# Optical-frequency conversion in gases using Gaussian laser beams with different confocal parameters

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This paper presents a general description of optical-frequency mixing of $n$ Gaussian laser beams ($n=1,2,3,\ldots$) with different confocal parameters focused at different positions in the gas medium. A detailed theoretical analysis of third-order sum- and difference-frequency mixing of the light of two laser beams indicates that under certain conditions the output power is enhanced by using unequal confocal beam parameters.

## I. INTRODUCTION

Frequency conversion by four- and six-wave mixing in gaseous media is a well-established experimental method to generate coherent light in the spectral region of the vacuum ultraviolet (vuv) at wavelengths between 105 and 200 nm and at wavelengths in the extreme ultraviolet ($\lambda_{\text{vuv}} < 105$ nm).[1-3]

Theoretically, the sum- and difference-frequency mixing of focused Gaussian beams has been analyzed using the Fourier-transform method[4,5] or the integral equation formalism.[6] More recently,[7] it was shown that the integral equation formalism is well suited to describe conversion processes of $n$th order in gases with arbitrary density distribution. This analysis was restricted, however, to the investigation of third- and fifth-order frequency mixing of laser beams with equal confocal parameters focused at the same position of the gas medium. As shown by the investigations presented in this paper the integral equation method is also appropriate to describe the frequency mixing of $n$th order of laser beams with different confocal parameters focused at different positions in the gas medium. The obtained results indicate that for two-photon enhanced conversion processes, for example, unequal confocal beam parameters—chosen in an appropriate ratio—could substantially improve the output power. These results thus provide new information which is important for the optimization of the efficiency of sum- and difference-frequency mixing in gases.

## II. THEORY

It is assumed that $n$ different Gaussian laser beams of lowest order propagate collinearly along the $z$ axis. The light beams with confocal parameters $b_j$ and wave vectors $k_j$ are focused at the positions $z_{0,j}$ of the nonlinear gaseous medium with arbitrary density distribution $N(\mathbf{r})$ (where $\mathbf{r}=x,y,z$). The electric fields $\mathbf{E}_j(\mathbf{r},\omega_j)$ of the laser light with the frequency $\omega_j$ induce in the gas a nonlinear polarization $\mathbf{P}(\mathbf{r},t)$. The Fourier component $\mathbf{P}_g(\mathbf{r},\omega_g)$ of this polarization generates an electric field $\mathbf{E}_g(t)$ oscillating with the frequency ($\omega_g=\omega_1\pm\cdots\pm\omega_n$). The field $\mathbf{E}_g(t)$ is observed at a position $\mathbf{r}'=x',y',z'$ outside of the medium. The power of the generated radiation detected at $z'$ is given by[7]

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

with

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

According to Eq. (1) the output is proportional to the squared modulus of the nonlinear susceptibility $\chi^{(n)}$ which is—in general—a tensor of $(n+1)$th order. It is a scalar if all fundamental light beams have the same linear polarization. The numerical factor $D_n$ contains the numbers $m$ and $n_K$. The quantity $m$ is the number of different frequencies which contribute to the conversion process, and the numbers $n_K$ indicate how many times each frequency is involved in the mixing process. The quantity $c$ denotes the speed of light. In Eq. (1) all parameters are taken in cgs units except the input powers $I_j$ and the output power $I_g$ which are taken in watts. The parameter $k_g$ is the wave vector of the generated radiation, and $F^{(n)}$ is a dimensionless generalized phase-matching function given by the following relation:

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

 

with

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

$$\Delta k_0=k_g-(k_1+\cdots+k_n) ,$$

where the confocal parameters were normalized to $b_1$. The function $S(z)$ represents the normalized density distribution of the nonlinear medium: $S(z)=N(z)/N_0$. The difference $\Delta k(z'')$ is the wave-vector mismatch between the generated radiation and the driving polarization. It has the same spatial dependence as the gas density. The phase mismatch $\Delta k_0$ is related to the gas density $N_0$ by the equation $\Delta k_0=C(\lambda_g,\lambda_1,\ldots,\lambda_n)N_0$, where $C$ accounts for the wavelength dependence of the wave-vector mismatch caused by the dispersion of the medium. The integrations over $r'$ $[r'=(x'^2+y'^2)^{1/2}]$ and the coordinate $z$ have to be performed numerically. For this purpose the functions $a(z,z')$ and $q(z,z')$ are defined in the following way:

$$a(z,z')=[1-i(\epsilon_1-\epsilon_1')h(z)]\prod_{j=1}^{n}(1+i\epsilon_j) ,$$
$$q(z,z')=\frac{k_g}{b_1}\frac{h(z)}{1-i(\epsilon_1-\epsilon_1')h(z)} , \tag{3}$$

with

$$h(z)=\sum_{j=1}^{n}\frac{\bar{b}_j\bar{k}_j}{1+i\epsilon_j} ,$$

$$\bar{b}_j=\frac{b_1}{b_j},\quad \bar{k}_j=\frac{k_j}{k_g} ,$$

$$\epsilon_1=\frac{2}{b_1}(z-z_{0,1}),\quad \epsilon_1'=\frac{2}{b_1}(z'-z_{0,1}) ,$$

$$\left.\begin{array}{l}\epsilon_j=\bar{b}_j(\epsilon_1-2\,\overline{\Delta z_j})\\ \overline{\Delta z_j}=\dfrac{\Delta z_j}{b_1}\end{array}\right\}j=2,\ldots,n .$$

Equation (3) is derived from the general definition of the functions $a(z,z')$ and $q(z,z')$ given in Ref. 7. For frequencies $\omega_j$ of the fundamental electric fields with negative sign (present in the case of difference-frequency mixing) the corresponding parameters $\epsilon_j$ and $\bar{k}_j$ have to be replaced by $-\epsilon_j$ and $-\bar{k}_j$, respectively. Different values of the confocal parameters $b_j$ result in different values of the ratios $\bar{b}_j$. The displacements $\Delta z_j$ $(j=2,\ldots,n)$ of the foci measured from the position $z_{0,1}$ of the focus of the laser beam with frequency $\omega_1$ are normalized to the confocal parameter $b_1$.

