FOCUSING EFFECTS FOR LASING PROCESSES ON FIFTH-ORDER NONLINEAR SUSCEPTIBILITY IN ISOTROPIC MEDIA

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Frequency mixing in focused beams on a fifth-order nonlinear susceptibility in isotropic media is discussed theoretically. A detailed investigation is made of the behavior of focusing functions, analytical solutions are found, and conditions under which these solutions are valid are ascertained. Power optimization in focusing is discussed for all processes involved. Cascade processes in focused beams are discussed cursorily.

Focusing of laser beams is an effective way to enhance conversion efficiency in nonlinear interactions, and this is particularly crucial for phase mismathcing processes when phase-lock conditions are impossible to maintain or are fraught with great experimental difficulties. It should be noted that beam focusing at the enter of a nonlinear medium is the only available way, in investigations of gaseous media, to avoid breakdown of cell windows while at the same time producing the required power density.

Conversion efficiency in focusing is enhanced by raising pump power density with simultaneous partial or complete (vector synchronism) compensation of phase mismatch via the noncollinearity of the interaction in the focused beam. The effect of focusing is determined first and foremost by the degree of focusing, i.e., by the ratio of the length of the nonlinear medium to the confocal parameter. The effect of focusing contrasts quite sharply in the limiting cases of loose and tight focusing, when this ratio is respectively much smaller than or much larger than unity. In the general case, a function taking into account the effect of focusing on the effectiveness of some process depends on a large array of parameters. In each specific case it is important to know the optimum focusing conditions in order to attain maximum conversion efficiency. But analysis of the behavior of the function aimed at ascertaining these conditions is possible only for the above limiting cases when analytical solutions can be found for the function, and in all remaining cases computer numerical calculations are the only option available.

The overwhelming majority of investigations carried out in nonlinear optics at the present time involves processes occurring on second-order and third-order nonlinear susceptibilities, but in recent years interest in research on many-quantum interactions on higher (than third-order) nonlinear susceptibilities has been on the increase for a number of reasons. Focusing effects for processes occurring on lower-order nonlinearities have been studied in detail in the literature [1-6]. The generation of higher-order harmonics in focused beams has also been studied [6,7]. In particular, close attention has been given [7] to the treatment of cascade processes (see also [8]) in fifth-harmonic generation and the problem has been solved numerically for the case of three coupled waves in an isotropic medium; the results so obtained argue for the feasibility of efficient conversion to the fifth harmonic. Conversion efficiency evaluations for some many-quantum processes are also cited in [9]. Finally, we take note of the first experimental research work [10,11] done on processes occurring on higher-order nonlinearities.

This article presents results of a theoretical investigation of frequency mixing processes occurring in focused beams on a fifth-order nonlinear susceptibility. Nonresonance, stationary (quasistationary) mixing processes in an isotropic medium are discussed. Expressions for the power of the radiation generated are derived in the prespecified-field approximation. The behavior of the focusing functions is analyzed in detail, and analytical solutions are found in the limiting cases of loose and tight focusing, while the range of applicability of the analytical solutions is discussed. Results of computer numerical integration of the focusing functions are plotted graphically in some instances. Power optimization is discussed for all types of processes involved. The theory of cascade processes in focused beams is discussed briefly.

THEORY

Frequency mixing on a fifth-order nonlinear susceptibility is a six-photon process and is characterized in general by the interaction of five distinct pump fields of frequency $\omega_n (n = 1, \ldots, 5)$ exciting nonlinear polarization waves of frequencies $\omega_6=\omega_i\pm\omega_k\pm\omega_l\pm\omega_m\pm\omega_p$ $(i, k, l, m, p = 1, \ldots, 5)$ in the medium. With no loss in generality, we can restrict the discussion in what follows to the following frequency-nondegenerate interactions:

$$\omega_6 = \begin{cases} \omega_1+\omega_2+\omega_3+\omega_4+\omega_5 & (1.1)\\ \omega_1+\omega_2+\omega_3+\omega_4-\omega_5 & (1.2)\\ \omega_1+\omega_2+\omega_3-\omega_4-\omega_5\,, & (1.3)\\ \omega_1+\omega_2-\omega_3-\omega_4-\omega_5 & (1.4)\\ \omega_1-\omega_2-\omega_3-\omega_4-\omega_5 & (1.5)\end{cases}$$

which represent all modes of interactions occurring in frequency mixing on fifth-order susceptibilities. (Here and in what follows, the type of process is indicated by the second digit in the numeration of the expresssions.) All of the remaining partially or totally frequency-degenerate interactions constitute particular cases of interactions (1).

It is also assumed below that the conversion process is stationary and nonresonant, and the pump fields are assumed to be specified functions of the coordinates (specified-field approximation).

We shall assume that pumping comprises lower-order Gaussian beams propagating down the z-axis, with field amplitudes E_{n0} and wave vectors k_n; all of the beams are focused with the same confocal ratio and same position of the beam-waist region (beam foci) along the z-axis. The total pump field in the medium is representable in this case in the form

$$\mathbf{E}(\mathbf{r}, t)=\frac{1}{2}\sum_{n=1}^{5}\{\mathbf{E}_n(\mathbf{r})e^{-i\omega_n t}+\mathbf{E}_n^*(\mathbf{r})e^{i\omega_n t}\}, \qquad (2)$$

$$\mathbf{E}_n(\mathbf{r})=\mathbf{E}_{n0}(1+i\xi)^{-1}e^{ik_n z}\exp\left[-\frac{k_n(x^2+y^2)}{b(1+i\xi)}\right], \qquad (3)$$

where $b = k_n w_{0n}^2$ is the confocal focusing ratio (here w_{0n} are the beam radii in the focal plane), f is the position of the focal plane along the z-axis, and $\xi=2(z-f)/b$ is a dimensionless normalized coordinate.

