Nonlinear zeros in second-harmonic generation at grating couplers

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In a study concerned with second-harmonic generation at grating couplers, Kull *et al.* [Opt. Lett. **16,** 1930 (1991)] observed a curious phenomenon in which the second-harmonic efficiency exhibits dips, instead of peaks, when the angle of incidence of the pump beam is scanned through its resonant value. This surprising result has not been explained. It is our aim to show that this effect is closely related to the existence of nonlinear zeros.

Organics have attracted a great deal of interest for second-harmonic generation (SHG) because they exhibit a large χ^2 nonlinearity. In addition, these materials can be easily spin coated, leading to a guided wave or surface plasmon geometry for SHG. The grating coupler technique enables one to investigate the influence of resonances of χ^2 on the second-harmonic (SH) efficiency.

Such a spectroscopic study was performed in Ref. 1, in which SHG at a metallic grating coupler coated by a polyurethane side chain (PUSC) thin film was studied (Fig. 1). Two pump wavelengths λ_1 were used in such a way that the SH wavelength λ_2 ($\lambda_2 = \lambda_1/2$) fell outside or inside the absorption band of the polymer: For $\lambda_1 = 1319$ nm, $\lambda_2 = \lambda_1/2 = 660$ nm. Both λ_1 and λ_2 do not belong to the absorption domain of PUSC. For $\lambda_1 = 1064$ nm, $\lambda_2 = \lambda_1/2 = 532$ nm; λ_1 is not absorbed but λ_2 is inside the absorption range of the PUSC, which starts at 0.630 nm. In this case an enhancement of the nonlinear susceptibility is expected, together with a strong absorption only at λ_2. In both cases ($\lambda_1 = 1319$ and 1064 nm) the incidence angle was varied such that the electromagnetic resonances take place at the pump frequency, i.e., resonant excitation of guided modes or surface plasmons leading to an increase of the electric field at λ_1 inside the nonlinear guiding layer. Because of this electromagnetic resonance at λ_1, an enhancement of the SH efficiency is expected when the angle of incidence is scanned through its resonant values. This was indeed the case for $\lambda_1 = 1319$ nm (i.e., outside the absorption band), but for $\lambda_1 = 1064$ nm (λ_2 inside the absorption band) minima were observed instead of maxima. This unexpected result is not understood.

The aim of this Letter is to explain why an increase of the pump field intensity as the result of an electromagnetic resonance can lead to either an increase or a decrease of the SH efficiency, depending on the value of the SH wavelength. As we show here, the key point is the existence of nonlinear zeros first introduced in Ref. 2. Thus it is appropriate to begin the Letter by presenting these nonlinear zeros. The geometry of interest is represented in Fig. 1.

Throughout this Letter it is assumed that the undepleted pump approximation holds. This assumption linearizes the SH problem. Let $B_{2\omega}$ be a column vector whose components are the Rayleigh coefficients of the SH field diffracted in reflection (i.e., in the upper medium). According to the linearity of the problem, there exists a scattering matrix[2] $(S_{2\omega})$ at the SH frequency 2ω, which links the vector $(B_{2\omega})$ to a source vector $(A_{2\omega})$:

$$(B_{2\omega}) = (S_{2\omega})(A_{2\omega})\,. \tag{1}$$

According to Ref. 2, the vector $(A_{2\omega})$ accounts for the existence of the nonlinear polarization at 2ω, $\mathbf{P}^{\mathrm{NL}}(2\omega)$, arising from the diffraction of the pump field.[2,3] Besides, these grating couplers are used in such a way as to achieve resonant excitation of normal modes (guided modes, surface plasmons) at the pump frequency. Therefore the amplitude g_ω of the

