of the energy states in a strong field can also be neglected. However, because of the difference between the relaxation constants Γ and Γ_m, the following condition may be satisfied:

$$\Gamma^2\varkappa = (4\Gamma/\Gamma_m)|G_p|^2 \gg \Omega_p^2, \quad (3.5)$$

and this means that there is a change in the populations in the pump field. Therefore, when (3.4) and (3.5) are satisfied, the main effect of saturation is the saturation of population difference.

If the level n is the ground state and the level m is unoccupied in the absence of the pump, then in (3.2) we must pass to the limit $\Gamma_n \to 0$, $(\Gamma_m - \gamma_{mn})/\Gamma_h \to 1$, $N_m \to 0$, $N_h \to N$. Using (2.2) and (3.2), and confining our attention in these expressions to terms of the order of $|G_p|^2/\Omega_p^2$, we then have the following equation for I_{th}:

$$I_{th} = \frac{\hbar^2 c\Gamma|\Omega_p|}{4\pi d^2}\left[1 + \frac{T_1}{D^0}\frac{(\Omega_p - \Delta_1)^2}{\Omega_p^2}\left(1 + \frac{I_{th}}{I_s}\right)\right]^{1/2} \times \left[1 + \frac{T_2}{D^0}\frac{(\Omega_p + \Delta_1)^2}{\Omega_p^2}\left(1 + \frac{I_{th}}{I_s}\right)\right]^{1/2}, \quad (3.6)$$

where

$$D^0 = \frac{4\pi d^2 \Gamma N l}{\hbar \lambdabar \Omega_p^2}; \quad \lambdabar = \frac{\lambda_{mn}}{2\pi}, \quad d^2 \equiv |d_{mn}|^2,$$

$$I_s = \frac{\hbar^2 c \Omega_p^2 \Gamma_m}{4\pi d^2 2\Gamma} = \frac{\hbar c \Gamma_m N l}{D^0 2\lambda\!\!\!^{-}} \quad \left(D_{1,2} = \frac{\Omega_p^2}{(\Omega_p \mp \Delta_1)^2} \frac{D^0}{1 + I/I_s} \right)$$

The quantity D^0 is the field unsaturated value of the pump absorption coefficient in the nonlinear medium, and D_j represents the saturated absorption at frequency ω_j. The factor $1 + I_{th}/I_s$ on the right of (3.6) is due to the equalization of the level populations in the strong field.

Equation (3.6) has physical solutions and, consequently, generation is possible only when

$$\begin{gathered} 0 < \Delta < |\Omega_p| \sqrt{1+f}, \text{ if } f > 1; \\ |\Omega_p| \sqrt{1-g} < \Delta < |\Omega_p| \sqrt{1+f}, \text{ if } f < 1; \\ f = \frac{4\pi d^2 N \Gamma_m l}{\sqrt{T_1 T_2}\, \hbar |\Omega_p| 2\Gamma \lambda\!\!\!^{-}} = D^0 |\Omega_p| \Gamma_m / 2\Gamma^2 \sqrt{T_1 T_2}, \quad \Delta = |\Delta_j|. \end{gathered} \tag{3.7}$$

The significance of these conditions is that, for sufficiently large frequency difference between pump and resonance, the generation is possible if the maximum departure from resonance of one of the generated waves is not too large (right boundary) and the frequency of the other is close to the resonance (left boundary). Generation stops for Δ_j in excess of a certain value because of the reduction in the parametric susceptibility due to the saturation as the pump increases. Saturation processes, in turn, have a strong influence on the effective relaxation time of the level populations in the nonlinear medium in the region of the interaction with the field [the factor Γ_m/Γ in (3.7)].

The effective relaxation rate may be substantially reduced by the trapping of spontaneous emission within the interaction volume[15]

$$\begin{gathered} \Gamma_m = F A_{mn}; \\ F = 1, \text{ if } k_0 a < 1.5 \text{ и } F = 1.6/k_0 a \sqrt{\pi \ln k_0 a}, \text{ if } k_0 a > 1{,}5. \end{gathered} \tag{3.8}$$

In these expressions A_{mn} is the probability of spontaneous transition from level m to level n, k_0 is the absorption coefficient at the center of the line due to the transition mn, and a is the minimum size of the interaction region (cross section of the pump beam). In the case of optical transitions and particle density $N \sim 10^{14}\ \text{cm}^{-3}$, we have $F \approx 0.005$. The homogeneous line half-width Γ is also a function of pressure and consists of two parts, namely, radiative and collisional: $\Gamma_r + \Gamma_{col} = \Gamma$. If the collisional broadening is due to resonance exchange on collision,[16] then

$$\Gamma \approx \frac{A_{mn}}{2}\left(1 + \frac{N}{N_0}\right), \quad N_0 = \frac{1}{11.4\lambda\!\!\!^{-3}} \tag{3.9}$$

It is clear from (3.3)–(3.7) that I_{th} is a complicated function of Δ, Ω_p, N, and l. These parameters are related by the additional conditions $D_i^0 \lesssim T_i$ which impose a restriction on the admissible absorption in the nonlinear medium. The admissible values of N for given Ω_p and Δ can be determined from the graphs given in Fig. 1, which were constructed with allowance for the relationships given by (3.9). The choice of N determines both the size of the threshold and the range of possible changes in Δ. The possibility of frequency tuning increases with increasing value of $\beta = |\Omega_p| f$, which is given by

$$\beta = |\Omega_p| f \approx \frac{l}{a} \frac{\sqrt{\ln 2}\, k\bar{v} + 0.5 A_{mn}(1 + N/N_0)}{\sqrt{T_1 T_2 \pi \ln k_0 a}\,(1 + N/N_0)}. \tag{3.10}$$

Since for optical transitions we always have $k\bar{v} \gg A_{mn}$, maximum tuning is acheved when $N < N_0$. It is clear from (3.6) that, as the pressure is reduced, when $N \ll N_0$ and $D^0 \ll T_1$, the threshold increases in proportion to $1/N$. When the pressure is increased and

