The Gouy phase shift in nonlinear interactions of waves

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Abstract: We theoretically analyze the influence of the Gouy phase shift on the nonlinear interaction between waves of different frequencies. We focus on $\chi^{(2)}$ interaction of optical fields, e.g. through birefringent crystals, and show that focussing, stronger than suggested by the Boyd-Kleinman factor, can further improve nonlinear processes. An increased value of 3.32 for the optimal focussing parameter for a single pass process is found. The new value builds on the compensation of the Gouy phase shift by a spatially varying, instead constant, wave vector phase mismatch. We analyze the single-ended, singly resonant standing wave nonlinear cavity and show that in this case the Gouy phase shift leads to an additional phase during backreflection. Our numerical simulations may explain ill-understood experimental observations in such devices.

OCIS codes: (190.2620) Frequency conversion; (190.4970) Parametric oscillators and amplifiers.

References and links

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1. Introduction

Nonlinear interactions of waves, in particular those of optical fields, have opened new research areas and have found various applications. In general, waves of different frequencies are coupled via nonlinear media, like birefringent crystals. Examples are the production of higher harmonics of laser radiation [1], the generation of tunable frequencies through optical parametric

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oscillation [2] and the generation of nonclassical light [3, 4] for high precision metrology [5, 6], fundamental tests of quantum mechanics and quantum information [7]. The efficiency of a nonlinear process depends on parameters of the nonlinear medium, and generally increases with higher intensities of the fields involved and with better phase matching of their wave fronts. To achieve strong nonlinear interactions, pulsed laser radiation, strong focussing and, especially for continuous wave radiation, intensity build-up in resonators are used. In plane wave theory perfect phase matching is achieved if the wave fronts of interacting fields propagate with the same velocity. For focussed laser beams, however, this is not true because of the well-known Gouy phase shift. This phase shift occurs due to the spatial confinement of a focussed wave and generally depends on the spatial mode as well as the frequency of the wave [8]. The influence of focussing into a nonlinear medium has been investigated by Boyd and Kleinman in great detail [9]. They discovered that the efficiency of the nonlinear process does not monotonically increase with decreasing focal size. They especially considered the lowest order nonlinearity that enables second harmonic generation (SHG) and optical parametric amplification (OPA) and is described by the susceptibility $\chi^{(2)}$, and numerically found an optimum factor between the length of the nonlinear crystal and the Rayleigh range of the focussed Gaussian beam for a single pass through the crystal.

In this Letter we show that focussing stronger than suggested by the Boyd Kleinman factor can further improve nonlinear processes. We show that this effect can be understood by considering the Gouy phase shift between the interacting waves. We also show that the Gouy phase shift results in a non-trivial phase mismatch problem in standing wave cavities.

2. Singlepass interaction

Boyd and Kleinman have found that the maximum nonlinear coupling between two Gaussian beams of fundamental (subscript 1) and second harmonic waves (subscript 2) is achieved for a positive wave vector phase mismatch $\Delta k = 2|k_1| - |k_2| > 0$ which increases with decreasing waist size of the beam. For a single pass through a nonlinear crystal of length L they numerically found the optimal focussing parameter given by the relation

$$\xi := \frac{L}{2z_R} = 2.84\,, \tag{1}$$

where $z_R = \pi w_0^2 n_1/\lambda$ is the Rayleigh range of the beams inside the crystal, and w_0, n_1 and λ are the beam's waist size, refractive index and wavelength, respectively. We first show that the Boyd-Kleinman factor according to Eq. (1) is a consequence of maximizing the intensity of the mean pump field inside the nonlinear medium under the constraint of the Gouy phase shift. In a $\chi^{(2)}$ medium the nonlinear interaction is described by the following set of differential equations

$$\partial_z E_{0,1}(z) \quad \propto \quad E^*_{0,1}(z) E_{0,2}(z) \cdot g^*(z)\,, \tag{2}$$

$$\partial_z E_{0,2}(z) \quad \propto \quad E^2_{0,1}(z) \cdot g(z)\,, \tag{3}$$

$$g(z) := \frac{e^{i\Delta k z}}{1 + i\frac{z-z_0}{z_R}} = \frac{w_0}{w(z)} e^{i(\Delta k z + \Delta\phi(z))}\,, \tag{4}$$