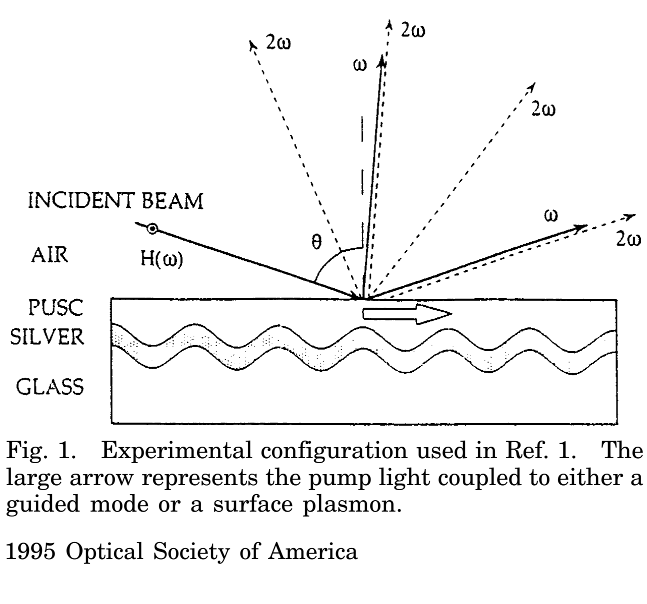

Fig. 1. Experimental configuration used in Ref. 1. The large arrow represents the pump light coupled to either a guided mode or a surface plasmon.

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resonantly excited evanescent diffracted order q at circular frequency ω can be written as[4,5]

$$g_\omega = \frac{t}{\beta - \beta_{1p}} A_i , \qquad (2a)$$

where β_{1p} is a complex pole[6] at frequency ω and

$$\beta = k_0 \sin \theta + q \frac{2\pi}{d} , \qquad (2b)$$

A_i is the amplitude of the pump field, and t is the in-coupling coefficient.

Recalling the expression of the nonlinear polarization,

$$P_i^{\mathrm{NL}}(2\omega) = \varepsilon_0 \chi_{ijh}(2\omega) E_i(\omega) E_h(\omega) , \qquad (3)$$

we see from Eqs. (3) and (2a) that β_{1p} is a double pole of $(A_{2\omega})$ and thus of $(B_{2\omega})$. In addition,[2] $(S_{2\omega})$ also presents poles at 2ω. Therefore an element $B_{2\omega,n}$ of $(B_{2\omega})$, which represents the amplitude of the nth diffracted order in reflection at circular frequency 2ω, can be written as

$$B_{2\omega,n}(h) = \frac{D_n(h)}{[\beta - \beta_{1p}(h)]^2 [2\beta - \beta_{2p}(h)]} . \qquad (4)$$

Let us now consider the associated plane structure that we deduce from the grating coupler by letting the groove depth h tend to zero. This plane device pumped by the ω field has a nonlinear reflectivity $R_{2\omega}$ that fulfills

$$\lim_{h \to 0} B_{2\omega,n}(h) = \delta_{0,n} R_{2\omega} , \qquad (5)$$

where $\delta_{0,n}$ is the Kronecker symbol ($\delta_{0,n} = 1$ if $n = 0$, $\delta_{0,n} = 0$ if $n \neq 0$).

According to Eqs. (4) and (5),

$$D_0(h = 0) = R_{2\omega}[\beta - \beta_{1p}(h = 0)]^2 [2\beta - \beta_{2p}(h = 0)] . \qquad (6)$$

Thus, in the limit for which $h = 0$, the numerator of $B_{2\omega,0}$ is proportional to

$$[\beta - \beta_{1p}(h = 0)]^2 [2\beta - \beta_{2p}(h = 0)] .$$

When the groove depth h increases from zero, computations[2] have shown that the poles existing in $B_{2\omega,0}$ move in the complex plane. But the poles of $B_{2\omega,0}$ have different trajectories whether they are in the numerator or in the denominator. In the numerator, the double pole β_{1p} gives rise to two different complex zeros, β_{11z} and β_{12z}, whereas the single pole β_{2p} leads to a single complex zero, β_{2z}. These nonlinear zeros fulfill

$$\beta_{11z}(h = 0) = \beta_{12z}(h = 0) = \beta_{1p}(h = 0) ,$$
$$\beta_{2z}(h = 0) = \beta_{2p}(h = 0) .$$

Thus $B_{2\omega,0}(h \neq 0)$ may be expressed as

$$B_{2\omega,0}(h) = R_{2\omega} \frac{[\beta - \beta_{11z}(h)][\beta - \beta_{12z}(h)][2\beta - \beta_{2z}(h)]}{[\beta - \beta_{1p}(h)]^2 [2\beta - \beta_{2p}(h)]} . \qquad (7)$$

