Second-harmonic generation in reflection from a metallic grating

G. A. Farias* and A. A. Maradudin
Department of Physics, University of California, Irvine, California 92717
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We present a nonperturbative theory of second-harmonic generation in reflection from a metallic grating. p-polarized light of frequency ω is assumed to be incident on the grating surface from the vacuum side, with the plane of incidence perpendicular to the grooves of the grating. We employ the method of reduced Rayleigh equations to solve Maxwell's equations in the presence of a nonlinear polarization and calculate the diffracted field at the frequency 2ω. In the vicinity of the frequency where the incident photon couples to surface polaritons on the grating we obtain an enhancement of the second-harmonic intensity by a factor of $\sim 10^4$ over its value for a flat surface. The effect of the surface current density in the nonlinear polarization on the enhancement is analyzed. Numerical results are presented for a grating of silver.

I. INTRODUCTION

The second-harmonic generation (SHG) of light in reflection from a metal surface[1,2] has attracted considerable interest recently due to the experimental discovery by Chen *et al.*[3] that SHG at a silver-air interface can be enhanced by a factor of $\sim 10^4$ by roughening the surface. The roughness-induced excitation of a surface polariton by the incident light resonantly enhances the magnitude of the nonlinear source term in Maxwell's equations responsible for the reflected light at twice the frequency of the incident light, and leads to the effect observed by Chen *et al.*[3]

A quantitative theory of this effect is difficult to construct because a randomly rough-surface profile is very difficult to treat, except in the small-roughness limit. However, the deterministic, periodic surface profile provided by a one-dimensional grating can be used as a model for a randomly rough surface.[4] Such a surface also allows the incident light to excite surface polaritons and thereby enhance the intensity of second-harmonic generation in reflection. Recently, using perturbation theory, Agarwal and Jha[5] calculated the enhancement of the SHG in reflection from a metal grating surface.

However, one of the advantages of using a grating surface instead of a randomly rough surface in studying optical interactions at rough surfaces is that such interactions can be studied exactly in this case without the amplitude of the grating being treated as a small perturbation.

In this paper we present an exact calculation of the SHG in reflection from a metallic grating, assuming p-polarized light of frequency ω to be incident on the grating surface from the vacuum side, with the plane of incidence perpendicular to the grooves. Since in p polarization the magnetic field has only one nonzero component, in Sec. II we obtain a scalar wave equation for this component of the magnetic field, at frequency 2ω, in the presence of the nonlinear polarization. For the nonlinear polarization, which consists of a bulk term and contributions from two surface current densities, one normal and one tangential to the surface, we use the results of Sipe *et al.*[6] In Sec. III we employ the method of reduced Rayleigh equations[7] to solve the equation for the magnetic field at frequency 2ω and obtain the amplitudes of the reflected waves. An advantage of this method is that only the electromagnetic fields directly required are calculated, which reduces the computations by a factor of 2. Thus, in the calculation of the first-order fields only the fields in the metal are obtained: The first-order fields in the vacuum are not required and enter the theory indirectly only through the boundary conditions satisfied by the fields in the medium. Similarly, only the second-harmonic fields in the vacuum are calculated: Those in the metal appear only indirectly through the boundary conditions satisfied by the fields in the vacuum. In Sec. IV we present numerical calculations for a grating of silver and discuss the results obtained. In Appendix A we obtain the amplitudes of the magnetic field—at the fundamental frequency ω—inside the metal.[8] These amplitudes are required for the calculation of the nonlinear polarization presented in the text. Finally, in Appendix B we present the boundary conditions at a corrugated surface, for the magnetic field at frequency 2ω, in the presence of the nonlinear polarization.

After the present work was completed a paper appeared in which a very general treatment of nonlinear optical interactions (viz., nonlinear mixing of two incident fields, optical rectification, and second-harmonic generation) at grating surfaces is presented.[9] In this work, as in the present paper, the surface profile function is not treated perturbatively, but the authors of Ref. 9 do not use the Rayleigh method: they use instead the differential method,[10] in which the Maxwell equations are integrated numerically across the selvedge region. With this method it is possible to study more strongly corrugated surfaces than is the case with the method of reduced Rayleigh equations used here. Fortunately, many of the most interesting effects occur for corrugation strengths that are sufficiently small for the method of reduced Rayleigh equations to be valid. Another difference between the present work and that presented in Ref. 9 is that in the latter work, in the case of a metal, the nonlinear polarization appears both as a source term in the Maxwell equations for the second-harmonic fields and in the boundary conditions they satisfy. In the present work the second-harmonic (magnetic) field satisfies a homogeneous equation, and the nonlinear polarization enters only through the boundary conditions. This formulation of the problem leads to some computational simplifications.

II. FIELD EQUATIONS IN THE PRESENCE OF A NONLINEAR POLARIZATION

In this section we present the basic equations for the electromagnetic field of the system that we are analyzing, which consists of a nonlinear medium (metal) filling the half-space $z<\zeta(x)$, while vacuum occupies the half-space $z>\zeta(x)$. The surface profile function $\zeta(x)$ is assumed to be a periodic function of x with a period a (Fig. 1). A p-polarized electromagnetic wave of frequency ω is incident on the vacuum-metal interface at $z=\zeta(x)$ from the vacuum side. If the plane of incidence is taken to be the xz plane, i.e., if it is perpendicular to the grooves of the grating, the electric and magnetic fields in our system can be written as

$$\vec{H}(\vec{x},t)=(0,H_y(x,z\mid t),0) \tag{2.1}$$

and

$$\vec{E}(\vec{x},t)=(E_x(x,z\mid t),0,E_z(x,z\mid t)) . \tag{2.2}$$

From Maxwell's equations, with $\mu=1$, we have

$$-\frac{\partial}{\partial z}H_y(x,z\mid t)=\frac{1}{c}\frac{\partial}{\partial t}D_x(x,t\mid t) , \tag{2.3a}$$

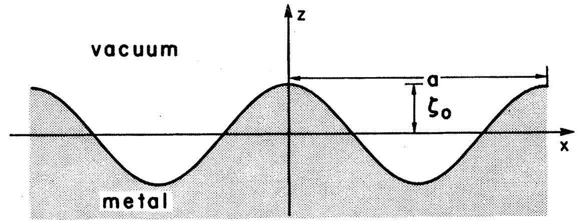

FIG. 1. Geometry analyzed in the present paper. The metal has a sinusoidal profile with period a and amplitude ζ_0.

$$\frac{\partial}{\partial x}H_y(x,z\mid t)=\frac{1}{c}\frac{\partial}{\partial t}D_z(x,z\mid t) , \tag{2.3b}$$

and

$$\frac{\partial}{\partial z}E_x(x,z\mid t)-\frac{\partial}{\partial x}E_z(x,z\mid t)=-\frac{1}{c}\frac{\partial}{\partial t}H_y(x,z\mid t) . \tag{2.4}$$

Since $\vec{H}(\vec{x},t)$ has only a single nonzero component it is convenient to work with it. Thus, expressing the displacement vector in Eqs. (2.3) and (2.4) as

$$\vec{D}(\vec{x},t)=\vec{E}(\vec{x},t)+4\pi\vec{P}^{L}(\vec{x},t)+4\pi\vec{P}^{NL}(\vec{x},t) , \tag{2.5}$$

where $\vec{P}^{L}(\vec{x},t)$ and $\vec{P}^{NL}(\vec{x},t)$ are the linear and nonlinear polarizations, respectively, we obtain a scalar wave equation for the magnetic field given by

$$\left[\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right]H_y(x,z\mid t)=0,\quad z>\zeta(x) \tag{2.6a}$$

and

$$\left[\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right]H_y(x,z\mid t)-\frac{4\pi}{c}\frac{\partial}{\partial t}\int_{-\infty}^{\infty}dt'\chi(t-t')\frac{\partial}{\partial t'}H_y(x,z\mid t')$$
$$=\frac{4\pi}{c}\frac{\partial}{\partial t}\left[\frac{\partial}{\partial x}P_z^{NL}(x,z\mid t)-\frac{\partial}{\partial z}P_x^{NL}(x,z\mid t)\right],\quad z<\zeta(x) , \tag{2.6b}$$

where $\chi(t-t')$ is the linear susceptibility. In order to obtain an equation for the harmonic components of the magnetic field we expand all fields in the usual way[1]

$$H_y(x,z\mid t)=H_y^{(1)}(x,z\mid\omega)e^{-i\omega t}+H_y^{(2)}(x,z\mid 2\omega)e^{-2i\omega t}+\cdots , \tag{2.7}$$

$$E_x(x,z\mid t)=E_x^{(1)}(x,z\mid\omega)e^{-i\omega t}+E_x^{(2)}(x,z\mid 2\omega)e^{-2i\omega t}+\cdots , \tag{2.8}$$

$$E_z(x,z\mid t)=E_z^{(1)}(x,z\mid\omega)e^{-i\omega t}+E_z^{(2)}(x,z\mid 2\omega)e^{-2i\omega t}+\cdots , \tag{2.9}$$

and substitute these expansions into Eqs. (2.6). In this way we obtain as the equations for the amplitude of the magnetic field at frequency ω

$$\left[\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}+\frac{\omega^2}{c^2}\right]H_y^{(1)}(x,z\mid\omega)=0,\quad z>\zeta(x) \tag{2.10a}$$

$$\left[\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}+\frac{\omega^2\epsilon(\omega)}{c^2}\right]H_y^{(1)}(x,z\mid\omega)=0,\quad z<\zeta(x) , \tag{2.10b}$$

where $\epsilon(\omega)$ is the frequency-dependent dielectric constant of the metal, while the equations for the amplitude of the second harmonic of the magnetic field take the forms

$$\left[\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}+\frac{4\omega^2}{c^2}\right]H_y^{(2)}(x,z\mid 2\omega)=0,\quad z>\zeta(x) \tag{2.11a}$$

$$\left[\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}+\frac{4\omega^2\epsilon(2\omega)}{c^2}\right]H_y^{(2)}(x,z\mid 2\omega)=\frac{8\pi i\omega}{c}[\vec{\nabla}\times\vec{P}^{NL}(x,z\mid 2\omega)]_y,\quad z<\zeta(x) , \tag{2.11b}$$

where we have used the fact that

$$\vec{P}^{NL}(\vec{x}\mid t)=\vec{P}^{NL}(\vec{x}\mid 2\omega)e^{-2i\omega t} . \tag{2.12}$$