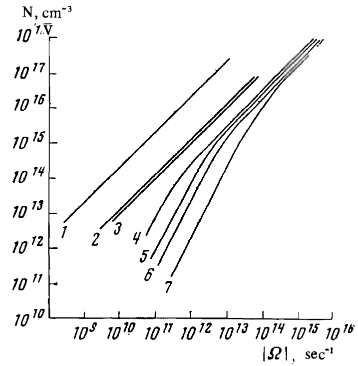

FIG. 1. Particle density as a function of departure from resonance for fixed unsaturated absorption: $l = 10$ cm, $d = 5 \times 10^{-18}$ esu, $D^0 = 0.1$ for curve 1, and $D^0 = 0.01$ for the others; (1) $\lambda = 100\ \mu$, ($k\bar{v} = 2.5 \times 10^{17}\ \text{sec}^{-1}$), (2) $\lambda = 10.6\ \mu$ ($k\bar{v} = 2.8 \times 10^8\ \text{sec}^{-1}$), (3) $\lambda = 5.3\ \mu$ ($k\bar{v} = 5.6 \times 10^8\ \text{sec}^{-1}$), (4) $\lambda = 6280$Å ($k\bar{v} = 5 \times 10^9\ \text{sec}^{-1}$), (5) $\lambda = 3140$Å ($k\bar{v} = 10^{10}\ \text{sec}^{-1}$), (6) $\lambda = 2512$Å ($k\bar{v} = 1.2 \times 10^{10}\ \text{sec}^{-1}$), (7) $\lambda = 1256$Å ($k\bar{v} = 2.5 \times 10^{10}\ \text{sec}^{-1}$).

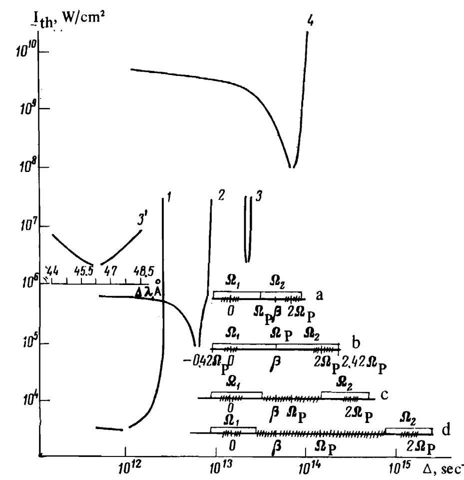

FIG. 2. Tuning curves and generation ranges for the two-level system (in the shaded region parametric generation is impossible). $l = 10$ cm, $a = 0.1$ cm, $T_{1,2} = 0.01$, $d = 5 \times 10^{-18}$ esu, (1-3) $\lambda = 6280$Å, $N = 10^{14}\ \text{cm}^{-3}$, $k_0 a = 90$, $F = 0.0047$, $\beta = 7.4 \times 10^{12}\ \text{sec}^{-1}$, (4) $\lambda = 1256$Å, $N = 5 \times 10^{13}\ \text{cm}^{-3}$, $k_0 a = 45$, $F = 0.01$, $\beta = 7.2 \times 10^{13}\ \text{sec}^{-1}$. Curve 3′ corresponds to the lower part of curve 3 with an expanded scale. $\Delta = |\Omega_p - \Omega_1| = |\Omega_p - \Omega_2|$. Curve (1) $|\Omega_p| = 10^{12}\ \text{sec}^{-1}$, (2) $|\Omega_p| = 6 \times 10^{12}\ \text{sec}^{-1}$, (3) $|\Omega_p| = 2.2 \times 10^{13}\ \text{sec}^{-1}$, (4) $|\Omega_p| = 7 \times 10^{13}\ \text{sec}^{-1}$.

$N > N_0$, i.e., in the region of collisional broadening, the range of possible values of Δ will contract. At the same time, the threshold will increase in proportion to $\Gamma|\Omega_p|$. It follows from the foregoing analysis of (3.6) that optimum densities lie in the region $N \sim N_0$.

It follows from (3.7) that for optimum densities and increasing $|\Omega_p|$, the range of possible changes in $|\Omega_j| = |\Omega_p \pm \Delta|$ at first increases and eventually reaches the maximum value $\Delta = \sqrt{2}\beta$ for $|\Omega_p| = \beta$ $(0 < \Delta < \sqrt{2}\beta = \sqrt{2}|\Omega_p|)$. The quantity f then becomes less than unity, and generation close to degenerate, $\Delta \ll |\Omega_p|$, becomes impossible, Moreover, the tuning range contracts somewhat and becomes equal to $\beta\,(|\Omega_p| - 1/2\beta < \Delta < |\Omega_p| + 1/2\beta)$. The change in the tuning range with increasing Ω_p is illustrated by graphs a–d in the lower part of Fig. 2. The upper part of the figure shows the tuning curves for different values of $|\Omega_p|$ and $\lambda\!\!\!^{-}_{mn}$. Curves 1–3 correspond to optical pumping and curve 4 to ultraviolet pumping. Curve 1 represents the case when $|\Omega_p| < \beta$ and a shows the tuning range. Curve 2 represents the case $|\Omega_p| \approx \beta$ and b shows the tuning range.

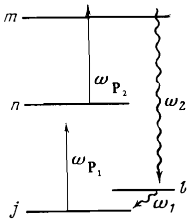

FIG. 3. Frequency conversion using a four-level system.

Curve 3 corresponds to the case $|\Omega_p| > \beta$, when the tuning range splits into two (c and d). Curve 4 represents the case $|\Omega_p| \approx \beta$. The breaks in the curves and the shaded parts on the diagrams of the tuning ranges correspond to values of $|\Omega_j|$, $\Delta \lesssim k\bar{v}$ for which the above formulas are not valid. When the graphs were constructed, steps were taken to ensure that (3.4) was satisfied. It is clear from Fig. 2 that the threshold is a relatively slowly varying function over a range of a few angstroms, and then rises sharply as the critical values of Δ are approached.

The generation threshold is a function of Ω_p. As $|\Omega_p|$ increases, the threshold at first decreases linearly, and then for $D^0 \ll T_1$ the variation is more rapid and the rate of variation is a function of Δ. It can be shown that when $|\Omega_p| \sim |\Omega_j| \sim 10k\bar{v}$, the generation threshold may be reduced down to 100 W/cm^2 for optical pumping ($\chi^{(4)} \sim 10^{-9}$ esu) and of the order of 10^5 W/cm^2 for ultraviolet pumping ($\lambda \approx 1200$ Å).