If the output power is optimized by varying, for example, the gas density of the nonlinear medium, the dependence of the generated power on the gas density is determined by the dimensionless function $G^{(n)}=(b_1\,\Delta k_0)^2F^{(n)}$. From Eq. (1) it follows that the output power $I_g$ is related to $G^{(n)}$ by the following expression:

$$I_g=(2^{2n-2}\pi D_n)^2C^{-2}\left[\frac{10^7}{c}\right]^{n-1}|\chi^{(n)}|^2\frac{k_g}{b_1^{n-1}}\times\left[\prod_{j=1}^{n}k_jI_j\right]G^{(n)} . \tag{4}$$

For applications of the generated radiation it is of interest to know the divergence of the produced light. If the nonlinear medium is negative dispersive $(b_1\Delta k_0<0)$ and all fundamental beams are focused at the same position $(\Delta z_j=0)$ at the center of the medium the divergence can be estimated in the following way. The integration over the coordinate $z$ in Eq. (2) is performed only over a small interval centered at $z=0$. This is justified because the main part of the radiation is generated by the interaction of the laser beams with the gas in the central area of their foci. According to this approximation the value of the coordinate $z$ in the expression

$$\exp[-r'^2q(z,z')]$$

of Eq. (2) is assumed to be zero. This expression determines the radial field distribution at the observation plane $z'$ generated by the dipoles located at the plane $z=0$. It can be shown that

$$\exp[-r'^2q(0,z')]$$

describes a Gaussian field distribution with a beam waist $w_g$ which is related to the beam waists $w_j$ of the fundamental beams in the following way:

$$w_g=(w_1^{-2}+\cdots+w_n^{-2})^{-1/2} , \tag{5}$$

where $w_j=(b_j/k_j)^{1/2}$. The full angle $\theta_g$ of the beam divergence can be obtained from $w_g$ by the known relation $\theta_g=2\lambda_g/(\pi w_g)$. For difference-frequency mixing in positive dispersive media $(b_1\Delta k_0>0)$, however, Eq. (5) is not valid because the beam profile of the generated radiation is not Gaussian, as will be discussed in detail in Sec. III.

## III. RESULTS

The influence of different values of the confocal parameters on the functions $F^{(3)}$ and $G^{(3)}$ is analyzed in detail for third-order processes of the type $\omega_g=2\omega_1\pm\omega_2$. In this case the parameter relevant for the variation of the functions $F^{(3)}$ and $G^{(3)}$ (due to different confocal parameters) is the ratio $\bar{b}=b_1/b_2$. The normalized distance of the positions of the foci is given by $\overline{\Delta z}=\Delta z/b_1$; the wavelength dependence of the phase-matching function is characterized by the parameter $\bar{k}=k_1/k_g$.

To calculate the function $F^{(3)}$ it is useful to substitute the variable $z$ in an appropriate way. Which substitution is the most appropriate one depends mainly on the density distribution $N(z)$. If the laser beams are focused, for example, in a gas cell of length $L$ the corresponding normalized homogeneous density distribution is given by

$$S(z)=\begin{cases}1 & \text{for } |z|\le\dfrac{L}{2}\\ 0 & \text{for } |z|>\dfrac{L}{2}.\end{cases} \tag{6}$$

In this case the substitutions $z=b_1/2u$ and $z'=L/2$ provide the following equation for the phase-matching function:

$$F^{(3)}=\bar{b}\frac{4k_g}{b_1}\int_0^{\infty}r'\left|\int_{-L/b_1}^{+L/b_1}\frac{1}{a(u,\epsilon_1')}\exp[-r'^2q(u,\epsilon_1')]\exp\left[-i\frac{b_1\Delta k_0}{2}u\right]du\right|^2 dr' , \quad (7)$$

with

$$\epsilon_1'=\frac{L}{b_1}\left[1-2\frac{z_{0,1}}{L}\right] .$$

If the frequency conversion is performed, however, in a free expanding gas jet, the density profile is given to a good approximation by a Lorentzian distribution:[8]

$$S(z)=\frac{1}{1+\left[\frac{2z}{L}\right]^2} . \quad (8)$$

The parameter $L$ is the full width at half maximum (FWHM) of the gas density distribution. With the substitutions $z=L/2\tan u$ and $z'=5L$ the phase-matching function $F^{(3)}$ is determined by the following expression:

$$F^{(3)}=\bar{b}\frac{4k_g}{b_1}\left[\frac{L}{b_1}\right]^2\int_0^{\infty}r'\left|\int_{-\pi/2}^{+\pi/2}\frac{1}{a(u,\epsilon_1')}\exp[-r'^2q(u,\epsilon_1')]\exp\left[-i\frac{L}{b_1}\frac{b_1\Delta k_0}{2}u\right]du\right|^2 dr' , \quad (9)$$

with

$$\epsilon_1'=2\frac{L}{b_1}\left[5-\frac{z_{0,1}}{L}\right] .$$

Equations (7) and (9) indicate that in both cases $F^{(3)}$ depends essentially on the dimensionless parameters $b_1\Delta k_0$, $b_1/L$, $\bar{b}$, $\bar{k}$, $z_{0,1}/L$, and $\overline{\Delta z}$.

From Eqs. (7) and (3) the phase-matching function is calculated as a function of $b_1\Delta k_0$ for different parameters $\bar{b}$. The values of the parameter $\bar{k}$ depend on the conversion process. In the calculations values of $\bar{k}=0.4$ (for $\omega_g=2\omega_1+\omega_2$) and $\bar{k}=0.6$ (for $\omega_g=2\omega_1-\omega_2$) have been assumed. If the laser light is strongly focused into the center of the cell (with identical positions of the foci of the two light beams) the remaining parameters have the following values: $b_1/L=0.01$, $z_{0,1}/L=0$, and $\overline{\Delta z}=0$. The corresponding phase-matching functions $F_+^{(3)}$ calculated for sum-frequency mixing and different values $\bar{b}$ are displayed in Fig. 1(a). These results indicate that for sum-frequency mixing of tightly focused radiation $F^{(3)}$ is nonzero only if $b_1\Delta k_0<0$. This result if well known for $\bar{b}=1$ but is valid also for frequency mixing of laser light with different confocal parameters. Figure 1(a) shows in addition that the decrease of $F_+^{(3)}$ at higher negative values of $b_1\Delta k_0$ is smaller for larger values of the parameter $\bar{b}$. This behavior is even more obvious for the function $F_-^{(3)}$—determined for difference-frequency mixing—in the positive region of $b_1\Delta k_0$ [Fig. 1(b)]. For this reason the maxima of the corresponding $G^{(3)}$ functions are shifted to higher values of $b_1\Delta k_0$ if $\bar{b}$ is increased. This fact is seen explicitly from the graphs of the $G^{(3)}$ functions calculated for sum- and difference-frequency mixing as a function of $b_1\Delta k_0$ (Fig. 2). To demonstrate the possible enhancement of $G^{(3)}$ with increasing values of $\bar{b}$ the optimum values of $G^{(3)}$ are calculated as a function of $\bar{b}$ (Fig. 3). The change of the corre-

