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Optical-frequency conversion in gaseous media

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This paper presents a general theoretical analysis of optical-frequency mixing of the radiation of n different laser beams ($n=1,2,3,\ldots$) in gaseous media with arbitrary density distribution. Phase-matching conditions between the induced nonlinear polarization and the generated optical radiation are calculated for third- and fifth-order sum- and difference-frequency mixing in free expanding gas jets taking into account absorption of the generated light and displacements of the foci of the laser radiation from the center of the gas jet.

INTRODUCTION

Four- and six-wave mixing of powerful laser radiation are well established methods[1–3] for the generation of intense coherent light in the vacuum ultraviolet spectral region (vuv, $\lambda_{vuv}=100-200$ nm) and at wavelengths in the extreme vacuum ultraviolet region (xuv) below the transmission cutoff of LiF ($\lambda_{xuv}<100$ nm). For third- and fifth-order frequency conversion ultraviolet and visible laser light is focused into gaseous nonlinear media like metal vapors or rare gases. For the generation of vuv light ($\lambda_{vuv}=105-200$ nm) the gases are contained in a simple glass or metal cell equipped with a vuv transmitting (LiF or MgF_2) output window. In the xuv spectral region, where transparent solid window materials are not available, the generation and detection of xuv radiation requires a differentially pumped, windowless system. In this case a pinhole (of 0.1–0.3 mm diameter),[4] a rotating disc combined with a pinhole,[5] or a capillary array[6,7] have been used as xuv transmitting windows.

In addition to these devices pulsed free expanding gas jets are well suited for this purpose.[8–12] Compared to a cell (equipped with a pinhole) the gas consumption and thus the pump capacity (required in differentially pumped systems) is reduced by up to two orders of magnitude. Since the gas jet is well defined in space (and time) it provides in a windowless environment a short region (0.5–3 mm) of high gas density (of up to 5×10^{18} atoms/cm^3). The small diameter of the gas jet strongly reduces possible absorption of the generated xuv radiation.

Experimental results of optical-frequency mixing in pulsed gas jets have been published by several research groups.[8–12] So far, however, no rigorous theoretical analysis of frequency mixing in inhomogeneous gas media (such as pulsed free jets) has been reported.

This paper presents a general description of optical-frequency mixing of the radiation of n different laser beams ($n=1,2,3,\ldots$) in gases of arbitrary density distribution. The derived results are used to calculate the phase matching and the power of the optical radiation generated by third- and fifth-order sum- and difference-frequency mixing in gas jets with different density distributions taking into account possible absorptions of the generated vuv light and displacements of the foci of the laser light from the center of the gas beam.

THEORY

For optical-frequency conversion intense laser radiation is focused into an appropriate nonlinear medium. The polarization induced by the laser light consists of a linear part which accounts for the refractive index and for possible absorptions of the laser radiation and a nonlinear part which causes all nonlinear effects such as frequency mixing. The induced nonlinear polarization is calculated as the expectation value of the induced atomic dipole moment by means of time-dependent perturbation theory. The result, multiplied by the density of the medium, provides the value of the macroscopic polarization.

The frequency spectrum of the nonlinear polarization contains frequencies which are a linear combination of the laser frequencies. The electric field (and thus the power) of the optical radiation generated by the induced oscillating polarization is calculated by solving the corresponding inhomogeneous Maxwell equations.

The first theoretical description of harmonic generation in nonlinear optical media has been published by Armstrong *et al.*,[13] Bloembergen and Pershan,[14] and Kleinman.[15] These authors solved the inhomogeneous Maxwell equation for the second harmonic generated in crystals by plane light waves. Second-harmonic generation by focused light beams was calculated by decomposing the electric light field into its plane-wave Fourier components. With the solutions known for plane waves the inverse Fourier transformation yielded the electric field generated at the second harmonic. This Fourier method was used also by Boyd *et al.*[16] to take into account the finite beam aperture in second-harmonic generation of plane light waves and by Bjorkholm[17] as well as by Kleinman *et al.*[18] to determine the second harmonic of focused Gaussian beams.

The same method was applied by Ward and New[19] to treat the third-harmonic generation in gases and by Bjorklund[20] to describe the third-order sum- and difference-frequency mixing in homogeneous isotropic media. It should also be mentioned that the influence of higher modes of Gaussian laser beams on third-harmonic generation has been investigated by Yiu *et al.*[21] In order to take saturation effects into account Vidal *et al.* solved a set of coupled inhomogeneous Maxwell equations. The results have been published in a series of papers.[22–25] Calculations of the power of the light generated by higher-order conversion processes have been reported, for example, by Tomov and Richardson[26] and by Reintjes *et al.*[27]

Another way to calculate the electric field generated by an induced nonlinear polarization is to use the integral equation formalism proposed by Bloembergen and Pershan.[14] With this formalism Franken and Ward[28] calculated the second harmonic generated by plane light waves in crystals. McMahon and Franklin[29] determined the second harmonic generated by focused beams using Huygen's construction of wave fronts which is, in principle, identical with the integral method. Boyd and Kleinman[30] applied the integral method (in a more heuristic way) to investigate the optimum conditions for second-harmonic generation and for parametric frequency conversion of focused Gaussian light beams. In this paper we use this integral equation formalism to calculate the phase matching and the output power of frequency mixing of nth order in inhomogeneous gaseous media.

The electric field $\mathbf{E}(\mathbf{r}',t)$ generated by a polarization $\mathbf{P}(\mathbf{r},t)$ is equal to the following integral:[31]

$$\mathbf{E}(\mathbf{r}',t)=\int_V \nabla\times\nabla\times\frac{[\mathbf{P}(\mathbf{r},t)]}{R}dV\ . \tag{1}$$

The brackets denote the retarded value of $\mathbf{P}$ and R is the distance between the volume element dV [located in the medium at $\mathbf{r}=(x,y,z)$] and the point of observation [at $\mathbf{r}'=(x',y',z')$]. According to this equation the electric field observed at the point $\mathbf{r}'$ is determined by the superposition of the components of the dipole radiation generated by the polarization $\mathbf{P}$ in all parts dV of the nonlinear medium. It should be noted that Eq. (1) can be derived by solving the inhomogeneous Maxwell equation using Green's method.[32]