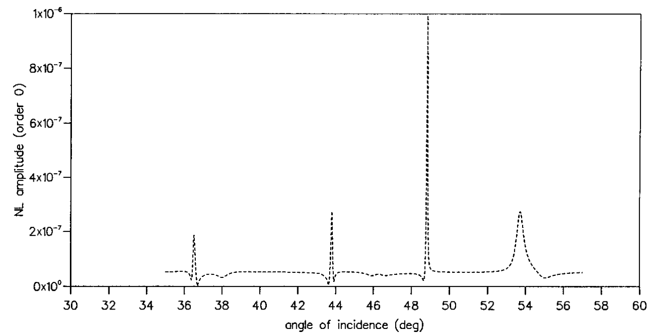
Equation (7) is the desired result. This equation can be rewritten as

$$B_{2\omega,0}(h) = a(h) + \frac{b(h)}{[\beta - \beta_{1p}(h)]^2} + \frac{c(h)}{[2\beta - \beta_{2p}(h)]} + \frac{d(h)}{[\beta - \beta_{1p}(h)]^2 [2\beta - \beta_{2p}(h)]} . \qquad (8)$$

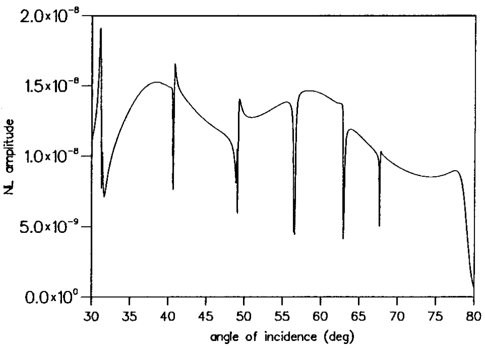
In Eq. (8) the coefficients a, b, c, and d depend on h but do not depend on the angle of incidence.

The physical meaning of Eq. (8) is clear; the specularly diffracted order at 2ω results from different contributions: a nonresonant term, a term (the second one) that has a resonance at ω only, a term (the third one) that has a resonance at 2ω only, and a resonant term (the last one) at both ω and 2ω. The last contribution corresponds to phase matching in which

$$2\mathrm{Re}[\beta_{1p}(h)] = \mathrm{Re}[\beta_{2p}(h)] . \qquad (9)$$

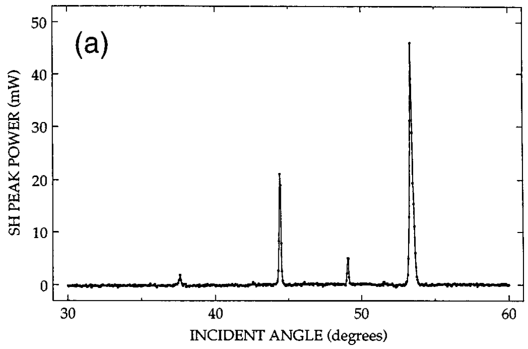


(a)

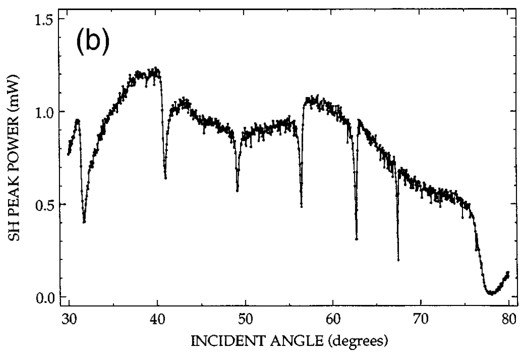


(b)

