15 September 2000

ELSEVIER

Optics Communications 183 (2000) 529–545

OPTICS COMMUNICATIONS

www.elsevier.com/locate/optcom

The surface enhanced second harmonic generation of light from a randomly rough metal surface in the Kretschmann geometry

T.A. Leskova [a], M. Leyva-Lucero [b], E.R. Méndez [c], A.A. Maradudin [d,*], I.V. Novikov [d,1]

[a] Institute of Spectroscopy, Russian Academy of Sciences, Troitsk 142092, Russia
[b] Escuela de Ciencias Físico-Matematicas, Universidad Autonoma de Sinaloa, Ciudad Universitaria, C.P. 80000 Culiacan, Sinaloa, Mexico
[c] División de Física Aplicada, Centro de Investigación Científica y de Educación Superior de Ensenada, Apartado Postal 2732, Ensenada, Baja California, Mexico
[d] Department of Physics and Astronomy, and Institute for Surface and Interface Science, University of California, Irvine, California, 92697, USA

Received 11 April 2000; received in revised form 17 July 2000; accepted 18 July 2000

Abstract

We present results of perturbative calculations of the second harmonic light generated in the transmission of p-polarized light through a thin metal film with a one-dimensional random surface in the Kretschmann attenuated total reflection (ATR) geometry. The metal film is deposited on the planar surface of a prism through which the light is incident. The back surface of the film is a one-dimensional random surface whose generators are perpendicular to the plane of incidence. It is in contact either with a semi-infinite vacuum or with a semi-infinite nonlinear crystal (quartz). It is shown that when the random surface separates the metal film from vacuum so that the nonlinearity of the film surfaces gives rise to the harmonic light, for a general angle of incidence a dip appears in the angular dependence of the intensity of the transmitted harmonic light in the direction normal to the mean surface. When the second harmonic generation is due to the nonlinearity of the crystal in contact with the metal film, a peak in the angular dependence of the intensity of the transmitted harmonic light occurs in this direction. These dips and peaks are multiple-scattering effects. However, when the angle of incidence is optimal for the excitation of surface plasmon polaritons at the film-vacuum/nonlinear crystal interface the nonlinear mixing of the incident light and the backward propagating surface plasmon polariton leads to an intense peak in the angular dependence of the intensity of the transmitted harmonic light in the direction normal to the mean surface. This peak is already present in the single-scattering approximation.

1. Introduction

Experimental and theoretical studies of second harmonic generation (SHG) of light in reflection from a metal surface go back at least three decades, to the first experimental observation [1] and the first theoretical

* Corresponding author. E-mail: aamaradu@uci.edu
[1] Present address: Stanford Research Systems, 1290 ReamWood Ave., Sunnyvale, California, 94089, USA.

PII: S0030-4018(00)00897-X

description of the phenomenon in [2,3]. At this early stage of the studies [2,3] it was established that this phenomenon is a surface effect. It is therefore very sensitive to anything happening on the surface. In the last several years interest in second harmonic generation from a rough metal surface has arisen due to the growing interest in interference effects occurring in the multiple scattering of electromagnetic waves from randomly rough metal surfaces and the related backscattering enhancement phenomenon [4]. It has been expected that the nonlinear optical interactions at a randomly rough metal surface should also produce new features owing to interference effects in the multiple scattering of electromagnetic waves. The results of a perturbative calculation carried out by McGurn et al. [5] predicted that enhanced second harmonic generation of light at a weakly rough, clean, metal surface occurs not only in the retroreflection direction but also in the direction normal to the mean scattering surface. Interference effects in the multiple scattering of surface plasmon polaritons of the fundamental frequency, excited by the incident light through the roughness of the surface, are responsible for the appearance of the peak in the direction normal to the mean surface, while those occurring in the multiple scattering of surface plasmon polaritons at the harmonic frequency are responsible for the appearance of the peak in the retroreflection direction.

This work stimulated several subsequent experimental studies of second-harmonic generation in the scattering of light from random metal surfaces [6–11], and enhanced second harmonic generation peaks in the direction normal to the mean surface and in the retroreflection direction were observed [6–9,11]. In these experiments, however, the scattering system was not a clean random interface between vacuum and a semi-infinite metal. To amplify the second harmonic signal the Kretschmann attenuated total reflection (ATR) geometry [12] was used. Therefore, the scattering system was the random interface with a dielectric or vacuum of a thin metal film deposited on the planar base of a dielectric prism through which the light was incident. In the experiments of Refs. [6,7,9] the scattering system was the random interface between a silver film and a nonlinear quartz crystal, so that the nonlinear interaction occurred in the quartz crystal rather than at the significantly more weakly nonlinear silver surfaces. A well-defined peak of the second harmonic generation in the direction normal to the mean interface was observed in transmission in [6]. When the experiment was carried out with long-range surface plasmon polaritons [13], peaks of the enhanced second harmonic generation were detected both in the retroreflection direction [7,9] and in the direction normal to the surface in transmission [9]. In Refs. [8,10,11] attempts to detect the peaks of the enhanced second harmonic generation at a silver film-vacuum interface were made. A well-defined peak in the direction normal to the mean surface was observed in transmission in [8,11], while only a broad depolarized background, but no peak in the direction normal to the mean surface, was observed in transmission in Ref. [10]. The peak of the intensity of the generated light in the direction normal to the mean surface observed in [6,8,9,11] was interpreted as due to the coherent nonlinear mixing of multiply-scattered contrapropagating surface plasmon polaritons. As is well known [14], the symmetry of the surface nonlinear polarization of a clean metal surface forbids second harmonic generation due to contrapropagating beams of surface plasmon polaritons. In [8,11] it was argued that the surface roughness breaks the symmetry and, as a result, makes the second harmonic generation by contrapropagating beams of surface plasmon polaritons possible. We believe that this explanation is incorrect, and present what we believe to be the correct explanation in the present work.

The main reason for using the Kretschmann ATR geometry in the experiments [6–11] was to excite surface plasmon polaritons associated with the film-vacuum (film-nonlinear crystal) interface through the ATR phenomenon, and thus to enhance the field at the metal film-vacuum (or metal film-nonlinear crystal) interface. For many years this experimental configuration was used to enhance the second harmonic generation in reflection from a metal surface [15].

If, however, the metal surface is rough the surface plasmon polaritons can be excited through the surface roughness without a prism coupler. The nonlinear interaction of the excited surface plasmon polaritons with the incident light also leads to a strong enhancement of the second harmonic generation [5,16,17]. Such processes lead to strong peaks, up to several orders of magnitude above the background, in the angular distribution of the intensity of light of frequency 2ω in the directions determined by the conditions $q = k \pm k_{sp}(\omega)$, where

$q=(2\omega/c)\sin\theta_s$, $k=(\omega/c)\sin\theta_0$, θ_s and θ_0 are the angles of scattering and incidence, and $k_{sp}(\omega)$ is the wavenumber of the surface plasmon polaritons of frequency ω that are excited through the surface roughness. These peaks arise already in single-scattering processes, and their positions depend on the angle of incidence.