The following considerations will be restricted to gaseous media. Since $\mathbf{E}(\mathbf{r}',t)$ is calculated at a point $\mathbf{r}'$ located outside of the nonlinear medium the sequence of the integration and of the $\nabla\times\nabla$ operator in Eq. (1) can be changed. Thus the field generated by the Fourier component $\mathbf{P}_g(\mathbf{r},\omega_g)$ of $\mathbf{P}(\mathbf{r},t)$ oscillating at the frequency ω_g is determined by the following expression:

$$\mathbf{E}(\mathbf{r}',t)=\nabla\times\nabla\times\int_V \frac{\mathbf{P}_g(\mathbf{r},\omega_g)}{R}\times\exp\left[-i\omega_g\left[t-\frac{l}{c}\right]\right]dV\ . \tag{2}$$

The parameter l is the optical length of the distance between the points $\mathbf{r}$ and $\mathbf{r}'$ and c is the speed of light. The real electric field is obtained by taking the real part of the complex field described by Eq. (2). The induced nonlinear polarization $\mathbf{P}_g(\mathbf{r},\omega_g)$ at the frequency ω_g is related to the electric fields $\mathbf{E}_j(j=1,\ldots,n)$ of the laser beams and to the gas density $N(\mathbf{r})$ by

$$\mathbf{P}_g(\mathbf{r},\omega_g)=D_nN(\mathbf{r})\chi^{(n)}\prod_{j=1}^{n}\mathbf{E}_j(\mathbf{r},\omega_j)$$

with (3)

$$D_n=\frac{n!}{2^{n-1}\prod_{k=1}^{m}n_k!}\ .$$

The factor $\chi^{(n)}$ is the tensor of the nonlinear susceptibility of $(n+1)$th order, m is equal to the number of different frequencies which contribute to the conversion process ($m\le n$), and the numbers n_k indicate how many times the different frequencies ω_k are involved in the mixing process.

In order to simplify the calculation it is assumed that the laser light consists of Gaussian beams of lowest order propagating collinearly along the z axis with confocal parameters b_j, wave vectors k_j, and focused at $z_{0,j}$. Under these conditions the amplitudes of the components of the electric field are given by

$$\mathbf{E}_j(\mathbf{r},t)=\mathbf{A}_{0,j}\frac{\exp\left[-\dfrac{k_j(x^2+y^2)}{b_j(1+i\epsilon_j)}\right]}{1+i\epsilon_j}\times\exp\left[i\int_{-\infty}^{z}k_j(z'')dz''-i\omega_jt\right]$$

$$\text{with } \epsilon_j=\frac{2}{b_j}(z-z_{0,j})\ . \tag{4}$$

Since the refractive index is assumed to be a function of z the phase factors $U_j=\exp(ik_jz)$ are written in the generalized form

$$U_j=\exp\left[i\int_{-\infty}^{z}k_j(z'')dz''\right]\ .$$

From Eqs. (3) and (4) the induced macroscopic polarization $\mathbf{P}_g(\mathbf{r},\omega_g)$ can be determined as

$$\mathbf{P}_g(\mathbf{r},\omega_g)=D_n\chi^{(n)}\prod_{j=1}^{n}\mathbf{A}_{0,j}\exp\left[-\frac{f(z)}{g(z)}(x^2+y^2)\right]\frac{1}{g(z)}\times\exp\left[i\int_{-\infty}^{z}k'(z'')dz''\right]$$

with (5)

$$k'=k_1+\cdots+k_n\ ,$$

$$f(z)=\sum_{s=1}^{n}\frac{k_s}{b_s}\prod_{\substack{j=1\\(j\neq s)}}^{n}(1+i\epsilon_j)\ ,$$

$$g(z)=\prod_{j=1}^{n}(1+i\epsilon_j)\ .$$

As for the fundamental light beams the phase factor of the generated radiation $U_g=\exp[i\omega_g(l/c)]$ is generalized by the expression

$$U_g = \exp\left[i\frac{\omega_g}{c}\int dl \right] .$$

The optical length can be calculated by the integral along a straight line between the points at $\mathbf{r}$ and $\mathbf{r}'$.[33] Since the vector components x' and y' are usually small compared to z' the line integral has the following value:

$$\frac{\omega_g}{c}\int_{\mathbf{r}}^{\mathbf{r}'} dl = \int_{-\infty}^{z'} k_g(z'')dz'' - \int_{-\infty}^{z} k_g(z'')dz'' + \frac{k_g}{2}\left[\frac{(x'-x)^2}{z'-z} + \frac{(y'-y)^2}{z'-z} \right] , \tag{6}$$

where k_g is the wave vector of the generated radiation in the nonlinear medium.

The generated electric field $\mathbf{E}_g(\mathbf{r}',t)$ is obtained by inserting (5) and (6) into Eq. (2). If all fundamental light beams have the same linear polarization $\chi^{(n)}$ is a scalar. In the far-field approximation[31] all terms in $1/R$ of higher than first order are neglected and the operator $\nabla\times\nabla$ is replaced by the expression $-\omega_g^2 c^{-2}\sin(\theta)$, where θ is the angle between $\mathbf{P}_g(\mathbf{r})$ and the vector $(\mathbf{r}'-\mathbf{r})$. Because θ is close to $\pi/2$ the value of $\sin(\theta)\approx 1$. Integrating over the x and y coordinates and using the approximation $(z'-z)\simeq R$ the following result is obtained for the generated electric field $E_g(\mathbf{r}',t)$,

$$E_g(\mathbf{r}',t) = -2\pi i D_n \chi^{(n)} \exp(i\phi)\exp(-i\omega_g t)\left[\prod_{j=1}^{n} A_{0,j} \right] k_g \times \int_{-\infty}^{\infty} N(z)\frac{\exp[-(x'^2+y'^2)q(z,z')]}{a(z,z')}\exp\left[-i\int_{-\infty}^{z} \Delta k(z'')dz'' \right] dz$$

with (7)

$$\Delta k(z'') = k_g(z'') - k'(z''),\quad a(z,z') = g(z) - i\frac{2(z-z')}{k_g}f(z),\quad q(z,z') = \frac{f(z)}{a(z,z')},\quad \phi = \int_{-\infty}^{z'} k_g(z'')dz'' .$$

The difference $\Delta k(z'')$ is the wave-vector mismatch between the generated radiation and the driving polarization. The wave-vector mismatch is related to the gas density by the equation $\Delta k(z'') = C(\lambda_g, \lambda_1 \cdots \lambda_n)N(z)$, where C accounts for the wavelength dependence of the wave-vector mismatch per atom caused by the dispersion of the medium. The wave-vector mismatch $\Delta k(z'')$ has the same spatial dependence as the gas density. For the integration over the x and y coordinates the gas density is assumed to vary slowly so that within the diameter of the light beams a constant value provides a good approximation. The power I_g of the generated radiation is determined by the amplitudes of the electric field $E_g(\mathbf{r}',t)$,

$$I_g = \frac{c}{8\pi}\int_0^{\infty} 2\pi r' \,|\, E_g(\mathbf{r}',t)\,|^2 dr' , \tag{8}$$

where $r' = (x'^2+y'^2)^{1/2}$. For the normalized density distribution $S(z) = N(z)/N_0$ the output power I_g is given by the following equation:

$$I_g = (2^{2n-2}\pi D_n N_0)^2 \left[\frac{10^7}{c} \right]^{n-1} |\,\chi^{(n)}\,|^2 \times \frac{k_g}{b_1^{n-3}}\left[\prod_{j=1}^{n} k_j I_j \right] F^{(n)} . \tag{9}$$