We may therefore conclude from the foregoing that when the synchronism condition is satisfied, the two-level gas systems can be used to transform the frequency of radiation by an amount of the order of Ω_p both in the upward and downward directions, and can be tuned within these ranges by an amount corresponding to a few thousand Doppler widths. This may be important for nonlinear and active laser spectroscopy at high resolution.

We must now analyze possible applications of four-level gas systems for frequency conversion to values very different from the pump frequency.

4. FREQUENCY CONVERSION IN FOUR-LEVEL SYSTEM

Consider the interaction between four electromagnetic fields and the four-level system shown in Fig. 3. The pump frequencies in this case are different and are close to the nj and mn transition frequencies. Solving the equations for the components of the density matrix in the first approximation in the generated field, and evaluating the polarization at the corresponding frequencies, we obtain the following expressions for the susceptibilities:

$$\tilde{\chi}(\omega_{p1},\omega_{p2},-\omega_1)=\left\langle \frac{id_{jn}d_{nm}d_{ml}d_{ij}}{\hbar^3 DP_{ml}}\left[\frac{r_j-r_l}{P_{nl}P_{lj}^*} - \left(1+\frac{|G_{nj}|^2}{P_{nl}P_{lj}^*}\right)\frac{r_{mj}}{G_{mn}G_{nj}}+i\frac{r_{nj}}{G_{nj}P_{nl}}\right]\right\rangle_v,$$

$$\tilde{\chi}(\omega_{p1},\omega_{p2},-\omega_2)=\left\langle \frac{id_{jn}d_{nm}d_{ml}d_{lj}}{\hbar^3 D^*P_{lj}}\left[\frac{r_l-r_m}{P_{ln}^*P_{ml}^*} + \left(1+\frac{|G_{mn}|^2}{P_{nl}^*P_{ml}^*}\right)\frac{r_{mj}}{G_{mn}G_{nj}}+i\frac{r_{mn}}{G_{mn}P_{nl}^*}\right]\right\rangle_v; \quad (4.1)$$

$$\chi(\omega_2)=\left\langle\frac{i|d_{ml}|^2}{\hbar DP_{ml}}\left[\left(1+\frac{|G_{nj}|^2}{P_{nl}P_{lj}^*}\right)(r_l-r_m)+\frac{G_{mn}G_{nj}}{P_{nl}P_{ml}^*}r_{jm}+i\frac{G_{mn}}{P_{nl}^*}r_{nm}\right]\right\rangle,$$
$$(4.2)$$
$$\chi(\omega_1)=\left\langle\frac{i|d_{lj}|^2}{\hbar D^*P_{lj}}\left[\left(1+\frac{|G_{mn}|^2}{P_{nl}^*P_{ml}^*}\right)(r_j-r_l)-\frac{G_{mn}G_{nj}}{P_{nl}^*P_{ml}^*}r_{jm}+i\frac{G_{nj}}{P_{nl}^*}r_{jn}\right]\right\rangle_v.$$

In these expressions

$$D=1+\frac{|G_{mn}|^2}{P_{ml}P_{nl}}+\frac{|G_{nj}|^2}{P_{lj}^*P_{nl}},\ P_{ml}=\Gamma_{ml}-i\Omega_{ml}'\ \ \Omega_{ml}'=\omega_2-\omega_{mn}-\mathbf{k}_2\mathbf{v}\quad\text{and so on}$$

$$\Omega_{mj}=\Omega_{mn}+\Omega_{nj},\ \ \Omega_{nl}=\Omega_{nj}-\Omega_{lj}=\Omega_{ml}-\Omega_{mn},\ G_{nj}=-\mathbf{E}_{p1}\mathbf{d}_{nj}/2\hbar\quad\text{and so on}$$

The diagonal elements and the pre-exponential factors in the diagonal elements of the density matrix, r_p and r_{pq}, are solutions of the problem of the interaction of the system with only strong fields:

$$r_{mj}=\frac{G_{mn}G_{nj}}{CP_{mj}}\left(\frac{r_n-r_m}{P_{mn}}-\frac{r_j-r_n}{P_{nj}}\right),$$
$$r_{nj}=-i\frac{G_{nj}}{CP_{mj}}\left[r_j-r_n+\frac{(r_j-r_n)|G_{nj}|^2+(r_n-r_m)|G_{mn}|^2}{P_{mn}P_{mj}}\right];$$
$$r_{mn}=-i\frac{G_{mn}}{CP_{mn}}\left[r_n-r_m+\frac{(r_n-r_m)|G_{mn}|^2+(r_j-r_n)|G_{nj}|^2}{P_{nj}P_{mj}}\right]; \quad (4.3)$$
$$\Gamma_m r_m=Q_{nm},\quad \Gamma_n r_n=Q_{jn}-\left(1-\frac{\gamma_{mn}}{\Gamma_{in}}\right)Q_{nm},\quad r_m+r_n+r_l+r_j=N,$$
$$(\Gamma_l+\gamma_{jl})r_l=\gamma_{jl}N-\frac{\gamma_{jl}}{\Gamma_n}Q_{jn}+\left[\frac{\gamma_{ml}}{\Gamma_m}-\frac{\gamma_{jl}}{\Gamma_m}\left(1-\frac{\Gamma_m-\gamma_{mn}}{\Gamma_n}\right)\right]Q_{nm},$$

where

$$C=1+\frac{|G_{mn}|^2}{P_{mj}P_{nj}}+\frac{|G_{nj}|^2}{P_{mj}P_{mn}},\qquad Q_{nm}=2\,\mathrm{Re}(iG_{nm}r_{mn}),\qquad Q_{jn}=2\,\mathrm{Re}(iG_{jn}r_{nj}).$$