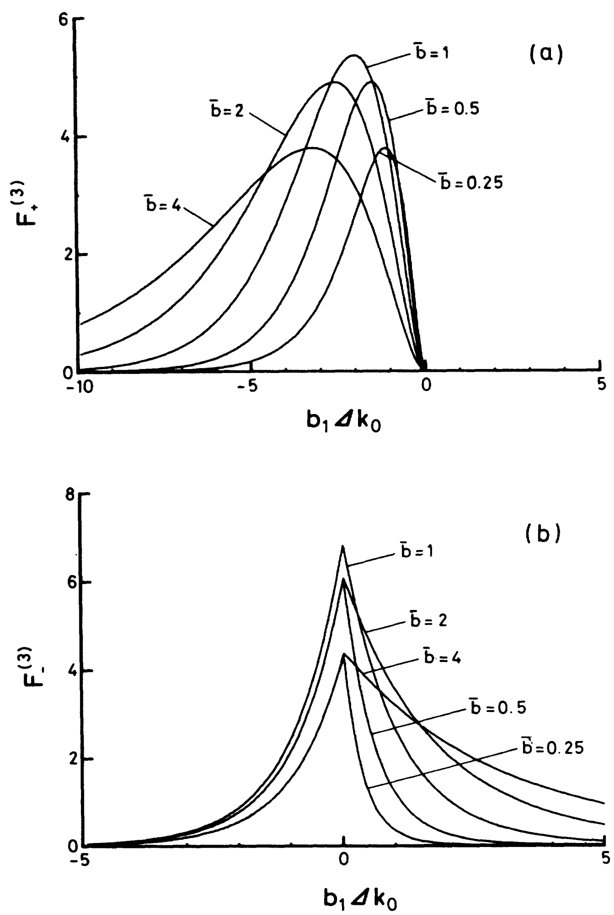


FIG. 1. $F^{(3)}$ as a function of $b_1\Delta k_0$ for different values of the ratio $\bar{b}$; $b_1/L=0.01$; $z_{0,1}/L=0$; $\overline{\Delta z}=0$; $L$ is the length of the gas cell; (a) $\omega_g=2\omega_1+\omega_2$, $\bar{k}=0.4$; (b) $\omega_g=2\omega_1-\omega_2$, $\bar{k}=0.6$.

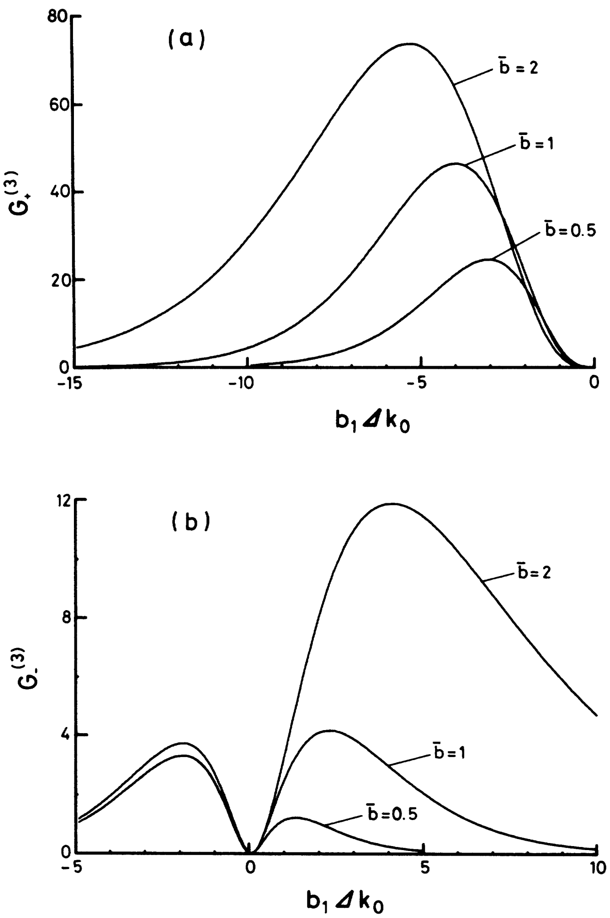


FIG. 2. $G^{(3)}$ as a function of $b_1\Delta k_0$ determined from the results displayed in Fig. 1 for three values of the ratio $\bar{b}$; (a) $\omega_g=2\omega_1+\omega_2$; (b) $\omega_g=2\omega_1-\omega_2$.

sponding values of $b_1\Delta k_0$ with $\bar{b}$ is shown in Fig. 4. The results of Fig. 3 indicate that the enhancement of the function $G^{(3)}$ is considerably larger for difference-frequency mixing than for sum-frequency generation. It should be noted that the wavelength dependence of the enhancement of $G^{(3)}$—characterized by $\bar{k}$—is different for both processes: For the sum frequency the enhancement of $G^{(3)}$ is larger for longer wavelengths of the generated radiation (and thus larger values of $\bar{k}$); for difference-frequency mixing, however, the improvement of $G^{(3)}$ is smaller for lower values of $\bar{k}$.

