# Electromagnetic theory of diffraction in nonlinear optics and surface-enhanced nonlinear optical effects

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We present the first rigorous electromagnetic theory of diffraction in nonlinear optics. This theory allows the study of any type of nonlinear grating: bare or coated, whatever the groove depth and the profile of grating and coatings may be. The formalism developed here is derived from Maxwell's equations. The existence of the excitation and its nonlinear feature on the one hand, and the diffraction of the pump beams and of the signal on the other hand, are fully taken into account. The calculation reported here is valid for all cases of polarization (TM or TE) of the pump beams and of the signal. Two expressions of the nonlinear polarization at the signal frequency are derived. One is valid below the modulated region; the other one, inside this region. These two expressions take into account all the diffracted orders at the pump frequencies: propagating and evanescent. We then get the expression of the electromagnetic field at the signal frequency everywhere: not only outside the modulated region, but also inside it. The results thus obtained show that this electromagnetic field is a superposition of a diffracted field, with radiated and evanescent orders, and an infinite number of elementary driven waves. We also derive the nonlinear grating equation which allows the determination of the directions of propagation of the radiated diffracted orders. This is achieved using a new geometrical construction. It is shown that the evanescent diffracted orders at the signal frequency and at the pump frequencies can be resonantly excited. The rigorous feature of the electromagnetic theory developed here allows us to get the following new and important result: There exists an optimal groove depth for which the electromagnetic resonance contribution to the enhancement of the nonlinear optical process is the strongest. These results can be applied to the study of different nonlinear optical processes, such as enhanced second-harmonic generation, surface-enhanced Raman scattering, Pockels effect, and optical rectification.

## I. INTRODUCTION

The existing theories of diffraction all have in common the following features: They deal with the *linear* diffraction of light. In these studies, a plane wave with frequency $\omega$ is incident on a grating ruled on a linear material (in the optical sense) and the interest is in the diffracted field at the *same* frequency $\omega$. A rigorous study of this problem, i.e., a study which does not consider the groove depth of the grating as a perturbation, was developed in the last ten years by several authors (for a review, see Ref. 1). The rigorous feature of the formalism developed by these authors allowed them to explain, among other things, what is usually termed as Wood anomalies of gratings.[2] They showed that these anomalies, exhibited with a TM incident wave, are closely related to the resonant excitation of normal modes (which correspond to surface plasmons or surface polaritons) of the interface obtained by allowing the groove depth of the grating to go to zero.

The authors of Ref. 1 also showed that, in the case of coated gratings, other anomalies may exist which are associated with the existence of different normal modes (corresponding to guided waves) from those considered above. These anomalies occur not only with TM incident waves but also with TE ones.

Now what happens if the grating, instead of being ruled on a linear optical material, is impressed on a nonlinear one? In that case, we are concerned with the diffraction of light by a nonlinear grating. Some interesting questions may be raised:

(i) What are the efficiencies of the diffracted orders at the new frequencies but also at the pump frequencies?

(ii) What about the anomalies described above?

It must be emphasized that the answer to this last question is of great importance. Indeed, it is usually believed that the enhanced feature of Raman effect by molecules adsorbed on a grating[3] and second-harmonic generation[4] (SHG) is due, to a large extent, to the resonant excitation of surface plasmons,[5] i.e., of one of the normal modes cited above.

It is the aim of this paper to present the first electromagnetic theory of diffraction in nonlinear optics. A forthcoming paper will be devoted to corresponding numerical calculations. For the sake of specificity, we only consider nonlinear optical processes of the kind

$$(\omega_1,\omega_2)\rightarrow\omega_3=\omega_1+\omega_2 ,$$

where $\omega_1,\omega_2$ are the two incident pump beam frequencies.

Our theory explicitly takes into account

(a) the existence of the excitation,

(b) its nonlinear feature,

(c) the diffraction of the pump beams and of the signal beam,

(d) the losses of the different media, and

(e) it does not consider the groove depth as a perturbation.

This theory is valid for any groove shape. It even applies to coated gratings, whatever the nature (linear or nonlinear) of each of the layers may be. The analysis allows dealing with the two fundamental cases of polarization (TE, TM) of the incident pump beams and also of the signal. The generalization of the study to TE $\omega_1,\omega_2,\omega_3$ waves is required by the answer to question (ii) of this section. Indeed, as previously noted, when dealing with coated gratings, the anomalies also occur with TE waves.

To our knowledge, such a theory has never been developed. The existing theories, which do not consider the groove depth as a perturbation,[6] take into account neither the existence of the excitation nor the damping of the different media.

## II. THEORY OF NONLINEAR DIFFRACTION

### A. General considerations

Two beams with frequencies $\omega_1$ and $\omega_2$ $(\omega_1>\omega_2)$ are incident on a grating (periodicity $d$, groove depth $\delta$) ruled on a dielectric or metallic nonlinear medium and coated by one or several dielectric or metallic layers constituted by linear or nonlinear materials (Fig. 1).

Above the modulated structure, space is filled with a linear material whose permittivity is real and positive. $\theta_\nu$ $(\nu=1,2)$ denotes the angle of incidence of each of the pump beams. The case of a nonlinear medium whose entrance face is constituted by a bare grating is dealt with by making $e=0$.

Throughout the paper $\partial/\partial z=0$. Thus the solutions at frequency $\omega_\nu$ $(\nu=1,2,3)$ are either TM or TE polarized. In the following, the concept of associated smooth structure will be useful: It is the system derived from that of Fig. 1 by allowing every modulation depth to tend to zero.

The method used to study the nonlinear diffraction is the following: First, we obtain the expression of the electric field of the diffracted beams at each frequency $\omega_\nu$ $(\nu=1,2)$ below and inside the modulated region. Then, we determine the expression of the nonlinear polarization at frequency $\omega_3=\omega_1+\omega_2$ below the modulated region but also inside it. Finally, we generalize the theory developed in Ref. 7 (henceforth referred to as I) to the case where the diffraction of the pump beams is taken into account and where the signal at frequency $\omega_3$ may be TE polarized.

### B. Expression of the electric field at frequencies $\omega_1$ and $\omega_2$

We make the usual undepleted pump approximation for the two pump beams. Thus, Maxwell's equations read

$$\vec{\nabla}\times\vec{\mathscr{E}}(\omega_\nu,\vec{r})=j\omega_\nu\mu_0\vec{\mathscr{H}}(\omega_\nu,\vec{r}) , \tag{1a}$$

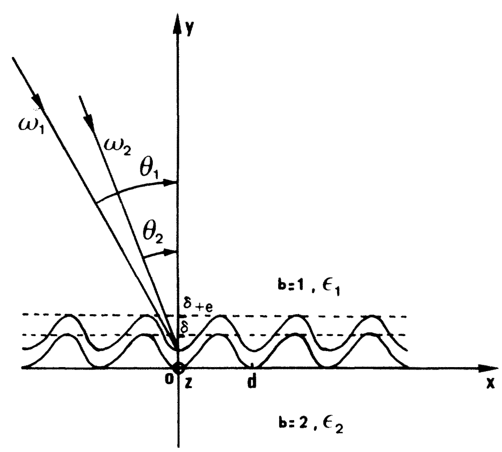


FIG. 1. Scattering geometry when the grating (periodicity $d$, groove depth $\delta$) is coated by a single dielectric or metallic layer. Index $b$ characterizes the medium above or below the modulated region.

$$\vec{\nabla}\times\vec{\mathscr{H}}(\omega_\nu,\vec{r})=-j\omega_\nu\epsilon_0\epsilon(\omega_\nu,x,y)\vec{\mathscr{E}}(\omega_\nu,\vec{r})\ ,\quad \nu=1,2\ . \tag{1b}$$

The $e^{-j\omega t}$ time dependence is assumed and the magnetic permittivity is equal to $\mu_0$ everywhere.

In Eqs. (1), $\epsilon(\omega_\nu,x,y)$ is the relative permittivity which, in the modulated region, depends on $x$ and $y$. Outside the modulated region,

$$\epsilon(\omega_\nu,x,y)=\epsilon_2(\omega_\nu)\quad \text{for } y<0\ ,$$

$$\epsilon(\omega_\nu,x,y)=\epsilon_1(\omega_\nu)\quad \text{for } y>\delta+e\ ,\quad \nu=1,2\ .$$

From Eqs. (1), we see that (a) the two pump beams are *linearly* diffracted by the grating; (b) their diffraction takes place independently of that of the signal at frequency $\omega_3$ since in Eqs. (1), $\nu=1,2$.

According to point (a), the expression of $\vec{\mathscr{E}}(\omega_\nu,r)$ $(\nu=1,2)$ can be obtained by using the rigorous linear electromagnetic theory of gratings.[1] For this theory to apply, we must assume that all the overcoatings have the same periodicity, but not necessarily the same profile. This assumption will be made throughout this paper. Let us consider separately each case of polarization of the incident pump beams.

### *1. TM case*

Since we deal with the TM case,

$$\vec{\mathscr{H}}(\omega_\nu,\vec{r})=\mathscr{H}(\omega_\nu,\vec{r})\vec{e}_z\quad (\nu=1,2)$$

where $\vec{e}_z$ is a unit vector directed along the $z$ axis.

*a. Expression of $\vec{\mathscr{E}}(\omega_\nu,\vec{r})$ below the modulated region ($y<0$).* The expression of $\mathscr{H}(\omega_\nu,\vec{r})$ may be written as[1]

$$\mathscr{H}(\omega_1,\vec{r})=\sum_{l=-\infty}^{+\infty} T_{l,1}e^{j(-\alpha_{2,l,1}y+\gamma_{l,1}x)}\ , \tag{2a}$$

$$\mathscr{H}(\omega_2,\vec{r})=\sum_{m=-\infty}^{+\infty} T_{m,2}e^{j(-\alpha_{2,m,2}y+\gamma_{m,2}x)}\ , \tag{2b}$$

with

$$\alpha_{2,l,1}^2+\gamma_{l,1}^2=\epsilon_2(\omega_1)\frac{\omega_1^2}{c^2}=k_2^2(\omega_1)\ , \tag{3a}$$

$$\alpha_{2,m,2}^2+\gamma_{m,2}^2=\epsilon_2(\omega_2)\frac{\omega_2^2}{c^2}=k_2^2(\omega_2)\ , \tag{3b}$$

and

$$\gamma_{l,1}=k_1(\omega_1)\sin\theta_1+l\sigma\ , \tag{3c}$$

$$\gamma_{m,2}=k_1(\omega_2)\sin\theta_2+m\sigma\ , \tag{3d}$$

where

$$\sigma=\frac{2\pi}{d}\ ,$$

$$k_1^2(\omega_\nu)=\epsilon_1(\omega_\nu)\frac{\omega_\nu^2}{c^2}\quad (\nu=1,2)\ .$$

From Eq. (1b), we get

$$\vec{\mathscr{E}}(\omega_\nu,\vec{r})=j\frac{\omega_\nu\beta_2(\omega_\nu)}{\epsilon_0 c^2}\vec{\nabla}\mathscr{H}(\omega_\nu,\vec{r})\times\vec{e}_z\ , \tag{4}$$

with

$$\beta_b(\omega_\nu)=\frac{c^2}{\omega_\nu^2\epsilon_b(\omega_\nu)}\quad (b=1,2,\ \nu=1,2)\ .$$

Putting Eqs. (2) into Eq. (4) yield

$$\vec{\mathscr{E}}(\omega_1,\vec{r})=\sum_{l=-\infty}^{+\infty}\vec{u}_{l,1}e^{j(-\alpha_{2,l,1}y+\gamma_{l,1}x)}\ , \tag{5a}$$

$$\vec{\mathscr{E}}(\omega_2,\vec{r})=\sum_{m=-\infty}^{+\infty}\vec{u}_{m,2}e^{j(-\alpha_{2,m,2}y+\gamma_{m,2}x)}\ , \tag{5b}$$

with

$$u_{l,1,x}=\frac{\omega_1\alpha_{2,l,1}\beta_2(\omega_1)}{\epsilon_0 c^2}T_{l,1}\ , \tag{6a}$$

$$u_{l,1,y}=\frac{\omega_1\gamma_{l,1}\beta_2(\omega_1)}{\epsilon_0 c^2}T_{l,1}\ , \tag{6b}$$

$$u_{l,1,z}=0\ .$$

The expressions of the components of $\vec{u}_{m,2}$ are derived from those of $\vec{u}_{l,1}$ by making the substitution $(l,1)\rightarrow(m,2)$.