In the general case of elliptical polarization of the vectors $\mathbf{E}_{n0}$, the relationship between nonlinear polarization of the medium at frequency ω_6 and the pump fields is expressed in terms of all of the nonlinear susceptibility tensor components and is exceedingly cumbersome. We shall assume that all of the pump fields are linearly polarized in one direction (say, along the x-axis). In this case the vectorial nature of the fields can be neglected, and we can state the nonlinear polarization of the medium in scalar form

$$\mathcal{P}_6(\mathbf{r}, t)=\frac{1}{2}\{\mathcal{P}_6(\mathbf{r})e^{-i\omega_6 t}+\mathcal{P}_6^*(\mathbf{r})e^{i\omega_6 t}\}, \qquad (4)$$

where $\mathcal{P}_6(\mathbf{r})$ is equal, in the case of an interaction of the first type (frequency addition), to

$$\mathcal{P}_6(\mathbf{r})=\frac{15}{2}N\chi^{(5)}(\omega_6)\prod_{n=1}^{5}E_n(\mathbf{r}). \qquad (5.1)$$

Here N is the density of the nonlinear medium, $\chi^{(5)}(\omega_6)\equiv\chi_{xxxxxx}(\omega_6)$ is the susceptibility of fifth order calculated per atom. The formulas for the remaining modes of interactions are analogous, with $E_n(\mathbf{r})$ replaced by $E_n^*(\mathbf{r})$, for each negative frequency ω_n.

For definiteness, we shall assume that the nonlinear medium occupies the half space $z > 0$, and that the half space $z < 0$ is a vacuum.

With this in mind, we obtain, from Eqs. (3) and (5),

$$\mathcal{P}_6(\mathbf{r}) = \frac{15}{2} N\chi^{(5)}(\omega_6) \prod_{n=1}^{5} E_{n0} W(\xi)(1+\xi^2)^{-1} \times \\ \times \exp(ik'z)\exp\left[-\frac{(k''-ik'\xi)(x^2+y^2)}{b(1+\xi^2)}\right] B(z), \tag{6}$$

where $k' = k_1 + k_2 + k_3 + k_4 + k_5$, $B(z) = \{1$ when $z > 0$ or 0 when $z < 0\}$, and the nonlinear polarization wave vector k' and the term $W(\xi)$ for these interaction modes take on the form

$$k' = \begin{cases} k_1+k_2+k_3+k_4+k_5 \equiv k'' \\ k_1+k_2+k_3+k_4-k_5 \\ k_1+k_2+k_3-k_4-k_5; \\ k_1+k_2-k_3-k_4-k_5 \\ k_1-k_2-k_3-k_4-k_5 \end{cases} \quad W(\xi) = \begin{cases} (1+i\xi)^{-4}(1-i\xi) & (7.1) \\ (1+i\xi)^{-3} & (7.2) \\ (1+i\xi)^{-2}(1-i\xi)^{-1} & (7.3) \\ (1+i\xi)^{-1}(1-i\xi)^{-2} & (7.4) \\ (1-i\xi)^{-3} & (7.5) \end{cases}$$

The truncated wave equation for the slow amplitude of the field of the wave generated $A_6(\mathbf{r})$ takes the form [12]:

$$2ik_6 \frac{\partial A_6(\mathbf{r})}{\partial z} + \Delta_\perp A_6(\mathbf{r}) = -4\pi k_{60}^2 \mathcal{P}_6(\mathbf{r}) e^{-ik_6 z}, \tag{8}$$

where k_{60} and k_6 are the wave vector of the wave generated in vacuum and the wave vector of the wave generated in the medium, $\Delta_\perp$ is the transverse Laplacian. The solution of Eq. (8) can be found either with the aid of the Green's function [1,3] or by the spectral method [2,5,6]. When $|\Delta k| \ll k_6, k'$, we have for $E_6(\mathbf{r}) = A_6(\mathbf{r}) e^{ik_6 z}$:

$$E_6(\mathbf{r}) = i\frac{15}{2}\pi \frac{k_{60}^2 b}{k_6} N\chi^{(5)}(\omega_6) \prod_{n=1}^{5} E_{n0} e^{ik'z} \times \\ \times \int_{-\xi}^{\xi} d\xi' \, e^{-i\frac{b\Delta k}{2}(\xi'-\xi)} \frac{W(\xi')}{(k''-ik'\xi')H(\xi,\xi')} e^{-\frac{(x^2+y^2)}{bH(\xi,\xi')}}, \tag{9}$$

where $\xi = \frac{2f}{b}$ is the dimensionless normalized coordinate of the beam focus (beam-waist region), $\Delta k = k_6 - k'$ is the mismatch of the wave vectors of the wave generated and of the nonlinear polarization of the medium:

$$H(\xi,\xi') = \frac{1+\xi'^2}{k''-ik'\xi'} - i\frac{\xi'-\xi}{k'}. \tag{10}$$

If we put $\xi = 2(L-f)/b$, where L is the length of the nonlinear medium, then Eq. (9) fully determines the field of the wave generated in the $z = L$ plane upon emergence from the nonlinear medium. In that case the power P_6 of the radiation generated will be determined by the expression

$$P_6 = (4{,}504 \cdot 10^{-7}) \frac{k_{60}^4 k_1 k_2 k_3 k_4 k_5}{b^2 k_6^2 k'} N^2 [\chi^{(5)}(\omega_6)]^2 P_1 P_2 P_3 P_4 P_5 \times \\ \times \frac{n_6}{n_1 n_2 n_3 n_4 n_5} F_j\left(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'}\right). \tag{11}$$