In the Kretschmann geometry the angle of incidence is tuned so that the surface polaritons on the rough metal film-vacuum interface or the rough metal film-nonlinear crystal interface are excited efficiently. This means that $k=k_{sp}(\omega)$, and the single-scattering peaks move to $q=0$, i.e. to the direction normal to the mean surface, and to $q=2k_{sp}(\omega)$, i.e. to the nonradiative region. Due to the single-scattering origin of the peak in the direction normal to the mean surface, it could be expected that its intensity is much higher than that of the peak predicted in [5], which is associated with multiple-scattering processes. As a result, the weak features associated with the coherent effects in the multiple scattering of surface plasmon polaritons are masked by the strong single-scattering contribution. The presence of roughness leads also to the excitation of the surface plasmon polaritons associated with the prism–metal film interface. As a result, two additional strong peaks appear in the angular distribution of the intensity of the generated light. Their positions are determined by the conditions $q=k\pm k_{sp}^{(p)}(\omega)$, where $k_{sp}^{(p)}(\omega)$ is the wavenumber of the surface plasmon polaritons associated with the prism–metal film interface.

In this paper we present results of perturbative calculations of the second harmonic generation of light in transmission in the Kretschmann ATR geometry when the interface of a thin metal film with vacuum or a nonlinear crystal is weakly rough. The emphasis will be on the existence of the peak of the enhanced second harmonic generation in the direction normal to the mean surface, which is due to the interference of multiply-scattered surface plasmon polaritons of the frequency of the incident light. The results of rigorous numerical simulations, applicable to more strongly corrugated surfaces, will be presented elsewhere.

2. Formulation of the scattering problem

The physical system we consider consists of a dielectric prism characterized by a positive dielectric constant ϵ_0 in the region $x_3>D$, a metal film characterized by an isotropic, complex, frequency-dependent dielectric function $\epsilon_1(\omega)=\epsilon'(\omega)+i\epsilon''(\omega)$ in the region $D>x_3>\zeta(x_1)$ and, generally, a nonlinear crystal characterized by a linear dielectric function $\epsilon_2(\omega)$ in the region $x_3<\zeta(x_1)$ (Fig. 1).

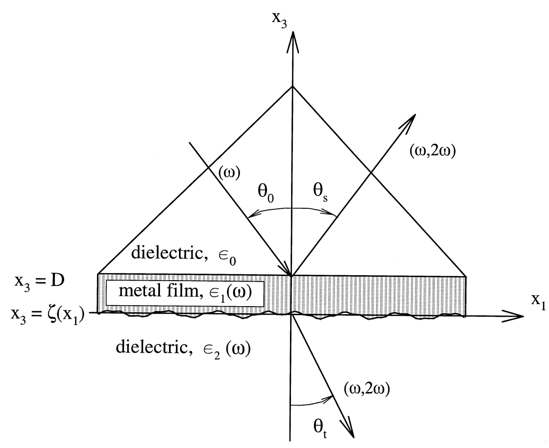

Fig. 1. The system studied in this paper.

The surface profile function $\zeta(x_1)$ is assumed to be a single-valued function of x_1 that is differentiable as many times as is necessary, and to constitute a zero-mean, stationary, Gaussian random process defined by

$$\langle \zeta(x_1) \rangle = 0, \tag{2.1a}$$

$$\langle \zeta(x_1)\zeta(x_1') \rangle = \delta^2 W(|x_1 - x_1'|). \tag{2.1b}$$

The angle brackets in Eqs. (2.1) denote an average over the ensemble of realizations of the surface profile function, and $\delta = \langle \zeta^2(x_1) \rangle^{1/2}$ is the rms height of the surface.

We also introduce the Fourier integral representation of the surface profile function,

$$\zeta(x_1) = \int_{-\infty}^{\infty} \frac{dQ}{2\pi} \hat{\zeta}(Q) e^{iQx_1}. \tag{2.2}$$

The Fourier coefficient $\hat{\zeta}(Q)$ is a zero-mean Gaussian random process defined by

$$\langle \hat{\zeta}(Q) \rangle = 0, \tag{2.3a}$$

$$\langle \hat{\zeta}(Q)\hat{\zeta}(Q') \rangle = 2\pi\delta(Q+Q')\delta^2 g(|Q|), \tag{2.3b}$$

where $g(|Q|)$, the power spectrum of the surface roughness, is given by

$$g(|Q|) = \int_{-\infty}^{\infty} dx_1 W(|x_1|) e^{-iQx_1}. \tag{2.4}$$

In this paper we will present results calculated for a random surface characterized by a Gaussian power spectrum given by

$$g(|Q|) = \sqrt{\pi}\, a e^{-Q^2a^2/4}. \tag{2.5}$$

In our treatment of second harmonic generation we neglect the influence of the nonlinearity on the fundamental fields (undepleted pump approximation). We treat the nonlinearity of the metal film through effective nonlinear boundary conditions, while the bulk nonlinearity of the crystal is taken into account through the solution of the Maxwell equations. Since we study the Kretchsmann ATR geometry, and the surface roughness is one-dimensional, only *p*-polarized incident waves are of interest to us. As is known, the nonlinear polarization of a metal surface has only *p*-polarized components independent of whether the incident light is *p*- or *s*-polarized. Therefore, in what follows we assume that a *p*-polarized plane wave of frequency ω is incident from the prism onto the planar prism–metal interface. The angle of incidence, measured counterclockwise from the normal to the mean surface is θ_0, and the plane of incidence is the x_1x_3-plane. We first solve the linear scattering problem, and with its solution in hand we determine the surface and bulk nonlinear polarizations at the harmonic frequency 2ω. For the sake of simplicity we assume that the orientation of the crystal is chosen so that the bulk nonlinear polarization in it has a nonzero tangential component [18]

$$P_1^{(\mathrm{NL})}(x_1,x_3|2\omega) = d_{11}\left(E_1^2(x_1,x_3|\omega) - E_3^2(x_1,x_3|\omega)\right), \tag{2.6}$$

where d_{11} is the second order nonlinear susceptibility. In solving the scattering problem for the harmonic fields we will use the nonlinear boundary conditions [19], whose general form is