where $E_{0,1}$, $E_{0,2}$ are the electrical fields of the fundamental and the harmonic mode in the focal center at position z_0, and $\Delta k = 4\pi\Delta n/\lambda$ is the phase mismatch between the two interacting modes. Although we look at the fields inside the waist, it is clear that the field strength at this position depends on the propagation length z through the nonlinear material. Here we use the following abbreviations

$$\Delta\phi(z) \quad = -\arctan\left(\tfrac{z-z_0}{z_R}\right)\,, \tag{5}$$

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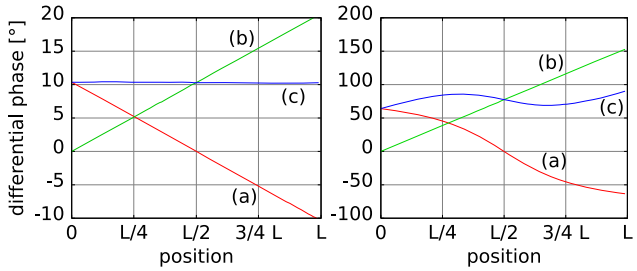

Fig. 1. (Color online) Left: For weak focussing into the nonlinear medium ($\xi = 0.18$) the Gouy phase shift can be compensated by choosing $\Delta k = 1/z_R$, and perfect phase matching can be realized over the full crystal length. Right: For stronger focussing ($\xi = 2.03$) a constant Δk can not provide perfect phase matching. (a) Gouy phase shift $\Delta\phi$, (b) compensating phase Δkz, (c) overall phase ϕ_0, where a constant value describes perfect phase matching.

$$w(z) \quad = w_0 \sqrt{1 + \left(\tfrac{z-z_0}{z_R}\right)^2}\,, \tag{6}$$

where $w(z)$ corresponds to the beam width at the position z. In plane wave theory one finds $g(z) = \exp(i\Delta kz)$ and $\Delta\phi = 0$, and equal indices of refraction for the two interacting modes provide the maximum nonlinearity. When focussing the beam into a nonlinear material however, there is a non zero phase difference $\Delta\phi$. Since the phase difference between a plane wave and a focussed Gaussian beam is given by the Gouy phase shift, $\Delta\phi$ should be the difference of two such phase shifts. When considering phase shifts between different oscillator frequencies, phases have to be frequency normalized. We therefore introduce the Gouy phase shift normalized to the optical frequency of mode i

$$\tilde{\phi}_G(\omega_i) := \frac{\phi_G}{\omega_i} = -\frac{(m+n+1)}{\omega_i} \arctan\left(\frac{z-z_0}{z_R}\right), \tag{7}$$

where m and n describe the spatial Hermite-Gaussian modes (TEM$_{mn}$). If we now normalize $\Delta\phi$ to the harmonic frequency we find

$$\frac{\Delta\phi}{\omega_2} = \tilde{\phi}_G(\omega_1) - \tilde{\phi}_G(\omega_2)\,. \tag{8}$$

We point out, that for an optimized nonlinear interaction of Gaussian beams the Rayleigh ranges are identical for all modes involved. In the case of frequency conversion of a single pump field this is automatically realized by the nonlinear process. For the $\chi^{(2)}$-processes considered in Eqs. (2)-(4) ($m = n = 0$) we find

$$\Delta\phi = \phi_G\,. \tag{9}$$

From this one can conclude that the Gouy phase shift leads to a nonperfect matching of the (nonplanar) phase fronts in nonlinear processes. To quantify this effect we define the *effective nonlinearity* κ of the process. This quantity is proportional to the conversion efficiency in SHG as well as to the optical gain of OPA. For weak interaction, i. e. the pump field is not depleted by the nonlinear interaction, κ is given by

$$\kappa := \left| \int dz\, \frac{g(z)}{w_0} \right|^2, \tag{10}$$

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where the integration is taken over the whole interaction length. For a single pass through a nonlinear medium of length L the effective nonlinearity is given by

$$\kappa_{sp} = \left| \int_0^L dz \, \frac{e^{i(\Delta k z + \phi_G(z))}}{w(z)} \right|^2 . \tag{11}$$

This quantity is maximized if the averaged field strength inside the crystal is maximized, i. e. if the focus is placed in the crystal center and if the condition

$$\Delta k z + \Delta\phi(z) = \phi_0 = \text{const.} \tag{12}$$

is satisfied. In this case all partial waves are produced exactly *in* phase to each other, and perfect phase matching is realized.