Fig. 2. Angular dependence of the SH specular amplitude. Periodicity, 1.66 μm; groove depth, 0.08 μm. (a) Pump wavelength $\lambda_1 = 1.319$ μm, SH wavelength $\lambda_2 = 0.6595$ μm. The thickness of the PUCS film is 2.92 μm. The refractive indices n have the following values: $n_{\mathrm{PUSC}}(\lambda_1 = 1.319\ \mu\mathrm{m}) = 1.57 + i0.00004$, $n_{Ag}(\lambda_1 = 1.319\ \mu\mathrm{m}) = 0.05976 + i8.3668$, $n_{\mathrm{PUSC}}(\lambda_2 = 0.6595\ \mu\mathrm{m}) = 1.585 + i0.0027$, $n_{Ag}(\lambda_2 = 0.6595\ \mu\mathrm{m}) = 0.09363 + i3.5361$. (b) Pump wavelength $\lambda_1 = 1.064$ μm, SH wavelength $\lambda_2 = 0.532$ μm. The thickness of the PUCS film is 3.05 μm. The refractive indices n have the following values: $n_{\mathrm{PUSC}}(\lambda_1 = 1.064\ \mu\mathrm{m}) = 1.574802 + i0.00004$, $n_{Ag}(\lambda_1 = 1.064\ \mu\mathrm{m}) = 0.13 + i7.474$, $n_{\mathrm{PUSC}}(\lambda_2 = 0.532\ \mu\mathrm{m}) = 1.6010998 + i0.0593343$, $n_{Ag}(\lambda_2 = 0.532\ \mu\mathrm{m}) = 0.051 + i3.16622$.



(a)



(b)

Fig. 3. Data from Ref. 1: (a) angular dependence of the specularly reflected SH peak power for $\lambda_1 = 1.319\ \mu m$ and $\lambda_2 = 0.6595\ \mu m$, (b) angular dependence of the specularly reflected SH peak power for $\lambda_1 = 1.064\ \mu m$ and $\lambda_2 = 0.532\ \mu m$.

The physical interpretation of Eqs. (7) and (8), as well as the existence of nonlinear zeros, extends to the SHG domain similar results already obtained in linear diffraction[6] and also in nonlinear optics for optical Kerr interactions at grating couplers.[7]

Equation (7) shows that the angular dependence of $B_{2\omega,0}(h \neq 0)$ depends on the respective positions of the zeros and the poles in the complex β plane. If the zeros are located close (at a distance of the order of the width of the resonance curve) to the poles, then minima are present instead of the maxima obtained when the zeros are far from the poles.

Let us now consider Fig. 1, which corresponds to the experimental configuration of Ref. 1.

The crystallographic group is $C_{\infty mm}$, and the pump field is TM polarized. Thus the SH signal is also TM polarized. According to the respective values of the pump wavelengths and grating periodicity, all the normal modes (guided modes or surface plasmon) at ω are resonantly excited through the $+1$ evanescent diffracted order. This means that $q = +1$ in Eq. (2b).

The angular distribution of the amplitude of the SH beam is plotted in Fig. 2(a) for $\lambda_1 = 1319$ nm and in Fig. 2(b) for $\lambda_1 = 1064$ nm. These curves have been obtained by new theoretical approaches[8] to study SHG at grating couplers. For convenience, the corresponding experimental results of Ref. 1 are shown in Fig. 3(a) ($\lambda_1 = 1319$ nm) and in Fig. 3(b) ($\lambda_1 = 1064$ nm).

The agreement between Figs. 2 and 3 is very good. The theoretical results reproduce well the angular intensity distribution at the SH frequency, i.e., maxima for $\lambda_1 = 1319$ nm, where λ_2 does not belong to the absorption band, and minima for $\lambda_1 = 1064$ nm, where λ_2 falls inside the absorption band.

It is the location of the nonlinear zeros and poles in the complex plane that determines the angular distribution of the SH signal. The curves in Fig. 2 demonstrate that not only the poles but also the nonlinear zeros may influence the angular distribution of the SH amplitude. For $\lambda_1 = 1319$ nm, the zeros are far from the poles, and the SH intensity exhibits maxima. For $\lambda_1 = 1064$ nm, owing to the proximity of zeros, a more complicated behavior of the SH intensity is observed.

In conclusion, this Letter shows that the authors of Ref. 1 have observed the influence of nonlinear zeros on the angular distribution of the SH intensity. To our knowledge it was the first time that such a phenomenon was reported. These nonlinear zeros are interesting not only on the level of fundamental physics but also in view of applications. Indeed, they may alter in a dramatic way the intensity distribution at the SH frequency leading to minima instead of maxima. Because these modifications result from the respective positions of the zeros and the poles, knowledge of their location in the complex plane is crucial when one is looking for efficient SH generators.

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