$F^{(n)}$ is a dimensionless generalized phase-matching function defined as

$$F^{(n)} = \left[\prod_{j=1}^{n} \frac{b_1}{b_j} \right] \frac{4k_g}{b_1} \left[\frac{2}{b_1} \right]^2 \int_0^{\infty} r' \left| \int_{-\infty}^{\infty} \frac{S(z)}{a(z,z')}\exp[-r'^2 q(z,z')]\exp\left[-i\int_{-\infty}^{z} \Delta k(z'')dz'' \right] dz \right|^2 dr'$$

with (10)

$$\Delta k(z'') = S(z'')\Delta k_0 ,$$

where the confocal parameters b_j were normalized to b_1. The phase mismatch Δk_0 corresponds to the density N_0. In Eqs. (9) and (10) all physical quantities are taken in cgs units, except the powers I_j and I_g which are taken in watts. Absorption by the medium can be considered by using the following complex wave vectors,

$$k_j = \frac{2\pi n_j}{\lambda_j} + i\frac{\alpha_j}{2}$$

with (11)

$$\alpha_j = N(z)\sigma(\lambda_j),\ j = g, 1, \ldots, n$$

where $\sigma(\lambda_j)$ is the absorption cross section at the wavelength λ_j. Equation (9) is valid for a mixing process of nth order of focused Gaussian light beams (of lowest order) in a medium of inhomogeneous density. The fundamental light beams have different confocal parameters and are focused at different positions at the common axis of the laser beams. Equation (9) includes the case of plane light waves which corresponds to the limit of $b_j \to \infty$ and hence $a(z,z') \to 1$ and $q(z,z') \to 0$.

The following investigation will be restricted to equal confocal parameters and identical positions of the foci. Under these conditions the mode of the electric field generated by sum-frequency mixing is a lowest-order Gaussian. The integration over r' [Eq. (10)] can be performed analytically with the following result:

$$F^{(n)} = \left| \frac{2}{b} \int_{-\infty}^{\infty} \frac{S(z)}{(1+i\epsilon)^{n-1}} \exp\left[-i \int_{-\infty}^{z} \Delta k(z'')dz'' \right] dz \right|^2 . \tag{12}$$

The output power I_g of a certain process can be optimized, for example, by varying the gas density of the nonlinear medium. In this case the density dependence of the generated power is determined by the dimensionless function $G^{(n)} = (b\Delta k_0)^2 F^{(n)}$.

RESULTS

In the following the dependence of the phase-matching function $F^{(n)}$ on different experimental parameters is investigated for conversion processes of third, fifth, and nth order in a gaseous medium with homogeneous and inhomogeneous density distribution. $F^{(n)}$ depends essentially on the parameters $b\Delta k_0$, b/L, L/l_0, and z_0/L. L is the full width at half maximum (FWHM) of the gas density distribution $N(z)$, l_0 is the absorption length at density N_0, and z_0 the distance between the position of the focus and the center of the nonlinear medium.

THIRD-ORDER SUM-FREQUENCY MIXING

The phase-matching function $F^{(3)}$ for third-order sum-frequency generation ($\omega_g = \omega_1 + \omega_2 + \omega_3$) is calculated assuming the following distributions S_1, S_2, and S_3 of the gas density,

$$S_1(z) = \frac{1}{1 + \left[\frac{2z}{L} \right]^2} ,$$

$$S_2(z) = \begin{cases} \cos\left[\frac{2\pi}{3} \frac{z}{L} \right] & \text{for} \quad |z| \le \frac{3L}{4} , \\ 0 \quad \text{for} \quad |z| > \frac{3L}{4} , \end{cases} \tag{13}$$

$$S_3(z) = \begin{cases} 1 \quad \text{for} \quad |z| \le \frac{L}{2} , \\ 0 \quad \text{for} \quad |z| > \frac{L}{2} . \end{cases}$$

The Lorentzian (S_1), the cosine (S_2), and the homogeneous (S_3) distribution have the same FWHM. S_1 and S_2 are similar—to a certain extent—to the density distribution of a free expanding gas jet, whereas S_3 describes the homogeneous density in a gas cell. For a numerical evaluation of the integrals [Eq. (12)], the variable z is substituted by u which is determined by the following equations:

$$S_1(z): \quad u = \arctan\left[2\frac{z}{L} \right] ,$$

$$S_z(z): \quad u = \sin\left[\frac{2\pi}{3} \frac{z}{L} \right] , \tag{14}$$

$$S_3(z): \quad u = \frac{2}{b} z .$$

These transformations of the variable parameter provide the following equations for the phase-matching functions $F^{(3)}$:

$$F_1^{(3)} = \left| \frac{b}{L} \int_{-\pi/2}^{\pi/2} \frac{1}{\left[\frac{b}{L} + i \left[\tan(u) - \frac{2z_0}{L} \right] \right]^2} \exp(-ia_1 u) du \right|^2 \quad \text{with} \quad a_1 = \frac{L}{b} \frac{b\Delta k_0}{2} ,$$

$$F_2^{(3)} = \left| \frac{3}{\pi} \frac{b}{L} \int_{-1}^{1} \frac{1}{\left[\frac{b}{L} + i \left[\frac{3}{\pi} \arcsin(u) - \frac{2z_0}{L} \right] \right]^2} \exp(-ia_2 u) du \right|^2 \quad \text{with} \quad a_2 = \frac{3}{\pi} \frac{L}{b} \frac{b\Delta k_0}{2} , \tag{15}$$

$$F_3^{(3)} = \left| \int_{-L/b}^{L/b} \frac{1}{\left[1 + i \left[u - \frac{L}{b} \frac{2z_0}{L} \right] \right]^2} \exp(-ia_3 u) du \right|^2 \quad \text{with} \quad a_3 = \frac{b\Delta k_0}{2} .$$

Figure 1 shows the values of $F_1^{(3)}$, $F_2^{(3)}$, and $F_3^{(3)}$ calculated as functions of $b\Delta k_0$ for three different ratios of b/L. The focus of the laser light is located in the center of the gas medium ($z_0/L=0$) and absorption is neglected. It should be noted that $F_3^{(3)}$ is identical with the result obtained by Bjorklund[20] for a gas contained in a cell of length L.