The solution of the first-order equations (2.10), together with the associated boundary conditions, for the electromagnetic fields in the metal $[z<\zeta(x)]$ that enter the nonlinear polarization is described in Appendix A. In the remainder of this section we focus our attention on the second-order fields.

In Eq. (2.11b) only the bulk contribution to the nonlinear polarization can contribute to the inhomogeneous term. However, as given by Eq. (B1) in Appendix B, this bulk contribution to the nonlinear polarization can be written in the form

$$\vec{P}_B^{NL}(\vec{x}\,|\,2\omega)=\vec{\nabla}\phi(\vec{x}\,|\,2\omega)\,, \tag{2.13}$$

where $\phi(\vec{x}\,|\,2\omega)$ is a scalar function. Since $\vec{\nabla}\times\vec{\nabla}\phi(\vec{x}\,|\,2\omega)=0$, $\vec{P}_B^{NL}(\vec{x}\,|\,2\omega)$ does not contribute to the inhomogeneous term in Eq. (2.11b), and consequently the wave equation for the second harmonic of the magnetic field becomes a homogeneous differential equation given by

$$\left[\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}+\frac{4\omega^2}{c^2}\right]H_y^{(2)}(x,z\,|\,2\omega)=0,\quad z>\zeta(x) \tag{2.14a}$$

$$\left[\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}+\frac{4\omega^2\epsilon(2\omega)}{c^2}\right]H_y^{(2)}(x,z\,|\,2\omega)=0,\quad z<\zeta(x)\,. \tag{2.14b}$$

The nonlinear polarization therefore contributes to the magnetic field only through the boundary conditions. As shown in Appendix B, they are given by

$$H_y^{(2)}(x,z\,|\,2\omega)\,\Big|_-^+=\frac{32\pi ic\gamma b_s}{\omega\epsilon(\omega)^2}\frac{1}{g}\left[\frac{\partial}{\partial n}H_y^{(1)}(x,z\,|\,\omega)\right]\left[\zeta'(x)\frac{\partial}{\partial z}H_y^{(1)}(x,z\,|\,\omega)+\frac{\partial}{\partial x}H_y^{(1)}(x,z\,|\,\omega)\right]\Bigg|_{z=\zeta(x)^-} \tag{2.15a}$$

and

$$\begin{aligned}\frac{1}{\epsilon(2\omega\,|\,z)}\frac{\partial}{\partial n}H_y^{(2)}(x,z\,|\,2\omega)\Big|_-^+=&\frac{1}{\epsilon(2\omega)}\frac{16\pi i\gamma c}{\omega\epsilon(\omega)^2}\frac{1}{g}\Bigg[\left[\frac{\partial}{\partial x}H_y^{(1)}(x,z\,|\,\omega)\right]\left[\frac{\partial^2}{\partial x^2}H_y^{(1)}(x,z\,|\,\omega)\right]\\&+\zeta'(x)\left[\frac{\partial}{\partial z}H_y^{(1)}(x,z\,|\,\omega)\right]\left[\frac{\partial^2}{\partial z^2}H_y^{(1)}(x,z\,|\,\omega)\right]\\&+\left[\frac{\partial^2}{\partial x\partial z}H_y^{(1)}(x,z\,|\,\omega)\right]\\&\times\left[\frac{\partial}{\partial z}H_y^{(1)}(x,z\,|\,\omega)+\zeta'(x)\frac{\partial}{\partial x}H_y^{(1)}(x,z\,|\,\omega)\right]\Bigg]\Bigg|_{z=\zeta(x)^-}\\&-\frac{32\pi i\gamma ca_s}{\omega\epsilon(\omega)^2}\frac{1}{g}\Bigg[\left[\frac{\partial}{\partial x}H_y^{(1)}(x,z\,|\,\omega)\right]\\&\times\Bigg[-\frac{1}{g^4}\zeta'(x)\zeta''(x)\frac{\partial}{\partial x}H_y^{(1)}(x,z\,|\,\omega)+\frac{1}{g^2}\frac{\partial^2}{\partial x^2}H_y^{(1)}(x,z\,|\,\omega)\\&+\frac{1}{g^4}\zeta''(x)[1-\zeta'(x)^2]\frac{\partial}{\partial z}H_y^{(1)}(x,z\,|\,\omega)\\&+\frac{2}{g^2}\zeta'(x)\frac{\partial^2}{\partial x\partial z}H_y^{(1)}(x,z\,|\,\omega)+\frac{1}{g^2}\zeta'(x)\frac{\partial^2}{\partial z^2}H_y^{(1)}(x,z\,|\,\omega)\Bigg]\\&+\left[\frac{\partial}{\partial z}H_y^{(1)}(x,z\,|\,\omega)\right]\\&\times\Bigg[\frac{1}{g^4}\zeta'(x)\zeta''(x)\frac{\partial}{\partial z}H_y^{(1)}(x,z\,|\,\omega)+\frac{2}{g^2}\zeta'(x)^2\frac{\partial^2}{\partial x\partial z}H_y^{(1)}(x,z\,|\,\omega)\\&+\frac{1}{g^2}\zeta'(x)\frac{\partial^2}{\partial x^2}H_y^{(1)}(x,z\,|\,\omega)\\&+\frac{1}{g^2}\zeta'(x)^3\frac{\partial^2}{\partial z^2}H_y^{(1)}(x,z\,|\,\omega)\Bigg]\Bigg]\Bigg|_{z=\zeta(x)^-}\,,\end{aligned} \tag{2.15b}$$

where we have used the notation

$$\phi(\vec{x})\,|\,^{+}_{-}=\phi(\vec{x})\,|_{z=\zeta(x)+}-\phi(\vec{x})\,|_{z=\zeta(x)-}\;, \tag{2.16}$$

and where the quantities γ, b_s, a_s, g, $\epsilon(2\omega\,|\,z)$, etc. are defined in Appendix B.

III. THE METHOD OF REDUCED RAYLEIGH EQUATIONS FOR THE SECOND-HARMONIC FIELD

In this section we obtain the amplitude of the second-harmonic field reflected from the grating, by the method of reduced Rayleigh equations.[7] We require the solutions of Eqs. (2.14) to be outgoing waves or exponentially decaying at infinity. Then the solutions of Eqs. (2.14) outside the grooves of the grating that satisfy these boundary conditions can be written as

$$H_y^{(2)}(x,z\,|\,2\omega)=\sum_{p=-\infty}^{\infty}A_p(2k\,|\,2\omega)\exp[ik_p^{(2)}x+i\alpha_p(2k\,|\,2\omega)z],\quad z>\zeta(x)_{\max} \tag{3.1a}$$

and

$$H_y^{(2)}(x,z\,|\,2\omega)=\sum_{p=-\infty}^{\infty}B_p(2k\,|\,2\omega)\exp[ik_p^{(2)}x-i\beta_p(2k\,|\,2\omega)z],\quad z<\zeta(x)_{\max'}\;, \tag{3.1b}$$

where we have defined

$$k_p^{(2)}=2k+\frac{2\pi}{a}p\;, \tag{3.2}$$

$$\alpha_p(2k\,|\,2\omega)=\left[\frac{4\omega^2}{c^2}-(k_p^{(2)})^2\right]^{1/2},\quad (k_p^{(2)})^2<4\omega^2/c^2 \tag{3.3a}$$

$$=i\left[(k_p^{(2)})^2-\frac{4\omega^2}{c^2}\right]^{1/2},\quad (k_p^{(2)})^2>4\omega^2/c^2\;, \tag{3.3b}$$

$$\beta_p(2k\,|\,2\omega)=\left[\frac{4\omega^2\epsilon(2\omega)}{c^2}-(k_p^{(2)})^2\right]^{1/2},\quad \mathrm{Re}\beta_p(2k\,|\,2\omega)>0,\ \ \mathrm{Im}\beta_p(2k\,|\,2\omega)>0\;, \tag{3.4}$$

and

$$k=(\omega/c)\sin\theta\;, \tag{3.5}$$

where θ is the angle of incidence. If $\alpha_p(2k\,|\,2\omega)$ is real, $A_p(2k\,|\,2\omega)$ is the amplitude of the pth-order beam diffracted from the grating at the frequency 2ω. If $\alpha_p(2k\,|\,2\omega)$ is imaginary, the corresponding contribution to $H_y^{(2)}(x,z\,|\,\omega)$ and the remaining electromagnetic field components are localized to the grating surface.