*b. Expression of $\vec{\mathscr{E}}(\omega_\nu,\vec{r})$ in the modulated region ($0<y<\delta+e$).* We now have[1]

$$\mathscr{H}(\omega_1,\vec{r})=\sum_{l=-\infty}^{+\infty}H_{l,1}(y)e^{j\gamma_{l,1}x}\ , \tag{7a}$$

$$\mathscr{H}(\omega_2,\vec{r})=\sum_{m=-\infty}^{+\infty}H_{m,2}(y)e^{j\gamma_{m,2}x}\ . \tag{7b}$$

Let

$$\beta(\omega_\nu,x,y)=\frac{c^2}{\omega_\nu^2\epsilon(\omega_\nu,x,y)}\ , \tag{8a}$$

i.e., $\beta(\omega_\nu,x,y)$ is now a function of $x$ and $y$ which is periodic with respect to $x$ with period $d$. Thus it may be expanded into its Fourier series,

$$\beta(\omega_1,x,y)=\sum_{l=-\infty}^{+\infty}\beta_{l,1}(y)e^{jl\sigma x}\ . \tag{8b}$$

For the expansion of $\beta(\omega_2,x,y)$, make the substitution $(l,1)\rightarrow(m,2)$. Putting Eqs. (7) and (8) into Eq. (4) leads to

$$\vec{\mathscr{E}}(\omega_1,\vec{r})=\sum_{l=-\infty}^{+\infty}\vec{E}_{l,1}(y)e^{j\gamma_{l,1}x}\ , \tag{9a}$$

$$\vec{\mathscr{E}}(\omega_2,\vec{r})=\sum_{m=-\infty}^{+\infty}\vec{E}_{m,2}(y)e^{j\gamma_{m,2}x}\ , \tag{9b}$$

with

$$E_{l,1,x}=j\frac{\omega_1}{\epsilon_0 c^2}\sum_{l'=-\infty}^{+\infty}\beta_{l-l',1}(y)\frac{dH_{l',1}(y)}{dy}\ , \quad (10a)$$

$$E_{l,1,y}=\frac{\omega_1}{\epsilon_0 c^2}\sum_{l'=-\infty}^{+\infty}\beta_{l-l',1}(y)\gamma_{l',1}H_{l',1}(y)\ , \quad (10b)$$

$$E_{l,1,z}=0\ .$$

The components of $\vec{E}_{m,2}$ are derived from those of $\vec{E}_{l,1}$ by making the substitution $(l,l',1)\rightarrow(m,m',2)$.

### 2. *TE case*

Here, the situation is much simpler since the electric field at frequency $\omega_\nu$ ($\nu=1,2$) is directed along the $z$ axis:

$$\vec{\mathscr{E}}(\omega_\nu,\vec{r})=\mathscr{E}(\omega_\nu,\vec{r})\vec{e}_z\ .$$

*a. Expression of $\mathscr{E}(\omega_\nu,\vec{r})$ below the modulated region*[1] *($y<0$).*

$$\mathscr{E}(\omega_1,\vec{r})=\sum_{l=-\infty}^{+\infty}u_{l,1,z}e^{j(-\alpha_{2,l,1}y+\gamma_{l,1}x)}\ , \quad (11a)$$

$$\mathscr{E}(\omega_2,\vec{r})=\sum_{m=-\infty}^{+\infty}u_{m,2,z}e^{j(-\alpha_{2,m,2}y+\gamma_{m,2}x)}\ , \quad (11b)$$

with $u_{l,1,z},u_{m,2,z}$ being the Rayleigh coefficients of $\mathscr{E}(\omega_1,\vec{r})$ and $\mathscr{E}(\omega_2,\vec{r})$.

*b. Expression of $\mathscr{E}(\omega_\nu,\vec{r})$ in the modulated region*[1] *($0<y<\delta+e$).*

$$\mathscr{E}(\omega_1,\vec{r})=\sum_{l=-\infty}^{+\infty}E_{l,1,z}(y)e^{j\gamma_{l,1}x}\ , \quad (12a)$$

$$\mathscr{E}(\omega_2,\vec{r})=\sum_{m=-\infty}^{+\infty}E_{m,2,z}(y)e^{j\gamma_{m,2}x}\ . \quad (12b)$$

### 3. *General expression of $\vec{\mathscr{E}}(\omega_\nu,\vec{r})$ ($\nu=1,2$)*

The coefficients of expansions (5), (9), (11), and (12) are determined with the aid of computer programs described in Ref. 1. From now on, $\vec{u}_{l,1}$ and $\vec{E}_{l,1}$ refer to vectors whose nonzero components are derived respectively from Eqs. (6a), (6b), (10a), and (10b) in the TM case and from $u_{l,1,z}$ and $E_{l,1,z}$, occurring in Eqs. (11a) and (12a), in the TE case. To get $\vec{u}_{m,2},\vec{E}_{m,2}$ make the substitution $(l,1)\rightarrow(m,2)$. Let us emphasize that $\vec{u}_{l,1},\vec{u}_{m,2},\vec{E}_{l,1},\vec{E}_{m,2}$ depend on $\delta$, but contrary to $\vec{E}_{l,1}$ and $\vec{E}_{m,2}$, $\vec{u}_{l,1}$ and $\vec{u}_{m,2}$ are independent of $y$. Finally, whatever the polarization of each of the incident beams may be, we may write the following expansion for the electric field, below the modulated region ($y<0$):

$$\vec{\mathscr{E}}(\omega_1,\vec{r})=\sum_{l=-\infty}^{+\infty}\vec{u}_{l,1}e^{j(-\alpha_{2,l,1}y+\gamma_{l,1}x)}\ , \quad (13a)$$

$$\vec{\mathscr{E}}(\omega_2,\vec{r})=\sum_{m=-\infty}^{+\infty}\vec{u}_{m,2}e^{j(-\alpha_{2,m,2}y+\gamma_{m,2}x)}\ , \quad (13b)$$

and in the modulated region ($0<y<\delta+e$),

$$\vec{\mathscr{E}}(\omega_1,\vec{r})=\sum_{l=-\infty}^{+\infty}\vec{E}_{l,1}(y)e^{j\gamma_{l,1}x}\ , \quad (13c)$$

$$\vec{\mathscr{E}}(\omega_2,\vec{r})=\sum_{m=-\infty}^{+\infty}\vec{E}_{m,2}(y)e^{j\gamma_{m,2}x}\ . \quad (13d)$$

Where the components of $\vec{u}_{l,1},\vec{u}_{m,2},\vec{E}_{l,1},\vec{E}_{m,2}$ are determined according to the above mentioned statement. We are now in a position to get the expression of the nonlinear polarization at frequency $\omega_3=\omega_1+\omega_2$.

### 4. *Nonlinear polarization below the modulated region ($y<0$)*

We have to distinguish between the two situations where the nonlinear material on which the grating is ruled is either a dielectric or a metal.

*a. Dielectric.* The nonlinear (NL) polarization at frequency $\omega_3$ is written as[8]

$$\mathscr{P}_h^{\mathrm{NL}}(\omega_3=\omega_1+\omega_2,\delta,\vec{r})=\chi_{h,i,j}(\omega_3=\omega_1+\omega_2)\mathscr{E}_i(\omega_1,\vec{r})\mathscr{E}_j(\omega_2,\vec{r})\ , \quad (14)$$

$[\chi(\omega_3)]$ is the nonlinear susceptibility tensor of the medium on which the grating is ruled.

According to Eqs. (3c), (3d), (13a), and (13b)

$$\vec{\mathscr{P}}^{\mathrm{NL}}(\omega_3,\delta,\vec{r})=\sum_{l,p}\vec{p}_{l,p-l}^{\,\mathrm{NL}}e^{j\vec{\kappa}_{l,p}\cdot\vec{r}}\ , \quad (15)$$

with

$$p=l+m\ ,$$

$$\kappa_{l,p,x}=\gamma_{l,1}+\gamma_{p-l,2}\ , \quad (16)$$

$$\kappa_{l,p,y}=-(\alpha_{2,l,1}+\alpha_{2,p-l,2})\ ,$$

$$p_{l,p-l,h}^{\mathrm{NL}}=\chi_{h,i,j}(\omega_3=\omega_1+\omega_2)u_{l,1,i}u_{p-l,2,j}\ . \quad (17)$$

Let us introduce

$$\vec{K}=\vec{\kappa}_{0,0}\ ,$$

which would be the wave vector of the nonlinear polarization if the two pump beams were not diffracted.

Equation (15) may be rewritten as

$$\vec{\mathscr{P}}^{\mathrm{NL}}(\omega_3,\delta,\vec{r})=e^{j\vec{K}\cdot\vec{r}}\sum_{p=-\infty}^{+\infty}\vec{P}_p^{\mathrm{NL}}(\omega_3,\delta,y)e^{jp\sigma x} \quad (18a)$$

with

$$\vec{P}_p^{NL}(\omega_3,\delta,y)=\sum_{l=-\infty}^{+\infty}\vec{P}_{l,p-l}^{NL}e^{j(\kappa_{l,p,y}-K_y)y}\ . \quad (18b)$$

Equations (17), (18a), and (18b) are valid for any polarization of the incident beams.