Curves (a) in Fig. 1 show the differential Gouy phase shifts $\Delta\phi(z) = \phi_G(z)$ for weak and strong focussing, respectively. For weak focussing the gouy phase shift evolves linearly inside the medium, and one can compensate this phase mismatch by choosing $\Delta k = 1/z_R > 0$, as found by Boyd and Kleinman [9]. For stronger focussing, however, it is not possible to achieve perfect compensation from Δk that is constant over the crystal. Curves (b) show the compensating linear phase Δkz that is due to the propagation inside the medium and curves (c) show the total phase ϕ_0. The value of Δk was chosen to provide the lowest variance of ϕ_0 over the whole interaction range.

We now show that it is possible to realize perfect phase matching for an arbitrary focussing by applying the following position dependent index of refraction

$$\Delta n_{sp}(z) = \frac{\lambda}{4\pi}\Delta k(z) \quad = \frac{\lambda}{4\pi}\frac{\phi_0 + \arctan(\frac{z-z_0}{z_R})}{z}, \tag{13}$$

where the constant value of the phase ϕ_0 is set by the Gouy phase at the entrance surface of the nonlinear medium

$$\phi_0 = \arctan\left(\frac{z_0}{z_R}\right) = \phi_G(0). \tag{14}$$

Figure 2 shows $\Delta n_{sp}(z)$ for a nonlinear crystal of length L for different focussing parameters ξ, with focus placed into the crystal's center. Curves (a) to (c) could experimentally be realized by applying an appropriate temperature gradient along z-direction. Alternatively, an electrical field applied to the crystal could be used. The temperature values on the right vertical axis constitute an example for 7 % magnesium-oxide-doped lithium niobate (MgO:$LiNbO_3$). Data was deduced from measurements of the nonlinear efficiency of crystals used in type I OPA in [6].

The effective nonlinearity for the single pass setup with perfect Gouy phase compensation can be written as

$$\kappa_{sp} = \frac{1}{\xi}\ln^2\left(\frac{\sqrt{1+\xi^2}+\xi}{\sqrt{1+\xi^2}-\xi}\right), \tag{15}$$

with $\xi = L/2z_R$. The numerical optimization of the above expression leads to

$$\xi_{opt} = 3.32. \tag{16}$$

Our result means that with optimum, position dependent phase matching, the optimal waist size is approximatly 7.5 % smaller than suggested by the Boyd-Kleinman factor, and according to this, the effective nonlinearity is further increased by 4.4 %.

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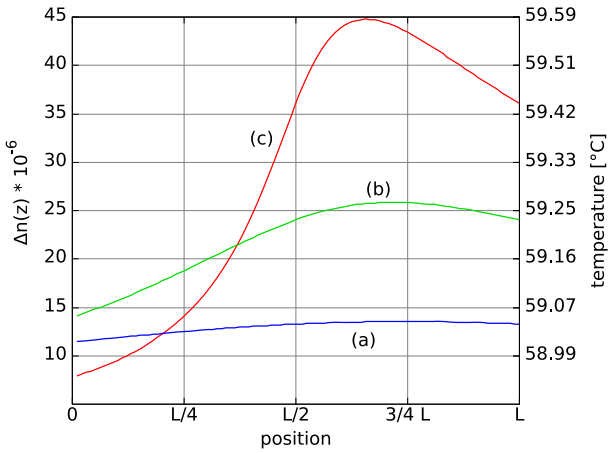

Fig. 2. (Color online) Change of refractive index along the crystal to compensate for the Gouy phase shift, for three different strengths of focussing. The temperature scale on the right corresponds to a $MgO(7\%):LiNbO_3$ crystal that has been used in [6]. (a) $\xi = 0.55$, (b) $\xi = 1.14$, (c) $\xi = 3.32$.