For strong focusing ($b/L=0.1$), the shape of $F_1^{(3)}$, $F_2^{(3)}$, and $F_3^{(3)}$ is almost the same. Even the values of the maximum of these three functions are not very different and the maxima are obtained at almost the same negative value of $b\Delta k_0$. If $b/L=0.5$ or $b=L$ the optimum values of $F_1^{(3)}$ and of $F_2^{(3)}$ are observed also at nearly the same value of $b\Delta k_0$, $F_3^{(3)}$, however, optimizes at a somewhat smaller mismatch. For these ratios of b/L the maximum values of $F_1^{(3)}$, $F_2^{(3)}$, and $F_3^{(3)}$ are rather different.

The results displayed in Fig. 1 indicate that each of the three phase-matching functions provides to a good approximation the same dependence of the phase matching on $b\Delta k_0$ if the laser radiation is strongly focused. For this reason a detailed analysis of the third-order sum-frequency conversion in a free expanding gas jet will be performed only for a Lorentzian distribution of the gas density.

In the limit $b/L\to 0$ the three functions $F^{(3)}$ converge to the same solution $F_A^{(3)}$ derived analytically for tight focusing ($b \ll L$),

$$F_A^{(3)}=\begin{cases}\pi^2(b\Delta k)^2\exp(b\Delta k) & \text{for } b\Delta k\le 0\ ,\\ 0 & \text{for } b\Delta k>0\ .\end{cases} \tag{16}$$

The dependence of $F_A^{(3)}$ on $b\Delta k$ is shown also in Fig. 1. $F_A^{(3)}$ is independent of the distribution of the density, but depends only on the value of $b\Delta k$ calculated at the z position of the focus and thus on the local density in this region of the focus.

$F^{(3)}$ can also be calculated analytically for the plane-wave limit ($b\gg L$)

$$F_p^{(3)}=\left[\frac{2L}{b}\right]^2\left[\frac{\sin\left[\frac{\beta}{2}\right]}{\frac{\beta}{2}}\right]^2 , \tag{17}$$

where β is the total integrated phase mismatch, given by the following integral

$$\beta=\Delta k_0\int_{-\infty}^{\infty} S(z)dz\ . \tag{18}$$

$F_p^{(3)}$ is identical with the phase-matching function for the frequency conversion of plane light waves in homogeneous media, generalized by the integrated phase mismatch β according to Eq. (18).

In Fig. 2 the function $G_1^{(3)}$ is shown for several values of the parameter b/L. As is seen from this figure $G_A^{(3)}=(b\Delta k)^2F_A^{(3)}$ provides a good approximation for $G_1^{(3)}$ as long as $b/L<0.3$.

The optimum values $F_{\rm opt}^{(3)}$ and $G_{\rm opt}^{(3)}$ of the functions $F_1^{(3)}$ and $G_1^{(3)}$ (which correspond to the optimum values of the parameter $b\Delta k_0$) were calculated as functions of the ratio b/L. The results of this calculation are displayed in Figs. 3(a) and 3(b) together with the optimum values $(b\Delta k_0)_{\rm opt}$. The functions $F_{\rm opt}^{(3)}$ and $G_{\rm opt}^{(3)}$ are normalized to their values at $b/L=0$. As seen from Fig. 3(a) optimum phase matching is obtained at $b/L=0$ which corresponds to the maximum value of $F_{\rm opt}^{(3)}$. For the function $G_{\rm opt}^{(3)}$, on the other hand, maximum values are obtained if the confocal parameter b is equal to L. It should be noted that a similar result has been reported by Bethune *et al.*[11]

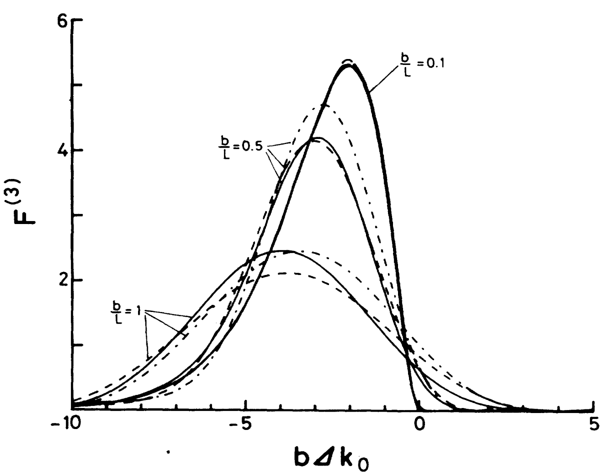

FIG. 1. $F^{(3)}$ as function of $b\Delta k_0$ for different values of the ratio b/L and different distributions of the gas density $S(z)$: ——, Lorentzian; — — —, cosine; and —·—·—·, homogeneous. For the ratio $b/L=0.1$ the values of $F^{(3)}$ calculated for the different density distributions coincide—except at the maximum—within the width of the line of the drawn curve. This is valid also for the analytical function $F_A^{(3)}$.

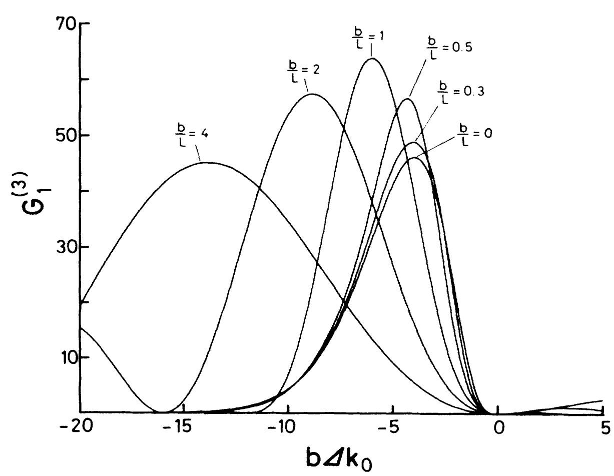

FIG. 2. $G_1^{(3)}$ as function of $b\Delta k_0$ calculated for a Lorentzian density profile of the gas jet and different values of b/L.