$$\begin{aligned} &H_2^{(f)}(x_1,x_3|2\omega)|_{x_3=D,\zeta(x_1)} - H_2^{(p,v)}(x_1,x_3|2\omega)|_{x_3=D,\zeta(x_1)} \\ &\quad = \frac{2ic}{\omega}\mu_3^{(p,v)}\frac{1}{\phi^2(x_1)}\frac{1}{\epsilon_1(\omega)}\frac{\partial}{\partial N}H_2^{(f)}(x_1,x_3|\omega)|_{x_3=D,\zeta(x_1)}\frac{d}{dx_1}H_2^{(f)}(x_1,x_3|\omega)|_{x_3=D,\zeta(x_1)} \\ &\quad \equiv A^{(p,v)}(x_1|2\omega), \end{aligned} \tag{2.7a}$$

$$\frac{1}{\epsilon_1(2\omega)}\left[\frac{\partial}{\partial N}H_2^{(f)}(x_1,x_3|2\omega)\right]\Bigg|_{x_3=D,\zeta(x_1)} - \frac{1}{\epsilon_{0,2}(2\omega)}\left[\frac{\partial}{\partial N}H_2^{(p,v)}(x_1,x_3|2\omega)\right]\Bigg|_{x_3=D,\zeta(x_1)}$$
$$= \frac{2ic}{\omega}\frac{d}{dx_1}\left\{\frac{1}{\phi^2(x_1)}\left[\mu_1^{(p,v)}\left(\frac{d}{dx_1}H_2^{(f)}(x_1,x_3|\omega)\Bigg|_{x_3=D,\zeta(x_1)}\right)^2\right.\right.$$
$$\left.\left. + \frac{\mu_2^{(p,v)}}{\epsilon_1^2(\omega)}\left(\frac{\partial}{\partial N}H_2^{(f)}(x_1,x_3|\omega)\Bigg|_{x_3=D,\zeta(x_1)}\right)^2\right]\right\} \equiv B^{(p,v)}(x_1|2\omega), \tag{2.7b}$$

where $\partial/\partial N = (-\zeta'(x_1)\partial/\partial x_1, 0, \partial/\partial x_3)$ is the unnormalized derivative along the normal to the surface, $\phi(x_1) = [1 + (\zeta'(x_1))^2]^{1/2}$ and the indices p, f, and v denote the fields in the prism, metal film, and vacuum, respectively. In Eqs. (2.7) $\mu_{1,2,3}^{(p;v)}$ are phenomenological nonlinear coefficients at the prism–film (p) and film-vacuum (v) interfaces, and are to be determined from experimental data or by using some particular model for the surface nonlinear polarization.

We assume that the metal film-crystal interface is weakly rough and satisfies the condition for the validity of the Rayleigh hypothesis, $|\zeta'(x_1)| \ll 1$ [20–22]. In this case we can seek the x_2-component of the magnetic fields of frequencies ω and 2ω in the form

$$H_2^{(p)}(x_1,x_3|\Omega) = H_0 e^{ikx_1} e^{-i\alpha_0(k,\Omega)(x_3-D)} + \int_{-\infty}^{\infty}\frac{dq}{2\pi}R(q,\Omega)e^{iqx_1}e^{i\alpha_0(q,\Omega)(x_3-D)} \tag{2.8a}$$

in the prism,

$$H_2^{(f)}(x_1,x_3|\Omega) = \int_{-\infty}^{\infty}\frac{dq}{2\pi}e^{iqx_1}\left[T_1(q,\Omega)e^{i\alpha_1(q,\Omega)x_3} + T_2(q,\Omega)e^{-i\alpha_1(q,\Omega)x_3}\right] \tag{2.8b}$$

in the metal film, and

$$H_2^{(v)}(x_1,x_3|\Omega) = \int_{-\infty}^{\infty}\frac{dq}{2\pi}e^{iqx_1}\left[T(q,\Omega)e^{-i\alpha_2(q,\Omega)x_3} + \mathscr{P}(q,\Omega|x_3)\right] \tag{2.8c}$$

in the crystal. In Eqs. (2.8) Ω stands for either ω or 2ω, $R(q,\Omega)$ and $T(q,\Omega)$ are the scattering and transmission amplitudes, respectively, $\alpha_0(q,\Omega) = \sqrt{\epsilon_0(\Omega^2/c^2) - q^2}$, $\mathrm{Re}(\alpha_0(q,\Omega)) > 0$, $\mathrm{Im}(\alpha_0(q,\Omega)) > 0$, $\alpha_1(q,\Omega) = \sqrt{\epsilon_1(\Omega)(\Omega^2/c^2) - q^2}$, $\mathrm{Re}(\alpha_1(q,\Omega)) > 0$, $\mathrm{Im}(\alpha_1(q,\Omega)) > 0$, and $\alpha_2(q,\Omega) = \sqrt{\epsilon_2(\Omega)(\Omega^2/c^2) - q^2}$, $\mathrm{Re}(\alpha_2(q,\Omega)) > 0$, $\mathrm{Im}(\alpha_2(q,\Omega))) > 0$.

When we consider fields of the fundamental frequency ω, H_0 is the amplitude of the incident field, $k = \sqrt{\epsilon_0}(\omega/c)\sin\theta_0$ and $\alpha_0(k,\omega) = \sqrt{\epsilon_0}(\omega/c)\cos\theta_0$ are the tangential and normal components of the wavector of the incident light, and $\mathscr{P}(q,\omega|x_3) = 0$. For the second harmonic fields $H_0 = 0$ and $\mathscr{P}(q,2\omega|x_3)$ describes the particular solution of the Maxwell equations in the nonlinear crystal:

$$\mathscr{P}(q,2\omega|x_3,) = -\frac{2cd_{11}}{\omega\epsilon_2^2(\omega)}\int_{-\infty}^{\infty}\frac{dp}{2\pi}e^{-i(\alpha_2(p,\omega)+\alpha_2(q-p,\omega))x_3}T(q-p,\omega)T(p,\omega)F(q,p), \tag{2.9}$$

where

$$F(q,p) = \frac{(\alpha_2(p,\omega) + \alpha_2(q-p,\omega))(p(q-p) - \alpha_2(p,\omega)\alpha_2(q-p,\omega))}{(\alpha_2(p,\omega) + \alpha_2(q-p,\omega))^2 - \alpha_2^2(q,2\omega)}. \tag{2.10}$$