The Rayleigh hypothesis[11] consists of the assumption that the expressions for the fields given by Eqs. (3.1), which are valid outside the selvedge region, can be continued in to the interface itself and used to satisfy the boundary conditions, Eqs. (2.15). When this is done we obtain the following pair of equations for the determination of the coefficients $\{A_p(2k\,|\,2\omega)\}$ and $\{B_p(2k\,|\,2\omega)\}$:

$$\begin{aligned}\sum_{p=-\infty}^{\infty}&\{A_p(2k\,|\,2\omega)\exp[i\alpha_p(2k\,|\,2\omega)\zeta(x)]-B_p(2k\,|\,2\omega)\exp[-i\beta_p(2k\,|\,2\omega)\zeta(x)]\}\exp(ik_p^{(2)}x)\\&=\frac{32\pi ic\gamma b_s}{\omega\epsilon(\omega)^2}\frac{1}{1+[\zeta'(x)]^2}\sum_{m,n=-\infty}^{\infty}\{[\beta_m(k\,|\,\omega)+\zeta(x)k_m][k_n-\zeta'(x)\beta_n(k\,|\,\omega)]\}\\&\qquad\times B_m(k\,|\,\omega)B_n(k\,|\,\omega)\exp\{i(k_m+k_n)x-i[\beta_m(k\,|\,\omega)+\beta_n(k\,|\,\omega)]\zeta(x)\}\;,\end{aligned} \tag{3.6a}$$

and

$$\sum_{p=-\infty}^{\infty}\Bigg\{[\alpha_p(2k\,|\,2\omega)-\zeta'(x)k_p^{(2)}]A_p(2k\,|\,2\omega)\exp[i\alpha_p(2k\,|\,2\omega)\zeta(x)]$$

$$+\frac{1}{\epsilon(2\omega)}[\beta_p(2k\,|\,2\omega)+\zeta'(x)k_p^{(2)}]B_p(2k\,|\,2\omega)\exp[-i\beta_p(2k\,|\,2\omega)\zeta(x)]\Bigg\}\exp(ik_p^{(2)}x)$$

$$=\frac{1}{\epsilon(2\omega)}\frac{16\pi ic\gamma}{\omega\epsilon(\omega)^2}\sum_{m,n=-\infty}^{\infty}(-k_m[(k_n)^2+\beta_m(k\,|\,\omega)\beta_n(k\,|\,\omega)]+\zeta'(x)\beta_m(k\,|\,\omega)\{[\beta_n(k\,|\,\omega)]^2+k_mk_n\})$$

$$\times B_m(k\,|\,\omega)B_n(k\,|\,\omega)\exp\{i(k_m+k_n)x-i[\beta_m(k\,|\,\omega)+\beta_n(k\,|\,\omega)]\zeta(x)\}$$

$$-\frac{32\pi i\gamma ca_s}{\omega\epsilon(\omega)^2}\sum_{m,n=-\infty}^{\infty}\Bigg[-\frac{\zeta''(x)k_m}{\{1+[\zeta'(x)]^2\}^2}\{\zeta'(x)k_n+[1-\zeta'(x)^2]\beta_m(k\,|\,\omega)\}$$

$$+\frac{ik_m}{1+[\zeta'(x)]^2}\{-(k_n)^2+2\zeta'(x)k_n\beta_n(k\,|\,\omega)-\zeta'(x)^2[\beta_n(k\,|\,\omega)]^2\}$$

$$-\frac{\zeta'(x)\zeta''(x)}{\{1+[\zeta'(x)]^2\}^2}\beta_m(k\,|\,\omega)\beta_n(k\,|\,\omega)$$

$$-\frac{i\zeta'(x)}{1+[\zeta'(x)]^2}\beta_m(k\,|\,\omega)\{2\zeta'(x)k_n\beta_n(k\,|\,\omega)-(k_n)^2-\zeta'(x)^2[\beta_n(k\,|\,\omega)]^2\}\Bigg]$$

$$\times B_m(k\,|\,\omega)B_n(k\,|\,\omega)\exp i(k_m+k_n)x-i[\beta_m(k\,|\,\omega)+\beta_n(k\,|\,\omega)]\zeta(x)\,, \tag{3.6b}$$

where the quantities k_i, $\beta_i(k\,|\,\omega)$, and $B_i(k\,|\,\omega)$ $(i=m,n)$ are associated with the field $H_y^{(1)}(x,z\,|\,\omega)$ and are defined in Appendix A.

Now we focus our attention on the coefficients $\{A_p(2k\,|\,2\omega)\}$ that give the amplitudes of the waves reflected back into the vacuum. We can eliminate the coefficients $\{B_p(2k\,|\,2\omega)\}$ from the pair of equations (3.6) to obtain an equation for the $\{A_p(2k\,|\,2\omega)\}$ alone. We do this by first multiplying Eq. (3.6a) by

$$[\beta_r(2k\,|\,2\omega)-\zeta'(x)k_r^{(2)}]\exp[ik_r^{(2)}x-i\beta_r(2k\,|\,2\omega)\zeta(x)]\,,$$

then multiplying Eq. (3.6b) by $\epsilon(2\omega)\exp[-ik_r^{(2)}x-i\beta_r(2k\,|\,2\omega)\zeta(x)]$, adding the resulting pair of equations and finally integrating the result with respect to x over the interval $(-a/2,\,a/2)$. In this way we obtain

$$\sum_{p=-\infty}^{\infty}\Bigg[\frac{1}{a}\int_{-a/2}^{a/2}dx\,\exp\{-i(k_r^{(2)}-k_p^{(2)})x-i[\beta_r(2k\,|\,2\omega)-\alpha_p(2k\,|\,2\omega)]\zeta(x)\}$$

$$\times\{[\beta_r(2k\,|\,2\omega)+\epsilon(2\omega)\alpha_p(2k\,|\,2\omega)]-\zeta'(x)[k_r^{(2)}+\epsilon(2\omega)k_p^{(2)}]\}A_p(2k\,|\,2\omega)$$

$$+\frac{1}{a}\int_{-a/2}^{a/2}dx\,\exp\{-i(k_r^{(2)}-k_p^{(2)})x-i[\beta_r(2k\,|\,2\omega)+\beta_p(2k\,|\,2\omega)]\zeta(x)\}$$

$$\times\{-[\beta_r(2k\,|\,2\omega)-\beta_p(2k\,|\,2\omega)]+\zeta'(x)(k_r^{(2)}+k_p^{(2)})\}B_p(2k\,|\,2\omega)\Bigg]$$

$$=\frac{32\pi ic\gamma b_s}{\omega\epsilon(\omega)^2}\sum_{m,n=-\infty}^{\infty}\frac{1}{a}\int_{-a/2}^{a/2}dx\,\exp\{-i(k_r^{(2)}-k_m-k_n)x-i[\beta_r(2k\,|\,2\omega)+\beta_m(k\,|\,\omega)+\beta_n(k\,|\,\omega)]\zeta(x)\}$$

$$\times\left[\frac{1}{1+[\zeta'(x)]^2}[\beta_r(2k\,|\,2\omega)-\zeta'(x)k_r^{(2)}][\beta_m(k\,|\,\omega)+\zeta'(x)k_m][k_n-\zeta'(x)\beta_n(k\,|\,\omega)]\right]$$

$$\times B_m(k\,|\,\omega)B_n(k\,|\,\omega)$$

$$+\frac{16\pi ic\gamma}{\omega\epsilon(\omega)^2}\sum_{m,n=-\infty}^{\infty}\frac{1}{a}\int_{-a/2}^{a/2}dx\,\exp\{-i(k_r^{(2)}-k_m-k_n)x-i[\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)]\zeta(x)\}$$

$$\times(-k_m[(k_n)^2+\beta_m(k\mid\omega)\beta_n(k\mid\omega)]$$

$$+\zeta'(x)\beta_m(k\mid\omega)\{[\beta_n(k\mid\omega)]^2+k_mk_n\})B_m(\mid\omega)B_n(k\mid\omega)$$

$$-\frac{32\pi i\gamma c a_s\epsilon(2\omega)}{\omega\epsilon(\omega)^2}\sum_{m,n=-\infty}^{\infty}\frac{1}{a}\int_{-a/2}^{a/2}dx\exp\{-i(k_r^{(2)}-k_m-k_n)x-i[\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)]\zeta(x)\}$$

$$\times\Bigg[\frac{\zeta''(x)k_m}{1+[\zeta'(x)]^2}(\zeta'(x)k_n+\{1-[\zeta'(x)]^2\}\beta_n(k\mid\omega))$$

$$+\frac{ik_m}{1+\zeta'(x)^2}\{-(k_n)^2+2\zeta'(x)k_n\beta_n(k\mid\omega)-\zeta'(x)^2[\beta_n(k\mid\omega)]^2\}$$