In (4.1) and (4.2), it is assumed that the synchronism condition $\Omega_{nj}+\Omega_{mn}=\Omega_{ml}+\Omega_{lj}$ is satisfied. We shall also assume henceforth that all the differences between field frequencies and the resonances, with the splitting of the energy levels in the pump field taken into account, are much greater than the lindwidths. In particular, the conditions for two- and three-photon resonances are also not satisfied. The averaging with respect to v is then removed from (4.1) and (4.2), and we may set v = 0. We shall also confine our attention to the case when

$$\frac{|G_{nj}|^2}{|\Omega_{mj}\Omega_{mn}|},\ \frac{|G_{nj}|^2}{|\Omega_{nl}\Omega_{nj}|},\ \frac{|G_{nj}|^2}{|\Omega_{mj}\Omega_{nj}|}\ll 1;\quad \frac{|G_{mn}|^2}{|\Omega_{mj}\Omega_{nj}|},\ \frac{|G_{mn}|^2}{|\Omega_{nl}\Omega_{ml}|}\ll 1, \quad (4.4)$$

for which level splitting in the case of optical transitions can be ignored. The change in the populations under the influence of the field can be neglected provided

$$\frac{\Gamma_{jn}}{\Gamma_n}\frac{|G_{jn}|^2}{\Omega_{nj}^2}\ll 1,\quad \frac{|G_{mn}G_{nj}|^2}{\Gamma_m}\mathrm{Re}\left\{\frac{1}{P_{mn}P_{nj}P_{mj}}\right\}\ll 1,$$
$$\frac{|G_{mn}G_{nj}|^2}{\Gamma_l+\gamma_{jl}}\mathrm{Re}\left\{\frac{1}{P_{mn}P_{nj}P_{mj}}\right\}\ll 1,\quad \frac{\gamma_{ml}}{\Gamma_m}\frac{|G_{mn}G_{nj}|^2\Gamma_{mn}\Gamma_{nj}}{\Gamma_n(\Gamma_l+\gamma_{jl})\Omega_{nm}^2\Omega_{nj}^2}\ll 1. \quad (4.5)$$

If the level j is the ground state and the transition lj corresponds to the submillimeter band, then when (4.5) are satisfied, only the levels j and l will be populated and $N_i/N_j=\eta=\exp\{-\hbar\omega_{ij}/k_BT\}$. The result is

$$\tilde{\chi}(\omega_{p1},\omega_{p2},-\omega_1)=\tilde{\chi}(\omega_{p1},\omega_{p2},-\omega_2)=-\frac{Nd_{jn}d_{nm}d_{ml}d_{lj}}{\hbar^3(1+I_1/I_s)F},$$
$$D_1=\frac{4\pi|d_{lj}|^2}{\lambda_1\hbar}\frac{1-\eta}{1+\eta}\frac{N\Gamma_{lj}}{\Omega_{lj}^2}\frac{1+\nu I_1/I_s}{(1+I_1/I_s)^2}l,\qquad D_2=\frac{4\pi|d_{ml}|^2}{\lambda_2\hbar}\frac{\eta}{1+\eta}\frac{N\Gamma_{ml}}{\Omega_{ml}^2}l; \quad (4.6)$$

where

$$F=(1+\eta)\Omega_{nl}\Omega_{ml}\Omega_{lj}\left(\frac{\Omega_{ml}\Omega_{nl}}{\Omega_{nj}\Omega_{mj}}-\eta\right)^{-1},$$
$$\nu=\frac{\Gamma_{nl}\Omega_{lj}}{\Gamma_{lj}\Omega_{nl}},\qquad \frac{I_1}{I_s}=\frac{|G_{nj}|^2}{\Omega_{nl}\Omega_{lj}}. \quad (4.7)$$

From (2.2), (4.6), and (4.7) we find that the necessary condition for the excitation of parametric generation is that the intensities satisfy the condition

$$I_1I_2>I_0^2\left(1+\xi\frac{I_1}{I_s}+\zeta\frac{I_1^2}{I_s^2}\right), \quad (4.8)$$

where

$$I_j=\frac{c}{8\pi}|E_{pj}|^2,\quad I_s=\frac{c\hbar^2}{2\pi|d_{lj}|^2}\Omega_{lj}\Omega_{nl},\quad \xi=\frac{2T_1+\nu D_1^0}{\Lambda_1^0 l},\quad \zeta=\frac{T_1}{\Lambda_1^0 l}$$
$$I_0^2=\left(\frac{c}{8\pi^2}\right)^2\frac{\lambda_1\lambda_2\hbar^6\Lambda_1^0\Lambda_2^0F^2}{|d_{jn}d_{nm}d_{ml}d_{lj}|^2N^2}\quad \Lambda_j^0=\frac{T_j+D_j^0}{l},\quad D_j^0=D_j|_{I_1=0}. \quad (4.9)$$

We shall assume throughout that

$$D_j \leqslant T_l. \tag{4.10}$$

If as a result of the trapping of the radiation in the n_j transition and small $\Gamma_l + \gamma_{jl}$, the last two conditions in (4.5), or one of them, are violated and are reversed, whilst the remaining conditions are satisfied, the particles are pumped from level j to level l through stepwise and two-photon processes involving the cascade $j \to n \to m$. At the same time, according to (4.3), $N_l \approx N \gg N_n, N_m, N_j$, i.e., we have population inversion on the transition l_j. In that case,

$$\tilde{\chi}(\omega_{p1}, \omega_{p2}, -\omega_1) = \tilde{\chi}(\omega_{p1}, \omega_{p2}, -\omega_2) = \frac{N|d_{jn}d_{nm}d_{ml}d_{lj}|}{\hbar^3(1+I_1/I_s)F'}, \tag{4.11}$$

$$D_1 = -\frac{4\pi|d_{lj}|^2 N\Gamma_{lj}}{\lambda_1 \hbar \Omega_{lj}^2} \frac{1+\nu I_1/I_s}{(1+I_1/I_s)^2} l, \quad D_2 = \frac{4\pi|d_{ml}|^2 N\Gamma_{ml}}{\lambda_2 \hbar \Omega_{ml}^2} l, \tag{4.12}$$

where $F' = |\Omega_{lj}\Omega_{nl}\Omega_{ml}|$.

The dependence of $\tilde{\chi}$ and D_1 on I_1 is due to the splitting of the level j in the pump field.[13,14] The condition for parametric generation in the case of the inverted population on the transition lj has a form analogous to (4.8), except that $D_{1,2}$ and F' are given by (4.12).

Let us now consider the case of nondegenerate pumping when different fields interact with the transitions nj and nm. The solution of (4.8) for I_1 can be written in the form $I_1^{(1)} < I_1 < I_1^{(2)}$;

$$I_1^{(1,2)} = \frac{1-\xi K \pm \sqrt{(1-\xi K)^2 - 4\xi K^2}}{2\xi K} I_s, \quad K = \frac{I_0^2}{I_s I_2}. \tag{4.13}$$

Figures 4a and 4b give a qualitative illustration of the generation region for the equilibrium population and for inversion in the case of the transition lj, respectively. In contrast to the situation when the susceptibility is field-independent, in the present case, the pump field intensities have both an upper and a lower limit. At a certain value of I_0, the solution of the quadratic equation given by (4.8) may become degenerate $[I_1^{(1)} = I_1^{(2)}]$ which corresponds to the end of generation. Further increase in I_0 prevents generation because the increase in the pump intensity does not ensure that the gain will exceed the saturated losses, due to the dependence of $\tilde{\chi}$ on I_1.