Since in the experiments [6,8–11] it was the transmitted signal that was measured, we solve the scattering problem for the transmission amplitude $T(q,\Omega)$. Using the boundary conditions for the tangential component of the magnetic field and the tangential component of the electric field across the interfaces at the fundamental and harmonic frequencies, we can obtain a single integral equation for the transmission amplitude $T(q,\Omega)$ that is analogous to the reduced Rayleigh equation of the scattering problem,

$$T(q,\Omega)=G_0(q,\Omega)\int_{-\infty}^{\infty}\frac{dp}{2\pi}V(q|p,\Omega)T(p,\Omega)+G_0(q,\Omega)Q(q,\Omega), \tag{2.11}$$

where the scattering potential $V(q|k,\Omega)$ is

$$V(q|k,\Omega)=i\frac{\epsilon_1(\Omega)-\epsilon_2(\Omega)}{\epsilon_2(\Omega)\epsilon_1(\Omega)}\left[m(q|k,\Omega)+r_{01}(q,\Omega)^{2i\alpha_1(q,\Omega)D}n(q|k,\Omega)\right], \tag{2.12}$$

with

$$m(q|k,\Omega)=\frac{qk+\alpha_1(q,\Omega)\alpha_2(k,\Omega)}{\alpha_1(q,\Omega)-\alpha_2(k,\Omega)}J\big(-(\alpha_1(q,\Omega)-\alpha_2(k,\Omega))|q-k\big), \tag{2.13a}$$

$$n(q|k,\Omega)=\frac{qk-\alpha_1(q,\Omega)\alpha_2(k,\Omega)}{\alpha_1(q,\Omega)+\alpha_2(k,\Omega)}J\big(\alpha_1(q,\Omega)+\alpha_2(k,\Omega)|q-k\big), \tag{2.13b}$$

$$J(\gamma|Q)=\int_{-\infty}^{\infty}dx_1e^{-iQx_1}(e^{-i\gamma\zeta(x_1)}-1), \tag{2.13c}$$

and where $G_0(q,\Omega)$ is the Green's function associated with a three-layer system with planar interfaces

$$G_0(k,\Omega)=\frac{i}{D(k,\Omega)}, \tag{2.14a}$$

with

$$D(k)=\left(\frac{\alpha_1(k,\Omega)}{\epsilon_1(\Omega)}+\frac{\alpha_2(k,\Omega)}{\epsilon_2(\Omega)}\right)\left[1+r_{01}(k)r_{12}(k)e^{2i\alpha_1(k,\Omega)D}\right], \tag{2.14b}$$

and

$$r_{ij}(q,\Omega)=\frac{\big(\alpha_i(q,\Omega)\epsilon_j(\Omega)-\alpha_j(q,\Omega)\epsilon_i(\Omega)\big)}{\big(\alpha_i(q,\Omega)\epsilon_j(\Omega)+\alpha_j(q,\Omega)\epsilon_i(\Omega)\big)},\quad i,j=0,1,2. \tag{2.15}$$

The driving term $Q(q,\Omega)$ in (2.11) has different forms for the fundamental and harmonic fields. For the fundamental field it is related to the incident field,

$$Q(q,\omega)=-2i\frac{\alpha_1(k,\omega)}{\epsilon_1(\omega)}t_{01}(k,\omega)e^{i\alpha_1(k,\omega)D}H_0 2\pi\delta(q-k), \tag{2.16}$$

where

$$t_{ij}(k,\Omega)=\frac{2\alpha_i(k,\Omega)\epsilon_j(\Omega)}{\alpha_i(k,\Omega)\epsilon_j(\Omega)+\alpha_j(k,\Omega)\epsilon_i(\Omega)},\quad i,j=0,1,2, \tag{2.17}$$

so that $G_0(q,\omega)Q(q,w)=T_0(k,\omega)2\pi\delta(q-k)$, where $T_0(k,\omega)$ is the Fresnel transmission coefficient for the layer system

$$T_0(k,\omega)=\frac{t_{12}(k,\omega)t_{01}(k,\omega)e^{i\alpha_1(k,\omega)D}}{1+r_{12}(k,\omega)r_{01}(k,\omega)e^{2i\alpha_1(k,\omega)D}}. \tag{2.18}$$

For the second harmonic field the nonlinear driving source $Q(q,2\omega)$ has three contributions,

$$Q(q,2\omega) = Q^{(p)}(q,2\omega) + Q^{(v)}(q,2\omega) + Q^{(b)}(q,2\omega), \tag{2.19}$$

where $Q^{(p)}(q,2\omega)$ is associated with the prism–metal film interface

$$Q^{(p)}(q,2\omega) = it_{10}(q,2\omega)e^{i\alpha_1(q,2\omega)D}\left(\frac{\alpha_0(q,2\omega)}{\epsilon_0}A^{(p)}(q) + B^{(p)}(q)\right), \tag{2.20a}$$

$Q^{(v)}(q,2\omega)$ is associated with the metal film-crystal interface

$$\begin{aligned} Q^{(v)}(q,2\omega) = i\int_{-\infty}^{\infty}\frac{dp}{2\pi}\Bigg\{\frac{1}{\epsilon_1(2\omega)}\left[\alpha_1(q,2\omega) + \frac{q(q-p)}{\alpha_1(q,2\omega)}\right]A^{(v)}(p)\big[I(-\alpha_1(q,2\omega)|q-p) \\ + r_{01}(q,2\omega)e^{2i\alpha_1(q,2\omega)D}I(\alpha_1(q,2\omega)|q-p)\big] + iB^{(v)}(p)\big[I(-\alpha_1(q,2\omega)|q-p) \\ - r_{01}(q,2\omega)e^{2i\alpha_1(q,2\omega)D}I(\alpha_1(q,2\omega)|q-p)\big]\Bigg\}, \end{aligned} \tag{2.20b}$$

and $Q^{(b)}(q,2\omega)$ arises from the nonlinearity of the crystal

$$\begin{aligned} Q^{(b)}(q,2\omega) = -i\int_{-\infty}^{\infty}\frac{dp}{2\pi}\Bigg\{\frac{1}{\epsilon_1(2\omega)}\left(\alpha_1(q,2\omega) + \frac{q(q-p)}{\alpha_1(q,2\omega)}\right)\mathscr{P}(p)\big[I(-\alpha_1(q,2\omega)|q-p) \\ + r_{01}(q,2\omega)I(\alpha_1(q,2\omega)|q-p)\big] + \frac{1}{\epsilon_2(2\omega)}\mathscr{P}_1(p)\big[I(-\alpha_1(q,2\omega)|q-p) \\ - r_{01}(q,2\omega)I(\alpha_1(q,2\omega)|q-p)\big]\Bigg\}, \end{aligned} \tag{2.21}$$

where $\mathscr{P}(p)$ and $\mathscr{P}_1(p)$ are the Fourier components of the driving term in the nonlinear medium and its normal derivative evaluated at the rough interface, and are given by

$$\mathscr{P}(p) = -\frac{2cd_{11}}{\omega\epsilon_2^2(\omega)}\int_{-\infty}^{\infty}\frac{dr}{2\pi}\int_{-\infty}^{\infty}\frac{dr'}{2\pi}I(\alpha_1(r,\omega) + \alpha_1(r-r',\omega)|p-r)F(r,r')T(r-r',\omega)T(r',\omega), \tag{2.22}$$

and

$$\begin{aligned} \mathscr{P}_1(p) = i\frac{2cd_{11}}{\omega\epsilon_2^2(\omega)}\int_{-\infty}^{\infty}\frac{dr}{2\pi}\int_{-\infty}^{\infty}\frac{dr'}{2\pi}I(\alpha_1(r,\omega) + \alpha_1(r-r',\omega)|p-r)\Bigg[\alpha_1(r,\omega) + \alpha_1(r-r',\omega) \\ - \frac{r(p-r)}{\alpha_1(r,\omega) + \alpha_1(r-r',\omega)}\Bigg]F(r,r')T(r-r',\omega)T(r',\omega). \end{aligned} \tag{2.23}$$

The solution of the reduced Rayleigh equation (2.11) and explicit expressions for the nonlinear driving term are presented in Appendices A and B, respectively.