3. Nonlinear cavities

We now analyze if the nonlinear interaction in standing-wave cavities can be similarily improved. Standing wave cavities, in particular in the form of a singly-resonant, single-ended cavity, i. e. with one mirror of almost perfect reflectivity, are frequently used in quantum and nonlinear optics [10, 6] . In such cavities waves that propagate in two different directions interfere with each other, and the differential phases introduced by the reflections at the cavity mirrors have to be considered. Paschotta *et al.* have investigated the phase difference $\Delta\varphi$ introduced from back reflection and have suggested an appropriate design of the high reflectivity dielectric multi-layer coating to annihilate any additional phase shift. It can be shown that the effective nonlinearity for a doublepass of *plane* waves through a crystal depends on the $\Delta\varphi$ in the following way

$$\kappa_{dp,pw} = \frac{\sin^2\left(\frac{\Delta kL}{2}\right)}{\left(\frac{\Delta kL}{2}\right)^2} \cdot \cos^2\left(\frac{\Delta kL}{2} + \frac{\Delta\varphi}{2}\right). \tag{17}$$

For the calculation of the doublepass effective nonlinearity in the case of focussed Gaussian beams we model the system with a nonlinear medium of length $2L$ with a thin lense at position L that refocuses the beam. In this way we obtain two waists at positions z_0 and $z_0' = 2L - z_0$ of size w_0 indicating the way to the endmirror and the way back, respectively. Now we integrate over $2L$ and find the following expression for the effective nonlinearity for a double pass of the fundamental Hermite-Gauss mode

$$\begin{aligned}
\kappa_{dp} &= \tfrac{1}{w_0^2} \cdot \left| \int_0^L dz\, g(z,z_0) + \int_L^{2L} dz\, g(z,z_0') e^{i\Delta\varphi} \right|^2 \\
&= \tfrac{1}{w_0^2} \cdot \left| \int_0^L dz\, \left[g(z,z_0) + g(z+L,z_0') e^{i\Delta\varphi} \right] \right|^2 \\
&= \left| \int_0^L dz\, \tfrac{e^{i(\Delta kz + \phi_G(z))}}{w(z)} \times \right. \\
&\quad \left. \left(1 + \tfrac{w(z)}{w'(z)} e^{i(\phi_G'(z) - \phi_G(z) + \Delta kL + \Delta\varphi)}\right) \right|^2,
\end{aligned} \tag{18}$$

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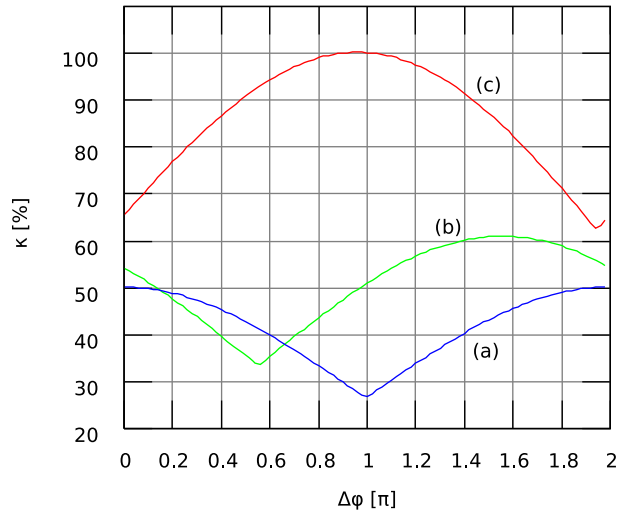

Fig. 3. (Color online) Effective nonlinearity κ_{dp} (normalized) versus the differential phase introduced by the back reflecting surface $\Delta\varphi$. For Gaussian beams with waist position on the reflecting surface, $\Delta\varphi = 0$ generally provides the highest κ_{dp}, (a) (here with strong focussing $\xi = 2.84$, $z_0 = L$); the same result is found for plane waves. Contrary, for $\xi = 2.84$ and $z_0 = L/2$ we find $\Delta\varphi \approx \pi$ (c). The result for weaker focussing is shown in (b) ($\xi = 0.775$, $z_0 = L/2$).