*b. Metal.* In Ref. 9, Bloembergen *et al.* determined the expression of the nonlinear polarization at the second harmonic frequency in a metallic medium. If, instead of dealing with SHG, we are concerned with the nonlinear interaction $(\omega_1,\omega_2)\rightarrow\omega_3=\omega_1+\omega_2$ the results of these authors may be generalized into

$$\vec{\mathscr{P}}^{NL}(\omega_3,\delta)=\tilde{a}\vec{\nabla}[\vec{\mathscr{E}}(\omega_1,\vec{r})\cdot\vec{\mathscr{E}}(\omega_2,\vec{r})]+\tilde{b}[\omega_2\vec{\mathscr{E}}(\omega_1,\vec{r})\vec{\nabla}\cdot\vec{\mathscr{E}}(\omega_2,\vec{r})+\omega_1\vec{\mathscr{E}}(\omega_2,\vec{r})\vec{\nabla}\cdot\vec{\mathscr{E}}(\omega_1,\vec{r})]\ . \quad (19)$$

Equation (19) is obtained in the same way as Eq. (14) of Ref. (9), except that now the expansion of the quantities occurring in Ref. 9 must be done with respect to the frequencies $\omega_1,\omega_2,\omega_1+\omega_2,\ldots$.

In Eq. (19)

$$\tilde{a}=\frac{n_0e^3}{m^{*2}\omega_1\omega_2\omega_3^2}\ ,$$

$$\tilde{b}=\frac{e\epsilon(0)}{m^*\omega_1\omega_2\omega_3}\ ,$$

with $n_0$ the unperturbed density of the electrons, $e$ the charge of the electron, $m^*$ the average effective mass of the electrons, and $\epsilon(0)$ the static permittivity.

The nonlinear polarization at frequency $\omega_3$ can still be put under the form given by Eqs. (18a) and (18b). But now, due to Eqs. (3c), (3d), (13a), and (13b), we get

$$\vec{P}_{l,p-l}^{NL}=j\tilde{a}(\vec{u}_{l,1}\cdot\vec{u}_{p-l,2})\vec{\kappa}_{l,p} \quad (20)$$

### 5. *Nonlinear polarization inside the modulated region* ($0<y<\delta+e$)

Here, we must take care of the fact that the layers are not necessarily of the same nature. We may find, for example, an overcoating of nonlinear dielectric on a nonlinear metallic grating or more generally a superposition of several layers, each of them being either a linear or a nonlinear dielectric, a linear or nonlinear metal. For a given $x$, when $y$ is varied, we meet a succession of linear or nonlinear media which can be either a dielectric or a metal. Thus, inside the modulated region, $\chi_{h,i,j}$, $\tilde{a}$, and $\tilde{b}$ are functions of $x$ and $y$.

The dependence of $\chi_{h,i,j}$, $\tilde{a}$, and $\tilde{b}$ on $x$ and $y$ is related to the characteristics of the modulated region: shape and groove spacing of the profile, nature of the material constituting the grating, and the eventual overcoatings.

The nonlinear polarization of a dielectric is written as

$$\mathscr{P}_h^{NL}(\omega_3,\delta,\vec{r})=\chi_{h,i,j}(\omega_3,x,y)\times\mathscr{E}_i(\omega_1,\vec{r})\mathscr{E}_j(\omega_2,\vec{r})\ , \quad (21a)$$

and that of a metal is written as

$$\vec{\mathscr{P}}^{NL}(\omega_3,\delta,\vec{r})=\tilde{a}(x,y)\vec{\nabla}[\vec{\mathscr{E}}(\omega_1,\vec{r})\cdot\vec{\mathscr{E}}(\omega_2,\vec{r})]+\tilde{b}(x,y)[\omega_2\vec{\mathscr{E}}(\omega_1,\vec{r})\vec{\nabla}\cdot\vec{\mathscr{E}}(\omega_2,\vec{r})+\omega_1\vec{\mathscr{E}}(\omega_2,\vec{r})\vec{\nabla}\cdot\vec{\mathscr{E}}(\omega_1,\vec{r})]\ . \quad (21b)$$

In Eqs. (21a) and (21b), $\vec{\mathscr{E}}(\omega_1)$ and $\vec{\mathscr{E}}(\omega_2)$ are the electric fields at the pump frequencies inside the modulated region. According to Eqs. (3c), (3d), (13c), and (13d), both Eqs. (21a) and (21b) can be put under the following form:

$$\mathscr{P}_h^{NL}=e^{j\vec{K}\cdot\vec{r}}\sum_{p=-\infty}^{+\infty}\mathscr{V}_{p,h}(\delta,x,y)e^{jp\sigma x}\ , \quad (22)$$

with

$$\mathscr{V}_{p,h}(\delta,x,y)=\chi_{h,i,j}(x,y)V_{p,i,j}^d(\delta,y)e^{-jK_yy}\ , \quad (23)$$

inside a nonlinear dielectric where

$$V_{p,i,j}^d(\delta,y)=\sum_{l=-\infty}^{+\infty}E_{l,1,i}(y)E_{p-l,2,j}(y)$$

and with

$$\vec{\mathscr{V}}_p(\delta,x,y)=\vec{\mathscr{V}}_p^a(\delta,x,y)+\vec{\mathscr{V}}_p^b(\delta,x,y) \quad (24)$$

inside a nonlinear metal with [according to Eqs. (3c), (3d), (13c), and (13d)]

$$\vec{\mathscr{V}}_p^a(\delta,x,y)=\tilde{a}(x,y)\vec{V}_p^a(\delta,y)e^{-jK_yy}\ ,$$

$$\vec{V}_p^a(\delta,y)=\vec{e}_xj(p\sigma+K_x)\sum_{l=-\infty}^{+\infty}\vec{E}_{l,1}(y)\cdot\vec{E}_{p-l,2}(y)+\vec{e}_y\sum_{l=-\infty}^{+\infty}\frac{d[\vec{E}_{l,1}(y)\cdot\vec{E}_{p-l,2}(y)]}{dy}\ ,$$

$\vec{e}_x,\vec{e}_y$ are unit vectors directed along the $x$ and $y$ axis and

$$\vec{\mathscr{V}}_p^b(\delta,x,y)=\tilde{b}(x,y)\vec{V}_p^b(\delta,y)e^{-jK_yy}\ ,$$

$$\vec{V}_p^b(\delta,y)=\sum_{l=-\infty}^{+\infty}\left[\omega_1\vec{E}_{p-l,2}\left[\frac{dE_{l,1,y}}{dy}+j\gamma_{l,1}E_{l,1,x}\right]+\omega_2\vec{E}_{l,1}\left[\frac{dE_{p-l,2,y}}{dy}+j\gamma_{p-l,2}E_{p-l,2,x}\right]\right]\ .$$

According to Eqs. (23) and (24), the $h$ coordinate $\mathscr{V}_{p,h}(x,y)$ of vector $\vec{\mathscr{V}}_p$ depends on $x$ only through $\chi_{h,i,j}(x,y)$, $\tilde{a}(x,y)$ and $\tilde{b}(x,y)$. Thus $\mathscr{V}_{p,h}(x,y)$ is a periodic function of $x$ with periodicity $d$. Therefore, we may expand it in Fourier series:

$$\mathscr{V}_{p,h}(\delta,x,y)=\sum_{q=-\infty}^{+\infty} v_{p,q,h}(\delta,y)e^{jq\sigma x} . \qquad (25)$$

Use of Eq. (25) shows that Eq. (22) may be rewritten as

$$\mathscr{P}_h^{\rm NL}(\omega_3,\delta,\vec{r})=e^{j\vec{K}\cdot\vec{r}}\sum_{n=-\infty}^{+\infty} P_{n,h}^{\rm NL}(\omega_3,\delta,y)e^{jn\sigma x} , \qquad (26a)$$

with

$$P_{n,h}^{\rm NL}(\omega_3,\delta,y)=\sum_{p=-\infty}^{+\infty} v_{p,n-p,h}(\delta,y) , \qquad (26b)$$

$$n=p+q=l+m+q . \qquad (26c)$$

Equations (18a) and (26a) give the expression of the nonlinear polarization at frequency $\omega_3=\omega_1+\omega_2$ below and inside the modulated region. Let us emphasize that these equations are valid for any grating profile and even for multicoated gratings (whatever the nature of the coatings may be), as well as for any polarization of the incident pump beams. These two equations are important and, before continuing, it is worth discussing them.

### *6. Discussion of Eqs. (18a) and (26a)*

It is seen from these equations that the diffraction of the two pump beams has the following consequence: Below the grating and inside the modulated region the nonlinear polarization at frequency $\omega_3=\omega_1+\omega_2$ is a superposition of an infinite number of elementary nonlinear polarizations, each of them having a longitudinal wave-vector component: $K_x+p\sigma$ below the modulated region and $K_x+n\sigma$ inside this region.

In order to understand this result, one has to remember that the nonlinear polarization at frequency $\omega_3$ comes from the nonlinear interaction between all the diffracted orders at frequencies $\omega_1$ and $\omega_2$. Besides, in region $y<0$, the interaction between a given $l$ and a given $m$ leads to what we may call a subelementary nonlinear polarization, i.e., $\vec{p}_{l,p-l}^{\rm NL}e^{j\vec{K}_{l,p}\cdot\vec{r}}$, with longitudinal wave-vector component:

$$\gamma_{l,1}+\gamma_{m,2}=K_x+(l+m)\sigma .$$

Thus the subelementary nonlinear polarizations may be ordered in sets characterized by the fact that all the subelementary nonlinear polarizations belonging to a given set have the same longitudinal wave-vector component. Any of these sets arises from the nonlinear interaction between all the diffracted orders $[l]$ at frequency $\omega_1$ and $[m]$ at frequency $\omega_2$ for which

$$l+m=p .$$

Each of these sets gives rise to an elementary nonlinear polarization with longitudinal wave-vector component $K_x+p\sigma$.

The same is true for the region $0<y<\delta+e$. But, in addition, one has the $\sigma$ periodicity along the $x$ axis which comes from the modulation of the nonlinear polarization. The longitudinal wave-vector component of each of the elementary nonlinear polarizations is equal to $K_x+(p+q)\sigma$. Now, the diffracted orders have to be classified with respect to the values of $n=p+q$, each set $n$ leading to an elementary nonlinear polarization in the modulated region.

Contrary to what one could think, all the subelementary nonlinear polarizations with the same longitudinal wave-vector component $K_x+p\sigma$ (or $K_x+n\sigma$) may have very different efficiencies. This will occur whenever the associated smooth structure will support guided normal modes at frequency $\omega_1$ and/or $\omega_2$.

Indeed, it has been shown in Ref. 10 that, fixing the frequency $\omega_\nu$ ($\nu=1,2$), there exists a specific value $\Theta_{l,1}$ of $\theta_1$ and $\Theta_{m,2}$ of $\theta_2$ for which the quantity $|T_{l,1}|^2, |T_{m,2}|^2$ of a given transmitted diffracted order, $l$ at frequency $\omega_1$, $m$ at frequency $\omega_2$, is maximum. It has also been shown in Ref. 10 that the value of this maximum strongly depends on $\delta$: There even exists a value $\delta_{\rm opt}(\omega_\nu)$ of $\delta$ for which this maximum value is itself maximum. If $\delta$ is increased beyond $\delta_{\rm opt}(\omega_\nu)$, then this peak value decreases. These results remain valid if, instead of fixing $\omega_\nu$, we fix $\theta_\nu$ and sweep $\omega_\nu$ ($\nu=1,2$).