The quantity we are interested in is the intensity of the second harmonic light, which we define as the total power of the harmonic light generated diffusely, normalized by the square of the power of the incident light and multiplied by the illuminated area S:

$$p_{2\omega} = \frac{P_{2\omega}}{(P_{\text{in}})^2}S, \tag{2.24}$$

where $S = L_1 L_2$, and L_1 and L_2 are the lengths of the surface along the x_1- and x_2-axes. If we define the angle of transmission θ_t explicitly by setting $q = (\sqrt{\epsilon_2(2\omega)}\, 2\omega/c)\sin\theta_t$, so that $\alpha_2(q,2\omega) = (\sqrt{\epsilon_2(2\omega)}\, 2\omega/c)\cos\theta_t$, the mean normalized intensity of second harmonic light takes the form

$$\langle I(\theta_t|2\omega)\rangle_{\mathrm{incoh}} = \frac{8\omega\epsilon_0}{L_1 c^2 |H_0|^4} \frac{\cos^2\theta_t}{\cos^2\theta_0} \left[\langle |T(q,2\omega)|^2 \rangle - |\langle T(q,2\omega)\rangle|^2 \right]. \tag{2.25}$$

3. Results and discussion

In Fig. 2(a), (b), and (c) we present the mean intensity of the second harmonic light, calculated by the use of the small-amplitude perturbation approach, when p-polarized light is incident through a prism on a one-dimen-

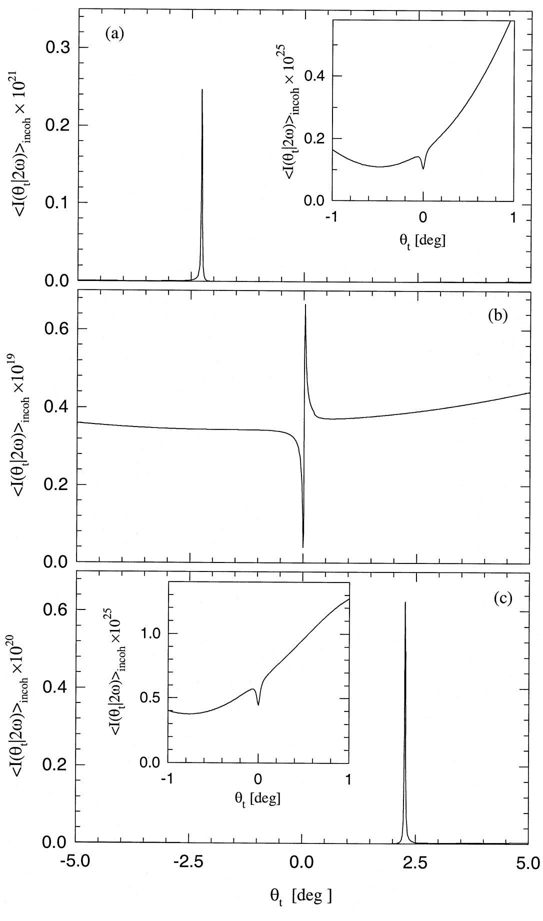

Fig. 2. The mean differential intensity of the second harmonic light as a function of the angle of transmission θ_t for the scattering of p-polarized light through a randomly rough silver film-vacuum interface. The interface roughness is characterized by the Gaussian power spectrum Eq. (2.5) with $\delta = 5$ nm and $a = 113.5$ nm. The nonlinear coefficients are given by the free-electron model. The angles of incidence are (a) $\theta_0 = 22°$, (b) $\theta_0 = 24°$, and (c) $\theta_0 = 26°$. The optimal angle for excitation of surface plasmon polaritons at the film-vacuum interface is 24°.

sional, random silver film-vacuum interface, characterized by the Gaussian power spectrum (2.5) with an rms height $\delta = 5$ nm and a transverse correlation length $a = 113.5$ nm. The mean thickness of the film is $D = 55$ nm, and the angles of incidence are $\theta_0 = 22°$ (a), $\theta_0 = 24°$ (b), and $\theta_0 = 26°$ (c). The refractive index of the prism is $n_0 = 2.479$, so that the optimal angle for the excitation of surface plasmon polaritons is $\theta_0 = 24°$. The nonlinear coefficients $\mu_{1,2,3}^{(p,v)}$ were calculated on the basis of the free-electron model [19].

In Fig. 3(a), (b), and (c) we present the mean intensity of the second harmonic light, calculated by the perturbative approach, when p-polarized light is incident through a prism on a one-dimensional, random silver film-quartz interface, characterized by the Gaussian power spectrum (2.5) with an rms height $\delta = 5$ nm and a transverse correlation length $a = 113.5$ nm. The mean thickness of the film is $D = 55$ nm, and the angles of incidence are $\theta_0 = 37.2°$ (a), $\theta_0 = 39.2°$ (b), and $\theta_0 = 41.2°$ (c). The refractive indices of the quartz are $n_2(\omega) = \sqrt{\epsilon_2(\omega)} = 1.536$ and $n_2(2\omega) = \sqrt{\epsilon_2(2\omega)} = 1.542$, and the optimal angle for the excitation of surface plasmon polaritons is $\theta_0 = 39.2°$.