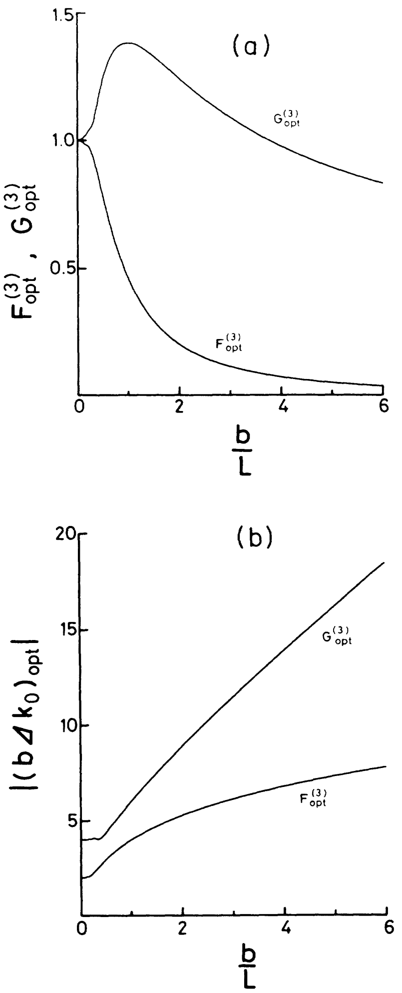

FIG. 3. (a) The optimum value of $F_1^{(3)}$ and $G_1^{(3)}$ as function of the ratio b/L normalized to $b/L=0$. (b) The values of $b\Delta k_0$ which provide the maximum values of the corresponding functions $F_1^{(3)}$ and $G_1^{(3)}$.

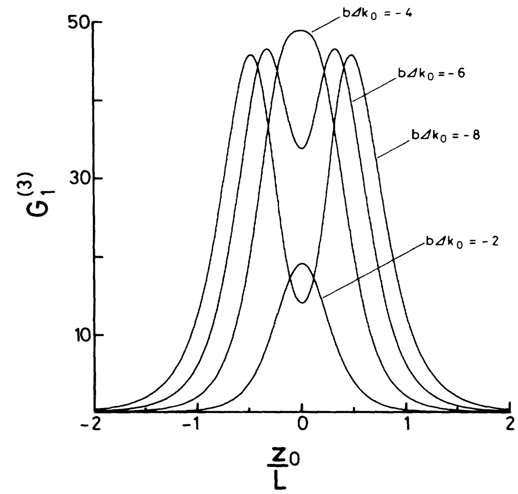

FIG. 4. $G_1^{(3)}$ as function of the normalized displacement z_0/L of the focus from the center of the gas jet for various values of the parameters $b\Delta k_0$ and $b/L=0.3$.

The influence of the normalized displacement z_0/L of the focus from the center of the gas jet on the values of $G_1^{(3)}$ (and thus on the power of the generated sum frequency) is shown in Fig. 4 for different values of the gas density at the jet center. If this density provides a mismatch $b\Delta k_0 > -4$ the maximum output power is achieved for a focus position at the jet center ($z_0=0$), the location of highest density. Any displacement from this position causes a decrease of the conversion efficiency. For $b\Delta k_0 < -4$ $G_1^{(3)}$ has a minimum at $z_0=0$ and two maxima located symmetrically to this position. If, for example, $b\Delta k_0=-8$ the two maxima occur at $z_0=L/2$ where the gas density is one half of the one at $z_0=0$.

The shape of $G_1^{(3)}$ has been measured for third-harmonic generation in an argon gas jet (Fig. 5). In the experiment the focus of the uv laser light is moved along the optical axis of the laser beam which crosses the gas jet at its center. The dependence of $G^{(3)}$ on the position of the focus provides a sensitive method for positioning the focus at the center of the gas jet. Moreover, it allows us to measure the local gas density and thus the density profile of a gas jet.[12]

In argon the generated third harmonic[12] ($\lambda_{vuv}=104.4$ nm) is not absorbed. In an absorbing gas the maximum of $G_1^{(3)}$ is obtained at a position of the focus outside of the center of the jet in direction of the light beam. This is shown in Fig. 6 for different gas densities for the ratio $b/L=0.3$ and for $L/l_0=3$. To account for absorption of the generated radiation the phase-matching function $F_1^{(3)}$ is extended in the following way:

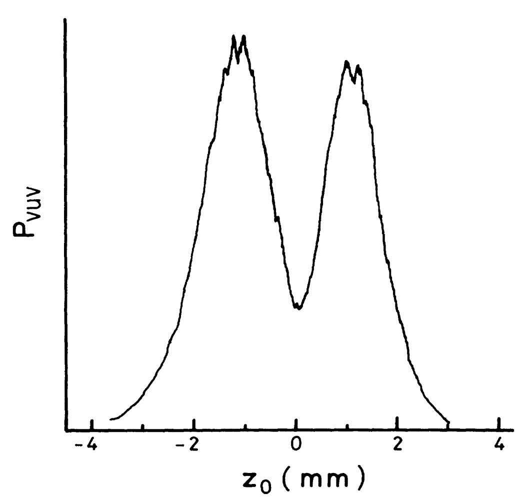

FIG. 5. Third harmonic generated in an argon gas jet at $\lambda_{vuv}=104.4$ nm as function of the focus displacement z_0 ($b/L=0.3$; $b\Delta k_0=-7.2$).

$$\hat{F}_1^{(3)}=\exp(-\pi y)\left|\frac{b}{L}\int_{-\pi/2}^{\pi/2}\frac{\exp(yu)}{\left[\frac{b}{L}+i\left[\tan(u)-\frac{2z_0}{L}\right]\right]^2}\exp(-ia_1u)du\right|^2 \quad \text{with } y=\frac{L}{4l_0} \tag{19}$$

The displacement of the maximum of $\hat{G}_1^{(3)}$ from the center is caused by the fact that most of the radiation is generated in the area of the focus, but is partly absorbed by the gas between the focus and the edge of the gas jet. The focus position which provides optimum output depends on $b\Delta k_0$ and on the strength of the absorption determined by L/l_0. If the absorption is sufficiently low ($L/l_0<1$) and $b\Delta k_0<-4$, $\hat{G}_1^{(3)}$ still exhibits two maxima. $\hat{G}_1^{(3)}$ has, however, only one maximum if the absorption is strong ($L/l_0>1$). This is demonstrated by the calculations shown in Fig. 6.

An experimental result obtained for frequency tripling in an absorbing gas is shown in Fig. 7. uv laser radiation ($\lambda_{uv}=280.5$ nm) is focused into a xenon gas jet. The third harmonic at $\lambda_{vuv}=93.5$ nm is partly absorbed by the autoionizing resonance. Since $b\Delta k_0$ is about -4 and L/l_0 is on the order of 8 only one maximum is observed for the generated vuv if the focus position is moved in the direction of the uv light beam.