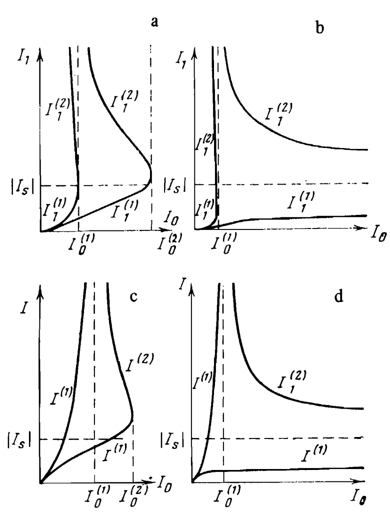

FIG. 4. Regions of four-photon parametric generation. $I_2/|I_S| = 2$, $D_1 = T_1/2$. (a) and (c)—equilibrium populations; (b) and (d)—$N_l > N_j$, (a) and (b)—$\omega_{P1} \neq \omega_{P2}$; (d) and (c)—$\omega_{P1} = \omega_{P2}$. When $I_S > 0$, generation is possible for $I_0 < I_0^{(1)}$ and for $I_S < 0$, generation is possible for $I_0 < I_0^{(2)}$. In Figs. c and d, the symbols next to the curves should be $I_1^{(1)}$ and $I_1^{(2)}$.

Therefore, parametric generation is possible for the following parameter ranges:

$$I_0^2 < (I_0^{(1)})^2, \text{ if } (I_0^{(1)})^2 > 0, \quad \text{and } I_0^2 < (I_0^{(2)})^2, \text{ if } (I_0^{(2)})^2 > 0 \tag{4.14}$$

[here $(I_0^{(1,2)})^2 = I_2 I_S/(\xi \pm 2\sqrt{\xi})$ for any I_0, provided $(I_0^{1,2})^2 < 0$]. In the absence of inversion on the transition lj$(T_1 < \Lambda_1^\circ l)$, generation is possible for $I_0 \leq I_0^{(1)}$ if $I_S > 0$, and $I_0 \leq I_0^{(2)}$ if $I_S < 0$ (Fig. 4a). Therefore, when a nonlinear resonance with one of the quasilevels is possible due to the increase in the parametric susceptibility, the range of parameters in which generation is possible expands $[I_0^{(2)} > I_0^{(1)}]$. If $D_1^0 \ll T_1$, then for $I_S < 0$ we have $I_0^{(2)} \to \infty$. The region near the nonlinear resonance $I_1 = -I_S$ must be excluded, since (4.6), (4.7), (4.11), and (4.12) were obtained under the assumption that

$$\left|1 + \frac{I_1}{I_s}\right| \gg \left|\frac{k_1 \bar{v}}{\Omega_{lj}} + \frac{(k_{p1} - k_1)\bar{v}}{\Omega_{nl}}\right|. \tag{4.15}$$

It is only when this condition is satisfied that we can neglect the Doppler effect in the neighborhood of the nonlinear resonance. Moreover, the equation for the threshold, given by (4.8), was obtained on the assumption that D_1 and $\sigma E_{p1}E_{p2}l$ were small, which is also not satisfied at the exact resonance. In the case of inverted populations $\xi - 2\sqrt{\xi} < 0$, so that if $I < 0$, generation is possible for any I_0 (Fig. 4b).

When the pump is degenerate ($I_1 = I_2 = I$), the solution of (4.8) has the form

$$I^{(1)} < I < I^{(2)},$$

$$I^{(1,2)} = \frac{\xi K \pm \sqrt{\xi^2 K^2 + 4K(1-\xi K)}}{2(1-\xi K)} I_s, \quad K = \frac{I_0^2}{I_s^2}. \tag{4.16}$$

This solution is illustrated graphically in Figs. 4c and 4d in the case of equilibrium and inverted populations on the transition lj, respectively. The generation region in the case of the degenerate pump with equilibrium populations is determined by the following inequalities:

$$I_0 < I_0^{(1)} = \frac{I_s}{\sqrt{\xi}}, \text{ if } I_s > 0, \quad I_0 < I_0^{(2)} = \frac{4I_s}{\sqrt{4\xi - \xi^2}}, \text{ if } I_s < 0. \tag{4.17}$$

Parametric generation has an upper bound only when $I_S < 0$ in the region $I_0 > I_0^{(1)}$. If the populations on the lj transition are inverted, then, as I_0 increase, generation does not terminate (Fig. 4c). The presence of a submillimeter transition can be used to reduce substantially the threshold intensity through a reduction in Ω_{ij}. Absorption at the frequency ω_1 then becomes quite small. If the submillimeter transition corresponds to the transition ml, then according to (4.1) this possibility of reducing the threshold is no longer available. The above discussion of the solutions for the threshold (4.8) enables us to go over to the analysis of specific conditions for frequency conversion.

Consider the possible transformation of frequencies ω_1 and ω_2 for a given degenerate pump frequency ($\omega_{P1} = \omega_{P2}$). Figure 5 shows the variation in the generation threshold with simultaneous variation in ω_1 and ω_2 when ω_1 lies in the infrared. With the chosen values of the parameters, the nonlinear resonance does not appear and $I \approx I_0$. By passing from one intermediate level to another, it is possible to achieve frequency conversion in the optical and infrared bands under quasiresonance conditions. Since for certain pump intensities all the particle may be concentrated on the level l, and this process is difficult to control numeric-