This phenomenon, i.e., the existence of $\delta_{\rm opt}$ and $\Theta$, arises from the resonant excitation, at $\omega_1$ and $\omega_2$, of the guided normal modes which propagate along the associated smooth structure.[1,10] For example, we have surface plasmons (surface polaritons) when dealing with a bare metallic (dielectric) grating; surface plasmons or guided waves (either TE or TM) in the case of a dielectric coated metallic grating.

The feature of the modulated region, namely bare or coated grating, is taken into account in the theory via the Fourier expansions, Eq. (8b), of $\beta(\omega_\nu,x,y)$ and Eq. (25) of $\mathscr{V}_{p,h}(x,y,\delta)$.

We are led to the following important conclusion: Among the numerous values $l$ of $[l]$ and $m$ of $[m]$ such that $l+m=p$, one may select two special couples $(l,\Theta_{l,1})$ and $(m,\Theta_{m,2})$ for which the transmitted intensity of the corresponding diffracted orders at

$\omega_1$ and $\omega_2$ is maximum, i.e., such that a resonant excitation of the guided normal modes occurs at frequencies $\omega_1$ and $\omega_2$. Then, the modulus of the corresponding subelementary nonlinear polarization is strongly enhanced when compared either to the case of flat interface between the two media $\epsilon_1$ and $\epsilon_2$ or to the case where no resonance takes place at the pump frequencies $\omega_1$ and $\omega_2$. This occurs, if $\theta_1 \neq \Theta_{l,1}$ and $\theta_2 \neq \Theta_{m,2}$ and if the associated smooth structure does not support any guided normal modes. Moreover, this enhancement can be *optimized* by choosing $\delta = \delta_{\text{opt}}$ either at frequency $\omega_1$ or at frequency $\omega_2$.

The special case of SHG (i.e., $\omega_1 = \omega_2$) is of interest because if the modulated structure is optimized ($\delta = \delta_{\text{opt}}$) at $\omega_1$ it is also optimized at $\omega_2$. Thus the case of SHG is the most favorable with respect to the enhancement of the nonlinear effect [Figs. 2(a) and 2(b)]. When $\delta = \delta_{\text{opt}}(\omega_1)$, the second-harmonic intensity will be strongly enhanced as compared to the case of a flat interface.

Let us now discuss the nature of the $\omega_1$ and $\omega_2$ diffracted orders occurring in the expressions (18a) and (26a) of the nonlinear polarization at frequency $\omega_3 = \omega_1 + \omega_2$.

According to the sign of $\text{Re}[\epsilon_2(\omega_\nu)]$ (where Re means the real part thereof) and to the values of $\gamma_{l,1}$ and $\gamma_{m,2}$ as compared to $\text{Re}[k_b(\omega_\nu)]$ ($b = 1,2$), a given diffracted order at $\omega_1$ or $\omega_2$ is almost one of the following cases: (a) evanescent above and below the modulated region; (b) evanescent below the grating and radiated above the modulated region (or vice versa); (c) radiated above and below the modulated region.

Expressions (18a) and (26a) of the nonlinear polarization at $\omega_3$ explicitly take into account the fact that the nonlinear excitation of the electromagnetic (EM) field comes from a nonlinear interaction between all the preceding types of diffracted orders at $\omega_1$ and $\omega_2$. All these diffracted orders, *regardless* of their nature, contribute to the nonlinear polarization. But, as pointed out previously, the efficiency of each contribution depends on the fact that a given diffracted order may or may not lead to a resonant excitation of one of the guided normal modes of the associated smooth structure. Notice that it is the diffracted orders of type (a) which may lead to this resonant excitation.

### C. Nonlinear diffraction of the $\omega_3 = \omega_1 + \omega_2$ electromagnetic field

The starting point is Maxwell's equation written at frequency $\omega_3$:

$$\vec{\nabla} \times \vec{\mathscr{E}}(\omega_3, \vec{r}) = j\omega_3\mu_0 \vec{\mathscr{H}}(\omega_3, \vec{r}) , \tag{27a}$$

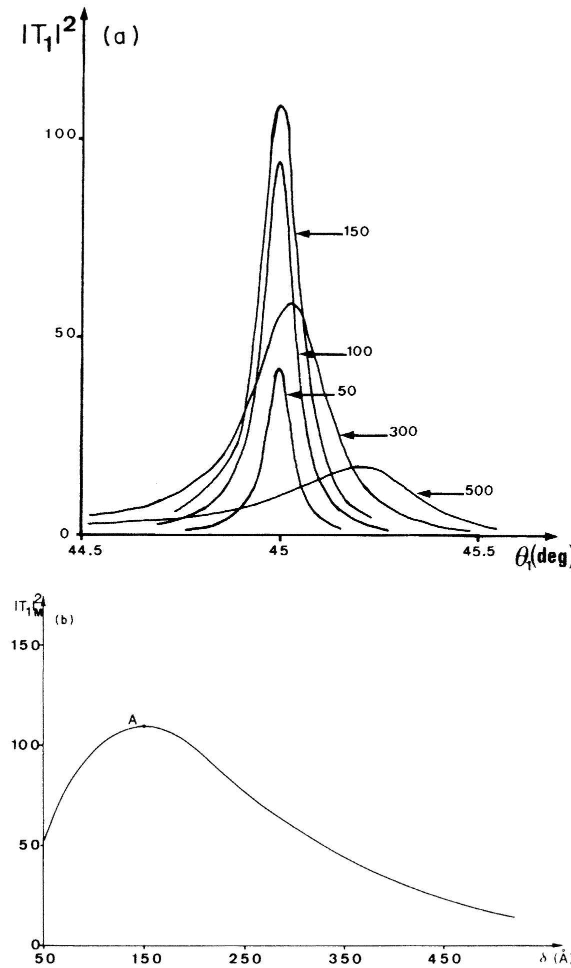


FIG. 2. (a) Relative transmitted intensity, $|T_1|^2$, of the $l = 1$ diffracted order at the pump wavelength $\lambda_1 = 1.06$ $\mu$m ($\lambda_1 = 2\pi c/\omega_1$) as a function of the incidence angle $\theta_1$ for different groove depths from 50 to 500 Å in the case of a bare sinusoidal grating with periodicity 6174 Å. Notice the shift of the $|T_1|^2$ peak position with $\delta$. Grating material: Ag. If the beam with wavelength $\lambda_1$ acts as a pump for SHG in such a medium, then the maximum increase of the square modulus of the nonlinear polarization at the second-harmonic frequency is of the order of $10^3$ as compared to the case of a flat air-Ag interface. (b) Peak value, $|T_1|^2_M$ of $|T_1|^2$, as a function of the groove depth $\delta$. The *maximum maximorum* of $|T_1|^2$ occurs at point $A$ for $\delta_{\text{opt}} = 150$ Å. $d = 6174$ Å, grating material: Ag.

$$\vec{\nabla} \times \vec{\mathscr{H}}(\omega_3, \vec{r}) = -j\omega_3 \vec{\mathscr{D}}(\omega_3, \vec{r}) , \tag{27b}$$

with

$$\vec{\mathscr{D}}(\omega_3, \vec{r}) = \epsilon_0 \epsilon(\omega_3, x, y) \vec{\mathscr{E}}(\omega_3, \vec{r}) + \vec{\mathscr{P}}^{\text{NL}}(\omega_3, \vec{r}) . \tag{28}$$

The determination of the nonlinear diffracted EM field at frequency $\omega_3$ is performed in the following way:

(a) We divide the space into three regions by means of the planes $y=0$, $y=\delta+e$ and obtain the expression of the propagation equation in each region.

(b) The solution of each propagation equation is looked for by means of a suitable expansion of the EM field.

(c) The coefficients of these expansions are linked to each other through the matching conditions at $y=0$ and $y=\delta+e$ applied to the tangential components of $\vec{\mathscr{E}}$ and $\vec{\mathscr{H}}$.

We then get a boundary value problem whose unknowns are the Fourier coefficients of the EM field inside the modulated region. According to the hypothesis $\partial/\partial z=0$, the solutions at frequency $\omega_3$ are either TM or TE polarized. Let us first consider the TM case.

### 1. TM case

In this case, $\vec{\mathscr{H}}(\omega_3,\vec{r})$ is directed along the $z$ axis:

$$\vec{\mathscr{H}}(\omega_3,\vec{r})=\mathscr{H}(\omega_3,\vec{r})\vec{e}_z \ . \tag{29}$$

According to Eqs. (27a), (27b), (28), and (29), $\mathscr{H}(\omega_3,\vec{r})$ fulfills the following equation:

$$\vec{\nabla}\cdot\left[\frac{1}{k^2(\omega_3,x,y)}\vec{\nabla}\mathscr{H}(\omega_3,\vec{r})\right]+\mathscr{H}(\omega_3,\vec{r}) =j\omega_3\left[\vec{\nabla}\times\left[\frac{\vec{\mathscr{P}}^{\,\mathrm{NL}}(\omega_3,\vec{r})}{k^2(\omega_3,x,y)}\right]\right]_z , \tag{30}$$

with

$$k^2(\omega_3,x,y)=\frac{\omega_3^2}{c^2}\epsilon(\omega_3,x,y)$$

and the subscript $z$ denotes the $z$ component of the curl operator.

It must be emphasized that (a) Eq. (30) is valid for any region, even in regions where $\epsilon$ and $\vec{\mathscr{P}}^{\,\mathrm{NL}}$ are $x$ and $y$ dependent, and (b) since Maxwell's equations are valid in the sense of generalized functions or distributions,[1] Eq. (30) is also valid in the sense of distributions. Thus, although the function $(1/k^2)\vec{\nabla}\mathscr{H}$ is discontinuous on the grating profile, its divergence is well defined. The same thing applies to $\vec{\nabla}\times(\vec{\mathscr{P}}^{\,\mathrm{NL}}/k^2)$. To solve this equation, we consider three regions: $y>\delta+e$, $y<0$, and $0<y<\delta+e$.

*a.* $y>\delta+e$. Equation (30) reduces to

$$\Delta\mathscr{H}(\omega_3,\vec{r})+k_1^2(\omega_3)\mathscr{H}(\omega_3,\vec{r})=0 \ .$$

Thus

$$\mathscr{H}(\omega_3,\vec{r})=\sum_{n=-\infty}^{+\infty}B_{n,3}^{\mathrm{TM}}e^{j(\alpha_{1,n,3}y+\gamma_{n,3}x)} \tag{31}$$

and

$$\alpha_{1,n,3}^2+\gamma_{n,3}^2=k_1^2(\omega_3) \ . \tag{32}$$

The constants $\gamma_{n,3}$ occurring in Eq. (32) will be derived later [see Eq. (40)].