For an absorption corresponding to $L/l_0\leq 4$ the main features of the dependence of $G^{(3)}$ on $b\Delta k_0$ are, in principle, the same as the one for a nonabsorbing gas. The values of $G^{(3)}$ are multiplied by an absorption factor which depends on the density profile $S(z)$ and on the position of the focus in the jet. If, however, the parameter $L/l_0>4$ and the focus is not shifted from the center of the nonlinear medium the shape of $G^{(3)}$ can considerably differ from the results shown in Fig. 2. Since the detected light is produced in a small range at the edge of the nonlinear medium, the effective length of this range may be small compared to b. Thus the dependence of $G^{(3)}$ on $b\Delta k_0$ will be similar to the one of $G_p^{(3)}=(b\Delta k_0)^2F_p^{(3)}$. It should be noted that the dependence of the generated power—and thus of $G^{(3)}$—on the position z_0 of the focus of the laser light is—under certain conditions—very similar for sum-frequency generation in a jet (Fig. 4) and in a cell (with homogeneous density distribution) displayed in Fig. 8.

The experimental results are obtained by focusing blue laser light ($\lambda_{uv}=430.2$ nm) into a cylindrical cell ($L=5$ cm) filled with xenon. The focus is moved along the cell axis in the direction of the input to the output window. For parameters $b/L=0.75$ and $b\Delta k_0=-6.2$ the output power has a maximum at $z_0=0$ [Fig. 8(a)], similar to the power of the third harmonic generated in a gas jet. If, for the same value of b/L, the mismatch $b\Delta k_0$ is increased to $b\Delta k_0=-10$ the output power showed a minimum at $z_0=0$ and two maxima located symmetrically to this central position. This result—not shown in Fig. 8—is very similar to the one measured for the sum frequency in a gas jet (Fig. 5). At even higher gas densities ($b\Delta k_0=-15.1$) the minimum at $z_0=0$ changed to a relative maximum [Fig. 8(b)]. It should be mentioned that a power dependence of this type cannot be observed for the sum frequency generated in a gas jet. If, finally, the ratio b/L is reduced ($b/L=0.06$) and the gas density is adjusted to provide, for example, a mismatch $b\Delta k_0=-7.8$ the output power is almost independent of the position z_0 except in the vicinity of the windows

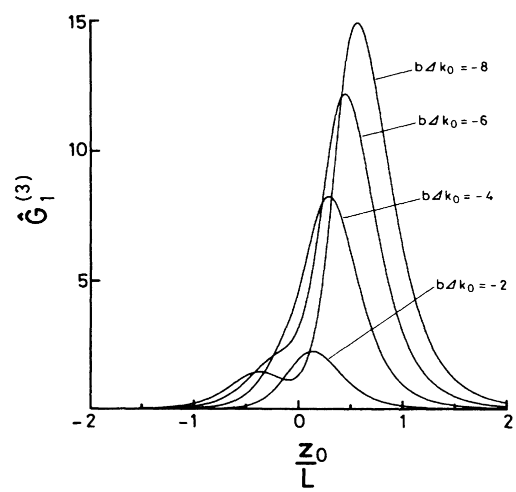

FIG. 6. Same as Fig. 4 but including absorption of the generated radiation ($L/l_0=3$).

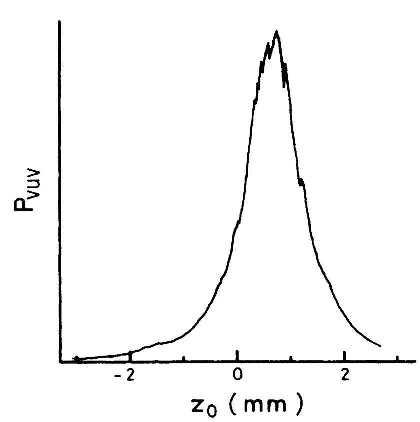

FIG. 7. Third harmonic generated in a xenon gas jet at $\lambda_{vuv}=93.5$ nm as function of the focus displacement z_0 ($b/L=0.3$; $b\Delta k_0=-4$; $L/l_0=8$).

where two narrow maxima are observed [Fig. 8(c)]. To compare the experimental results with the theoretical predictions $G_3^{(3)}$ has been calculated for the corresponding parameters b/L and $b\Delta k_0$. The dependence of the vuv output on z_0 determined in this way (displayed also in Fig. 8) is in good agreement with the measured results.

SUM-FREQUENCY MIXING OF *n*th ORDER

For sum-frequency mixing of nth order ($\omega_g=\omega_1+\cdots+\omega_n$) the phase-matching function $F^{(n)}$ is calculated in a similar way as for the third-order process. For a Lorentzian density profile of a nonabsorbing gas, $F_1^{(n)}$ is given by the following equation:

$$F_1^{(n)}=\left|\left[\frac{b}{L}\right]^{n-2}\int_{-\pi/2}^{\pi/2}\frac{1}{\left[\frac{b}{L}+i\left[\tan(u)-\frac{2z_0}{L}\right]\right]^{n-1}}\exp(-ia_1u)du\right|^2 . \tag{20}$$

As an example Fig. 9 shows the values of the phase-matching function $F_1^{(5)}$ for sum-frequency mixing of fifth order as functions of $b\Delta k_0$ for four different ratios b/L. If $b/L<0.1$, $F_1^{(5)}$ has a maximum at $b\Delta k_0=-6$. The position of the maximum shifts to larger negative values of $b\Delta k_0$ if b/L is increased. A similar dependence on $b\Delta k_0$ and b/L is obtained for sum-frequency conversions of even higher order. The maximum of $F_1^{(7)}$ occurs, for example, at $b\Delta k_0=-10$ and the one of $F_1^{(9)}$ at $b\Delta k_0=-14$. The dependence of these phase-matching functions on absorption of the generated light and on displacements of the focus from the center of the nonlinear medium are very similar to those described for third-order conversion. In the limit of strong focusing ($b \ll L$) the function $F^{(n)}$ can be calculated analytically,

$$F_A^{(n)}=\begin{cases}\left[\frac{2\pi}{(n-2)!}\right]^2\left[\frac{b\Delta k}{2}\right]^{2n-4}\exp(b\Delta k) \\ \text{for } b\Delta k\le 0 , \\ 0 \text{ for } b\Delta k>0 .\end{cases} \tag{21}$$

The maximum of $F_A^{(n)}$ is obtained for $b\Delta k=-2(n-2)$, whereas $G_A^{(n)}$ has a maximum value for $b\Delta k=-2(n-1)$. This result is the same as the one reported by Ward and New.[19]

DIFFERENCE-FREQUENCY MIXING

The difference-frequency mixing in gas jets will be investigated only for the fifth-order process $\omega_g=\omega_1+\omega_2+\omega_3+\omega_4-\omega_5$. The reason for this restriction is the fact that third-order difference-frequency conversion generates radiation in the region of the vuv. For this conversion gas cells with homogeneous density distribution are thus very appropriate.