Here n_i are the refraction indices of the waves in the medium and the powers of all the waves are expressed in watts, while the remaining quantities are expressed in the CGSE system. The dimensionless functions $F_j\left(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'}\right)$ take into account focusing effects and exhibit the form (subscript j denoting the type of process):

$$F_j\left(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'}\right) = \frac{2k'}{\pi b} \int_0^\infty 2\pi R\, dR \times \tag{12}$$

$$\times \left| \int_{-\zeta}^{\xi} d\xi' \, e^{-i \frac{b\Delta k}{2}(\xi'-\xi)} \frac{W(\xi')}{(k'' - ik'\xi')\, H(\xi', \xi)} \, e^{-\frac{R^2}{bH}} \right|^2. \tag{12}$$

In the case of partially or completely frequency-degenerate processes, the numerical coefficient in Eq. (11) will be different. This coefficient can be represented in the form $(8.007 \cdot 10^{-9})\alpha^2$, where α is the numerical coefficient in the expression of type 5 for the corresponding process. In particular, when a fifth harmonic is generated, $\alpha = 1/16$ and the numerical coefficient in Eq. (11) is $3.128 \cdot 10^{-11}$.

INVESTIGATION OF FUNCTIONS $F_j(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'})$

Formulas (12) can be treated most conveniently as dependences on the parameter $b\Delta k$ at fixed values of the remaining parameters. The general nature of this dependence is determined by the size of the focusing parameter L/b held fixed, and differs substantially in the limiting cases of loose and tight focusing. In those cases, Eqs. (12) lend themselves to simplification and analytical solutions can be found.

At the outset, we consider the loose focusing case $(b \gg L)$ which, generally speaking, always occurs even in unfocused beams, because of beam divergence. In that case F_j is representable in the form

$$F_j\left(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'}\right)_{b>L} = 4\frac{k'}{k''}\left(\frac{L}{b}\right)^2 \operatorname{sinc}^2\left[\left(-\frac{b\Delta k}{2} + m_j\right)\frac{L}{b}\right], \tag{13}$$

where

$$m_j = -\frac{k''}{k'} + \begin{cases} 5 & (14.1) \\ 3 & (14.2) \\ 1 & (14.3) \\ -1 & (14.4) \\ -3 & (14.5) \end{cases}$$

Recalling that $\xi' \leqslant \xi \ll 1$, terms linear in ξ and ξ' are retained everywhere in the derivation of Eq. (13). Clearly, we see from Eq. (13), in this approximation the position of the beam focus does not affect the value of F_j (of course, within the framework of the approximation, i.e., when $f \ll b$). Estimates show that Eqs. (13) are valid when $\frac{L}{b}\frac{k''}{k'} \leqslant 0{,}1$.

In the case of tight focusing at the center of the nonlinear medium ($\frac{f}{L} = 0{,}5;\ \frac{b}{L} \to 0,$ $\zeta = \xi \to \infty$), we can state

$$F_j\left(b\Delta k;\ 0;\ 0{,}5;\ \frac{k''}{k'}\right) = \left| \int_{-\infty}^{+\infty} d\xi' \frac{W(\xi')}{\left(\frac{k''}{k'} - i\xi'\right)} e^{-i\frac{b\Delta k}{2}\xi'} \right|^2, \tag{15}$$

and hence, utilizing residue theory, we arrive at

$$F_1(b\Delta k;\ 0;\ 0{,}5;\ 1) = \begin{cases} \frac{\pi^2}{9}\left(\frac{b\Delta k}{2}\right)^6 e^{b\Delta k}, & \Delta k < 0, \\ 0, & \Delta k > 0 \end{cases} \tag{16.1}$$

$$F_2\left(b\Delta k;\ 0;\ 0{,}5;\ \frac{k''}{k'}\right) = \begin{cases} \frac{\pi^2}{16\left(\frac{k''}{k'}+1\right)^2}\left\{\left[b\Delta k\left(\frac{k''}{k'}+1\right)^2 - 2\right]^2 + 4\right\}^2 e^{b\Delta k}, & \Delta k \leqslant 0 \\ \frac{4\pi^2}{\left(\frac{k''}{k'}+1\right)^6} e^{-b\Delta k \frac{k''}{k'}}, & \Delta k \geqslant 0 \end{cases} \tag{16.2}$$