*b.* $y<0$. Equation (30) becomes

$$\Delta\mathscr{H}(\omega_3,\vec{r})+k_2^2(\omega_3)\mathscr{H}(\omega_3,\vec{r}) =j\omega_3[\vec{\nabla}\times\vec{\mathscr{P}}^{\,\mathrm{NL}}(\omega_3,\vec{r})]_z \ .$$

In this equation, the nonlinear polarization is given by Eqs. (18).

The solution is now a superposition of plane waves (propagating and evanescent) and driven waves produced by the source term $j\omega_3[\vec{\nabla}\times\vec{\mathscr{P}}^{\,\mathrm{NL}}(\omega_3,\vec{r})]_z$.

$$\mathscr{H}(\omega_3,\vec{r})=\sum_{n=-\infty}^{+\infty}T_{n,3}^{\mathrm{TM}}e^{j(-\alpha_{2,n,3}y+\gamma_{n,3}x)} +e^{j\vec{K}\cdot\vec{r}}\sum_{p=-\infty}^{+\infty}A_{\mathrm{TM,p}}^{\mathrm{NL}}e^{jp\sigma x} \tag{33}$$

with

$$\alpha_{2,n,3}^2+\gamma_{n,3}^2=k_2^2(\omega_3) \ , \tag{34}$$

$$A_{\mathrm{TM,p}}^{\mathrm{NL}}=\sum_{l=-\infty}^{+\infty}-\omega_3\frac{(\vec{\kappa}_{l,p}\times\vec{\mathrm{P}}_{l,p-l}^{\mathrm{NL}})_z}{k_2^2(\omega_3)-\kappa_{l,p}^2} \times e^{j(\kappa_{l,p,y}-K_y)y} , \tag{35}$$

where the subscript $z$ denotes the $z$ component of the cross (vector) product.

*c.* $0<y<\delta+e$. In this region, since $\epsilon$ depends on $x$ and $y$, we have to deal with Eq. (30) whose solution must be looked for in the following form[1,7]:

$$\mathscr{H}(\omega_3,\vec{r})=\sum_{n=-\infty}^{+\infty}H_{n,3}(y)e^{j\gamma_{n,3}x} \ . \tag{36}$$

The determination of the Fourier coefficients $H_{n,3}(y)$ is done as follows: The periodicity of the modulated region with respect to $x$ implies the periodicity of $\epsilon(\omega_3,x,y)$, $\xi(\omega_3,x,y)=(\omega_3^2/c^2)\times\epsilon(\omega_3,x,y)$, and $\beta(\omega_3,x,y)=1/\xi(\omega_3,x,y)$. Thus $\xi(\omega_3,x,y)$ and $\beta(\omega_3,x,y)$ can be expanded into Fourier series:

$$\xi(\omega_3,x,y)=\sum_{n=-\infty}^{+\infty}\xi_{n,3}(y)e^{jn\sigma x} , \tag{37a}$$

$$\beta(\omega_3,x,y)=\sum_{n=-\infty}^{+\infty}\beta_{n,3}(y)e^{jn\sigma x} \ . \tag{37b}$$

It is convenient to introduce the functions

$$\tilde{K}_{n,3}(y)=\sum_{u=-\infty}^{+\infty}\beta_{n-u,3}(y)\left[\frac{dH_{u,3}(y)}{dy}+j\omega_3 P_{u,x}^{\mathrm{NL}}e^{jK_y y}\right], \tag{38}$$

where $P_{u,x}^{\mathrm{NL}}$ is given by Eq. (26b).

According to Eqs. (26a), (30), (37a), (37b), and (38), we get

$$\frac{dH_{n,3}(y)}{dy}=\sum_{u=-\infty}^{+\infty}\xi_{n-u,3}(y)\tilde{K}_{u,3}(y)-j\omega_3 P_{n,x}^{\mathrm{NL}}(y,\delta)e^{jK_y y}, \tag{39a}$$

$$\frac{d\tilde{K}_{n,3}(y)}{dy}=\gamma_{n,3}\sum_{u=-\infty}^{+\infty}\gamma_{u,3}\beta_{n-u,3}(y)H_{u,3}-H_{n,3}(y)-\omega_3\gamma_{n,3}\sum_{u=-\infty}^{+\infty}\beta_{n-u,3}(y)P_{u,y}^{\mathrm{NL}}(y,\delta)e^{jK_y y}, \tag{39b}$$

together with

$$\gamma_{n,3}=K_x+n\sigma . \tag{40}$$

The Fourier coefficients $H_{n,3}(y)$ not only fulfill Eqs. (39a) and (39b), but also the boundary conditions at the limits of the modulated region, i.e., the tangential components of the electric and magnetic fields must be continuous at $y=0$ and $y=\delta+e$.

At $y=0$, from the continuity of $\mathscr{H}(\omega_3,\vec{r})$, we get

$$T_{n,3}^{\mathrm{TM}}+A_{\mathrm{TM},p}^{\mathrm{NL}}(0)\delta_{n,p}=H_{n,3}(0), \tag{41a}$$

where $\delta_{n,p}$ is the Kronecker symbol. The continuity of $\mathscr{E}_x(\omega_3,\vec{r})$ leads to

$$-j\alpha_{2,n,3}T_{n,3}^{\mathrm{TM}}+jK_yA_{\mathrm{TM},p}^{\mathrm{NL}}(0)\delta_{n,p}=\left.\frac{dH_{n,3}(y)}{dy}\right|_{y=0}. \tag{41b}$$

Similarly, at $y=\delta+e$, we get

$$B_{n,3}^{\mathrm{TM}}e^{j\alpha_{1,n,3}(\delta+e)}=H_{n,3}(\delta+e), \tag{42a}$$

$$j\alpha_{1,n,3}B_{n,3}^{\mathrm{TM}}e^{j\alpha_{1,n,3}(\delta+e)}=\left.\frac{dH_{n,3}(y)}{dy}\right|_{y=\delta+e}. \tag{42b}$$

Equations (41a) and (41b) on the one hand, and Eqs. (42a) and (42b) on the other hand lead to

$$\left.\frac{dH_{n,3}(y)}{dy}\right|_{y=0}+j\alpha_{2,n,3}H_{n,3}(0)=j(\alpha_{2,n,3}+K_y)A_{\mathrm{TM},p}^{\mathrm{NL}}(0)\delta_{n,p}, \tag{43a}$$

$$\left.\frac{dH_{n,3}(y)}{dy}\right|_{y=\delta+e}-j\alpha_{1,n,3}H_{n,3}(\delta+e)=0 . \tag{43b}$$

Equations (39a), (39b), (43a), and (43b) constitute the TM boundary value problem (BVP) whose unknowns are the Fourier coefficients $H_{n,3}(y)$. The numerical determination of these Fourier coefficients allows knowing the Rayleigh coefficients $T_{n,3}^{\mathrm{TM}}$ and $B_{n,3}^{\mathrm{TM}}$ via Eqs. (41a) and (42a). We then know the TM nonlinear diffracted EM field at frequency $\omega_3=\omega_1+\omega_2$ everywhere, not only outside the modulated region, but also inside it.

Before considering the solution of the TM BVP, let us deal with the TE case at frequency $\omega_3=\omega_1+\omega_2$.

### 2. TE case

The method is similar to that of the TM case. Now $\vec{\mathscr{E}}(\omega_3,\vec{r})$ is directed along the $z$ axis:

$$\vec{\mathscr{E}}(\omega_3,\vec{r})=\mathscr{E}(\omega_3,\vec{r})\vec{e}_z . \tag{44}$$

Thus $\vec{\nabla}\cdot\vec{\mathscr{E}}=0$ (remember $\partial/\partial z=0$) and, from Eqs. (27a), (27b), (28), and (44), we get in the following different regions:

*a. $y>\delta+e$. We have*

$$\Delta\mathscr{E}(\omega_3,\vec{r})+k_1^2(\omega_3)\mathscr{E}(\omega_3,\vec{r})=0 .$$

Thus

$$\mathscr{E}(\omega_3,\vec{r})=\sum_{n=-\infty}^{+\infty}B_{n,3}^{\mathrm{TE}}e^{j(\alpha_{1,n,3}y+\gamma_{n,3}x)} . \tag{45}$$

*b. $y<0$. We have*

$$\Delta\mathscr{E}(\omega_3,\vec{r})+k_2^2(\omega_3)\mathscr{E}(\omega_3,\vec{r})=-\frac{\omega_3^2}{\epsilon_0c^2}\mathscr{P}_z^{\mathrm{NL}}(\omega_3,\vec{r})$$

with $\mathscr{P}_z^{\mathrm{NL}}(\omega_3,\vec{r})$ derived from Eqs. (18). We get

$$\mathscr{E}(\omega_3,\vec{r})=\sum_{n=-\infty}^{+\infty}T_{n,3}^{\mathrm{TE}}e^{j(-\alpha_{2,n,3}y+\gamma_{n,3}x)}+e^{j\vec{K}\cdot\vec{r}}\sum_{p=-\infty}^{+\infty}A_{\mathrm{TE},p}^{\mathrm{NL}}e^{jp\sigma x}, \tag{46}$$

where

$$A_{\mathrm{TE},p}^{\mathrm{NL}}=-\frac{\omega_3^2}{\epsilon_0c^2}\sum_{l=-\infty}^{+\infty}\frac{\mathrm{p}_{l,p-l,z}^{\mathrm{NL}}}{k_2^2(\omega_3)-\kappa_{l,p}^2}\times e^{j(\kappa_{l,p,y}-K_y)y} . \tag{47}$$

*c. $0<y<\delta+e$.* In this region, the propagation equation takes the form

$$\Delta\mathscr{E}(\omega_3,\vec{r})+k^2(\omega_3,x,y)\mathscr{E}(\omega_3,\vec{r})$$
$$=-\frac{\omega_3^2}{\epsilon_0 c^2}\mathscr{P}_z^{\rm NL}(\omega_3,\vec{r})$$

with $\mathscr{P}_z^{\rm NL}(\omega_3,\vec{r})$ derived from Eq. (26a).