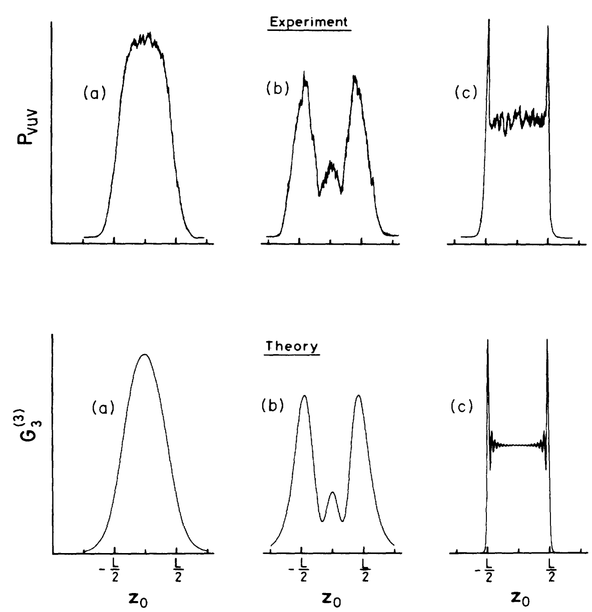

FIG. 8. Experiment: Third harmonic at $\lambda_{vuv}=143.4$ nm as function of the focus displacement z_0 in a gas cell ($L=5$ cm). (a) $b/L=0.75$, $b\Delta k_0=-6.2$; (b) $b/L=0.75$, $b\Delta k_0=-15.1$; and (c) $b/L=0.06$, $b\Delta k_0=-7.8$. Theory: $G_3^{(3)}$ calculated for the experimental parameters and a homogeneous density distribution.

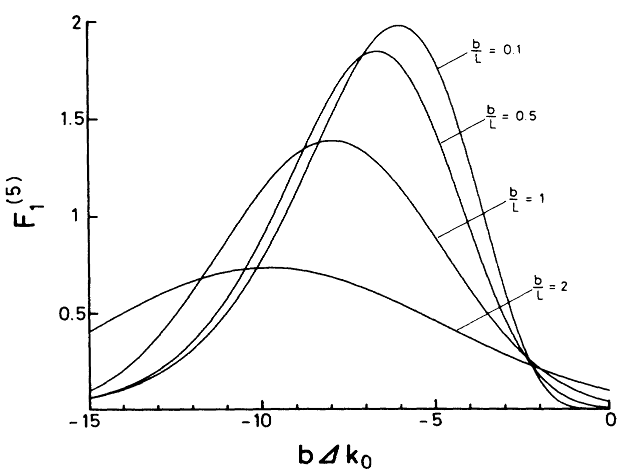

FIG. 9. $F_1^{(5)}$ as function of $b\Delta k_0$ for a Lorentzian density distribution of the gas jet and different values of b/L.

In general, the intensity distribution of the radiation generated by difference-frequency mixing is not a Gaussian mode. It is therefore not possible to derive an equation for the corresponding phase-matching function $F^{(5)}$ which is similar to Eq. (12). The calculation of the intensity distribution in the far field requires a numerical integration of Eq. (10) over the radius r'. For the additional integration over the coordinate z the functions $a(z,z')$ and $q(z,z')$ defined by Eq. (7) are expressed in the following way:

$$a(z,z')=(1+i\epsilon)^3(1+\epsilon^2)[1-i(\epsilon-\epsilon')h(z)] ,$$

$$q(z,z')=\frac{k_g}{b}\frac{h(z)}{1-i(\epsilon-\epsilon')h(z)} ,$$

with (22)

$$h(z)=\frac{1}{1+\epsilon^2}\left[\frac{\hat{k}}{k_g}-i\epsilon\right] , \quad \epsilon=\frac{2}{b}(2-z_0), \; \epsilon'=\frac{2}{b}(z'-z_0).$$

The phase-matching function depends in addition on the ratio $\hat{k}/k_g$ where $\hat{k}=k_1+k_2+k_3+k_4+k_5$ and $k_g=k_1+k_2+k_3+k_4-k_5$; k_g is the wave vector at the generated wavelength. The intensity distribution in the far field of the generated radiation is almost a Gaussian mode if $b\Delta k_0<0$, but a ring pattern if $b\Delta k_0>0$. This ring pattern is symmetric to the optical axis of the light beam. The radius of the ring increases for larger positive values of $b\Delta k_0$. It should be noted that the same result is obtained for difference-frequency mixing of third order in a homogeneous gas medium.[20]

The function $F_{1-}^{(5)}$ is calculated for $\hat{k}/k_g=1.5$ and different parameters of b/L. The position of the focus is on the jet axis. The results shown in Fig. 10 indicate that the maxima of $F_{1-}^{(5)}$ occur at negative values of $b\Delta k_0$. This result is different from $F_{1-}^{(3)}$ calculated for third-order difference-frequency mixing. The maximum of $F_{1-}^{(3)}$ is obtained for $b\Delta k_0=0$.

For tight focusing ($b/L\to 0$) and $\hat{k}/k_g=1$ from Eqs. (10) and (22), the following analytical phase-matching function is derived for fifth-order frequency mixing,

$$F_{A-}^{(5)}=\begin{cases}\dfrac{\pi^2}{64}[(b\Delta k-1)^2+1]^2\exp(b\Delta k) & \text{for } b\Delta k\le 0 ,\\[2mm] \dfrac{\pi^2}{16}\exp(-b\Delta k) & \text{for } b\Delta k>0 .\end{cases} \tag{23}$$

The dependence of this function on $b\Delta k$ is also shown in Fig. 10. Although difference-frequency mixing with $\hat{k}/k_g=1$ is physically impossible, Eq. (23) allows an estimate of the principle dependence of the phase matching of focused laser light on the dispersion and the gas density.