For a more detailed understanding of the enhancement of $G^{(3)}$ it is interesting to calculate the values of $F^{(3)*}$ which correspond to the maximum values of the $G^{(3)}$ functions $[F^{(3)*}=G^{(3)}_{\max}/(b_1\Delta k_0)^2_{\text{opt}}]$. In Fig. 5 these values are plotted as a function of the parameter $\bar{b}$. For the sum frequency as well as for the difference frequency $F^{(3)*}$ is optimum for $\bar{b}=1$. This result—which proves that optimum phase matching is obtained for equal confocal parameters (see also Fig. 1)—is in agreement with the calculation performed by Tomov and Richardson.[9] These authors analyzed third-order frequency conversion of Gaussian laser beams with different confocal parameters. Their calculations were restricted, however, to a homogeneous medium and did not provide information on the enhancement of the function $G^{(3)}$ achievable with different confocal parameters.

Despite the decrease of the values of $F^{(3)}$ the maxima of $G^{(3)}$ are shifted to larger negative values (in the case of sum-frequency mixing) or to larger positive values (for difference-frequency conversion) of the product $b_1\Delta k_0$. This increase of the optimum values of $|b_1\Delta k_0|$ (Fig. 4)

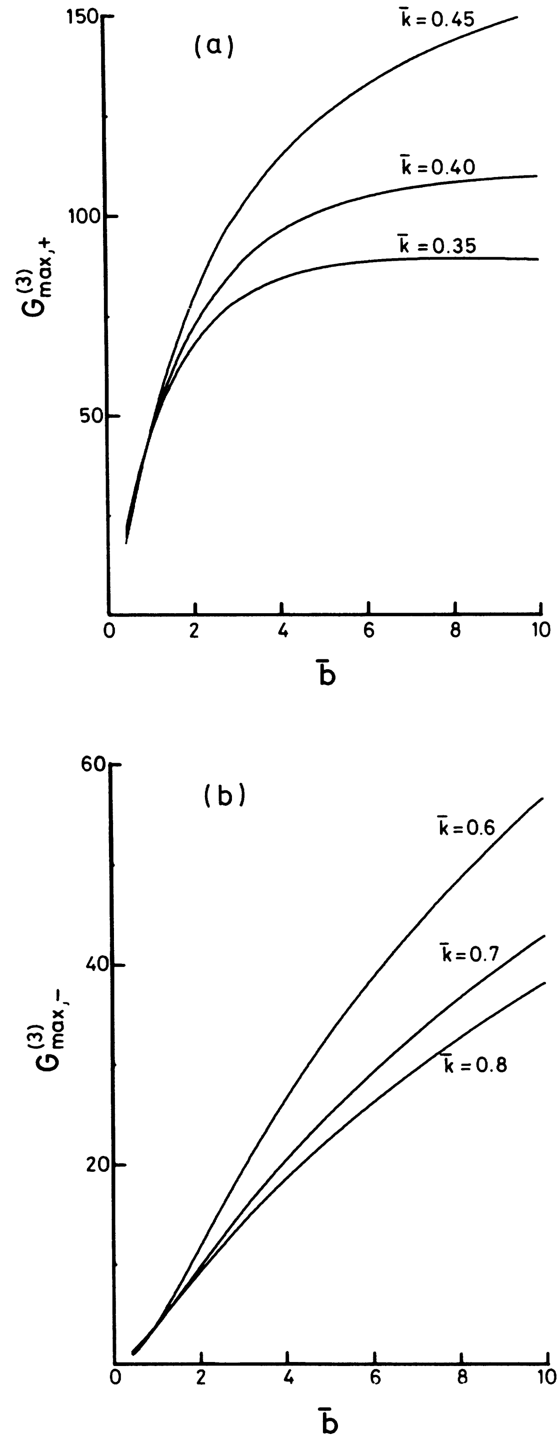


FIG. 3. Maximum values $G^{(3)}_{\max}$ of $G^{(3)}$ as a function of the ratio $\bar{b}$ for a homogeneous gas medium and three values of the parameter $\bar{k}$; (a) $\omega_g=2\omega_1+\omega_2$; (b) $\omega_g=2\omega_1-\omega_2$.

is the reason for the enhancement of $G^{(3)}$.

The results discussed so far indicate that the values of the confocal beam parameters $b_1$ and $b_2$ strongly influence the optimum value of $G^{(3)}$ and thus of the achievable output power and efficiency. This influence of $b_1$ and $b_2$ on the output power is illustrated in more detail by the following examples.

If $b_1$ if constant and $\bar{b}$ is increased by reducing $b_2$ the output power, which is proportional to $G^{(3)}$, is enhanced (see Fig. 3). This improvement in output power requires, however, that the value of $b_1 \Delta k_0$ is equal to $(b_1 \Delta k_0)_{opt}$. Since $(b_1 \Delta k_0)_{opt}$ increases with $\bar{b}$ (see Fig. 4) larger values