We have to look for the solution of the form

$$\mathscr{E}(\omega_3,\vec{r})=\sum_{n=-\infty}^{+\infty}E_{n,3}(y)e^{j\gamma_{n,3}x}\ . \tag{48}$$

Then the Fourier coefficients $E_{n,3}(y)$ fulfill

$$\frac{d^2E_{n,3}}{dy^2}-\gamma_{n,3}^2E_{n,3}+\sum_{u=\infty}^{+\infty}\xi_{n-u,3}(y)E_{u,3}$$
$$=-\frac{\omega_3^2}{\epsilon_0 c^2}P_{n,z}^{\rm NL}e^{jK_y y}\ . \tag{49}$$

The continuity of $\mathscr{E}_z$ and $\mathscr{H}_x$ at $y=0$ and $y=\delta+e$ leads to the following set of equations:

$$T_{n,3}^{\rm TE}+A_{{\rm TE},p}^{\rm NL}(0)\delta_{n,p}=E_{n,3}(0)\ , \tag{50a}$$

$$-j\alpha_{2,n,3}T_{n,3}^{\rm TE}+jK_yA_{{\rm TE},p}^{\rm NL}(0)\delta_{n,p}=\left.\frac{dE_{n,3}(y)}{dy}\right|_{y=0}, \tag{50b}$$

$$B_{n,3}^{\rm TE}e^{j\alpha_{1,n,3}(\delta+e)}=E_{n,3}(\delta+e)\ , \tag{50c}$$

$$j\alpha_{1,n,3}B_{n,3}^{\rm TE}e^{j\alpha_{1,n,3}(\delta+e)}=\left.\frac{dE_{n,3}(y)}{dy}\right|_{y=\delta+e}. \tag{50d}$$

From Eqs. (50a)–(50d), we get

$$\left.\frac{dE_{n,3}}{dy}\right|_{y=0}+j\alpha_{2,n,3}E_{n,3}(0)$$
$$=j(\alpha_{2,n,3}+K_y)A_{{\rm TE},p}^{\rm NL}(0)\delta_{n,p}\ , \tag{51a}$$

$$\left.\frac{dE_{n,3}}{dy}\right|_{y=\delta+e}-j\alpha_{1,n,3}E_{n,3}(\delta+e)=0\ . \tag{51b}$$

Equations (49), (51a), and (51b) constitute the TE BVP whose unknowns are the Fourier coefficients $E_{n,3}(y)$.

### 3. *Solution of the BVP in the TM and TE cases*

From Eqs. (36) and (48), the magnetic field in the TM case, the electric field in the TE case, can be written as

$$\sum_{n=-\infty}^{+\infty}\varphi_n(y)e^{j\gamma_{n,3}x}$$

with

$$\varphi_n(y)=\begin{cases}H_{n,3}(y)\\ E_{n,3}(y)\end{cases}$$

for the TM and TE cases, respectively.

According to the discussion of Secs. II C 1 and II C 2, the functions $\varphi_n(y)$ are solutions of the following set of equations which constitute the BVP common to the TM and TE cases:

$$\left.\frac{d\varphi_n}{dy}\right|_{y=0}+j\alpha_{2,n,3}\varphi_n(0)=j(\alpha_{2,n,3}+K_y)A_p^{\rm NL}(0)\delta_{n,p}\ , \tag{52a}$$

$$\left.\frac{d\varphi_n}{dy}\right|_{y=\delta+e}-j\alpha_{1,n,3}\varphi_n(\delta+e)=0\ , \tag{52b}$$

together with Eqs. (39a) and (39b) in the TM case and with Eq. (49) for the TE case.

In Eq. (52a), $A_p^{\rm NL}(0)=A_{{\rm TM},p}^{\rm NL}(0)$ in the TM case and $A_p^{\rm NL}(0)=A_{{\rm TE},p}^{\rm NL}(0)$ in the TE one.

In order to get the solution of this new BVP, let us consider the following expansion:

$$[\Phi(y)]=\sum_{t=-\infty}^{+\infty}C_t[\tilde{\Phi}_t(y)]+[\Phi^{\rm NL}(y)]\ . \tag{53}$$

$[\Phi(y)]$ is a column matrix with elements $\varphi_n(y)$; $[\tilde{\Phi}_t(y)]$ is an infinite set of particular column matrices with elements $\tilde{\varphi}_{n,t}(y)$ which fulfill Eq. (52b) and are solution of equations derived from Eqs. (39a) and (39b) for TM waves, and Eqs. (49) for TE waves, by putting $\vec{\mathscr{P}}^{\rm NL}=0$ (which leads to $\vec{P}_u^{\rm NL}=0$, $\vec{P}_n^{\rm NL}=0$); $[\Phi^{\rm NL}(y)]$ is a column matrix with elements $\varphi_n^{\rm NL}(y)$ which fulfill Eq. (52b) and is a particular solution of Eqs. (39a) and (39b) in the TM case and of Eq. (49) in the TE case.

By construction the column matrix $[\Phi(y)]$ defined by Eq. (53) fulfills Eqs. (39a) and (39b) in the TM case, Eq. (49) in the TE one, together with Eq. (52b). Thus $[\Phi(y)]$ will be the solution of the BVP common to the TM and TE cases provided the unknown coefficients $C_t$ are determined in such a way as to fulfill Eq. (52a). If $[C]$ is a column matrix with elements $C_t$, this implies

$$[M][C]=[G]$$

or

$$[C]=[M]^{-1}[G]\ ,$$

where $[M]$ is a square matrix with elements

$$\left.\frac{d\tilde{\varphi}_{n,t}}{dy}\right|_{y=0}+j\alpha_{2,n,3}\tilde{\varphi}_{n,t}(0)$$

and $[G]$ is a column matrix with elements

$$j(\alpha_{2,n,3}+K_y)A_p^{\rm NL}(0)\delta_{n,p} - \left[ \left. \frac{d\varphi_n^{\rm NL}(y)}{dy} \right|_{y=0} + j\alpha_{2,n,3}\varphi_n^{\rm NL}(0) \right] .$$

The Rayleigh coefficients $B_{n,3}^{\rm TM}$ and $T_{n,3}^{\rm TM}$ are derived from Eqs. (41a) and (42a), whereas $B_{n,3}^{\rm TE}$ and $T_{n,3}^{\rm TE}$ are obtained from Eqs. (50a) and (50c). Thus the EM field is determined everywhere.

Notice the following:

*a. For the TM case,* matrix [*M*] is the same as in I since it is constituted by an infinite set of particular column matrices whose elements are solutions of Eq. (52b) and (39a) and (39b) in which $\vec{\mathscr{P}}^{\rm NL}=0$; matrix [*G*] is different from that computed in I. Indeed, because of the diffraction of the pump beams, we deal with a different nonlinear polarization at frequency $\omega_3$.

*b. For the TE case,* the elements $\tilde{\varphi}_{n,t}(y)$ and $\varphi_n^{\rm NL}(y)$ are different from those corresponding to the TM case since the basic propagation equations are different.

This method allows us to determine the EM field at frequency $\omega_3=\omega_1+\omega_2$ outside, but also *inside*, the modulated region whatever the polarization of the $\omega_1$, $\omega_2$, and $\omega_3$ beams may be.

This is achieved with the aid of a computational method which is a generalization of that used in I. Corresponding numerical results will be given in a forthcoming publication. They will fully include the frequency dependence of $\epsilon$, $\chi_{h,i,j}$, $\tilde{a}$, and $\tilde{b}$. Effects considered in this study are such that no wave-vector dependence of these quantities occurs. This is the usual situation in diffraction theory.[1,6,7,10,11]

Before taking up the next section, let us say a few words about the convergence of the series occurring in this formalism. Computer calculations valid in the case of linear diffraction[1,10,11] (i.e., concerning expansions of the fields at frequencies $\omega_1$ and $\omega_2$) have shown that the pump fields are represented with an accuracy at least equal to 1% when 15 terms are taken into account (from $l=-7$ to $l=+7$). Early computer results show that the rapidity of convergence is about the same for the expansions of the EM field at frequency $\omega_3$.

### *4. Description of the nonlinear diffracted EM field at frequency $\omega_3=\omega_1+\omega_2$ outside the modulated region*

The pertinent equations are Eqs. (31)–(35), (40), and (45)–(47). We have to consider separately the regions $y>\delta+e$ and $y<0$.

*a.* $y>\delta+e$. The transverse variation of each diffracted order at frequency $\omega_3$ depends on the sign of $\alpha_{1,n,3}^2$. The diffracted orders for which $\alpha_{1,n,3}^2<0$ ($\gamma_{n,3}>k_1(\omega_3)$) are evanescent above the modulated region.

Note that for sufficiently large $K_x$ ($K_x>k_1(\omega_3)$) even the diffracted order $n=0$ may be evanescent. This is a specific feature of nonlinear diffraction. As is well known, in linear diffraction the specularly reflected wave (corresponding to $l=0$) is *always* radiated.

The diffracted orders $n$ for which $\alpha_{1,n,3}^2>0$ ($\gamma_{n,3}<k_1(\omega_3)$) are radiated in the outside medium $y>\delta+e$.

*b.* $y<0$. Below the modulated region, the EM field at frequency $\omega_3$ is a superposition of a diffracted field and of a driven field. The diffracted field is described by the first term of Eqs. (33) or (46).

If $\mathrm{Re}[\epsilon_2(\omega_3)]<0$, all the diffracted orders are evanescent in the region $y<0$. In this case, the orders $n$ for which $\alpha_{1,n,3}^2<0$, correspond to diffracted orders which remain bounded to the modulated region, i.e., these orders $n$ correspond to surface waves; the other diffracted orders correspond to radiated waves.

If $\mathrm{Re}[\epsilon_2(\omega_3)]>0$, one has to compare $\gamma_{n,3}$ with $k_2(\omega_3)$. Diffracted orders $n$ for which $\gamma_{n,3}>k_2(\omega_3)$ are evanescent in the region $y<0$, whereas those for which $\gamma_{n,3}<k_2(\omega_3)$ are radiated in this medium.

The driven field is a superposition of an infinite number of subelementary driven fields, each of them having a wave vector $\vec{\kappa}_{l,p}$. The longitudinal component of $\vec{\kappa}_{l,p}$ is always real and, as a consequence, all the subelementary driven fields, regardless of the indices $l$ and $m$, propagate along the $x$ axis. The transverse variation of each of these driven fields depends on the nature, real or imaginary, of $\alpha_{2,l,1}$ and $\alpha_{2,p-l,2}$. The results are summarized in Table I.

In fact, due to the damping of the nonlinear medium at frequencies $\omega_1$ and $\omega_2$, $\alpha_{2,l,1}$ and $\alpha_{2,p-l,2}$ are complex. But, since this damping is usually small, $\alpha_{2,l,1}$ and $\alpha_{2,p-l,2}$ are either almost real or almost imaginary. This justifies the classification of Table I.

For metallic gratings $A_{{\rm TM},p}^{\rm NL}=A_{{\rm TE},p}^{\rm NL}=0$. For these gratings there is no driven field in the TE case.