$$F_3\left(b\Delta k;\ 0;\ 0{,}5;\ \frac{k''}{k'}\right)=$$

$$=\begin{cases}\dfrac{\pi^2}{4\left(\frac{k''}{k'}+1\right)^4}\left[(b\Delta k-1)\left(\frac{k''}{k'}+1\right)-2\right]^2 e^{b\Delta k},\ \Delta k\leqslant 0\\[2ex] \dfrac{\pi^2}{4\left(\frac{k''}{k'}+1\right)^4\left(\frac{k''}{k'}-1\right)^2}\left[\left(\frac{k''}{k'}+1\right)^2-4e^{-\frac{b\Delta k}{2}\left(\frac{k''}{k'}-1\right)}\right]^2 e^{-b\Delta k},\ \Delta k\geqslant 0\end{cases} \tag{16.3}$$

$$F_4\left(b\Delta k;\ 0;\ 0{,}5;\ \frac{k''}{k'}\right)=$$

$$=\begin{cases}\dfrac{\pi^2}{4\left(\frac{k''}{k'}+1\right)^2}e^{b\Delta k},\ \Delta k\leqslant 0\\[2ex] \dfrac{\pi^2}{4\left(\frac{k''}{k'}-1\right)^4}\left\{\left[(b\Delta k+1)\left(\frac{k''}{k'}-1\right)-2\right]+\right.\\[1ex] \left.+\dfrac{4}{\left(\frac{k''}{k'}+1\right)}e^{-\frac{b\Delta k}{2}\left(\frac{k''}{k'}-1\right)}\right\}^2 e^{-b\Delta k},\ \Delta k>0,\end{cases} \tag{16.4}$$

$$F_5\left(b\Delta k;\ 0;\ 0{,}5;\ \frac{k''}{k'}\right)=$$

$$=\begin{cases}0,\ \ \Delta k\leqslant 0\\[1ex] \dfrac{\pi^2}{16\left(\frac{k''}{k'}-1\right)^6}\left\{\left[b\Delta k\left(\frac{k''}{k'}-1\right)-2\right]^2+4-8e^{-\frac{b\Delta k}{2}\left(\frac{k''}{k'}-1\right)}\right\}^2 e^{-b\Delta k},\ \Delta k>0.\end{cases} \tag{16.5}$$

In tight focusing onto the entrance ($f=0$) or exit ($f=L$) boundary of the medium, one of the limits of the integral in Eq. (15) goes to zero, so that

$$F_j\left(b\Delta k,\ 0,\ 0,\ \frac{k''}{k'}\right)=F_j\left(b\Delta k,\ 0,\ 1,\ \frac{k''}{k'}\right)=$$
$$=\frac{1}{4}F_j\left(b\Delta k;\ 0;\ 0{,}5;\ \frac{k''}{k'}\right)+F_j',$$

where F_j' is the contribution made by the imaginary part of the integral. In particular, for the frequency addition process we have

$$F_1'=\begin{cases}\dfrac{1}{36}\left[\left(\dfrac{b\Delta k}{2}\right)^3 e^{\frac{b\Delta k}{2}}\,\overline{Ei}\left(-\dfrac{b\Delta k}{2}\right)+\right.\\[1ex] \left.+\left(\dfrac{b\Delta k}{2}\right)^2-\dfrac{b\Delta k}{2}+2\right]^2,\ \Delta k\leqslant 0\\[2ex] \dfrac{1}{36}\left[\left(\dfrac{b\Delta k}{2}\right)^3 e^{\frac{b\Delta k}{2}}\,\overline{Fi}(-b\Delta k)+\right.\\[1ex] \left.+\left(\dfrac{b\Delta k}{2}\right)^2-\dfrac{b\Delta k}{2}+2\right]^2,\ \Delta k\geqslant 0\end{cases} \tag{17.1}$$

The maximum value of $F_j' = 0.3153$ at $b\Delta k=-2{,}44$. Here $E_i(x)$, $\overline{E}_i(x)$ are the integral exponential functions. The formulas for the remaining F_j' contain similar, but more cumbersome, functions, and need not be cited here.

When the focus of the beams is displaced from the boundary of the medium toward the center of the medium, the contribution made by the imaginary part of the integral, odd with respect to ξ', declines to zero in short order, while the contribution made by the real part of the integral tends rapidly to the value of F_j ($b\Delta k$; 0; 0.5; k''/k'). In physical terms, the explanation is that the basic contribution made to the amplitude of the field generated is that made by the region near the focus of the beams with a high pump field intensity, the dimensions of the region are relatively modest, and as long as it is accommodated entirely within the cell F_j remain weakly dependent on the position of the beams' focus and equal their value for focusing at the center of the medium. Estimates show that the difference from Eqs. (16) does not exceed one percent for any value of $b\Delta k$, provided $\frac{k''}{k'}b\ll f\ll\frac{k''}{k'}(L-b)$.