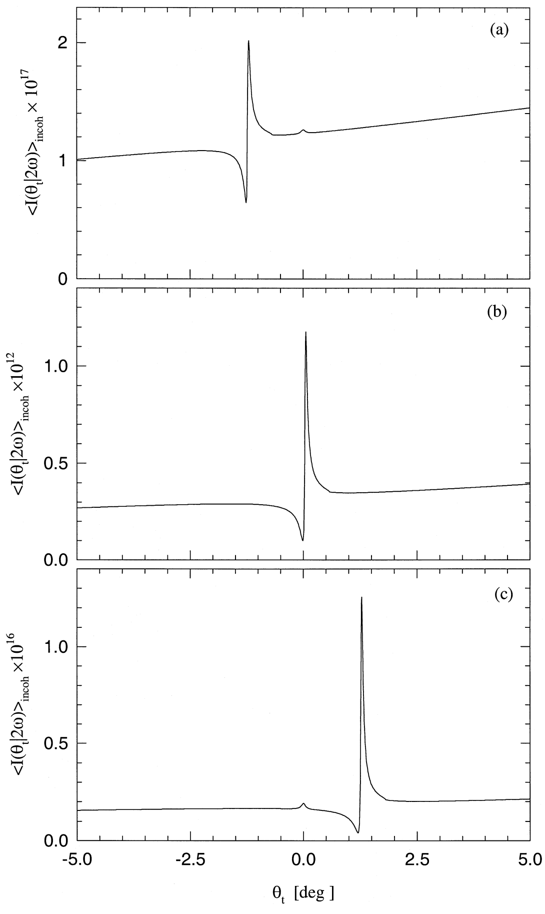

Fig. 3. The mean differential intensity of the second harmonic light as a function of the angle of transmission θ_t for the scattering of p-polarized light from a randomly rough silver film-nonlinear quartz interface, whose roughness is characterized by the Gaussian power spectrum Eq. (2.5) with $\delta = 5$ nm and $a = 113.5$ nm. The nonlinear susceptibility of the crystal is $d_{11} = 0.93 \times 10^{-9}$ esu. The angles of incidence are (a) $\theta_0 = 37.2°$, (b) $\theta_0 = 39.2°$, and (c) $\theta_0 = 41.2°$. The optimal angle for excitation of surface plasmon polaritons at the film-quartz interface is 39.2°.

To analyze the main features of the angular dependence of the intensity of the generated light we need explicit expressions for the different contributions to the mean differential intensity of the harmonic light. The effects of the multiple scattering of surface plasmon polaritons of frequency ω, which are of interest to us here, are contained in the the nonlinear driving term $Q(q,\Omega)$ on the right hand side of Eq. (2.11) evaluated at $\Omega = 2\omega$. The integral term in Eq. (A.13) of Appendix A describes the multiple scattering of the waves of frequency 2ω and, in particular, contains the effects of the multiple scattering of surface plasmon polaritons of frequency 2ω. Since these effects do not include the peak in the direction normal to the mean surface, which is of primary interest to us here, we omit their contribution to the mean differential intensity $\langle I(\theta_t|2\omega)\rangle_{\mathrm{incoh}}$. Since all three contributions to $Q(q,2\omega)$, Eq. (2.19), contain the product of the transmission amplitudes $T(p,\omega)T(p',\omega)$, by using the expression for the transmission amplitude $T(q,\omega) = \mathscr{T}_0(k,\omega)H_0[2\pi\delta(q-k) + G(q,\omega)t(q|k)]$ (see, Eq. (A.9)) we can subdivide each term in $Q(q,2\omega)$ into three contributions with different physical meanings. The first contributions to $Q^{(p,v,b)}(q,2\omega)$, which contain only the product of δ-functions, $(2\pi)^2\delta(p-k)\delta(p'-k)$, describe the nonlinear mixing of the fields of frequency ω which would be specular if the film-crystal interface were planar. The parts of $Q^{(p,v,b)}(q,2\omega)$ that contain the product of a δ-function and a term with the Green's function, $2\pi\delta(p-k)G(p',2\omega)t(p'|k) + 2\pi\delta(p'-k)G(p,2\omega)t(p|k)$, describe the interaction of the 'specular' and scattered fields, including the nonlinear mixing of the excited surface plasmon polaritons with the incident light. Finally, the parts of $Q^{(p,v,b)}(q,2\omega)$ which contain the product of two Green's functions, $G(p,2\omega)t(p|k)G(p',2\omega)t(p'|k)$, describe the nonlinear mixing of the scattered fields, and include the mixing of co- and contrapropagating surface plasmon polaritons. According to the classification we have just given we separate explicitly these three contributions to the nonlinear driving source, and include all the roughness induced corrections to them into the additional source function $\mathscr{Q}_{\mathrm{sc}}(q,2\omega)$, so that we obtain

$$Q(q,2\omega) = \frac{2ic}{\omega}\mathscr{T}_0^2(k,\omega)H_0^2\Big[\Gamma_0(k)2\pi\delta(q-2k) + \Gamma_1(q,k)G(q-k)t(q-k|k)$$
$$+\int_{-\infty}^{\infty}\frac{dp}{4\pi}\Gamma_2(q,p)G(q-p,\omega)G(p,\omega)t(q-p|k)t(p|k) + \mathscr{Q}_{\mathrm{sc}}(q,2\omega)\Big], \qquad (3.1)$$

where the nonlinear coefficients Γ_i are given by $\Gamma_i = \gamma_i^{(p)} + \gamma_i^{(v)} + \gamma_i^{(b)}$, and the explicit expressions for $\gamma_i^{(p,v,b)}$ are presented in Appendix B. The most resonant contributions to the intensity of the harmonic light have the form

$$\langle I(\theta_s|2\omega)\rangle_{\mathrm{res}} = \frac{512\omega^3}{\epsilon_0 c^4}\left|t_{10}(k,\omega)e^{2i\alpha_1(k,\omega)D}\right|^2\cos^2\theta_s\cos^2\theta_0|G(q,2\omega)|^2|G(k,\omega)|^4|$$
$$\times\Big[|\Gamma_1(q,k)|^2|G(q-k)|^2\tau(q-k|k)$$
$$+\int_{-\infty}^{\infty}\frac{dp}{2\pi}|\Gamma_2(q,p)|^2|G(q-p,\omega)G(p,\omega)|^2\tau(q-p|k)\tau(p|k)\Big], \qquad (3.2)$$

where we have defined $\langle t(q|k)t^*(p|k)\rangle = 2\pi\delta(q-p)\tau(q|k)$, and $\tau(q|k)$ is the averaged reducible vertex function that can be calculated by using, for example, the pole approximation for the Green's function [4].

The presence of the Green's functions $|G(q,2\omega)|^2$ and $|G(k,\omega)|^4$ in Eq. (3.2) leads to the amplification of nonlinear processes at the metal film-crystal interface in the Kretschmann geometry [23], because the Green's function $G(q,2\omega)$ has poles at the wavenumbers of the surface plasmon polaritons, $k_{sp}(2\omega)$, and $G(k,\omega)$ has poles at the wavenumbers of the surface plasmon polaritons, $k_{sp}(\omega)$. Thus, when the angle of incidence is the optimal one for the excitation of surface plasmon polaritons the enhancement of the intensity of the second harmonic light can reach several orders of magnitude compared with that for scattering from a single surface. This is true for both the coherent and incoherent parts of the intensity. When the angle of scattering is the optimal one for the detection of surface plasmon polaritons of frequency 2ω resonance enhancement of the intensity can also occur [24]. However, this enhancement can be observed only in the intensity of the reflected light.