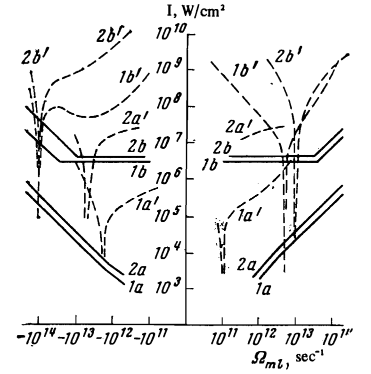

FIG. 5. Tuning of uv and ir frequencies for fixed degenerate pump frequencies. (1) $\lambda_p = 6280$Å, $\lambda_2 \approx 3140$Å, $\lambda_1 \approx 10.6\ \mu$ ($\eta \ll 1$), (a) $\Omega_{nj} = 1.8 \times 10^{12}\ \text{sec}^{-1}$, $\Omega_{mj} = 10^{11}\ \text{sec}^{-1}$, (b) $\Omega_{nj} = 10^{14}\ \text{sec}^{-1}$, $\Omega_{mj} = 5 \times 10^{12}\ \text{sec}^{-1}$; (2) $\lambda_p = 2512$Å, $\lambda_2 = 1256$Å, $\lambda_1 = 10.6\ \mu$, (a) $\Omega_{nj} = 5 \times 10^{12}\ \text{sec}^{-1}$, $\Omega_{mj} = 10^{11}\ \text{sec}^{-1}$, (b) $\Omega_{nj} = 10^{14}$, $\Omega_{mj} = 10^{13}\ \text{sec}^{-1}$. For curves (a): $N_j = N = 5 \times 10^{14}\ \text{cm}^{-3}$, (b) $N_j = N$—curve 2, Fig. 1, $D_p = \leqslant 0.01$. For curves (1a′) and (1b′) $N_l = N$—curve 5, Fig. 1; remaining parameters as for 1a, 1b and 2a, 2b, respectively.

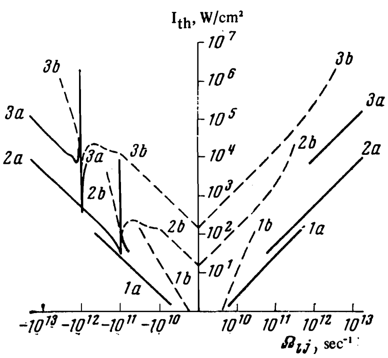

FIG. 6. Tuning of ir and submillimeter radiation for fixed degenerate ir pump frequencies. $\lambda_p = 10.6\ \mu$, $\lambda_1 \approx 100\ \mu$, $\lambda_2 \approx 5.3\ \mu$, (1) $\Omega_{nj} = 2.8 \times 10^9\ \text{sec}^{-1}$, $\Omega_{mj} = -2.8 \times 10^9\ \text{sec}^{-1}$, (2) $\Omega_{nj} = -10^{11}\ \text{sec}^{-1}$, $\Omega_{mj} = 6 \times 10^9\ \text{sec}^{-1}$, (3) $\Omega_{nj} = -10^{12}\ \text{sec}^{-1}$, $\Omega_{mj} = -10^{11}\ \text{sec}^{-1}$. Curves a—equilibrium populations. (1a) $N = 3.2 \times 10^{12}\ \text{cm}^{-3}$, (2a) $N = 1.3 \times 10^{14}\ \text{cm}^{-3}$, (3a) $N = 1.3 \times 10^{15}\ \text{cm}^{-3}$. Curves b—$N_l = N$, curve 3, Fig. 1.

ally, the transformation conditions in the case involving population inversion have been considered separately (curves a′ and b′). We have chosen those cases which are consistent with (4.4) and (4.10) and illustrate the possibilities of reaching low thresholds or large tuning ranges. The rapid reduction in the threshold intensity in the case of inversion for $\Omega_{ml} > 0$ is explained by the resonance on the transition lj, and for $\Omega_{ml} < 0$ by resonance on the forbidden transition nl. The break on the curves 1b and 2b in the region of the lj resonance is explained by the violation of (4.10) in this region. All the curves begin with values of Ω_{ml} for which inhomogeneous broadening can be neglected. The break on curves 1a and 2a is connected with the violation of (4.4). The remaining curves break off when $|\Omega_{ml}| \sim \omega_1 \sim 2 \times 10^{14}\ \text{sec}^{-1}$.

Figure 6 illustrates the generation of frequency-tuned submillimeter radiation using an infrared pump, which means that a higher quantum conversion coefficient can be achieved than for the optical pump. These curves were constructed subject to the same conditions as in Fig. 5. The sharp maxima on curves 3a and 2a for $\Omega_{lj} < 0$ in Fig. 6 are due to the population of the level l. The contributions of the states l and j to the parametric susceptibility (these levels are populated in accordance with the Boltzmann distribution) are mutually compensated in the immediate neighborhood of the ml resonance. This leads to a sharp increase in the threshold intensity and a possible end of generation.