$$-\frac{\zeta'(x)\zeta''(x)}{\{1+[\zeta'(x)]^2\}^2}\beta_m(k\mid\omega)\beta_n(k\mid\omega)$$

$$-\frac{i\zeta'(x)}{1+\zeta'(x)^2}\beta_m(k\mid\omega)\{2\zeta'(x)k_n\beta_n(k\mid\omega)-(k_n)^2-\zeta'(x)^2[\beta_n(k\mid\omega)]^2\}\Bigg]$$

$$\times B_m(k\mid\omega)B_n(k\mid\omega)\,. \tag{3.7}$$

When we integrate by parts the terms containing $\zeta'(x)$ on the left-hand side of Eq. (3.7), the coefficient of $\{B_p(2k\mid 2\omega)\}$ vanishes, and we obtain

$$\sum_{p=-\infty}^{\infty}\left[\frac{\beta_r(2k\mid 2\omega)\alpha_p(2k\mid 2\omega)+k_r^{(2)}k_p^{(2)}}{\beta_r(2k\mid 2\omega)-\alpha_p(2k\mid 2\omega)}\right]\frac{1}{a}$$

$$\times\int_{-a/2}^{1/2}dx\exp\{-i(k_r^{(2)}-k_p^{(2)})x-i[\beta_r(2k\mid 2\omega)-\alpha_p(2k\mid 2\omega)]\zeta(x)\}A_p(2k\mid 2\omega)$$

$$=\frac{1}{\epsilon(2\omega)-1}\frac{32\pi ic\gamma b_s}{\omega\epsilon(\omega)^2}\sum_{m,n=-\infty}^{\infty}[\beta_r(2k\mid 2\omega)\beta_m(k\mid\omega)k_nY^{(0)}_{r-m-n}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))$$

$$+\{\beta_r(2k\mid 2\omega)[k_mk_n-\beta_m(k\mid\omega)\beta_n(k\mid\omega)]-k_r^{(2)}\beta_m(k\mid\omega)k_n\}$$

$$\times Y^{(1)}_{r-m-n}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))$$

$$-\{\beta_r(2k\mid 2\omega)k_m\beta_n(k\mid\omega)+k_r^{(2)}[k_mk_n-\beta_m(k\mid\omega)\beta_n(k\mid\omega)]\}$$

$$\times Y^{(2)}_{r-m-n}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))$$

$$+k_r^{(2)}k_m\beta_n(k\mid\omega)Y^{(3)}_{r-m-n}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))]B_m(k\mid\omega)B_n(k\mid\omega)$$

$$-\frac{1}{\epsilon(2\omega)-1}\frac{16\pi ic\gamma}{\omega\epsilon(\omega)^2}$$

$$\times \sum_{m,n=-\infty}^{\infty} \left[k_m[(k_n)^2+\beta_m(k\mid\omega)\beta_n(k\mid\omega)] + \frac{\beta_m(k\mid\omega)\{[\beta_n(k\mid\omega)]^2+k_mk_n\}(k_r^{(2)}-k_m-k_n)}{\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)} \right]$$

$$\times \frac{1}{a}\int_{-a/2}^{a/2} dx \exp\{-i(k_r^{(2)}-k_m-k_n)x$$

$$-i[\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)]\zeta(x)\}B_m(k\mid\omega)B_n(k\mid\omega)$$

$$-\frac{1}{\epsilon(2\omega)-1}\frac{32\pi i\gamma c a_s\epsilon(2\omega)}{\omega\epsilon(\omega)^2}$$

$$\times \sum_{m,n=-\infty}^{\infty} [-i\{k_m(k_n)^2+\tfrac{1}{2}[k_mk_n-\beta_m(k\mid\omega)\beta_n(k\mid\omega)](k_r^{(2)}-k_m-k_n)\}$$
$$\times Y_{r-m-n}^{(0)}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))$$
$$+i\{[2k_m\beta_n(k\mid\omega)+\beta_m(k\mid\omega)k_n]k_n$$
$$-\tfrac{1}{2}[k_mk_n-\beta_m(k\mid\omega)\beta_n(k\mid\omega)][\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)]$$
$$+k_mk_n(k_r^{(2)}-k_m-k_n)\}Y_{r-m-n}^{(1)}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))$$
$$+i\{-[2\beta_m(k\mid\omega)k_n+k_m\beta_n(k\mid\omega)]\beta_n(k\mid\omega)+k_mk_n[\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)]\}$$
$$\times Y_{r-m-n}^{(2)}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))$$
$$-i\beta_m(k\mid\omega)[\beta_n(k\mid\omega)]^2Y_{r-m-n}^{(3)}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))$$
$$+[k_m\beta_n(k\mid\omega)-k_mk_n]U_{r-m-n}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))], \tag{3.8}$$

where

$$Y_{r-m-n}^{(l)}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))$$
$$=\frac{1}{a}\int_{-a/2}^{a/2} dx\exp\{-i(k_r^{(2)}-k_m-k_n)x-i[\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)]\zeta(x)\}\frac{[\zeta'(x)]^l}{1+[\zeta'(x)]^2}, \tag{3.9}$$

with $l=0$, 1, 2, 3, and

$$U_{r-m-n}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))$$
$$=\frac{1}{a}\int_{-a/2}^{a/2} dx\exp\{-i(k_r^{(2)}-k_m-k_n)x-i[\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)]\zeta(x)\}\frac{\zeta''(x)}{1+[\zeta'(x)]^2}. \tag{3.10}$$

IV. DISCUSSION OF NUMERICAL RESULTS

In this section we present and discuss the numerical results obtained for the intensity of the second-harmonic field reflected from a grating characterized by a sinusoidal profile, given by

$$\zeta(x)=\zeta_0\cos\left[\frac{2\pi}{a}x\right]. \tag{4.1}$$

For this profile function we can rewrite Eq. (3.8) as

$$\sum_{p=-\infty}^{\infty} M_{r,p}(2k\mid 2\omega)A_p(2k\mid 2\omega)=\frac{1}{\epsilon(2\omega)-1}\frac{16\pi i c\gamma}{\omega\epsilon(\omega)^2}\sum_{m,n=-\infty}^{\infty} C_{r,m,n}(2k\mid 2\omega)B_m(k\mid\omega)B_n(k\mid\omega),\quad r=0,\pm1,\pm2,\ldots \tag{4.2}$$

with

$$M_{r,p}(2k\mid 2\omega)=\frac{\beta_r(2k\mid 2\omega)\alpha_p(2k\mid 2\omega)+k_r^{(2)}k_p^{(2)}}{\beta_r(2k\mid 2\omega)-\alpha_p(2k\mid 2\omega)}(i)^{r-p}J_{r-p}([\alpha_p(2k\mid 2\omega)-\beta_r(2k\mid 2\omega)]\zeta_0)\;, \tag{4.3}$$

and

$$\begin{aligned}C_{r,m,n}(2k\mid 2\omega)=-&\left[k_m[(k_n)^2+\beta_m(k\mid\omega)\beta_n(k\mid\omega)]+\frac{\beta_m(k\mid\omega)\{[\beta_n(k\mid\omega)]^2+k_mk_n\}(k_r^{(2)}-k_m-k_n)}{\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)}\right]\\
&\times(i)^{r-m-n}J_{r-m-n}(-[\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)]\zeta_0)\\
&+(2b_s\beta_r(2k\mid 2\omega)\beta_m(k\mid\omega)k_n\\
&\quad+ia_s\epsilon(2\omega)\{2k_m(k_n)^2+[k_mk_n-\beta_m(k\mid\omega)\beta_n(k\mid\omega)](k_r^{(2)}-k_m-k_n)\})\\
&\times Y_{r-m-n}^{(0)}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))\\
&+(2b_s\{\beta_r(2k\mid 2\omega)[k_mk_n-\beta_m(k\mid\omega)\beta_n(k\mid\omega)]-k_r^{(2)}\beta_m(k\mid\omega)k_n\}\\
&\quad-ia_s\epsilon(2\omega)\{2[k_m\beta_n(k\mid\omega)+\beta_m(k\mid\omega)k_n]k_n-[k_mk_n-\beta_m(k\mid\omega)\beta_n(k\mid\omega)]\\
&\qquad\times[\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)]+2k_mk_n(k_r^{(2)}-k_m-k_n)\})\\
&\times Y_{r-m-n}^{(1)}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))\\
&-(2b_s\{\beta_r(2k\mid 2\omega)k_m\beta_n(k\mid\omega)+k_r^{(2)}[k_mk_n-\beta_m(k\mid\omega)\beta_n(k\mid\omega)]\}\\
&\quad+ia_s\epsilon(2\omega)\{-2[2\beta_m(k\mid\omega)k_n+k_m\beta_n(k\mid\omega)]\beta_n(k\mid\omega)\\
&\qquad+2k_mk_n[\beta_r(2k\mid 2\omega)+\beta_m(k\mid\omega)+\beta_n(k\mid\omega)]\})\\
&\times Y_{r-m-n}^{(2)}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))\\
&+2\{b_sk_r^{(2)}k_m\beta_n(k\mid\omega)+ia_s\epsilon(2\omega)\beta_m(k\mid\omega)[\beta_n(k\mid\omega)]^2\}\,Y_{r-m-n}^{(3)}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))\\
&-2ia_s\epsilon(2\omega)[k_m\beta_n(k\mid\omega)-k_mk_n]U_{r-m-n}(\beta_r(2k\mid 2\omega),\beta_m(k\mid\omega),\beta_n(k\mid\omega))\;,\end{aligned} \tag{4.4}$$

where $J_n(x)$ is a Bessel function.