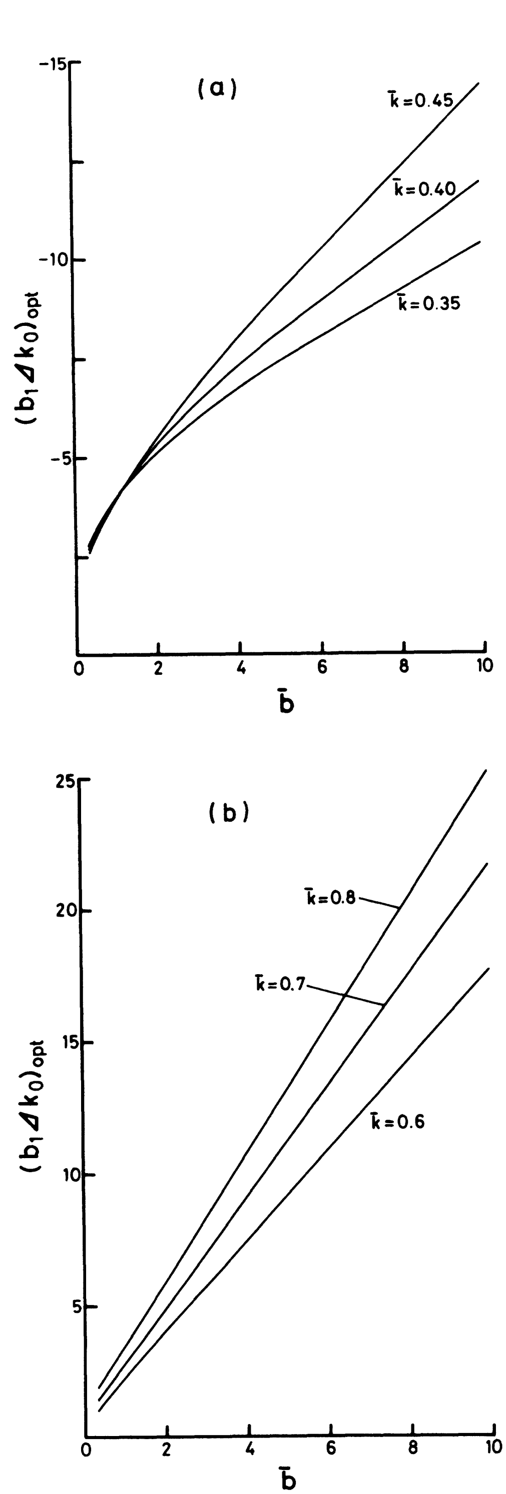


FIG. 4. Optimum values $(b_1 \Delta k_0)_{opt}$ of $b_1 \Delta k_0$ which correspond to the values of $G^{(3)}_{max}$ shown in Fig. 3; (a) $\omega_g = 2\omega_1 + \omega_2$; (b) $\omega_g = 2\omega_1 - \omega_2$.

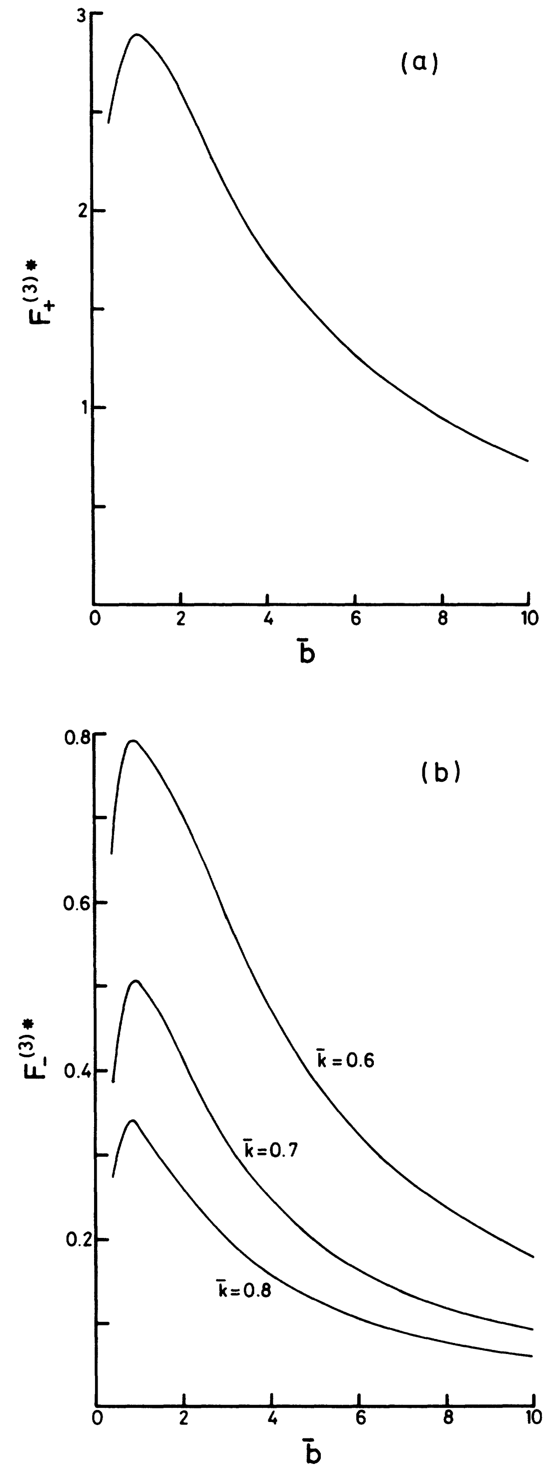


FIG. 5. Values $F^{(3)*}$ calculated from the values $G^{(3)}_{max}$ of Fig. 3; (a) $\omega_g = 2\omega_1 + \omega_2$, $\bar{k} = 0.45$ (the values for $\bar{k} = 0.35$ and 0.40 are almost identical to those for 0.45 and are thus not plotted); (b) $\omega_g = 2\omega_1 - \omega_2$.

of $\bar{b}$ require higher gas densities.

On the other hand, larger values of $b_2$ reduce the output power even if the gas density has the optimum value (see Figs. 2–4).

If the parameter $b_2$ is constant and $b_1$ is reduced we have to consider that the output is proportional to the ratio $G^{(3)}/b_1^2$. A detailed calculation indicates that in most cases the decrease of $G^{(3)}$ is smaller than the increase of $1/b_1^2$. Although the optimum value of the parameter $b_1\Delta k_0$ decreases (Fig. 4), the smaller value of $b_1$ still requires an increase of the gas density. As a result the output power is improved. The improvement in output power achievable in this way depends very much on the actual experimental conditions. If, for example, in a sum-mixing experiment $b_1$ is reduced by a factor of 0.5 the value of $G_{\max}^{(3)}$ decreases by a factor of 0.53 [see Fig. 2(a)]. The expected improvement in output (which is proportional to $G_{\max}^{(3)}/b_1^2$) is thus 2.12.

It should be mentioned that (at constant $b_2$) an increase of $b_1$ usually reduces the output because $G_{\max}^{(3)}$ increases less than $b_1^2$.

The examples discussed so far have to be compared with the case that both confocal parameters are equal and are reduced by the same amount. The power of the generated radiation is proportional to $1/b_1^2$. To obtain the optimum output power the gas density has to be increased. This result is well known from the analysis of the frequency conversion with equal confocal parameters.[5,7]

A detailed comparison of these different ways to change the values of $b_1$ and $b_2$ indicates that the largest enhancement of the output is obtained by reducing both confocal parameters $b_1$ and $b_2$. In practice, however, the possible reduction of the confocal parameters is limited. If, for example, a two-photon transition (with transition energy $2\hbar\omega_1$) is used to enhance the conversion efficiency small values of $b_1$ cause saturation of the conversion efficiency because of depopulation of the atomic ground state, intensity-dependent changes of the refractive index, and multiphoton ionization.[10–13] For these reasons $b_1$ should not be reduced below a certain value. In this case stronger focusing of the laser beam with frequency $\omega_2$ could improve the conversion efficiency since this radiation contributes to saturation by a smaller amount.