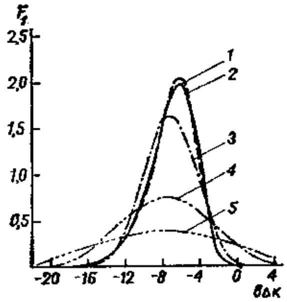

Fig. 1

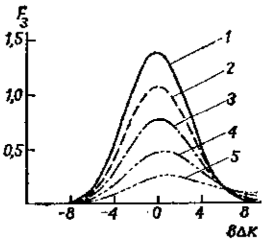

Fig. 2

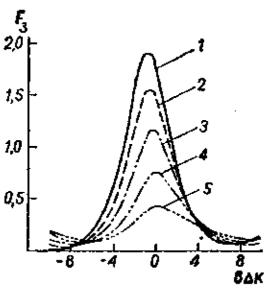

Fig. 3

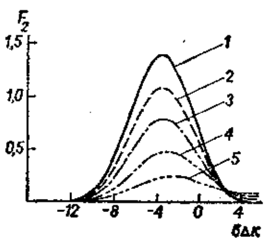

Fig. 4

Fig. 1. $F_1(b\Delta k, b/L, 1/2, 1)$ as a function of $b\Delta k$ for: 1) $b/L = 3$, 2) $b/L = 2$, 3) $b/L = 1$, 4) $b/L = 0.5$ and 5) $b/L = 1/3$.

Fig. 2. $F_2(b\Delta k, 1, 1/2, k''/k')$ as a function of $b\Delta k$ for: 1) $k''/k' = 1.2$; 2) $k''/k' = 1.5$; 3) $k''/k' = 2$; 4) $k''/k' = 3$ and 5) $k''/k' = 5$.

Fig. 3. $F_2(b\Delta k, 1/3, 1/2, k''/k')$ as a function of $b\Delta k$ for k''/k'. Notation same as in Fig. 2.

Fig. 4. $F_3(b\Delta k, 1, 1/2, k''/k')$ as a function of $b\Delta k$ for k''/k'. Notation same as in Fig. 2.

The condition for validity of Eqs. (16) can be estimated most simply for the function F_1 (frequency addition). In this case $k' \equiv k''$ and the transition from the exact expression (12) to the approximate expression (15) involves no more than a replacement of the finite limits $\xi = \zeta$ by infinite limits. The error in the value of F_1 incurred in that replacement satisfies at any value of $b\Delta k$ the inequality

$$\Delta F_1 < 4\left[1+\left(\frac{L}{b}\right)^2\right]^{-2}\times \{F_1(b\Delta k;\ 0;\ 0{,}5;\ 1)\}^{1/2}. \tag{18}$$

For all the remaining functions, at $k' \neq k''$, the conversion to (15) involves, in addition to substitution of limits, passage to the limit under the integral sign in Eq. (12) as $\xi \to \infty$, and an additional constraint is imposed on the validity of this conversion. In these cases we can resort to the following approximate estimate: for any value of $b\Delta k$ we have

$$\Delta F_i \leqslant 4\left[1+\left(\frac{L}{b}\,\frac{k'}{k''}\right)^2\right]^{-2}\left\{F_i\left(b\Delta k;\ 0;\ 0{,}5;\ \frac{k''}{k'}\right)\right\}^{1/2}. \tag{19}$$

We infer from formulas (18) and (19) that Eqs. (16) can be used to good advantage right up to $(k'/k'')(L/b) = 3$, whereupon $\Delta F_i \leqslant 0{,}06$ for all values of $b\Delta k$.

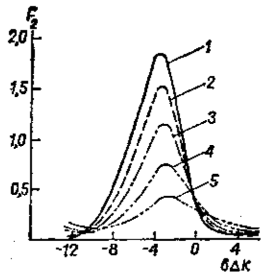

Fig. 5. $F_3(b\Delta k, 1/3, 1/2, k''/k')$ as a function of $b\Delta k$ for k''/k'. Notation same as in Fig. 2.

At values of the focusing parameter $L/b \sim 1$, approximations (13) and (16) are unacceptable, and the only recourse open is to computer numerical calculations of Eq. (12). Graphs of the dependence of function F_1 on the parameter $b\Delta k$ for focusing at the center of the medium are plotted in Fig. 1 for values of the focusing parameter $L/b = 1/3, 1/2, 1, 2, 3$; note that when $L/b = 3$ the difference from values computed using Eq. (16.1) agree quite closely with the estimate (18) at any $b\Delta k$ value. Graphs of F_2 as a function of $b\Delta k$ are plotted in Fig. 2 and Fig. 3 for focusing at the center of the medium in the cases $L/b=1$ and $L/b=3$, if $k''/k' = 1.2, 1.5, 2.0, 3.0, 5.0$. Similar graphs appear in Fig. 4 and Fig. 5 for the function F_3.

POWER OPTIMIZATION IN FOCUSING

Under conditions where the effect exerted by limiting factors (absorption, self-focusing, saturation, breakdown) can be safely neglected, Eq. (11) is valid for the power and

$$P_6 \sim [\chi^{(5)}(\omega_6)]^2 \left(\prod_{n=1}^{5} P_n\right) \frac{N^2}{b^2} F_j \left(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'}\right).$$

If the values of P_n, $\chi^{(5)}$ and k''/k' can be assigned, then power optimization reduces to optimization of the ratio $N^2b^{-2}F_j$ as the parameters $N, \Delta k, b, \frac{b}{L}$ and f/L are allowed to vary. Let us consider some characteristic patterns.

1. From the results of the preceding section, we infer that the most efficient focusing in any case will occur when

$$\frac{k''}{k'} b/L \ll f/L \ll \frac{k''}{k'}\left(1 - \frac{b}{L}\right).$$

Focusing is considered optimal when at the center of the nonlinear medium $\left(f = \frac{L}{2}, \xi = \zeta\right)$.