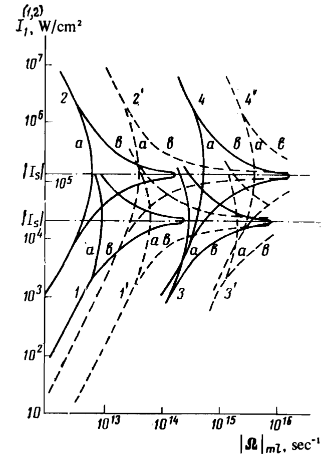

FIG. 7

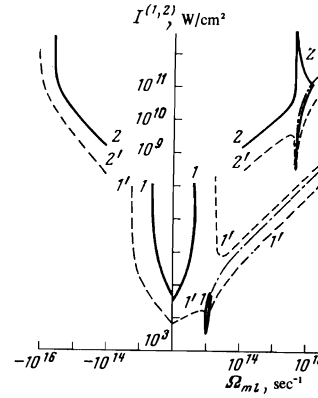

FIG. 8

FIG. 7. Variation in uv frequency due to non-degenerate pump frequency variation (ω_{p2}). The frequencies ω_{p1} and ω_1 are fixed. $\lambda_1 = 100\ \mu$, $\eta = 0.63$, $|\Omega_{lj}| = 10^{10}\ \text{sec}^{-1}$, $N_j + N_l = N = 10^{13}\ \text{cm}^{-3}$. (1) $\lambda_{p1} \approx \lambda_{p2} \approx 6280$Å, $\lambda_2 \approx 3140$Å, $\Omega_{nj} = 10^{11}\ \text{sec}^{-1}$, $I_2 = 10^6\ \text{W/cm}^2$. (2) $\lambda_{p1} \approx \lambda_{p2} \approx 2512$Å, $\lambda_2 = 1256$Å, $\Omega_{nj} = 6 \times 10^{11}\ \text{sec}^{-1}$, $I_2 = 10^{11}\ \text{W/cm}^2$. (3) $\lambda_{p1} \approx \lambda_{p2} \approx 6280$Å, $\lambda_2 \approx 3140$Å, $\Omega_{nj} = 10^{11}\ \text{sec}^{-1}$, $I_2 = 10^9\ \text{W/cm}^2$. (4) $\lambda_{p1} \approx \lambda_{p2} \approx 2512$Å, $\lambda_2 \approx 1256$Å, $\Omega_{nj} = 6 \times 10^{11}\ \text{sec}^{-1}$, $I_2 = 10^{11}\ \text{W/cm}^2$. For curves 1′, 2′, 3′, 4′ the parameters are the same as for 1, 2, 3, 4, respectively, but $N_l = N$. (a) $I_s > 0$ ($\Omega_{lj} > 0$); (b) $I_s < 0$ ($\Omega_{lj} < 0$).

FIG. 8. Variation in the uv frequency for varying degenerate-pump frequency. The frequency ω_1 is fixed. $\lambda_p \approx 2512$Å, $\lambda_2 \approx 1256$Å, $\lambda_1 = 100\ \mu$, $\eta = 0.63$, $\Omega_{lj} = -1.6 \times 10^9\ \text{sec}^{-1}$. (1) $N = 10^{13}\ \text{cm}^{-3}$, $\omega_{mn} - \omega_{nj} = 4 \times 10^{12}\ \text{sec}^{-1}$, (2) $N = 10^{16}\ \text{cm}^{-3}$, $\omega_{mn} - \omega_{nj} = 4 \times 10^{15}\ \text{sec}^{-1}$. For curves 1′ and 2′ the parameters are the same as for $N_l = N$. In the figure read ($-I_s$).

We now consider the transformation of radiation of frequency ω_2 by tuning the pump frequency ω_{p2} with fixed ω_{p1} and ω_1. In this case, the splitting of the level j in a strong pump field becomes important for large frequency changes. In the absence of inversion, we can use (4.14) and (4.9) together with the condition $|\Omega_{lj}| \ll |\Omega_{nj}|, |\Omega_{ml}|$ to show the following inequality which defines the range of variation of Ω_{ml}

$$\Omega_{ml}^2 < (\Omega_{ml}^2)_{\text{crit}} = \frac{32\pi^5 d^6 (1-\eta)^2 N^2 I_2}{c\Lambda_1^0 \Lambda_2^0 ƛ_1 ƛ_2 \hbar^4 (1+\eta)^2 |\Omega_{nl}\Omega_{lj}| \, |\xi \pm 2\sqrt{\zeta}|}. \quad (4.18)$$

The positive sign corresponds to $I_s > 0$ and the lower sign to $I_s < 0$. Figure 7 illustrates the tuning of the frequency ω_2 with the optimum ratio of the parameters and different intensities of the tunable pump. It is clear that, for equilibrium populations the tuning range for $I_s < 0$ will be greater by an order of magnitude than for $I_s > 0$. As I_2 increases, the relative retuning of ω_2 increases from a few percent up to several tens of percent.

If under the action of the pump field there is a population inversion on the lj transition, generation will not cease during frequency retuning due to the splitting of the level j when $I_s < 0$ (curves 1′a–4′a). When $I_s > 0$, the quantity $\Omega_{ml}^{\text{crit}}$ is given by (4.18) with $\eta = 0$.

In the case of a degenerate pump, the conversion of ω_2 at a low fixed value of Ω_{lj} satisfies the condition $\Omega_{ml} \approx \Omega_{nj} + \Omega_{mn}$. According to (4.17) and (4.9), the tuning range depends on N and Ω_{lj} as follows:

$$\Omega_{ml}^2 < (\Omega_{ml}^2)_{\text{crit}} \approx \frac{16\pi^4 d^4 (1-\eta)^2}{ƛ_1 ƛ_2 \Lambda_1^0 \Lambda_2^0 \hbar^2 (1+\eta)^2} N^2 \begin{cases} 4[\zeta - \xi^2/4]^{-1}, & \text{if } I_s < 0, \\ \zeta^{-1}, & \text{if } I_s > 0. \end{cases} \quad (4.19)$$

Figure 8 illustrates the conversion of radiation in the far uv. The curves were plotted for the optimum situation when the maximum possible tuning range [according to (4.19)] corresponded to the nj resonance. The break on curve 1 is due to the change in the sign of Ω_{nj} during the tuning process, so that $I_s < 0$ for $\Omega_{ml} > \Omega_{ml}^{crit}$. For the remaining curves, Ω_{nj} and I_s change sign for $\Omega_{ml} < \Omega_{ml}^{crit}$. As before, (4.4) and (4.10) were satisfied for these graphs.

We may thus conclude that the above analysis and illustrations show that, when spatial synchronism can be achieved, gases can be effectively employed for the vacuum uv and far ir in the case of four-photon parametric generation under quasiresonance conditions.

5. CONDITIONS FOR PHASE SYNCHRONISM

We shall now consider the conditions for precise phase matching ($\Delta k = 0$). Synchronous interaction between waves may be ensured either by specifying the angles between the vectors k_j, or with the aid of a synchronizing buffer gas. The refractive indices at the frequency ω_j have the form

$$n_j \approx 1 + 2\pi \operatorname{Re} \chi_s(\omega_j),$$

where $\chi_s(\omega_j)$ is the susceptibility at frequency ω_j which, in general, is a function of both $|G_p|^2$ and $|G_{1,2}|^2$. The terms which depend on $|G_{1,2}|^2$ determine the stationary generation states and, since we are concerned with the analysis of only the generation threshold, they will not be considered. For $I \ll I_s$, the refractive indices are determined by linear dispersion.