To investigate the dependence of the beam profile of the produced radiation on the ratio $\bar{b}$ the squared modulus of the electric field strength of the generated light is calculated for different values $\bar{b}$ as a function of the beam radius $r'$. The results obtained at the position $z'=L/2$ are shown in Fig. 6 for the sum frequency (with $\bar{k}=0.4$) as well as for the difference frequency (with $\bar{k}=0.7$). In this calculation the optimum values $(b_1\Delta k_0)_{\rm opt}$ were used as values of the parameter $b_1\Delta k_0$. It should be noted that the phase-matching function $F^{(3)}$ was calculated by integrating over the expression $r'\,|E_g(r')|^2$. Thus it is obvious from Fig. 6 that the decrease of $|E_g(r')|^2$ with increasing $\bar{b}$ causes—for both mixing processes—the observed decrease of the values of $F^{(3)*}$ (Fig. 5). The shape of the beam profile is almost Gaussian for the sum-frequency conversion but ring-

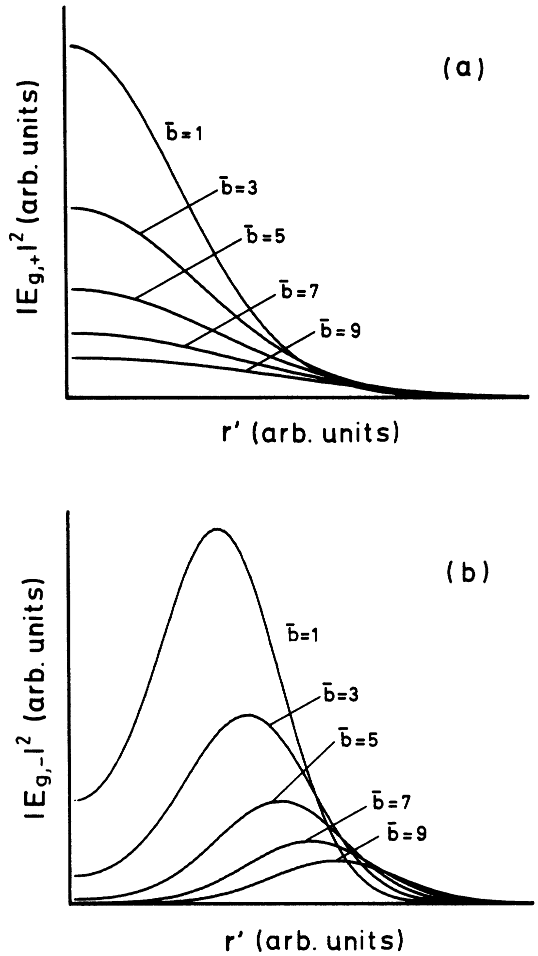


FIG. 6. Squared modulus of the field strength $E_g$ of the generated radiation as a function of the beam radius $r'$ at the position $z'=L/2$ for different values of the ratio $\bar{b}$ and $b_1\Delta k_0=(b_1\Delta k_0)_{\rm opt}$ (Fig. 4); $L$ is the length of the gas cell; (a) $\omega_g=2\omega_1+\omega_2$, $\bar{k}=0.4$; (b) $\omega_g=2\omega_1-\omega_2$, $\bar{k}=0.7$.

shaped for the difference-frequency mixing. As seen from Fig. 6(b) the diameter of the ring pattern—and thus of the divergence of the radiation—increases for larger values of $\bar{b}$. For negative dispersive media $(b_1\Delta k_0<0)$, however, the beam profile of the difference frequency is also nearly Gaussian. For $b_1\Delta k_0<0$ the divergence of the radiation generated by sum- or difference-frequency mixing can be estimated by applying Eq. (5). It can be shown that for sum-frequency mixing the values obtained by Eq. (5) agree very well with the results of the precise calculation [Fig. 6(a)]; in the case of difference-frequency mixing Eq. (5) provides at least a useful estimation of the divergence of the produced light beam.

Besides the study of the dependence of the vuv output on the ratio $\bar{b}$ of the confocal beam parameters it is of interest to analyze the influence of the relative positions of the foci of the two laser beams on the output power. This information should provide useful information on the precision of the axial overlap of the laser beams required for optimum conversion efficiencies. We assume that the focus of the laser beam with frequency $\omega_1$ is at the center of the gas cell. The focus of the laser beam with frequency $\omega_2$ is moved along the beam axis. Since for this investigation only the relative change of the output power is of interest the $G^{(3)}$ functions are calculated as a function of $\overline{\Delta z}$ and are normalized to their values at $\Delta z = 0$. In Fig. 7 the functions $G^{(3)}$ calculated for sum- and difference-frequency mixing are displayed for three different ratios $\bar{b}$ as a function of $\overline{\Delta z}$. Again the optimum values $(b_1 \Delta k_0)_{\text{opt}}$ are taken for the parameter $b_1 \Delta k_0$. The results shown in this figure indicate that the axial overlap of the foci is more critical for larger values of $\bar{b}$. Thus, if one intends to enhance the output power by increasing $\bar{b}$, the axial overlap of the foci has to be more precise. The wavelength dependence of the focus displacement is negligible. The results shown in Fig. 7 are calculated for $\bar{k} = 0.4$ (sum frequency) and $\bar{k} = 0.6$ (difference frequency). The $G^{(3)}$ functions calculated for other values of $\bar{k}$ are almost identical to those displayed in Fig. 7. It should be noted that in contrast to the results reported by Tomov and Richardson[9] the present analysis indicates that the precision required for the coincidence of the focus positions depends on the parameter $\bar{b}$. Moreover, it allows us to determine directly the dependence of the output power on the normalized focus displacement $\overline{\Delta z}$.