2. Mismatch of the wave vectors Δk in liquid and gaseous media can be varied independently of the density N via the introduction of buffering impurities to alter the refraction indices without affecting the nonlinear properties of the medium. In that case the dependence on N will be obvious, and it will be sufficient to vary Δk with b held fixed in order to optimize function F_j.

If no special measures are taken to vary Δk, then $\Delta k \sim N$ and the value of the product $(b\Delta k)^2 F_j$ will have to be optimized by allowing Δk to vary with b held fixed, whereupon the optimum wave vector mismatch $(\Delta k)_{opt}$ will also determine the optimum density N_{opt} of the medium.

3. While the value of Δk is held fixed, the optimum value of the confocal parameter b can be determined from the maximization condition for $(b\Delta k)^{-2}F_j$.

When $\Delta k \neq 0$, power optimization is possible in the limit as $b \to 0$ only in the case of processes $\omega_6 = \omega_1 + \omega_2 + \omega_3 + \omega_4 + \omega_5$ and $\omega_6 = \omega_1 - \omega_2 - \omega_3 - \omega_4 - \omega_5$. The power increases without bound as the confocal parameter is decreased, for all the remaining types of processes at $\Delta k \neq 0$ (and if Δk is independent of N, the same holds for $\Delta k = 0$ also). In these cases, power optimization reduces to an initial assignment of minimum b value at which all of the limiting processes can be safely neglected, with further optimization based on varying Δk (see section 2).

The selection of optimum conditions is simplified in those cases where analytical solutions are valid for the functions F_j.

In the loose focusing case $\left(\frac{k''}{k'}\frac{L}{b} \ll 10^{-1}\right)$, we infer from formula (13) that $P_6 \sim b^{-4}\,\mathrm{sinc}^2 \cdot \left[\left(\frac{b\Delta k}{2} + m_j\right)\frac{L}{b}\right]$. Since the value of the confocal parameter is always finite ($b = kw_0^2$) in the case of confined beams, even if unfocused, synchronous operation in such beams is achieved at some small mismatch of the wave vectors $\Delta k_{opt} = -2m_j/b$, and this condition goes over into the condition $\Delta k_{opt} = 0$ for the case of an infinite plane wave in the limit as $b\to\infty$.

In the tight focusing case, Eqs. (16) are valid with sufficient accuracy all the way to $(k'/k'')(L/b) \geq 3$. Consider here the optimization conditions for the frequency addition process. With $\Delta k\neq 0$ held fixed, and b allowed to vary (see section 3), power is optimized at $b\Delta k=-4$. With b held fixed and Δk allowed to vary (see section 2), power is optimized at $b\Delta k=-6$, provided Δk is a parameter independent of N, and at $b\Delta k=-8$ when $\Delta k\sim N$.

Note further that all the focusing effects were discussed for the case where the confocal parameters b_n and the positions of the beam foci f_n are identical for all of the pump beams. This choice is not a coincidence. In discussing the general case where the b_n and f_n differ for all the beams, we can show that the focusing functions become maximized as $b_n\to b$ and as $f_n\to f$, other things being equal, and the same goes for the power. We do not present here any general formulas for the functions F_j, given their cumbersomeness. As an example, we consider the case of loose focusing when the positions of the foci are identical $(f_n=f)$, but the confocal parameters b_n differ; putting $b_n=b/\beta_n$, where $b=\max\{b_n\}$ and $\beta_n\geqslant 1$, we obtain

$$F_j = 4\frac{k'}{k_\beta''}\left(\frac{L}{b}\right)^2 \operatorname{sinc}^2\left[\left(\frac{b\Delta k}{2}+m_j^{(\beta)}\right)\frac{L}{b}\right], \tag{20}$$

where

$$k_\beta'' = \sum_{n=1}^{5} k_n\beta_n \quad \text{и} \quad m_j^{(\beta)} = -\frac{k_\beta''}{k'} + \sum_{n=1}^{5}\beta_n \operatorname{sgn}\omega_n.$$

It is readily seen that Eq. (20) is maximized and coincides with Eq. (13) when $\beta_n=1$. In the tight focusing case, discrepancies between the values of b_n and f_n result in a shift of the poles of the integrand toward larger values on the imaginary axis, which in turn means a decrease in the F_j. The above imposes stringent requirements on the selection of the confocal parameters and on matching of the foci of the beams. Estimates show that for mismatches $\Delta b_n, \Delta f_n \leqslant 0{,}05$ b, conversion efficiency loss is not greater than 20%.

TREATMENT OF CASCADE PROCESSES

In the case of six-photon interactions in an isotropic medium it is very characteristic that, simultaneously with the direct process at work on the nonlinear susceptibility $\chi^{(5)}$, generation of a field of frequency ω_6 is brought about by a whole series of cascade processes on a third-order nonlinear susceptibility $\chi^{(3)}$ [8]. For example, in the case of the interaction $\omega_6=\omega_1+\omega_2+\omega_3-\omega_4-\omega_5$ the following cascade processes are possible

$$\omega_1+\omega_2+\omega_3\to\omega_s', \quad \omega_s'-\omega_4-\omega_5\to\omega_6,$$
$$\omega_1+\omega_2-\omega_4\to\omega_s'', \quad \omega_3+\omega_s''-\omega_5\to\omega_6,$$
$$\omega_1-\omega_4-\omega_5\to\omega_s''', \quad \omega_2+\omega_3+\omega_s'''\to\omega_6$$