where $w'(z)$ and $\phi'_G(z)$ belong to the focus at position z'_0. $\Delta\varphi$ is again the differential phase that may be introduced by the coating of the back reflecting mirror. We first consider the special case of weak focussing, i. e. $|z - z_0|/z_R \ll 1 \quad \forall z \in [0, L]$, and simplify the above expression as follows

$$\kappa_{dp} = \frac{\sin^2\left(\frac{\Delta k' L}{2}\right)}{\left(\frac{\Delta k' L}{2}\right)^2} \cdot \cos^2\left(\frac{\Delta k' L}{2} + \frac{\Delta\varphi'}{2}\right), \tag{19}$$

where $\Delta k' := \Delta k - 1/z_R$ and

$$\Delta\varphi' := \Delta\varphi + 2(L - z_0)/z_R\,. \tag{20}$$

This expression has the same form as the one for plane waves as given in Eq. (17). However, an additional phase shift appears. This phase shift is a result of spatial confinement and the swapping in sign of the wave front's radius of curvature during reflection, and corresponds to minus twice the Gouy phase in the limit considered here. From the expression of $\Delta\varphi'$ in Eq. (20) it follows that this additional phase jump vanishes if the waist is located exactly at the back reflecting surface. In this case we have plane wave fronts at the end mirror and therefore the system is similar to a single pass through a nonlinear medium of length $2L$.

We now examine κ_{dp} for reflected Gaussian beams of arbitrary focussing parameters ξ and the waist located in the center of the medium. Eq. (18) yields

$$\kappa_{dp} = 4\cos^2\left(\frac{\Delta kL + \Delta\varphi}{2}\right)\left|\int_0^L dz\,\frac{e^{i(\Delta kz + \phi_G(z))}}{w(z)}\right|^2. \tag{21}$$

The first term provides the optimal phase of the end mirror of $\Delta\varphi = -\Delta kL$. In turn Δk is again found by minimizing the variance of the term $\Delta kz + \phi_G(z)$. We obtain the following expression

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for the optimal differential phase $\Delta\varphi$ of the end mirror versus focussing parameter

$$\Delta\varphi = -\frac{3}{\xi^2}\left[(1+\xi^2)\cdot\arctan(\xi)-\xi\right]. \tag{22}$$

Figure 3 shows the effective nonlinearity versus $\Delta\varphi$ for three different standing wave cavity arrangements. In all cases the second harmonic wave is not resonant but simply back reflected. Curves (a) and (c) use the focussing parameter $\xi = 2.84$. This value optimizes the effective nonlinearity of the cavity if the refractive index of the medium does not depend on direction of propagation of waves. This is evident from Fig. 2 because the position dependent refractive indices are not symmetric with respect to the focal position and the back reflected wave would require different values. However, if one transfers the results from a single pass through the medium and realizes the required propagation direction dependent refractive index the optimum focussing parameter is again $\xi = 3.32$. Curve (a) represents the case for focussing directly onto the back reflecting surface. Curve (c) shows the effective nonlinearity for a waist position at the crystal's centre. In the latter case the best choice of the differential phase at the back reflecting surface is $\Delta\varphi \approx \pi$. This is exactly the opposite of what one might expect from plane wave theory, where the optimum phase is $\Delta\varphi = 0$, similar to curve (a). Trace (b) shows the effective nonlinearity for the focussing parameter that was chosen by Paschotta *et. al.* [11]. In that paper a full *quantitative* comparison between experiment and theory of the nonlinearity in standing wave cavities was conducted. In their experiment the back reflecting mirror was designed to prevent a differential phase shift between the two interacting modes and a value of $\Delta\varphi = 0$ was chosen. However, their experimental data revealed the effective nonlinearity to be $\approx 10\,\%$ smaller than expected from their calculations. From our calculation it follows that the optimum phase for their setup was $\Delta\varphi \approx 1.55\,\pi$ and that the chosen value of $\Delta\varphi = 0$ decreased the effective nonlinearity to about 90 % of the maximum value. Our considerations are therefore in excellent agreement with experimental results given in that paper and can solve the observed discrepancy.

4. Conclusion

In conclusion we have shown how for focussed waves the Gouy phase shift produces nonideal phase matching in case of $\Delta k = 0$. For a single pass through a nonlinear medium the optimum focussing parameter is found to be $\xi = 3.32$. In this case a position dependent refractive index is required to further improve the effective nonlinearity by 4.4 %. For a double pass and for cavities an optimum focussing parameter above $\xi = 2.84$ can only be achieved with a refractive index that also depends on propagation direction. We have also shown that the Gouy phase shift effects the optimum value for the phases introduced by cavity mirrors, with a significant effect on the achievable effective nonlinearity. Our theoretical analysis shows exact agreement with experimental data published elsewhere, and may lead to improved quantitative descriptions of nonlinear cavities.

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