In order to calculate the enhancement of the second-harmonic field we must normalize all Fourier components $\{A_p(2k\mid 2\omega)\}$ by the amplitude of the reflected field at a flat surface. This amplitude can be calculated by taking $\zeta(x)$, $\zeta'(x)$, and $\zeta''(x)$ equal to zero in Eq. (4.4). In this way we obtain

$$\frac{\beta_0(2k\mid 2\omega)\alpha_0(2k\mid 2\omega)+4k^2}{\beta_0(2k\mid 2\omega)-\alpha_0(2k\mid 2\omega)}A_f(2k\mid 2\omega)=\frac{1}{\epsilon(2\omega)-1}\frac{16\pi ic\gamma}{\omega\epsilon(\omega)^2}C_{0,0,0}(2k\mid 2\omega)[B_f(k\mid\omega)]^2\;, \tag{4.5}$$

where

$$C_{0,0,0}(2k\mid 2\omega)=k(-\{k^2+[\beta_0(k\omega)]^2\}+2b_s\beta_0(2k\mid 2\omega)\beta_0(k\mid\omega)+2ia_s\epsilon(2\omega)k^2)\;. \tag{4.6}$$

From Eqs. (4.5) and (4.6) we find that

$$\begin{aligned}A_f(2k\mid 2\omega)=-16\pi i\frac{\omega}{c}k&\left[\frac{B_f(k\mid\omega)}{\sqrt{\epsilon}}\right]^2\frac{\gamma}{\epsilon(2\omega)\alpha_0(2k\mid 2\omega)+\beta_0(2k\mid 2\omega)}\\
&\times\left[1-\frac{2b_sc^2}{\epsilon(\omega)\omega^2}\beta_0(2k\mid 2\omega)\beta_0(k\mid\omega)-2ia_s\epsilon(2\omega)\frac{c^2k^2}{\epsilon(\omega)\omega^2}\right]\;,\end{aligned} \tag{4.7}$$

where $A_f(2k\,|\,2\omega)$ and $B_f(k\,|\,\omega)$ are the amplitudes of the fields at the flat surface.

We define the efficiency of the nth-order beam diffracted from the grating at the frequency 2ω in the same way that this is done for the nth-order beam diffracted at the frequency ω,[12] viz., by dividing the total energy in the diffracted beam crossing a plane $z=\text{const}$ in the $+z$ direction by the total energy in the incident beam crossing the same plane in the $-z$ direction, and picking off the contribution proportional to $|A_n(2k\,|\,2\omega)|^2$. If we now normalize this efficiency by the corresponding one for the flat surface, we obtain the grating induced enhancement of the second-harmonic efficiency,

$$I_n=\frac{\alpha_n(2k\,|\,2\omega)\,|A_n(2k\,|\,2\omega)|^2}{\alpha_0(2k\,|\,2\omega)\,|A_f(2k\,|\,2\omega)|^2}\ . \tag{4.8}$$

We will call this quantity the *intensity* of the nth-order diffracted beam at frequency 2ω. We will be concerned only with the values of I_n associated with diffracted beams, i.e., those for which $\alpha_n(2k\,|\,2\omega)$ is real.

In order to present the numerical results illustrating the grating induced enhancement of second-harmonic generation, we assume a grating of silver with a period $a=8000$ Å and the frequency of the incident radiation corresponding to 1.17 eV. This frequency corresponds to the one used by Chen *et al.*[3] in their study of the enhancement of SHG by a randomly rough surface. The dielectric functions at the fundamental frequency and at the second-harmonic have the values

$$\epsilon(\omega)=(0.04+7.5i)^2 \tag{4.9a}$$

and

$$\epsilon(2\omega)=(0.054+3.4i)^2\ , \tag{4.9b}$$

respectively.

The coefficients $\{A_p(2k\,|\,2\omega)\}$ are obtained by solving Eq. (4.2) with a matrix $\overleftrightarrow{M}$ of finite dimension N (corresponding to $r,p=-N/2,\ldots,0,\ldots,N/2-1$) and increasing N until a convergent solution is obtained. Since in the method of reduced Rayleigh equations the solutions converge only for small values of the corrugation strength ζ_0/a, we consider values of $\zeta_0/a\lesssim 0.03$. For these values of the corrugation strength we obtained convergence for $N\lesssim 20$.

The dispersion curve for a surface polariton on a grating surface depends on the corrugation strength. However, for small values of this corrugation strength, the difference between the dispersion curves for a surface polariton on a grating at a flat surface is not significant.[13] Thus, we can analyze the coupling between the incident radiation and the surface polariton on a grating by using the dispersion curve for a flat surface.

In Fig. 2 we present the dispersion curve for a surface polariton on a flat surface. For an angle of incidence $\theta\cong 18.39°$ we observe that at the fundamental frequency $k_{-1}\cong -k_{\rm pol}(\omega)$, where $k_{\rm pol}(\omega)$ is the real part of the wave vector of the surface polariton at the frequency ω. Thus, at this angle the incident photon excites a surface polariton, with frequency ω, on the grating surface, and enhances the amplitudes $A_{-1}(k\,|\,\omega)$ and $B_{-1}(k\,|\,\omega)$, Eq.

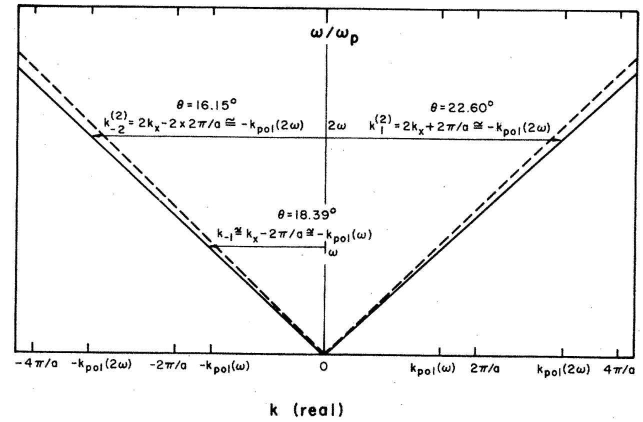

FIG. 2. Dispersion relation for surface polaritons on a flat surface (solid line). The dashed line is the light line.

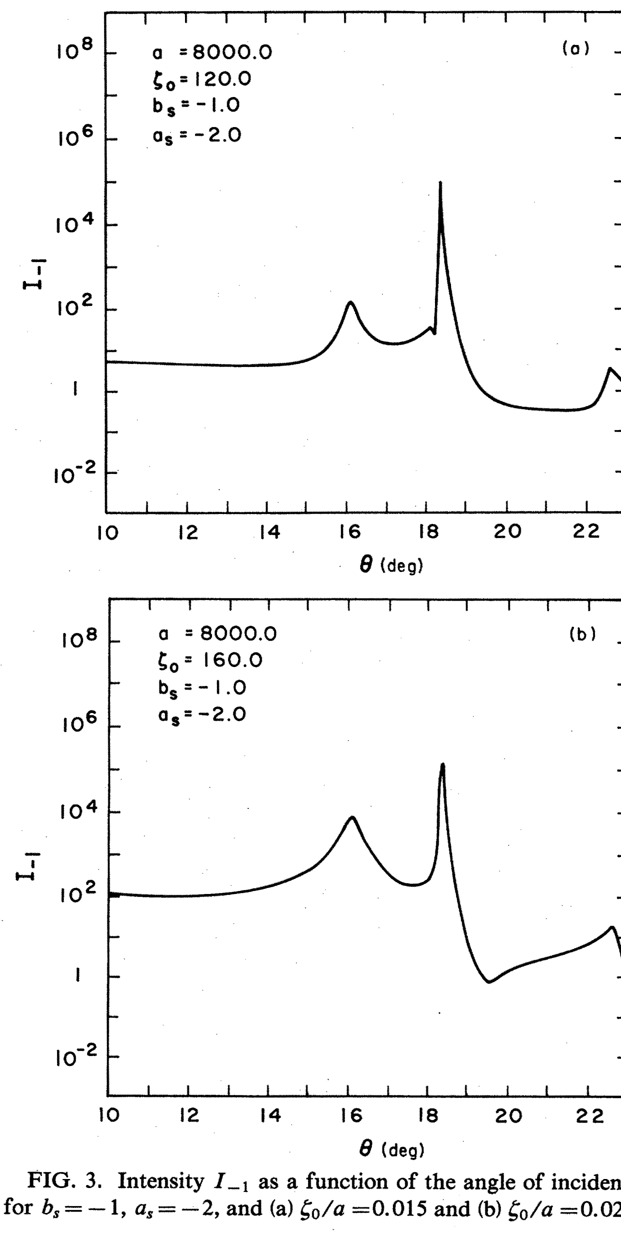

FIG. 3. Intensity I_{-1} as a function of the angle of incidence for $b_s=-1$, $a_s=-2$, and (a) $\zeta_0/a=0.015$ and (b) $\zeta_0/a=0.02$.