and so on. In the prespecified-field approximation, dealing with each cascade process leads to solution of a system of two truncated equations for the slow amplitudes of the fields $A_s(\mathbf{r})$ and $A_6^{(s)}(\mathbf{r})$ of respective frequencies ω_s and ω_6:

$$\begin{aligned} 2ik_s\frac{\partial A_s(\mathbf{r})}{\partial z} + \Delta_\perp A_s(\mathbf{r}) &= -4\pi k_{s0}^2 \mathscr{P}_s(\mathbf{r})\, e^{-ik_s z}, \\ 2ik_6\frac{\partial A_6^{(s)}(\mathbf{r})}{\partial z} + \Delta_\perp A_6^{(s)}(\mathbf{r}) &= -4\pi k_{60}^2 \mathscr{P}_6^{(s)}(\mathbf{r})\, e^{-ik_6 z}. \end{aligned} \tag{21}$$

A formula for $\mathscr{P}_6^{(s)}(\mathbf{r})$ can be derived in explicit form from the solution of the first equation, and this makes it possible to solve the second equation as well. If pumping is specified in form (2), the solution for the field $E_6^{(s)}(\mathbf{r}) = A_6^{(s)} e^{ik_6 z}$ excited in cascade fashion can be represented, in the case of nondegenerate interactions (1), in the form

$$E_6^{(s)}(\mathbf{r}) = -\frac{9}{4}\pi^2 \frac{k_{s0}^2 k_{60}^2}{k_s k_6} b^2 N^2 [\chi^{(3)}(\omega_s)\, \chi_s^{(3)}(\omega_6)] e^{ik'z} \times$$
$$\times \prod_{n=1}^{6} E_{n0}\, I\left(b\Delta k,\ b\Delta k_s,\ \frac{b}{L},\ \frac{f}{L},\ \frac{k''}{k'},\ \frac{k_s''}{k_s'}\right). \tag{22}$$

In the case of the cascade processes $\omega_i \pm \omega_j \pm \omega_l \to \omega_s$, $\omega_s \pm \omega_m \pm \omega_p \to \omega_6$, I $\left(b\Delta k,\ b\Delta k_s,\ \frac{b}{L},\ \frac{f}{L},\ \frac{k''}{k'},\ \frac{k_s''}{k_s'}\right)$ exhibits the form

$$I = \int_{-\zeta}^{\xi} d\xi' \frac{e^{-i\frac{b\Delta k}{2}(\xi'-\xi)} Q_6^{(s)}(\xi')}{(1 - i\xi')} \times$$
$$\times \int_{-\zeta}^{\xi'} d\xi'' \frac{e^{-i\frac{b\Delta k_s}{2}(\xi''-\xi')} Q_s(\xi'')\, e^{-\frac{R^2}{b\Phi_s}}}{(k_s'' - ik_s'\xi'')\, H_s(\xi'', \xi')\, V_s(\xi'', \xi')\, \Phi_s(\xi'', \xi', \xi)}, \tag{23}$$

with the notation

$$H_s(\xi'', \xi') = \frac{1 + \xi''^2}{k_s'' - ik_s'\xi''} - i\,\frac{\xi'' - \xi'}{k_s'},$$

$$V_s(\xi'', \xi') = \frac{1}{H_s} + \frac{(k'' - k_s'') - i(k' - k_s')\xi'}{(1 + \xi'^2)},$$

$$\Phi_s(\xi'', \xi', \xi) = \frac{1}{V_s} - i\,\frac{\xi' - \xi}{k'}, \quad \Delta k_s = k_s - k_s'.$$

Here k_s, k_s' are the wave vectors of the field and of the nonlinear polarization of the medium of frequency ω_s and $k_s'' = k_i + k_j + k_e$. For $Q_s(\xi'')$, equations

$$Q_s(\xi'') = (1 - i\xi'') \begin{cases} (1 + i\xi'')^{-2}, & \omega_s = \omega_i + \omega_j + \omega_e, \\ (1 + \xi''^2)^{-1}, & \omega_s = \omega_i + \omega_j - \omega_e, \\ (1 - i\xi'')^{-2}, & \omega_s = \omega_i - \omega_j - \omega_e. \end{cases}$$

are valid [5].

The expressions for $Q_6^{(s)}(\xi')$ are similar when the corresponding frequency constraints are fulfilled for $\omega_6 = \omega_s \pm \omega_m \pm \omega_p$. In the case of cascade addition of frequencies $\omega_i + \omega_j + \omega_l \to \omega_s$, $\omega_s + \omega_m + \omega_p \to \omega_6$, Eq. (23) simplifies appreciably; in this case we have $k' = k''$, $k_s' = k_s''$, and

$$I\left(b\Delta k;\ b\Delta k_s;\ \frac{b}{L};\ \frac{f}{L};\ 1;\ 1\right) = (1 + i\xi)^{-1} \exp\left[\frac{k''R^2}{b(1 + i\xi)}\right] I_k;$$
$$I_k = \int_{-\zeta}^{\xi} d\xi' \frac{e^{-i\frac{b\Delta k}{2}(\xi'-\xi)}}{(1 + i\xi')^2} \int_{-\zeta}^{\xi'} d\xi'' \frac{e^{-i\frac{b\Delta k_s}{2}(\xi''-\xi'-}}{(1 + i\xi'')^2}. \tag{24}$$