The guided EM waves at frequency $\omega_3$ (surface plasmons, surface polaritons, guided waves) must be looked for in the diffracted field at that frequency. They correspond to diffracted orders $n$ which are evanescent both above and below the modulated region. For example, we have the following: surface waves for a bare grating with $\mathrm{Re}[\epsilon_2(\omega_3)]<0$ and orders $n$ such that $\alpha_{1,n,3}^2<0$; guided waves when $\mathrm{Re}[\epsilon_2(\omega_3)]\gtrless 0$ together with a high index coating, i.e., whose permittivity $\epsilon_c(\omega_3)$ fulfills $\mathrm{Re}[\epsilon_c(\omega_3)]>\mathrm{Re}[\epsilon_b(\omega_3)]$ ($b=1,2$), and $n$ such that $\alpha_{b,n,3}^2<0$. These guided diffracted orders at fre-

TABLE I. Discussion of the nature of the driven fields.

| $\alpha_{2,l,1}$ | $\alpha_{2,p-l,2}$ | Characteristic of an $(l,m)$ subelementary driven field |
|---|---|---|
| Re | Re | Propagates in the bulk in the direction $\kappa_{l,p}$ |
| Re | Im | Propagates in the direction $(\kappa_{l,p,x},\alpha_{2,l,1})$. Evanescent in the $y$ direction. Attenuation length: $(\alpha_{2,p-l,2})^{-1}$ |
| Im | Re | Propagates in the direction $(\kappa_{l,p,x},\alpha_{2,p-l,2})$. Evanescent in the $y$ direction. Attenuation length: $(\alpha_{2,l,1})^{-1}$. |
| Im | Im | Propagates in the $x$ direction. Evanescent in the $y$ direction. Attenuation length: $(\alpha_{2,l,1}+\alpha_{2,p-l,2})^{-1}$. |

quency $\omega_3$ can be resonantly excited for suitable values of $\theta_1,\theta_2,d,\omega_3,n$ [see Eqs. (32), (34), and (40)]. In fact, due to the damping of the nonlinear medium $\epsilon_2(\omega_3)$ is complex and these guided waves do not propagate strictly parallel to the $x$ axis, but this does not change significantly the results stated above.

*c. Direction of propagation of the nonlinear diffracted orders at frequency* $\omega_3=\omega_1+\omega_2$. As already pointed out, the diffracted field contains propagating and evanescent waves. The direction of propagation of the radiated waves can be deduced from Eqs. (32) and (40) for the medium $y>\delta+e$ and from Eqs. (34) and (40) for the medium $y<0$.

We get the following result:

$$\frac{n_b(\lambda_3)}{\lambda_3}\sin\psi_{b,n}=\frac{n_1(\lambda_1)}{\lambda_1}\sin\theta_1+\frac{n_1(\lambda_2)}{\lambda_2}\sin\theta_2+\frac{n}{d} \qquad (54)$$

with $n=0,\pm1,\pm2,\ldots$, $n_b(\lambda_\nu)$ $(b=1,2,\ \nu=1,2,3)$ is the refractive index of medium $b$ at wavelength $\lambda_\nu$ $(\lambda_\nu=2\pi c/\omega_\nu)$, and $\psi_{b,n}$ is the diffraction angle of the diffracted order $n$ at wavelength $\lambda_3$ in medium $b$ $(b=1,2)$.

Equation (54) is very important since it represents the nonlinear grating equation. This equation allows determining which diffracted orders $n$ (at the signal frequency $\omega_3$) are radiated in medium $b$ $(b=1,2)$ (by $|\sin\psi_{b,n}|<1$). For these orders, their direction of propagation is given by $\psi_{b,n}$.

Equation (54) expresses the conservation of the longitudinal component of the total impulsion of the system constituted by the two incident photons, the signal photon and the grating through the nonlinear interaction $(\omega_1,\omega_2)\rightarrow\omega_3=\omega_1+\omega_2$.

The nonlinear grating equation is valid even for dispersive media. Notice that the nonlinear diffraction angles $\psi_{b,n}$ are different from the diffraction angles corresponding to the linear diffraction of the pump beams. Thus this novel grating effect allows separating pump fields and signal.

The geometrical construction, which leads to the determination of the direction of propagation of the radiated diffracted orders at frequency $\omega_3$, is deduced from Eq. (54) in the following way (Fig. 3): Draw two circles $C_1,C_2$ and two half circles $C_3,C_4$ with radius, respectively, equal to $n_1(\lambda_1)/\lambda_1$, $n_1(\lambda_2)/\lambda_2$, $n_1(\lambda_3)/\lambda_3$, $n_2(\lambda_3)/\lambda_3$; add the lengths $0A_2$ to $0A_1$ to get $0B_0$. Then, $0B_n$

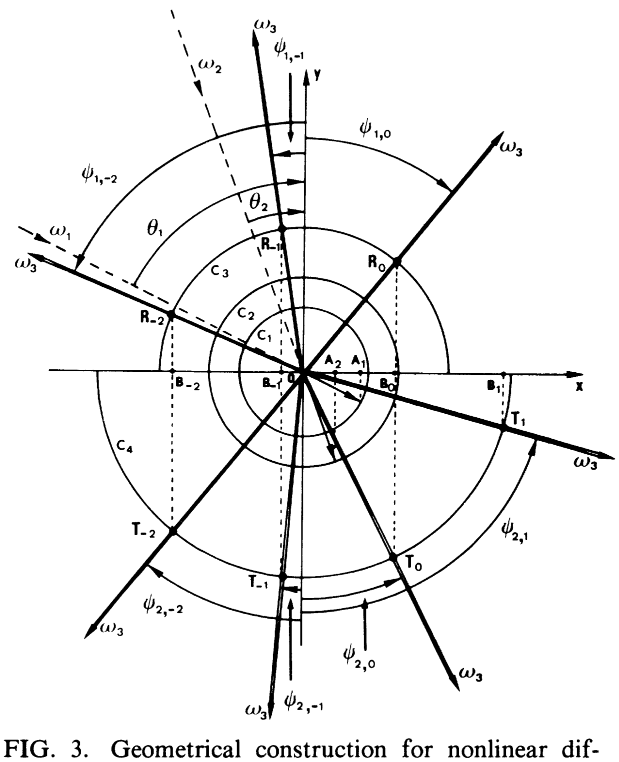


FIG. 3. Geometrical construction for nonlinear diffraction when the lower medium is a nonlinear dielectric. Note that the $n=1$ diffracted order at $\omega_3$ is radiated only in medium 2; it is evanescent inside medium 1. All the diffracted orders $n\geq2$ and $n\leq-3$ are evanescent in media 1 and 2. $B_{-2}B_{-1}=B_{-1}B_0=B_0B_1=1/d$. With the arbitrary numerical values corresponding to the figure, the only propagating orders at the pump frequencies $\omega_1$ and $\omega_2$ are the orders 0 and $-1$ (not reported on the figure).

($n = 0, \pm 1, \pm 2, \ldots$) is the longitudinal wave-vector component divided by $2\pi$, of the $n$th diffracted order. The direction of propagation of the $n$th reflected (transmitted) nonlinear diffracted order in $0R_n$ ($0T_n$).

Circle $C_4$ exists only in the case of a nonlinear dielectric. When dealing with a nonlinear metallic grating there is no circle $C_4$: All the nonlinear diffracted orders are evanescent in region $y < 0$. When $\theta_1, \theta_2, \lambda_1, \lambda_2$ are such that

$$\frac{n_1(\lambda_1)}{\lambda_1}\sin\theta_1 + \frac{n_1(\lambda_2)}{\lambda_2}\sin\theta_2 > \frac{n_b(\lambda_3)}{\lambda_3} .$$

Then, even the $n=0$ nonlinear diffracted order is evanescent in medium $b$ ($b=1,2$).

This case, which has been already considered for region $b=1$, corresponds to $K_x > k_b(\omega_3)$ ($b=1,2$). It is of special interest since, in medium $b=1$, it corresponds to the nonlinear specular reflection.

When $n=0$, the nonlinear grating equation, Eq. (54), becomes

$$n_b(\lambda_3)\left[\frac{1}{\lambda_1}+\frac{1}{\lambda_2}\right]\sin\psi_{b,0} = \frac{n_1(\lambda_1)}{\lambda_1}\sin\theta_1 + \frac{n_1(\lambda_2)}{\lambda_2}\sin\theta_2 . \tag{55a}$$

Equation (55a) determines the direction of nonlinear reflection when ($b=1$), or nonlinear transmission ($b=2$), in the case of a plane interface[8] separating two media with permittivity $\epsilon_1, \epsilon_2$. It is worth noticing that only the direction of propagation, and not the intensity, of the $n=0$ diffracted order is the same as that deduced from the theory of nonlinear reflection.[8] From Eq. (55a), it is seen that $\psi_{1,0}$ is different from $\theta_1$ and $\theta_2$ when the upper medium is dispersive.

If $n_1(\lambda_3) = n_1(\lambda_2) = n_1(\lambda_1)$, we get

$$\left[\frac{1}{\lambda_1}+\frac{1}{\lambda_2}\right]\sin\psi_{1,0} = \frac{1}{\lambda_1}\sin\theta_1 + \frac{1}{\lambda_2}\sin\theta_2 . \tag{55b}$$

If in addition $\theta_1 = \theta_2$, Eq. (55b) shows that $\psi_{1,0} = \theta_1 = \theta_2$, i.e., the nonlinear specular reflected beam and the specular reflected pump beams merge together. Figure 4 represents a possible aspect of the EM field at frequency $\omega_3$ in the case of SHG in a silver grating.

### *5. Summary of the EM theory of diffraction in nonlinear optics*

The formalism developed in the previous sections corresponds to a three-step theory of nonlinear diffraction (Fig. 5):

(1) The diffraction of the two pump beams is accounted for by using the rigorous linear theory of gratings: This allows getting the expression of the electric field of the diffracted pump beams below and inside the modulated region.

(2) We then derive the expression of the nonlinear polarization at frequency $\omega_3 = \omega_1 + \omega_2$, not only below the grating but also inside the modulated region, in terms of all the diffracted orders at frequencies $\omega_1$ and $\omega_2$, whatever their nature, radiated or evanescent, may be.

(3) The expressions (18a) and (26a) of the nonlinear polarization at frequency $\omega_3$ then act as source terms in the nonlinear diffraction problem at frequency $\omega_3$.

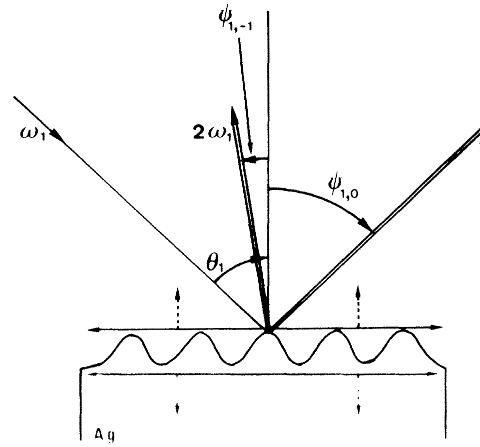


FIG. 4. SHG in the bare grating of Figs. 2(a) and 2(b): $\omega_2 = \omega_1$ $\lambda_2 = \lambda_1 = 1.06$ $\mu$m; $\theta_2 = \theta_1 = 45°$; $n_1(\lambda_1) = n_1(\lambda_3) = 1$. In this case, the nonlinear grating equation, Eq. (40), reduces to $(d/\lambda_3)(\sin\psi_{n,3} - \sin\theta_1) = n$ ($\lambda_3 = \lambda_1/2$). The specularly second-harmonic reflected light ($n=0$) is emitted in the direction $\psi_{1,0} = \theta_1$. Aside from this order, only the $n=-1$ second-harmonic diffracted order is propagating, with a back-diffracted angle $\psi_{1,-1} = -8.63°$. Concerning the diffraction of the fundamental light, $\omega_1$, only the $l=0$ (specular reflection) is radiated in direction $\theta_1$. ——; incident pump beam at $\omega_1$; ═══; radiated diffracted orders at $\omega_3 = 2\omega_1$; - - -; evanescent diffracted orders at $\omega_3 = 2\omega_1$. Since the grating material is a metal all the transmitted diffracted orders are evanescent.