(A3). In fact, the propagation constant $\alpha_{-1}(k\,|\,\omega)$, Eq. (A5a), at this angle of incidence is imaginary, which means that $A_{-1}(k\,|\,\omega)$ and $B_{-1}(k\,|\,\omega)$ correspond to localized fields. However, the corresponding $\alpha_{-1}(2k\,|\,2\omega)$, Eq. (3.3a), is real, and the second-harmonic beam associated with $A_{-1}(2k\,|\,2\omega)$ is an outgoing wave. Since from Eqs. (4.2) and (4.8) the intensity I_{-1} is approximately proportional to $|B_{-1}(k\,|\,\omega)|^4$, we expect that an increase in the amplitude $B_{-1}(k\,|\,\omega)$ corresponds to an enhancement of the intensity I_{-1} of the second harmonic. In fact, we observed that not only I_{-1} is enhanced, but other diffracted beams can have significant amplitudes at this angle of incidence.

Now, if we consider an angle of incidence $\theta \cong 16.15°$ we observe that $k_{-2}^{(2)} \cong -k_{\text{pol}}(2\omega)$, where $k_{\text{pol}}(2\omega)$ is the real part of the wave vector of the surface polariton at the frequency 2ω. Thus, at this angle the incident photon excites a surface polariton, with frequency 2ω, on the grating surface and enhances the amplitude $A_{-2}(2k\,|\,2\omega)$. However, the propagation constant $\alpha_{-2}(2k\,|\,2\omega)$, Eq. (3.3b), is imaginary, and we cannot observe the diffracted intensity I_{-2} because $A_{-2}(2k\,|\,2\omega)$ is the amplitude of a localized field. However, the excitation of the surface polariton at the frequency 2ω is associated with a decrease of the determinant of the matrix $\overleftrightarrow{M}$ in Eq. (4.2), because the vanishing of its determinant is the dispersion relation for the surface polariton at the frequency 2ω. Consequently not only $A_{-2}(2k\,|\,2\omega)$ increases but $A_{-1}(2k\,|\,2\omega)$ also can increase, and in this way we can observe an enhancement of the intensity I_{-1} of the mode (-1) and of other modes at this angle. At an angle of incidence $\theta = 22.6°$ we have $k_1^{(2)} \simeq k_{\text{pol}}(2\omega)$, but $\alpha_{-1}(2k\,|\,2\omega)$ is imaginary and I_1 corresponds to an evanescent wave. However, as is the case at the angle $\theta = 16.15°$, we expect an enhancement of the

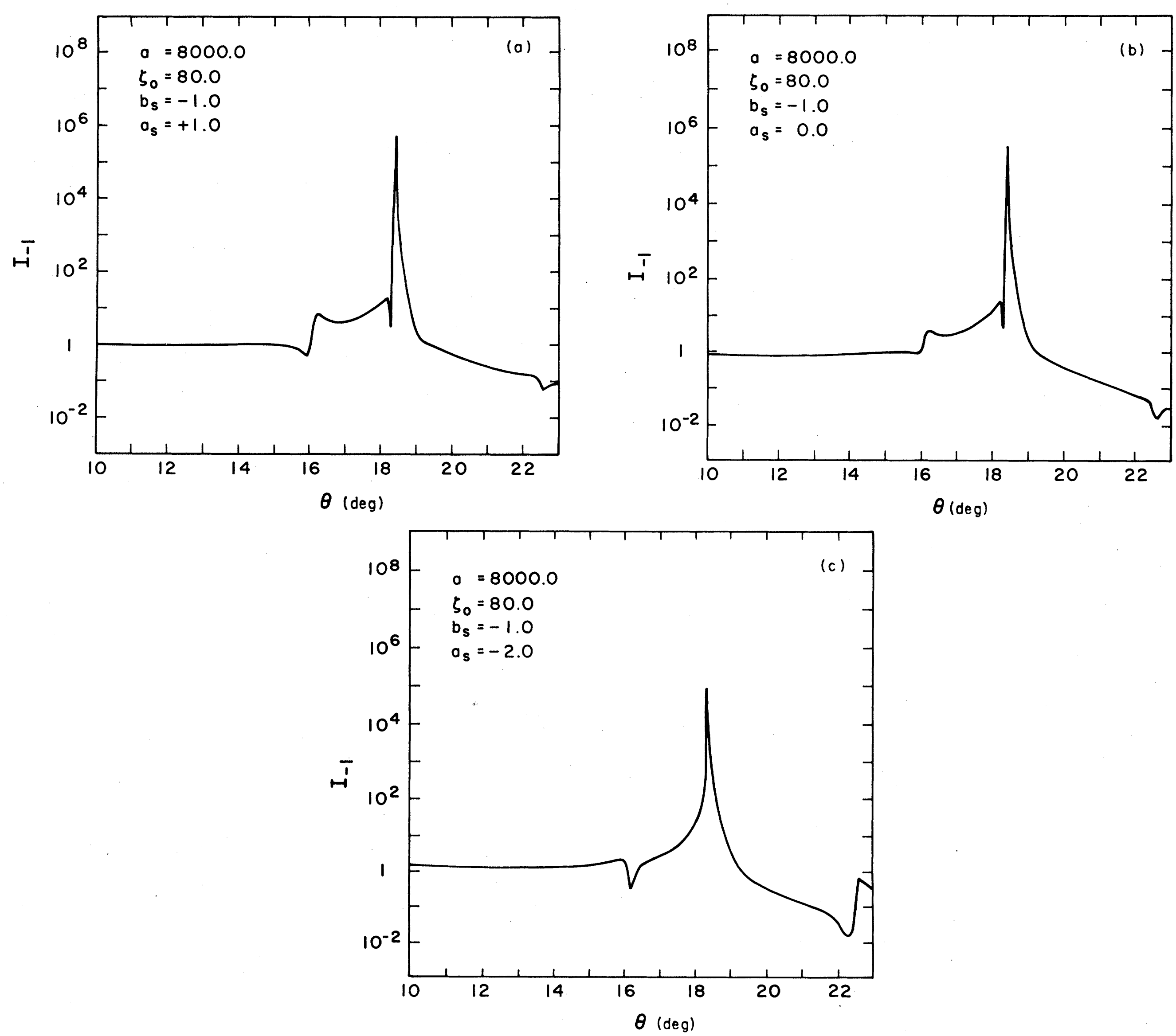

FIG. 4. Intensity I_{-1} as a function of the angle of incidence for $\zeta_0/a = 0.01$; $b_s = -1$; and (a) $a_s = 1$, (b) $a_s = 0$, and (c) $a_s = -2$.

intensities of other modes associated with outgoing waves.

In Fig. 3(a) we present the intensity I_{-1} as a function of the angle of incidence, for a corrugation strength $\zeta_0/a=0.015$ and surface current-density parameters $b_s=-1$ and $a_s=-2$. We observe an enhancement ($I_{-1}=1.09\times10^5$) at the angle $\theta=18.39°$ which is associated with excitation of the surface polariton on the grating with frequency ω. At the angles 16.15° and 22.6°, I_{-1} increases due to the excitation of the surface polariton at the frequency 2ω. However, the enhancement of I_{-1} at an angle of 16.15°, due to the increase of I_{-2}, is larger than the enhancement at the angle 22.6°, due to the increase of I_1, because the mode (−2) is closer to the mode (−1) than to the mode (1). In fact, the increase of I_1 has a larger effect on the intensity of the outgoing beam (0).

In Fig. 3(b) we present the intensity I_{-1} as a function of the angle of incidence, for the same parameters b_s and a_s, but with the corrugation strength $\zeta_0/a=0.02$. Again, we observe enhancements of I_{-1}, but the angles at which they occur differ from those obtained in Fig. 3(a) by $\Delta\theta\cong0.005°$. This difference is due to the fact that the dispersion relation for a surface polariton on a grating surface depends on the corrugation strength, and consequently the relations $k_{-1}=-k_{\text{pol}}(\omega)$, $k_{(n)}^{(2)}=|k_{\text{pol}}(2\omega)|$, $n=-2,1$, depend on the ratio ζ_0/a. The enhancement at the angle 18.39° is now 1.49×10^5, which means that the enhancement of I_{-1} increases with the corrugation strength, at least for the values of ζ_0/a considered in this paper. In fact, at the fundamental frequency ω the amplitudes $A_{-1}(k\,|\,\omega)$ and $B_{-1}(k\,|\,\omega)$ initially increase when we increase the corrugation strength, reach a maximum value at a critical value of the corrugation strength, and then decrease. This effect does not occur for the amplitude $A_{-1}(2k\,|\,2\omega)$ in SHG for the values of ζ_0/a we have considered, although it is not ruled out for larger values of ζ_0/a.