In the tight focusing case, with focusing at the center of the nonlinear medium ($\xi, \zeta \to \infty$), the double integral $I_k = \mathrm{Re} I_k + i \mathrm{Im} I_k$ is

$$\mathrm{Re}\, I_k = \begin{cases} \frac{\pi^2}{2} \cdot b^2 \Delta k_s (\Delta k - \Delta k_s) e^{\frac{b\Delta k}{2}}, & \Delta k < \Delta k_s < 0 \\ 0,\ \Delta k_s \leqslant \Delta k \leqslant 0; & \Delta k \leqslant 0,\ \Delta k_s \geqslant 0;\ \Delta k \geqslant 0,\ \Delta k_s - \text{arbitrary} \end{cases}$$

$$I_m I_k = \begin{cases} \frac{\pi}{4} b^2 e^{\frac{b\Delta k}{2}} \left[\Delta k^2 - 2\Delta k \Delta k_s + 2\Delta k_s (\Delta k - \Delta k_s) \ln\left|\frac{\Delta k - \Delta k_s}{\Delta k_s}\right|\right], & \Delta k < 0,\ \Delta k_s - \text{arbitrary} \\ 0,\quad \Delta k \geqslant 0,\quad \Delta k_s - \text{любое} \end{cases}$$

As a function of two variables $b\Delta k$, $b\Delta k_s$, $\mathrm{Re} I_k$ has an absolute maximum at $b\Delta k = 2b\Delta k_s = -4$; Im I_k has an absolute maximum at $b\Delta k_s = -4$; $b\Delta k_s = -0{,}333$, and an absolute minimum at $b\Delta k = -4$, $b\Delta k_s = -3{,}667$, equal in absolute value.

In the case of the cascade processes $\omega_i \pm \omega_j \pm \omega_l \to \omega_s$, $\omega_m \pm \omega_p - \omega_s \to \omega_6$ (with the frequency ω_s subtracted in the second stage),

$$e^{-i\frac{b\Delta k_s}{2}(\xi'' - \xi')}, \quad (k_s'' - ik_s'\xi''),\ Q_s(\xi''),\ H_s(\xi', \xi')$$

have to be replaced in Eq. (23) by their complex conjugates, and a minus sign must be prefixed to the entire expression; then

$$Q_6^{(s)}(\xi') = (1 - i\xi')\begin{cases} (1 + i\xi')^{-2}, & \omega_6 = \omega_m + \omega_p - \omega_s \\ (1 + \xi'^2)^{-1}, & \omega_6 = \omega_m - \omega_p - \omega_s \end{cases}$$

The total field generated $\tilde{E}_6(\mathbf{r})$ at frequency ω_6 can be stated in the form $\tilde{E}_6(\mathbf{r}) = E_6(\mathbf{r}) + \sum_s E_6^{(s)}(\mathbf{r}) = \mathrm{Re}\,\tilde{E}_6 + i\,\mathrm{Im}\tilde{E}_6$, where Eq. (9) is valid for $E_6(\mathbf{r})$ and summation over s encompasses all possible cascade processes (22). Note that the value of $E_6^{(s)}$ can be of the same order of magnitude as the value of E_6 [8], and in some instances can even exceed that value, as for example when a fifth-order harmonic is generated under two-photon resonance conditions. In that case the presence of double resonance gain is typical of the cascade process $\omega + \omega + \omega \to 3\omega$, $\omega + \omega + 3\omega \to 5\omega$. Discussion of resonance processes goes beyond the scope of the present article and will be published subsequently.

In summary, the article discusses characteristic features of frequency shift in focused beams on a fifth-order nonlinear susceptibility, with derivation of some results and drawing of some inferences. Comparison with similar results arrived at for processes occurring on lower-order nonlinearities [1-6] shows that focusing efficiency is enhanced with increasing order of the nonlinear process, while the dependence of the power output on the confocal parameter becomes more pronounced, and the basic contribution to laser action in tight focusing is made by the shortest region of all near the focus of the beams. When $n \geq 3$, where n is the order of the nonlinear susceptibility, the length of this region $L_{eff}^{(n)} \sim (10^{\frac{4}{n-1}} - 1)^{\frac{1}{2}} \frac{k''}{k'} b$. In contrast to [5], it is demonstrated in this paper that in loose focusing (Eq. (13)) conversion efficiency depends on the ratio k''/k'; this conclusion holds good for processes at work on third-order susceptibilities, as discussed by Bjorklund [5]. Note that analytical solutions of type (16), valid when the length of the nonlinear medium $L \geq L_{eff}^{(3)} \sim 10\ k''b/k'$, can be arrived at for those processes in the hard focusing case.

We present a concrete estimate of the power of the fifth harmonic of radiation emitted by a neodymium-doped laser in sodium vapor. Calculations yield $\chi^{(5)} \sim 10^{-44}$ CGSE units$\cdot$cm^3, $\Delta k \sim -7.2\cdot 10^{-17}$ N cm^{-1}. Assigning the values $N \sim 5\cdot 10^{16}$ cm^{-3}, $P_1 \sim 10^8$ W, and b ~ 3 cm ($b\Delta k \sim -11$), and cell length $L \geqslant 10$ cm from Eqs. (11) and (16.1), we obtain $P_6(5\omega) \sim 3\cdot 10^3$ W.

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