The second interesting feature of Eq. (3.2) is the presence of the term that contains the factor $|G(q-k,\omega)|^2$. This term describes the second harmonic generation due to the nonlinear mixing of the incident light with the surface plasmon polaritons which are excited due to the surface roughness. Its presence leads to the appearance of strong resonant peaks at $q=k\pm k_{sp}(\omega)$ and $q=k\pm k_{sp}^{(p)}(\omega)$. This is the origin of the peak of the strong enhancement of second harmonic generation in reflection from a randomly rough metal surface that was first observed in [16], and analyzed by Deck and Grygier [17] in the framework of the first order perturbation theory in the small roughness. These peaks of the enhanced second harmonic generation are due to the resonant interaction of the incident light and forward/backward propagating surface plasmon polaritons excited in a single scattering event. No interference effects are involved in the formation of the peaks. The main reason for using the Kretschmann geometry in the experiments [6,8–11] was to excite surface plasmon polaritons associated with the metal–vacuum (metal–nonlinear crystal) interface through the ATR phenomenon. Thus, the angle of incidence was tuned so that $k=k_{sp}(\omega)$. In this case the peaks due to the poles of $G(q-k,\omega)$ move so that the peak due to the mixing of the incident light with the forward propagating surface plasmon polariton moves into the nonradiative region, while that due to the mixing of the incident light with the backward propagating surface plasmon polariton moves to the direction of the normal to the mean surface, $q=k-k_{sp}(\omega)=0$. The strength and the shape of the peak is determined by the effective nonlinear coefficient $\Gamma_1(q,k)$. In the case when the nonlinearity of the system is due to the film interfaces only, this effective nonlinear coefficient is linear in q for small q. This is the result of the symmetry of the surface nonlinear polarization that forbids second harmonic generation by contrapropagating surface plasmon polaritons [14]. Since in the case where the angle of incidence is optimal for exciting surface plasmon polaritons the incident light is the surface plasmon polariton propagating in the forward direction, its nonlinear mixing with the scattered surface plasmon polaritons propagating in the backward direction is forbidden by this symmetry. As a result, the resonant peak has an antiresonant shape. The width of the peak is determined by the decay rate of the surface plasmon polaritons on the rough interface $\mathrm{Im}(k_{sp}(\omega))=\Delta_{tot}(\omega)=\Delta_{\epsilon}(\omega)+\Delta_{sc}(\omega)$, where $\Delta_{\epsilon}(\omega)$ is the decay rate of the surface plasmon polaritons of frequency ω due to ohmic losses, while $\Delta_{sc}(\omega)$ is their decay rate due to their roughness-induced scattering into other surface plasmon polaritons.

The effects of the multiple scattering of surface plasmon polaritons are contained in the function $\tau(q|k)$. As was shown in [4,25,26], because our scattering system supports two surface plasmon polaritons at the frequency ω, the function $\tau(q|k)$ contains a superposition of two Lorentzian peaks centered at $q+k=0$ (enhanced backscattering) with halfwidths $\Delta_{tot}(\omega)$and $\Delta_{tot}^{(p)}(\omega)$. It also contains Lorentzian peaks centered at $q+k\pm(k_{sp}(\omega)-k_{sp}^{(p)}(\omega))=0$ (satellite peaks) with a half-width $\Delta_{tot}(\omega)+\Delta_{tot}^{(p)}(\omega)$. In these results $\Delta_{tot}^{(p)}(\omega)$ is the decay rate of the surface plasmon polariton associated with the prism–metal interface. Therefore, $\tau(q-k|k)$ entering the second term in Eq. (3.2) has a peak at $q=0$. Since this contribution arises due to the nonlinear mixing of the incident and scattered waves, it describes the coherent generation of the second harmonic light by the incident and backscattered radiation, enhanced due to the multiple scattering of surface plasmon polaritons by the roughness.

The strongest contribution associated with the multiple scattering of surface plasmon polaritons of frequency ω comes, however, from the second term in Eq. (3.2), which describes the nonlinear mixing of the multiply-scattered surface plasmon polaritons of frequency ω propagating in opposite directions. In the pole approximation for the Green's function [4,25] this contribution has the form

$$\begin{aligned}\langle I(\theta_s|2\omega)\rangle_{res} = {} & \frac{512\,\omega^3}{\epsilon_0 c^4}\left|t_{10}(k,\omega)e^{2i\alpha_1(k,\omega)D}\right|^2|G(q,2\omega)|^2|G(k,\omega)|^2\left\{\frac{C^4(\omega)}{\Delta_{tot}(\omega)}\frac{1}{q^2+4\Delta_{tot}^2(\omega)}\right.\\ & \times\Big[|\Gamma_2(q,k_{sp}(\omega))|^2\tau(k_{sp}(\omega)|k)\tau(q-k_{sp}(\omega)|k)+|\Gamma_2(q,-k_{sp}(\omega))|^2\tau(-k_{sp}(\omega)|k)\\ & \times\tau(q+k_{sp}(\omega)|k)\Big]+C_p^4(\omega)\Delta_{tot}^{(p)}(\omega)\frac{1}{q^2+4(\Delta_{tot}^{(p)}(\omega))^2}\Big[|\Gamma_2(q,k_{sp}^{(p)}(\omega))|^2\\ & \times\tau(k_{sp}^{(p)}(\omega)|k)\tau(q-k_{sp}^{(p)}(\omega)|k)+|\Gamma_2(q,-k_{sp}^{(p)}(\omega))|^2\\ & \left.\times\tau(-k_{sp}^{(p)}(\omega)|k)\tau(q+k_{sp}^{(p)}(\omega)|k)\Big]\right\}, \qquad (3.3)\end{aligned}$$

where $C(\omega)$ and $C_p(\omega)$ are the residues of the Green's function at the poles $\pm k_{sp}(\omega)$ and $\pm k_{sp}^{(p)}(\omega)$, respectively. We have not included in Eq. (3.3) the contribution from the possible satellite peaks [26], since their positions are far from the direction normal to the mean surface. As expected, the result given by Eq. (3.3) contains Lorentzian factors centered at $q=0$. However, the efficiency of the nonlinear mixing of the contrapropagating surface plasmon polaritons is determined by the effective nonlinear coefficients $\Gamma_2(q,\pm k_{sp}(\omega))$ and $\Gamma_2(q,\pm k_{sp}^{(p)}(\omega))$. In the case where the nonlinearity of the system is due to the film interfaces only, these effective nonlinear coefficients are linear in q for small q. This is the manifestation of the well known fact that the symmetry of the nonlinear polarization of a metal surface forbids such processes [14]. As a result, the contribution given by Eq. (3.3) displays a dip rather than a peak in the direction normal to the mean surface. The depth of this dip depends strongly on the values of the material parameters and the angle of incidence of the fundamental light. However, when the metal film is in contact with a nonlinear quartz crystal, a suitable choice of the orientation of the crystal [18] makes possible second harmonic generation by contrapropagating surface plasmon polaritons, and a peak of the enhanced second harmonic generation occurs in the direction normal to the mean surface.