In the present paper we have used the result of Sipe *et al.*[6] for the surface current density, which contains two phenomenological parameters. One is associated with the current parallel to the surface, b_s, and the other is associated with the current perpendicular to the surface, a_s. They have argued that b_s has the universal value −1, while a_s is of order unity but could have either sign. In Figs. 4(a)–4(c) we present the effect of the perpendicular current on the enhancement of I_{-1} when $\zeta_0/a=0.01$ and $b_s=-1$ for $a_s=1$, 0, and −2, respectively. The enhancement of I_{-1} at the angle 18.39° is 4.26×10^5, 3.5×10^5, and 8.5×10^4, corresponding to the values $a_s=1$, 0, and −2, respectively. Since $a_s=0$ means that we neglect the effect of the perpendicular component of the surface current, from the results obtained we conclude that the contribution of this component is significant and it increases or decreases the value of I_{-1} for a_s positive or negative, respectively.

Recently Agarwal and Jha,[5] using perturbation theory, calculated the SHG from a grating. In this calculation they assumed that the tangential component of the electric field was continuous across the boundary surface. However, from the results obtained in the present paper, we find that the perpendicular component of the current cannot be neglected, that is, we have to take into account the fact that the tangential component of the electric field is discontinuous across the boundary surface.

In our problem we generalized the surface current densities, assuming that the surface was locally flat. For a rough surface Rudnick and Stern[14] suggested a value of $b_s=\frac{1}{2}$ to take into account the diffuse scattering of the electrons from it. It is therefore of interest to see the effect of changing b_s, particularly in the direction of values closer to $\frac{1}{2}$. If we consider $\zeta_0/a=0.01$, $a_s=-2$, and $b_s=-\frac{1}{2}$, the enhancement of I_{-1} at the angle 18.39° is 1.1×10^4, which is smaller than the result shown in Fig. 4(c) for $b_s=-1$. However, since we used small corrugation strengths, we expect that we can use the flat surface value $b_s=-1$, but for larger corrugation strengths we would have to take into account the fact that the surface is nonflat.

Finally, we note that the method of reduced Rayleigh equations can be used to obtain an exact solution for the SHG at a grating surface with small corrugation strengths. Since we can control the period and the amplitude of the grating, we expect that this system can be used to obtain the coefficients b_s and a_s experimentally.

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APPENDIX A: MAGNETIC FIELD AT THE FUNDAMENTAL FREQUENCY

A calculation of the magnetic field at the frequency ω, for a p-polarized electromagnetic wave incident on a dielectric grating, is presented in Ref. 8. In this appendix we summarize the results needed in the present paper.

The scalar wave equation (2.10) satisfied by the magnetic field has to be supplemented by the following boundary conditions at the interface $z=\zeta(x)$:

$$H_y^{(1)}(x,z\,|\,\omega)\,\big|_{z=\zeta(x)-}=H_y^{(1)}(x,z\,|\,\omega)\,\big|_{z=\zeta(x)+} \tag{A1a}$$

and

$$\frac{1}{\epsilon(\omega)}\frac{\partial}{\partial n}H_y^{(1)}(x,z\,|\,\omega)\big|_{z=\zeta(x)-}=\frac{\partial}{\partial n}H_y^{(1)}(x,z\,|\,\omega)\,\big|_{z=\zeta(x)+}\,, \tag{A1b}$$

where $\partial/\partial n$ is the derivative along the normal to the interface and is given by

$$\frac{\partial}{\partial n}=\{1+[\zeta'(x)]^2\}^{-1/2}\left[-\zeta'(x)\frac{\partial}{\partial x}+\frac{\partial}{\partial z}\right]. \tag{A2}$$

The solutions of Eq. (2.10), outside the grooves of the grating, that are outgoing waves or exponentially decaying waves can be written in the forms

$$H_y^{(1)}(x,z\mid\omega)=\exp[ikx-i\alpha_0(k\mid\omega)z]+\sum_{p=-\infty}^{\infty}A_p(k\mid\omega)\exp[ik_px+i\alpha_p(k\mid\omega)z],\quad z>\zeta_{\max} \tag{A3a}$$

$$=\sum_{p=-\infty}^{\infty}B_p(k\mid\omega)\exp[ik_px-i\beta_p(k\mid\omega)z],\quad z<\zeta_{\min}\ , \tag{A3b}$$

where we have defined

$$\alpha_p(k\mid\omega)=[(\omega^2/c^2)-k_p^2]^{1/2},\quad k_p^2<\omega^2/c^2 \tag{A4a}$$

$$=i[k_p^2-(\omega^2/c^2)]^{1/2},\quad k_p^2>\omega^2/c^2\ , \tag{A4b}$$

and

$$\beta_p(k\mid\omega)=\left[\epsilon(\omega)\frac{\omega^2}{c^2}-k_p^2\right]^{1/2},\quad \mathrm{Re}\beta_p(k\mid\omega)>0,\quad \mathrm{Im}\beta_p(k\mid\omega)>0\ , \tag{A5}$$

with

$$k_p=k+\frac{2\pi}{a},\quad p=0,\pm1,\pm2,\ldots\ . \tag{A6}$$

The Rayleigh hypothesis[11] is the assumption that the solutions given by Eqs. (A4) can be continued in to the interface itself and used to satisfy the boundary conditions, Eqs. (A1). When this is done we obtain a pair of equations for the determination of the coefficients $\{A_p(k\mid\omega)\}$ and $\{B_p(k\mid\omega)\}$:

$$\sum_{p=-\infty}^{\infty}\{-A_p(k\mid\omega)\exp[ik_px+i\alpha_p(k\mid\omega)\zeta(x)]+B_p(k\mid\omega)\exp[ik_px-i\beta_p(k\mid\omega)\zeta(x)]\}=\exp[ikx-i\alpha_0(k\mid\omega)\zeta(x)]\ , \tag{A7a}$$

$$\sum_{p=-\infty}^{\infty}\left\{[-i\alpha_p(k\mid\omega)+ik_p\zeta'(x)]A_p(k\mid\omega)\exp[ik_px+i\alpha_p(k\mid\omega)\zeta(x)]\right.$$
$$\left.+\frac{1}{\epsilon(\omega)}[-i\beta_p(k\mid\omega)-ik_p\zeta'(x)]B_p(k\mid\omega)\exp[ik_px-i\beta_p(k\mid\omega)\zeta(x)]\right\}$$
$$=-[i\alpha_0(k\mid\omega)+ik\zeta'(x)]\exp[ikx-i\alpha_0(k\mid\omega)\zeta(x)]\ . \tag{A7b}$$

Since the second-harmonic field depends on the field inside the metal, we can eliminate the coefficients $\{A_p(k\mid\omega)\}$ from the pair of equations (A7) to obtain an equation for the $\{B_p(k\mid\omega)\}$ alone. We do this by first multiplying Eq. (A7a) by $[-i\alpha_r(k\mid\omega)-ik_r\zeta'(x)]\exp[ik_rx+i\alpha_r(k\mid\omega)\xi(x)]$ and Eq. (B7b) by $\exp[-ik_rx+i\alpha_r(k\mid\omega)\zeta(x)]$; we integrate each of the resulting equations with respect to x over the interval $(-a/2,\ a/2)$; and finally we add the two equations so obtained. The result can be written in the form

$$\sum_{p=-\infty}^{\infty}\frac{\alpha_r(k\mid\omega)\beta_p(k\mid\omega)+k_rk_p}{\alpha_r(k\mid\omega)-\beta_p(k\mid\omega)}Y_{r-p}(\alpha_r(k\mid\omega),\beta_p(k\mid\omega))B_p(k\mid\omega)=\frac{2\epsilon(\omega)\alpha_0(k\mid\omega)}{1-\epsilon(\omega)}\delta_{r0}\ , \tag{A8}$$

where

$$Y_{r-p}(\alpha_r(k\mid\omega),\beta_p(k\mid\omega))=\frac{1}{a}\int_{-a/2}^{a/2}dx\,\exp\{-i(k_r-k_p)x+i[\alpha_r(k\mid\omega)-\beta_p(k\mid\omega)]\zeta(x)\}\ . \tag{A9}$$

In particular, if we assume

$$\zeta(x)=\zeta_0\cos\left[\frac{2\pi}{a}x\right], \tag{A10}$$

the function defined in Eq. (A9) is

$$Y_{r-p}(\alpha_r(k\mid\omega),\beta_p(k\mid\omega))=(i)^{r-p}J_{r-p}([\alpha_r(k\mid\omega)-\beta_p(k\mid\omega)]\zeta_0)\ , \tag{A11}$$

where $J_n(x)$ is a Bessel function.

APPENDIX B: BOUNDARY CONDITIONS IN THE PRESENCE OF A NONLINEAR POLARIZATION

In this appendix we present the boundary conditions for the magnetic field at the frequency 2ω in the presence of a nonlinear polarization. Before we analyze the boundary conditions at a grating surface let us first consider the boundary conditions at a flat surface of a metal occupying the region $z<0$.