In the two-level system, where the main saturation effect is the saturation of the population difference, the conditions for synchronism for $I \gg I_s$ are merely less stringent. Using (3.2) and (3.6), we have for the collinear interaction

$$\delta kl = (k_1 + k_2 - 2k_p) l = -\frac{4\pi N |d|^2 l \Delta^2}{\hbar \lambda_p \Omega_1 \Omega_2 \Omega_p} \left(1 + \frac{I}{I_s}\right)^{-1} = -D^0 \frac{\Omega_p}{\Gamma} \frac{\Delta^2}{\Omega_p^2 - \Delta^2} \left(1 + \frac{I}{I_s}\right)^{-1}. \quad (5.1)$$

For fixed Ω_p and pump absorption D^0, the quantity δkl increases as Δ approaches Ω_p. Precise synchronism ($\Delta \mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 - 2\mathbf{k}_p = 0$) can be ensured for the noncollinear interaction when $\delta kl > 0$. The latter occurs when

$$\Omega_p < 0 \text{ for } \Delta < |\Omega_p|; \quad \Omega_p > 0 \text{ for } \Delta > \Omega_p. \quad (5.2)$$

In this case, the angles $\Theta_{1,2}$ between $\mathbf{k}_{1,2}$ and $\mathbf{k}_p$ in the region of small values $\Theta_{1,2} \ll 1$ are given by

$$\Theta_1 = \frac{\lambda_1}{\lambda_2} \Theta_2 \approx \left(\frac{2\lambda_1}{l} \frac{\delta k_1 l}{1 + \lambda_2/\lambda_1}\right)^{1/2}. \quad (5.3)$$

Assuming that $\Delta = 10^{12}$ sec^{-1} in the case corresponding to curve 2 in Fig. 2, and $\Delta = 10^{13}$ sec^{-1} in the case of curve 4, we have $\delta kl = 0.14$ and $\delta kl = 4 \times 10^{-2}$, respectively. In both cases, $\delta kl < \pi/2$, which allows collinear interaction.

We must now consider the possible application of the synchronizing gas as a means of ensuring collinear interaction when $|\delta kl| > \pi/2$. The synchronizing impurity $\delta k_b l$ is given by

$$\delta k_b l = -\frac{4\pi N_b d_b^2 l \Delta^2}{\hbar \lambda_p \Omega_1^b \Omega_2^b \Omega_p^b} \left(1 + \frac{I}{I_s^b}\right)^{-1} = -D^0 \frac{\Omega_p^b}{\Gamma_b} \frac{\Delta^2}{\Omega_p^{b2} - \Delta^2} \left(1 + \frac{I}{I_s^b}\right)^{-1}, \quad (5.4)$$

where $\Omega_{j,p}^b = \omega_{j,p} - \omega_b$, ω_b is the frequency of the resonance transition in the buffer gas, which is closest to the frequency ω_p, and N_b is the density of the synchronizing impurity. It is clear from (5.1) and (5.4) that the synchronizing condition $\delta k_b = -\delta k$ can be satisfied for $|\Omega_p^b| \sim |\Omega_p|$, $D_b^0 \lesssim D^0$. The ratio of the signs of Ω_p^b and Ω_p depends on Δ (for $\Delta < |\Omega_p|$, $\Omega_p^b/\Omega_p < 0$). The parametric susceptibility due to the interaction between the fields and both subsystems is given by the expression

$$\tilde{\chi} \simeq -\frac{2d^4 N}{\Omega_p(\Omega_p^2 - \Delta^2)} \left(1 + \frac{I}{I_s}\right)^{-1} - \frac{2d_b^4 N_b}{\Omega_p^b(\Omega_p^{b2} - \Delta^2)} \left(1 + \frac{I}{I_s^b}\right)^{-1}$$

and does not vanish when $d \neq d_b$.

Let us now consider phase synchronism for parametric conversion in the case of the four-level system. For the degenerate pump, and with (4.4) satisfied, we have

$$\delta k = -\frac{2\pi}{\hbar} \left[\frac{|d_{ml}|^2 r_l}{\lambda_2 \Omega_{ml}} + \frac{|d_{lj}|^2 (r_j - r_l)}{\lambda_1 \Omega_{lj}} \left(1 + \frac{I}{I_s}\right)^{-1} - 2\frac{|d_{nj}|^2 r_j}{\lambda_p \Omega_{nj}}\right]. \quad (5.5)$$

For a nondegenerate pump, the factor 2 in front of the last term is replaced by unity. The contribution of the submillimeter transition lj to δk for equilibrium populations exceeds the contribution of the pump only for

$$|\Omega_{lj}(1 + I/I_s)| < 10^{-3} |\Omega_{nj}|.$$

Therefore, for the cases considered in Figs. 7 and 8, the main contribution to δk is due to the pump, and

$$|\delta kl| \approx {}^1/_2 D^0 |\Omega_{nj}| / \Gamma_{nj} \sim 1 \div 10.$$

Analysis of the cases illustrated in Figs. 5 and 6 also shows that, except for some individual and very narrow frequency regions, the main contribution to δk in the absence of population inversion on the lj transition is due to the pump, whilst in the case of inversion this is due to radiation at frequency ω_2. In both cases, $|\delta kl| \leq 1 - 20$. According to (5.3), this detuning can be compensated when $\Omega_{nj} > 0$ ($\Omega_{ml} < 0$ in the case of inversion) for angles $\Theta_1 \sim 0.1$ when $\lambda_1 = 100\ \mu$ and $\Theta_1 < 0.01$ when $\lambda_1 = 10\ \mu$. Another possibility is to use a buffer gas, the main contribution of which to the refractive index

$$\delta k_b l = -\frac{2\pi}{\hbar} \frac{d_b^2 N_b}{\lambda_p \Omega_p^b} l = -D_b^0 \frac{\Omega_p^b}{2\Gamma^b}.$$

Synchronism is ensured during absorption in the buffer gas, which is not greater than in the nonlinear medium, provided

$$\left|\frac{d_{nj}}{d_b}\right|^2 \frac{N}{N_b} \frac{\Omega_p^b}{\Omega_{nj}} \approx -1, \qquad \left|\frac{d_b}{d_{nj}}\right|^2 \frac{\Gamma^b N_b \Omega_{nj}^2}{N \Gamma_{nj} \Omega_p^{b2}} \leqslant 1.$$

When collisional broadening in both media exceeds radiative broadening, i.e., $\Gamma \sim 22.8 N d^2/\hbar$, the last two equations are satisfied simultaneously.

Thus, phase synchronism during parametric interaction of the above type can be achieved, and the specific synchronism conditions are determined by each particular situation.

Analysis of time-independent generation in the case of an internal pump, based on the assumption that synchronism is accurately maintained during the generation process,[12] shows that this is possible and the results are analogous to those obtained in[17]. The generated power may then reach a few tens of percent of the pump power.

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