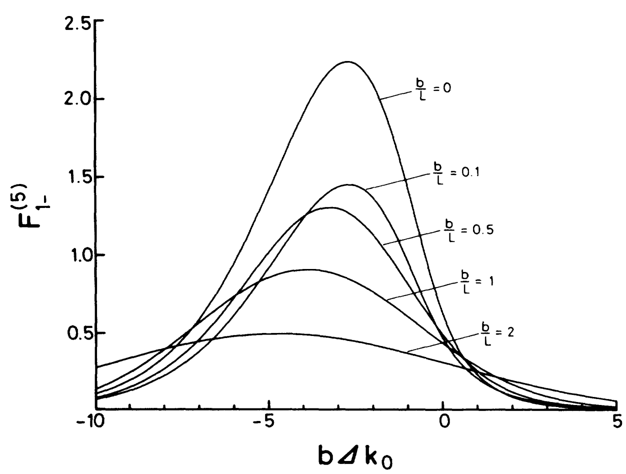

FIG. 10. $F_{1-}^{(5)}$ as function of $b\Delta k_0$ for a Lorentzian density distribution of the gas jet and different values of b/L; $\hat{k}/k_g=1.5$; the analytical phase-matching function $F_{A-}^{(5)}$ is shown also ($b/L=0$, $\hat{k}/k_g=1$).

CONCLUSIONS

It has been demonstrated that the integral equation formalism is very appropriate for the calculation of the electric field—and thus the output power—of the light generated by optical conversion processes in nonlinear media. This method has been used to determine the general phase-matching conditions between the induced nonlinear polarization and the generated optical radiation if n different laser beams are focused in a gaseous medium with arbitrary density distribution. The detailed calculations presented in this paper provide the phase-matching conditions for third- and fifth-order sum- and difference-frequency mixing and their dependence on the displacement of the foci in a free expanding gas jet as well as in a gas cell taking absorption of the generated radiation into account.

This analysis was restricted to laser beams with equal confocal parameters and identical positions of the foci. It should be mentioned that the results of a detailed investigation of effects caused by different confocal parameters of the laser beams (which was performed with the same method[34]) will be presented in a paper published elsewhere.

In addition, it should be noted that the integral method presented in this paper is also very useful for the investigation of the phase matching in anisotropic media. By changing the integration path of the line integral of Eq. (6) in order to account for double refraction present in a nonlinear crystal the electric field of the second harmonic, for example, is obtained straightforward. The result is identical to the one reported by Boyd and Kleinman [Eq. (2.9) in Ref. 30] which is the main equation for the theoretical studies performed by these authors on the optimization of second-harmonic generation in crystals. Because of the wide validity of the theoretical results obtainable by the integral equation formalism, it should be considered as a general and powerful method for a detailed description of optical conversion processes.

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[1]W. Jamroz and B. P. Stoicheff, in *Progress in Optics,* edited by E. Wolf (North-Holland, Amsterdam, 1983), Vol. 20, pp. 324–380, and references therein.

[2]C. R. Vidal, in *Tunable Lasers, Topics in Applied Physics,* edited by L. F. Mollenauer and J. C. White (Springer, Heidelberg, in press), and references therein.

[3]R. Hilbig, G. Hilber, A. Lago, B. Wolff, and R. Wallenstein, Comments At. Mol. Phys. **18**, 157 (1986).

[4]R. Hilbig and R. Wallenstein, Opt. Commun. **44**, 283 (1983).

[5]K. D. Bonin and T. J. McIlrath, Rev. Sci. Instrum. **55**, 1666 (1984).

[6]T. B. Lucatorto, T. J. McIlrath, and J. R. Roberts, Appl. Opt. **18**, 2505 (1979).

[7]B. P. Stoicheff, P. R. Herman, P. E. La Rocque, and R. H. Lipson, in *Laser Spectroscopy VII*, Vol. 49 of *Springer Series in Optical Sciences,* edited by T. W. Hansch and Y. R. Shen (Springer-Verlag, Berlin, 1985), pp. 174–178; Opt. Lett. **10**, 502 (1985).

[8]A. H. Kung, Opt. Lett. **8**, 24 (1983).

[9]J. Bokor, P. H. Bucksbaum, and R. R. Freeman, Opt. Lett. **8**, 217 (1983).

[10]E. E. Marinero, C. T. Rettner, R. N. Zare, and A. H. Kung, Chem. Phys. Lett. **95**, 486 (1983); J. Phys. Chem. **88**, 4459 (1984).

[11]D. S. Bethune, C. T. Rettner, and A. H. Kung (unpublished).

[12]A. Lago, G. Hilber, R. Hilbig, and R. Wallenstein, Laser Optoelektr. **17**, 357 (1985).

[13]J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962).

[14]N. Bloembergen and P. S. Pershan, Phys. Rev. **128**, 606 (1962).

[15]D. A. Kleinman, Phys. Rev. **128**, 1761 (1962).

[16]G. D. Boyd, A. Ashkin, J. M. Dziedzic, and D. A. Kleinman, Phys. Rev. A **137**, 1305 (1956).

[17]J. E. Bjorkholm, Phys. Rev. **142**, 126 (1966).

[18]D. A. Kleinman, A. Ashkin, and G. D. Boyd, Phys. Rev. **145**, 338 (1966).

[19]J. F. Ward and G. H. C. New, Phys. Rev. **185**, 57 (1969).

[20]G. J. Bjorklund, IEEE J. Quantum Electron. **QE-11**, 287 (1975).

[21]Y. M. Yiu, T. J. McIlrath, and R. Mahon, Phys. Rev. A **20**, 2470 (1979).

[22]H. Puell, K. Spanner, W. Falkenstein, W. Kaiser, and C. R. Vidal, Phys. Rev. A **14**, 2240 (1976).

[23]H. Puell and C. R. Vidal, IEEE J. Quantum Electron. **QE-14**, 364 (1978).

[24]H. Junginger, H. B. Puell, H. Scheingraber, and C. R. Vidal, IEEE J. Quantum Electron. **QE-16**, 1132 (1980).

[25]H. Scheingraber and C. R. Vidal, Opt. Commun. **38**, 75 (1981).

[26]I. V. Tomov and M. C. Richardson, IEEE J. Quantum Electron. **QE-12**, 521 (1976).

[27]J. T. Reintjes, Ch. Y. She, and R. C. Eckardt, IEEE J. Quantum Electron. **QE-14**, 581 (1978).

[28]P. A. Franken and J. F. Ward, Rev. Mod. Phys. **35**, 23 (1963).

[29]D. H. McMahon and A. R. Franklin, Appl. Phys. Lett. **6**, 14 (1965).

[30]G. D. Boyd and D. A. Kleinman, J. Appl. Phys. **39**, 3597 (1968).

[31]*Principles of Optics,* edited by M. Born and E. Wolff (Pergamon, New York, 1975).

[32]*Classical Electrodynamics,* edited by J. D. Jackson (Wiley, New York, 1974).

[33]A. Sommerfeld and J. Junge, Ann. Phys. (N.Y.) **35**, 277 (1911).

[34]G. Hilber, Ph.D. thesis, University of Bielefeld, Federal Republic of Germany, 1986.