In their investigation of second-harmonic generation at flat metal surfaces Sipe *et al.*[6] expressed the nonlinear polarization as the sum of a bulk contribution and a surface contribution. The bulk contribution was written in the form

$$\vec{P}_B^{NL}(\vec{x}\,|\,2\omega)=\gamma\vec{\nabla}[\vec{E}^{(1)}(\vec{x}\,|\,\omega)\cdot\vec{E}^{(1)}(\vec{x}\,|\,\omega)]\ , \tag{B1}$$

where the expression

$$\gamma=\frac{e}{8m\omega^2}\frac{1-\epsilon(\omega)}{4\pi} \tag{B2}$$

was adopted for the coefficient γ. In the work of Jha and Warke[15] the bulk contribution to the nonlinear polarization when nonresonant interband transitions are taken into account is given by Eq. (B1) but with

$$\gamma=\frac{e}{2m\omega^2}\frac{1-\epsilon(2\omega)}{4\pi}\ . \tag{B3}$$

Equations (B2) and (B3) are identical if we use the free-electron model for $\epsilon(\omega)$, but they differ by a non-negligible amount for silver at the frequency which we are considering. Since Eq. (B2) represents a better choice for the bulk contribution to the nonlinear polarization,[16] we will use it in Eqs. (B1) and (B5) below.

The surface contribution to the nonlinear polarization was expressed by Sipe *et al.*[16] in terms of an effective current density

$$\vec{J}^{NL}(\vec{x}\,|\,\omega)=(-2i\omega)\vec{Q}(\vec{x}\,|\,2\omega)\delta(z-0^+)\ , \tag{B4}$$

where for a p-polarized wave we have

$$Q_x(\vec{x}\,|\,2\omega)=4\gamma b_s E_z^{(1)}(\vec{x}\,|\,\omega)E_x^{(1)}(\vec{x}\,|\,\omega)\,|_{z=0^-}\ , \tag{B5a}$$

$$Q_z(\vec{x}\,|\,2\omega)=2\gamma a_s E_z^{(1)}(\vec{x}\,|\,\omega)E_z^{(1)}(\vec{x}\,|\,\omega)\,|_{z=0^-}\ , \tag{B5b}$$

and a_s and b_s are phenomenological coefficients. The notation $\delta(z-0^+)$ in Eq. (B4) indicates that this effective current density is placed just outside the metal, while the notation $z=0^-$ in Eq. (B5) means that the electric fields entering $\vec{J}^{NL}(\vec{x}\,|\,\omega)$ are evaluated just inside the metal.

The existence of a current at the surface implies that the tangential component of the magnetic field is discontinuous, and the boundary condition is given by

$$\hat{k}\times\vec{H}^{(2)}(\vec{x}\,|\,2\omega)\,|\,^+_-=\frac{4\pi}{c}K_x(\vec{x}\,|\,2\omega)\hat{i}\ , \tag{B6}$$

where

$$\phi(\vec{x})\,|\,^+_-=\phi(\vec{x})\,|_{z=0^+}-\phi(\vec{x})\,|_{z=0^-}\ . \tag{B7}$$

In Eq. (B6) K_x is the surface current, which in the case of p polarization is given by

$$K_x(\vec{x}\,|\,2\omega) =(-2i\omega)[4\gamma b_s E_z^{(1)}(\vec{x}\,|\,\omega)][E_x^{(1)}(\vec{x}\,|\,\omega)]\,|_{z=0^-}\ . \tag{B8}$$

Associated with $J_z^{NL}(\vec{x}\,|\,2\omega)$ there is a dipole moment density at the surface given by

$$\tau_z=Q_z(\vec{x}\,|\,2\omega)\ , \tag{B9}$$

and the boundary condition for the tangential component of the electric field can be written as

$$\hat{t}\cdot\vec{E}^{(2)}(\vec{x}\,|\,2\omega)\,|\,^+_-=4\pi(\vec{\nabla}\tau_z)\cdot\hat{t}\ , \tag{B10}$$

where $\hat{t}$ is the unit vector tangential to the surface. For p polarization $\hat{t}=\hat{i}$ and Eq. (B10) can be written as

$$E_x^{(2)}(\vec{x}\,|\,2\omega)\,|\,^+_-=4\pi\frac{\partial}{\partial x}Q_z(\vec{x}\,|\,2\omega)\ . \tag{B11}$$

In general, the values of the parameters a_s and b_s change from their values at a flat surface when the interface is described by an arbitrary profile function $\zeta(x)$.[14] However, if we assume that the parameters a_s and b_s do not change significantly from their values at a flat surface when we consider an interface with slowly varying profile function $z=\zeta(x)$, we can generalize the current K_x, Eq. (B8), and the dipole moment density τ_z, Eq. (B9), by changing the x and z components of the fields at the interface into the tangential and normal components, respectively. In this way the boundary condition for the tangential component of the magnetic field is obtained as

$$\hat{n}\times\vec{H}^{(2)}(\vec{x}\,|\,2\omega)\,|\,^+_-=\frac{4\pi}{c}\vec{K}_{\parallel}(\vec{x}\,|\,2\omega)\ , \tag{B12}$$

where now

$$\phi(\vec{x})\,|\,^+_-=\phi(\vec{x})\,|_{z=\zeta(x)^+}-\phi(\vec{x})\,|_{z=\zeta(x)^-}\ , \tag{B13}$$

$\hat{n}$ is the unit vector normal to the surface,

$$\hat{n}=\frac{1}{g}[-\zeta'(x)\hat{i}+\hat{k}]\ , \tag{B14}$$

with

$$g=\{1+[\zeta'(x)]^2\}^{1/2}\ , \tag{B15}$$

and $\vec{K}_{\parallel}(\vec{x}\,|\,2\omega)$ is the tangential component of the current at the surface, which is given by

$$\vec{K}_{\parallel}(\vec{x}\,|\,2\omega)=(-2i\omega)\hat{t}[4\gamma b_s\hat{n}\cdot\vec{E}^{(1)}(\vec{x}\,|\,\omega)] \times[\hat{t}\cdot\vec{E}^{(1)}(\vec{x}\,|\,\omega)]\,|_{z=\zeta(x)^-}\ , \tag{B16}$$

where $\hat{t}[\hat{t}\cdot\vec{E}^{(1)}(\vec{x}\,|\,\omega)]$ is the tangential component of the electric field at the surface. Using the free-electron model for $\epsilon(\omega)$ in Eq. (B16) and the value of b_s for a flat surface, the boundary condition given by Eq. (B12) coincides with the corresponding conditions used by Agarwal and Jha.[5]

The boundary condition for the tangential component of the electric field is[17]

$$\hat{t}\cdot\vec{E}^{(2)}(\vec{x}\,|\,2\omega)\,|\,^+_-=4\pi[\vec{\nabla}\tau_N(\vec{x}\,|\,2\omega)]\cdot\hat{t}\ , \tag{B17}$$

where $\hat{t}$ is the unit vector tangential to the surface,

$$\hat{t}=\frac{1}{g}[\hat{i}+\zeta'(x)\hat{k}]\ , \tag{B18}$$

and

$$\tau_N(\vec{x}\,|\,2\omega)=2\gamma a_s[\hat{n}\cdot\vec{E}^{(1)}(\vec{x}\,|\,\omega)]^2\,|_{z=\zeta(x)^-}\ , \tag{B19}$$

is the component of the dipole moment density normal to the surface. For a flat surface $\tau_N=\tau_z$ as given by Eq. (B9).

Since we are considering p polarization, the magnetic field has only a y component, and we can rewrite the boundary conditions in terms of $H_y^{(2)}(x,z\mid 2\omega)$ and $H_y^{(1)}(x,z\mid\omega)$. For this we use the fact that inside the metal

$$\vec{E}^{(1)}(\vec{x}\mid\omega)=\frac{ic}{\omega\epsilon(\omega)}\vec{\nabla}\times\vec{H}^{(1)}(\vec{x}\mid\omega) \tag{B20}$$

and

$$\vec{E}^{(2)}(\vec{x}\mid 2\omega)=\frac{ic}{2\omega\epsilon(2\omega)}\vec{\nabla}\times\vec{H}^{(2)}(\vec{x}\mid 2\omega)-\frac{4\pi}{\epsilon(2\omega)}\vec{P}_B^{NL}(\vec{x}\mid 2\omega)\,, \tag{B21}$$

and from Eqs. (B12) and (B16) we obtain

$$H_y^{(2)}(x,z\mid 2\omega)\mid_-^+=\frac{32\pi ic\gamma b_s}{\omega\epsilon(\omega)^2}\left[\frac{\partial}{\partial n}H_y^{(1)}(x,z\mid\omega)\right]\times\left.\left[\frac{\partial}{\partial t}H_y^{(1)}(x,z\mid\omega)\right]\right|_{z=\zeta(x)-}, \tag{B22}$$

where $\partial/\partial n$ has been defined in Eq. (A2) and

$$\frac{\partial}{\partial t}=\frac{1}{g}\left[\frac{\partial}{\partial x}+\zeta'(x)\frac{\partial}{\partial z}\right] \tag{B23}$$

is the derivative along the tangent to the interface.

Now substituting Eqs. (B20) and (B21) in Eqs. (B17) and (B19), we obtain Eq. (2.15b), where

$$\epsilon(2\omega\mid z)=\begin{cases}1, & z>\zeta(x)\\ \epsilon(2\omega), & z<\zeta(x)\,.\end{cases} \tag{B24}$$

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*Permanent address: Departamento de Fisica, Universidade Federal do Ceara, 60000, Fortaleza, Ceara, Brasil.

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