The analysis of the phase-matching functions presented so far is valid for a medium with homogeneous density distribution. However, the same calculations are performed easily for media with inhomogeneous density distributions. As an example, sum-frequency mixing ($\omega_g = 2\omega_1 + \omega_2$) is investigated in a free expanding gas jet. From the results given in Ref. 7 it is known that for $\bar{b} = 1$ the optimum output power is obtained if $b_1 = L$. The numerical evaluation of Eq. (9) (assuming $b_1 / L = 1$) provides the results displayed in Fig. 8. These results indicate that with increasing ratio $\bar{b}$ the output power is enhanced in the same way as the one obtained in a medium with homogeneous density distribution. It should be considered, however, that the gas density in a free expanding jet is usually limited to values of less than $10^{19}$ atoms/cm$^3$ and therefore the gas densities required for the enhancement of the output power may not be obtainable. Figure 9 displays the results for a displacement of the foci of the two laser beams. The decrease of the output power with increasing values of $\Delta z / b_1$ calculated for different ratios $\bar{b}$ is very similar to the results shown in Fig. 7 for a homogeneous density distribution. The results in Fig. 9 indicate that most of the converted radiation is generated within the focal region of the laser beam with frequency $\omega_1$. At $\Delta z = b_1 / 2$ the gas density is only one-half of its maximum value at the center of the jet.

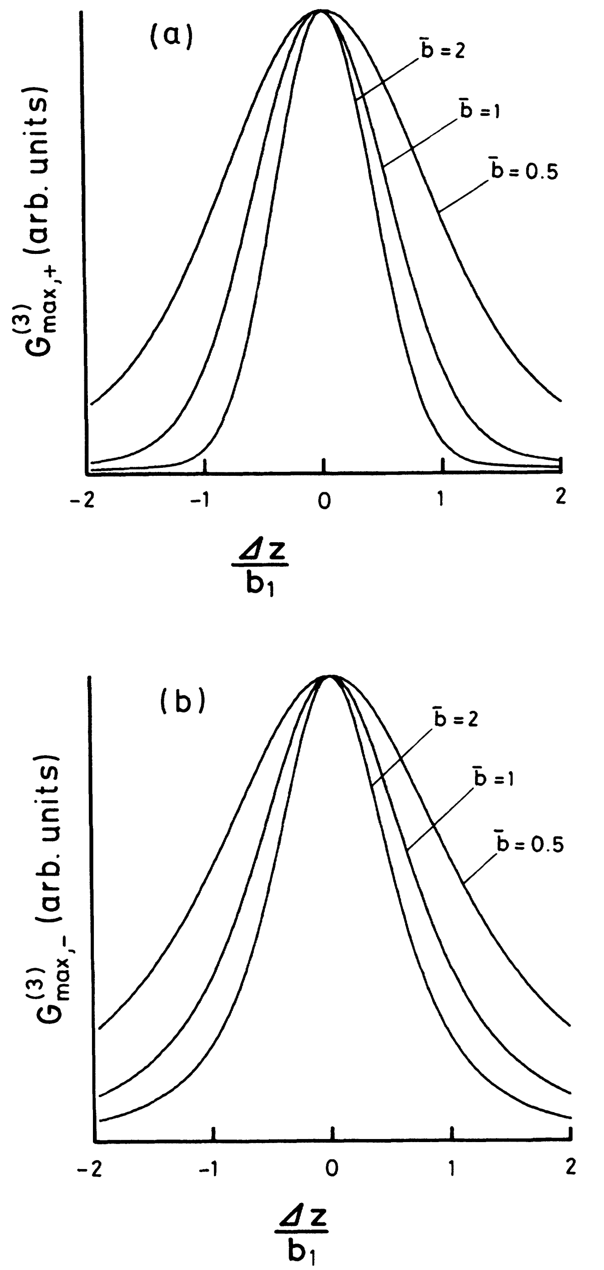


FIG. 7. $G^{(3)}_{\text{max}}$ as a function of $\Delta z / b_1$ for a homogeneous distribution of the gas density and three values of the ratio $\bar{b}$; the different functions $G^{(3)}$ are normalized to their values at $\Delta z / b_1 = 0$; (a) $\omega_g = 2\omega_1 + \omega_2$, $\bar{k} = 0.4$; (b) $\omega_g = 2\omega_1 - \omega_2$, $\bar{k} = 0.6$.

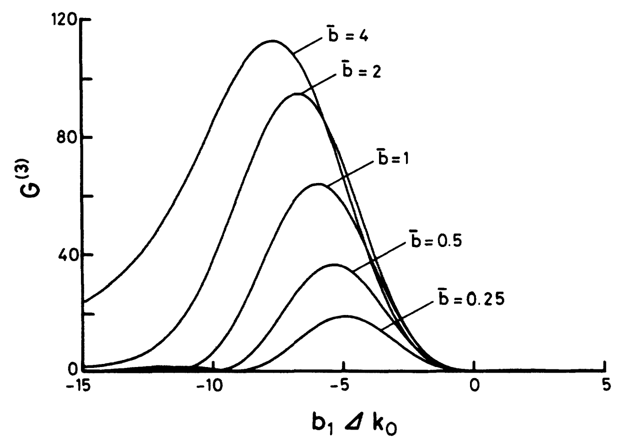


FIG. 8. $G^{(3)}$ as a function of $b_1 \Delta k_0$ for a Lorentzian density distribution of the gas jet and different values of the ratio $\bar{b}$; $b_1 / L = 1$; $z_{0,1} / L = 0$; $\overline{\Delta z} = 0$; $\bar{k} = 0.35$; $L$ is the FWHM of the Lorentzian gas profile; the conversion process is $\omega_g = 2\omega_1 + \omega_2$.

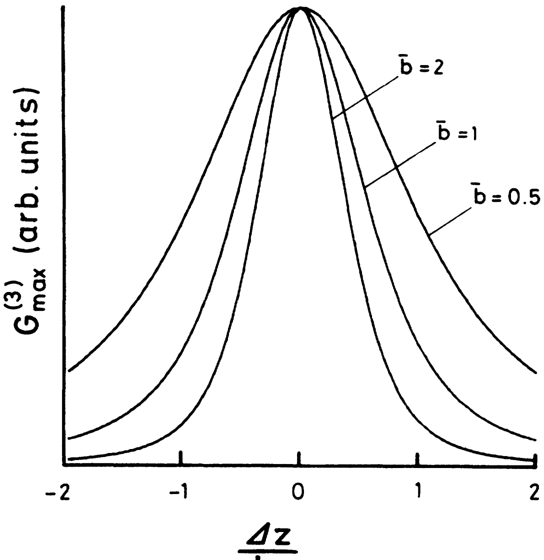


FIG. 9. $G^{(3)}_{max}$ as a function of $\Delta z/b_1$ for a Lorentzian density distribution of the gas jet and three values of the ratio $\bar{b}$; the different functions $G^{(3)}_{max}$ are normalized to their values at $\Delta z/b_1=0$; $\bar{k}=0.35$; conversion process: $\omega_g=2\omega_1+\omega_2$.