### *6. Comparison with the results of paper I*

Contrary to what occurs in I, the nonlinear polarization at frequency $\omega_3$ depends on the groove depth $\delta$. This allows us to account for an enhancement of the $\omega_3$ EM field through electromagnetic resonance at the pump frequencies.

Among these EM resonances is the well-known surface-plasmon or surface-polariton resonance. But, the theory of nonlinear diffraction presented

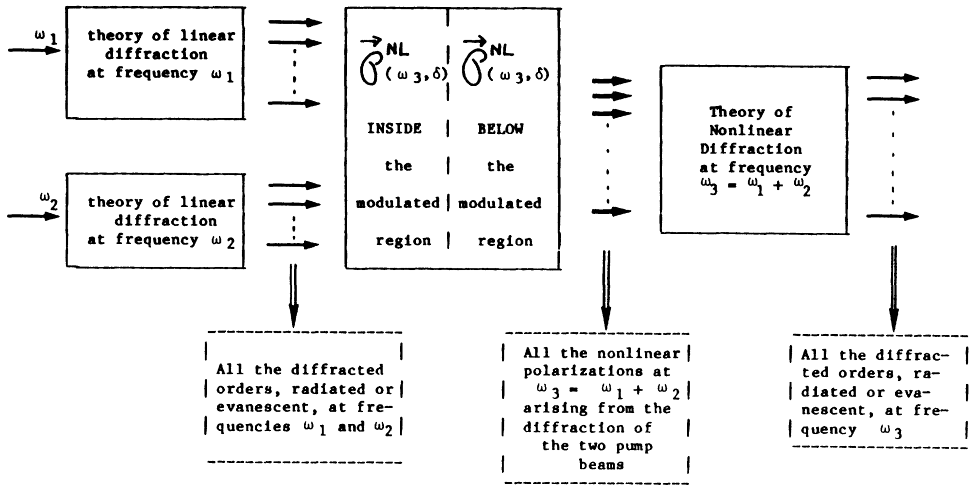


FIG. 5. Successive steps of the theory of diffraction in nonlinear optics. $\mathscr{P}^{NL}(\omega_3,\delta)$: nonlinear polarization at frequency $\omega_3=\omega_1+\omega_2$.

here, since it also applies to coated gratings, allows us to predict that nonlinear effects can be enhanced by using, at least, another kind of EM resonance, i.e., the guided wave one.

This is demonstrated in Fig. 6 where we consider SHG in a silver grating ($d=5556$ Å, $\delta=150$ Å) coated by a dielectric layer of thickness $e$; the pump wave $\omega_1$ is TE polarized. In that case, the optimal thickness, $e_{opt}$ is equal to 5700 Å and $|u_{1,1,z}e^{j\alpha_{1,1,1}(\delta+e)}|^2_{MM}=186$ (MM denotes *maximum maximorum*). Consequently, the square modulus of the nonlinear polarization at $2\omega_1$ is increased by a factor of the order of 2100 as compared

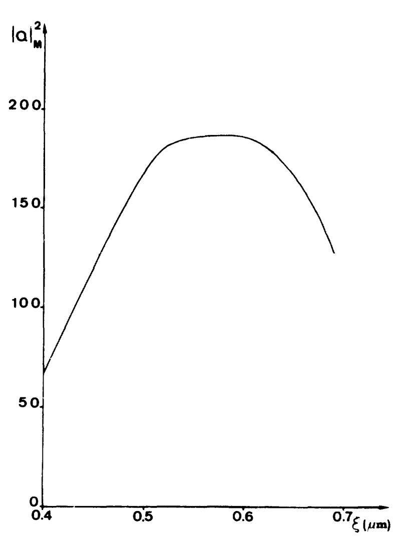


FIG. 6. Peak value, $|Q|^2_M$, of $Q=|u_{1,1,z}e^{j\alpha_{1,1,1}(\delta+e)}|^2$, as a function of the thickness $e$ of the dielectric layer. $\lambda_1=1.06$ μm, index of the layer, 1.49, $d=5556$ Å, $\delta=150$ Å.

to the case where a TM incident beam of frequency $\omega_1$ acts as a pump for SHG in a bare Ag medium with flat interface.[11]

This result has to be compared with the enhancement factor corresponding to a bare silver grating (i.e., the situation of Fig. 2) with $d=5556$ Å. Computer calculations show that $\delta_{opt}=114$ Å and $|T_{1,1}|^2_{MM}=67.05$ leading to an enhancement factor of the square modulus of the nonlinear polarization at frequency $2\omega_1$ of the order of 300 (the comparison is still performed with a flat air-Ag interface).

Thus the guided wave resonance leads to a stronger enhancement than the surface plasmon-polariton does. With the numerical values of Fig. 6, the enhancement factor corresponding to the guided-wave resonance is 7 times greater than that due to the surface-plasmon resonance.

We also see that the surface-plasmon resonance contribution depends on $d$. Indeed, in Fig. 2(b), $d=6174$ Å, $\delta_{opt}=150$ Å, and $|T_{1,1}|^2_{MM}=110$ whereas for a bare silver grating for which $d=5556$ Å, $\delta=\delta_{opt}=114$ Å, we get $|T_{1,1}|^2_{MM}=67.05$. This result has been already pointed out in Ref. 12 where it has been shown that, in the low modulation range, the highest modulation $\delta/d$ leads to the strongest EM resonance.

This guided-wave resonance at frequencies $\omega_1$ and $\omega_2$ is associated with guided EM waves in a dielectric wave guide (the associated smooth structure is a high index coated plane interface). Consequently, most of the pump energy is stored into the dielectric layer. If, in addition, this layer is constituted by a nonlinear medium, then, with regard to the nonlinear efficiency at frequency $\omega_3$, guided-wave resonance should lead to a greater enhancement of nonlinear optical effects than does surface-plasmon–polariton resonance. Indeed, the enhancement due to the guided-wave resonance is stronger than that corresponding to the surface-plasmon one, and in the case of bare metallic gratings most of the pump fields lie above the grating, i.e., in the *linear* region.

One may wonder if surface-plasmon–polariton resonance can occur simultaneously at the three frequencies $\omega_1$, $\omega_2$, and $\omega_3$. In this case, the EM resonance contribution to the enhancement of the signal at frequency $\omega_3$ would be the greatest.

If this happens, one has

$$\gamma_{l,1}=\beta_S(\omega_1)\ ,$$

$$\gamma_{m,2}=\beta_S(\omega_2)\ ,$$

$$\gamma_{n,3}=\beta_S(\omega_3)\ ,$$

where $\beta_S(\omega_\nu)$ is the surface-plasmon–polariton dispersion relation at frequency $\omega_\nu$ ($\nu=1,2,3$) corresponding to a flat interface $\epsilon_1,\epsilon_2$.

For the sake of simplicity, we neglect the small depression of the surface-plasmon dispersion curve when $\delta$ is different from zero (see Refs. 7 and 10).

From Eqs. (3c), (3d), (16), and (40), we get

$$k_1(\omega_1)\sin\Theta_{l,1}+l\sigma=\beta_S(\omega_1)\ ,$$

$$k_1(\omega_2)\sin\Theta_{m,2}+m\sigma=\beta_S(\omega_2)\ ,$$

$$k_1(\omega_1)\sin\Theta_{l,1}+k_1(\omega_2)\sin\Theta_{m,2}+n\sigma=\beta_S(\omega_3)\ .$$

Thus

$$\beta_S(\omega_1)+\beta_S(\omega_2)+q\sigma=\beta_S(\omega_1+\omega_2)\ . \tag{56}$$

From this equation, we see that if the law $\beta_S(\omega)$ is linear (for example, metal when $\omega \ll \omega_p$, where $\omega_p$ is the plasma frequency), then it is the Fourier component $q=0$ of $\mathscr{V}_{p,h}(x,y)$ which leads to the possibility of a simultaneous resonant excitation of the diffracted orders $l$ at $\omega_1$, $m$ at $\omega_2$, and $l+m$ at $\omega_3$.

If $\beta_S(\omega)$ is not a linear function of $\omega$ (which is usually the case), then, for a given integer value of $q$, there exists a value of $d$ such that Eq. (56) is fulfilled. In that case, the diffracted orders $l$ at $\omega_1$, $m$ at $\omega_2$, and $l+m+q$ at $\omega_3$ are simultaneously resonantly excited.

It is worth noticing that the EM resonance at frequencies $\omega_1$ and $\omega_2$ leads to an increase of the nonlinear polarization at frequency $\omega_3$ and, therefore, to an enhancement of the *whole* EM field at frequency $\omega_3$ (radiated diffracted orders included).

## III. CONCLUSION

We have presented in this paper the first rigorous electromagnetic theory of diffraction in nonlinear optics. This formalism is quite general. It applies to any type of gratings, whatever the profile and the groove depth of the grating and coatings may be. The coatings themselves can be made with either a linear or nonlinear material.

This new theory has a wide range of applications. When no EM resonance takes place at $\omega_\nu$ ($\nu=1,2,3$), we are concerned with what we may call the usual nonlinear diffraction. When $\omega_2=-\omega_1$ ($\omega_2=0$, $\omega_1\neq 0$), we deal with optical rectification (Pockels effect) in a nonlinear medium whose entrance face is constituted by a periodic rough surface. When $\omega_\nu$ ($\nu=1$ and/or 2 and/or 3) is chosen such that $\mathrm{Re}[\epsilon_2(\omega_\nu)]<0$, we study the surface-plasmon–polariton resonance contribution, at these frequencies, to Raman effect if $\omega_1>0$, $\omega_2<0$, and to SHG if $\omega_1=\omega_2$. In the case of a coated grating, we evaluate the guided wave resonance contribution to enhanced nonlinear effects.

Two important and new results must be emphasized:

(1) Each time the associated smooth structure supports guided normal modes, then, through a modulation of this structure, the possible EM resonances will lead to the enhancement of nonlinear effects taking place in this structure. In other words, depending on the structure which is used, the enhancement of nonlinear effects can be achieved by using surface plasmon-polariton or guided waves. Thus the surface-plasmon resonance is not the only one and, as pointed out previously, probably not the more suitable EM resonance to be used when one is interested in enhanced nonlinear effects.

(2) The contribution of these EM resonances to the enhancement of nonlinear effects can be *optimized* when $\delta=\delta_{\mathrm{opt}}$. It is worth noticing that this optimization is achieved for low modulation, i.e., $\delta_{\mathrm{opt}}/d \simeq 0.02$.

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