Despite this decrease of the gas density the output power calculated for a focus displacement of this size ($\Delta z=0.50$ is still 65% of the one expected for $\Delta z=0$ and $\bar{b}=1$.

It is of course of special interest to verify experimentally the enhancement of the power of the generated radiation expected for ratios $\bar{b}>1$. In a first experiment we investigated the difference-frequency mixing in xenon at $\lambda_{vuv}=134$ nm ($\omega_{vuv}=2\omega_1-\omega_2$) for different values of $\bar{b}$. At this vuv wavelength xenon is positive dispersive ($\Delta k_0>0$). Laser light at the frequencies $\omega_1$ and $\omega_2$ was generated by two Nd–yttrium aluminum garnet (YAG) laser-pumped dye lasers. The laser light was focused into a 300-mm-long stainless-steel cell filled with xenon. The radiation at $\omega_1$ ($\lambda_1=222.5$ nm) is generated by sum-frequency mixing ($\omega_1=\omega_{uv}+\omega_{ir}$) of the frequency-doubled dye laser output at $\lambda_{uv}=281.3$ nm and the infrared radiation of the Nd-YAG laser ($\lambda_{ir}=1.06$ $\mu$m). Visible dye laser light with frequency $\omega_2$ ($\lambda_2=650$ nm) was produced by the second dye laser. The input powers of the ultraviolet and visible laser light were $I_1=20$ kW and $I_2=600$ kW, respectively. To increase the efficiency of the conversion process the frequency $\omega_1$ was tuned close to the $6p'[1/2,0]$ two-photon resonance. The generated vuv radiation was spectrally separated from the fundamental laser light by a 0.2-m vacuum monochromator (McPherson model 234) and detected with a solar-blind photomultiplier (EMR 541G-08-18). The confocal parameter $b_1$ of the uv laser light was kept constant ($b_1=8$ mm) while the visible light ($\omega_2$) was focused with lenses of focal lengths of 500, 300, 200, and 100 mm. The values of the confocal parameters $b_1$ and $b_2$ of the input beams were determined from the beam divergence. The values of $\bar{b}$ obtained in this way were approximately 1, 2, 5, and 10. Considerable attention was given to the quality of the laser beams in order to obtain almost Gaussian intensity distributions. For the comparison of the experimental and theoretical results it is of importance to consider the increase of the divergence of the generated radiation with increasing ratio $\bar{b}$ (see Fig. 6). In the present experimental setup only part of the generated vuv light beam is detected. For this reason it was not possible to measure directly the increase of the vuv output power. In the performed measurements it was possible, however, to determine the optimum gas density—which is related to the parameter $(b_1\Delta k_0)_{opt}$—as a function of the parameter $\bar{b}$. The experimental results indicated an increase of the optimum gas pressure from 8 to 45 Torr. Figure 10 displays the corresponding increase of $(b_1\Delta k_0)_{opt}$ with increasing ratio $\bar{b}$. A comparison with the theoretical predictions (see Fig. 4) shows that the experimentally determined increase of $(b_1\Delta k_0)_{opt}$ is smaller by about 20–30%. There are several possible reasons for this difference. For a fixed value of the ratio $\bar{b}$ the diameter of the ring-shaped profile of the generated beam increases with gas density. Since part of the beam is cut off by the entrance slit of the detection system the measured increase of the output power with increasing gas density should be somewhat smaller than the actual value. In addition, deviations from the Gaussian intensity distribution of the laser beams should have the same influence on the experimental results. A new experimental setup which accepts the whole vuv beam (presently under construction) should allow a more precise test of the theoretical predictions.

## IV. CONCLUSIONS

The integral equation formalism has been used to obtain the phase-matching conditions for third-order sum- and difference-frequency mixing of laser beams with

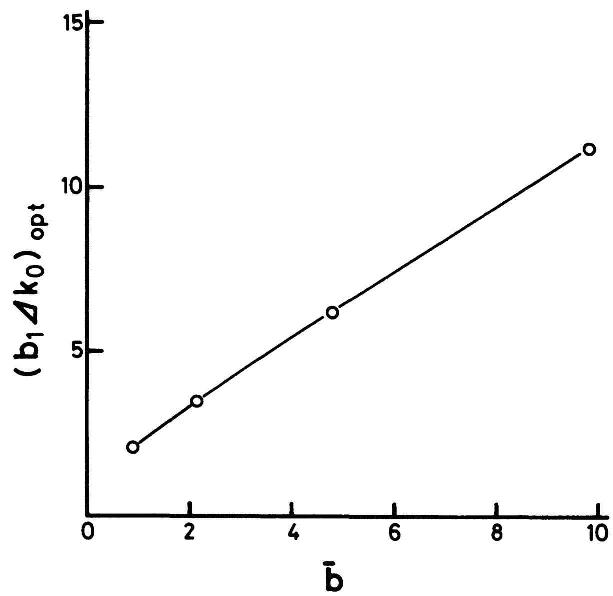


FIG. 10. The product $(b_1\Delta k_0)_{opt}$ as a function of the ratio $\bar{b}$ for vuv radiation generated at $\lambda_{vuv}=134$ nm by difference-frequency mixing in xenon ($\omega_{vuv}=2\omega_1-\omega_2$); the values of $(b_1\Delta k_0)_{opt}$ were determined from the gas density required for optimum vuv output.

different confocal parameters focused at different positions of the gaseous medium. The performed analysis provides new information important for the optimization of the efficiency of light sources in the vacuum ultraviolet based on the nonlinear frequency conversion in gases. The theoretical results determine the experimental conditions required for an optimum output of a certain conversion process. For this reason the derived results should be a very useful contribution to the further development of powerful, coherent vuv light sources.

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