Recently the first experimental studies of multiple-scattering effects in the second harmonic generation of light scattered from a clean one-dimensional vacuum-metal interface were carried out in a series of papers by O'Donnell and his colleagues [27–29]. It was found that, for weakly rough silver surfaces a dip is present in the retroreflection direction in the angular dependence of the intensity of the scattered second harmonic light rather than the peak that occurs in scattering at the fundamental frequency [27,29], while no well-defined peak or dip in the direction normal to the mean surface was observed [28]. In the latter experiment the random surfaces were fabricated in a special way to produce a strong excitation of surface plasmon polaritons of the fundamental frequency, propagating in both the forward and backward directions. The results of Ref. [28] suggest a surface nonlinear polarization of a form different from those usually used in studies of second harmonic generation in scattering from an isotropic metal surface. The possible anisotropy of the surface could be the reason for the absence of the dip in the direction normal to the mean surface. If, for some reason, the nonlinear surface polarization is such that no peak or dip in the direction normal to the mean surface, which is due to the multiple scattering of surface plasmon polaritons by the surface roughness, can be formed, in this case in experiments in the Kretschmann geometry the resonant peak at $q=0$ will also be absent. Finally, the different qualitative results of [8,11] and [10] suggest that the mechanism for the nonlinear polarization might be different for the strongly rough surfaces studied in [8,11] and for the weakly rough surfaces studied in [10]. As an example, in Fig. 4 we present the mean intensity of the second harmonic light, calculated by the small-amplitude perturbation approach, assuming that the surface nonlinearity arises from a thin subsurface layer of the metal

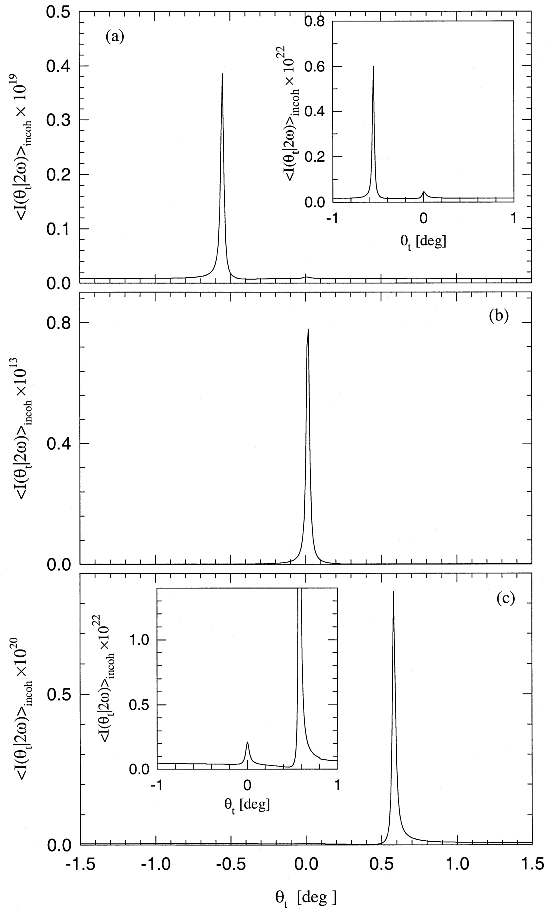

Fig. 4. The mean differential intensity of the second harmonic light as a function of the angle of transmission θ_t for the transmission of p-polarized light through the same film as in Fig. 2, but for the case where the surface nonlinear polarization has an artificial nature that is described in the text. The inset presents the contribution to $\langle I(\theta_t|2\omega)\rangle_{incoh}$ from double-scattering processes only. The angles of incidence are (a) $\theta_0 = 23.5°$, (b) $\theta_0 = 24°$, and (c) $\theta_0 = 24.5°$. The optimal angle for excitation of surface plasmon polaritons at the film-vacuum interface is 24°.

film, or from a monolayer of nonlinear molecules covering the metal film. In either case the surface nonlinear polarization is associated not with the surface itself but is given by the 'bulk' nonlinear polarization of the layer, which is assumed to be given by Eq. (2.6). In this case both the nonlinear mixing of the incident light with the backward propagating surface plasmon polariton and the nonlinear mixing of the multiply-scattered surface plasmon polaritons lead to the appearance of the peak in transmission in the direction normal to the mean surface.

4. Conclusions

In this work we have discussed the second harmonic generation of light in transmission in the Kretschmann ATR geometry, in the case where a metal film-vacuum or a metal film-nonlinear quartz crystal interface is weakly rough. We have analyzed the origin of the peak of the enhanced second harmonic generation observed in the direction normal to the mean surface in [6,8,11]. We have shown that in the case where the metal film is in contact with vacuum, so that the nonlinearity of the film surfaces gives rise to the harmonic light, the interference effects in the multiple scattering of surface plasmon polaritons lead, due to the symmetry of the nonlinear polarization, to the appearance of a dip rather than a peak in the direction normal to the mean surface. When the second harmonic generation is due to the nonlinearity of the crystal adjacent to the metal film a peak of the enhanced second harmonic generation occurs in this direction.

However, when the angle of incidence is chosen to be optimal for the excitation of surface plasmon polaritons localized at the film-vacuum/nonlinear crystal interface, a much stronger mechanism leads to the appearance of a peak of the enhanced second harmonic generation in the direction normal to the mean interface. It is the nonlinear mixing of the incident light, which in this case is the resonantly excited surface plasmon polariton, and the backward-propagating surface plasmon polariton excited through the roughness that forms the peak. No interference effects are involved in the formation of the peak. Since this peak appears already in the single-scattering processes, it is considerably stronger than the weak features associated with multiple scattering. When the angle of incidence is shifted from the optimal one, the peak moves away from the direction normal to the mean interface, so that the weak peak/dip which is due to the multiple-scattering effects, can be observed. However, the efficiency of second harmonic generation decreases considerably as the angle of incidence shifts from the optimal one.

Acknowledgements

The work of A.A.M., I.V. N., and T.A.L. was supported by Army Research Office Grant DAAG 55-98-C-0034. The work of E.R.M., and M. L.-L. was supported by CONACYT Grant 32254–E.

Appendix A.

To solve the linear scattering problem we seek the solution of Eq. (2.11) in the form

$$T(q,\omega) = -2iH_0 G_\omega(q|k)\frac{\alpha_1(k,\omega)}{\epsilon_1(\omega)}t_{01}(k,\omega)e^{i\alpha_1(k,\omega)D}, \tag{A.1}$$

where we have introduced the Green's function $G_\omega(q|k)$ associated with the randomly rough interface between the vacuum and the metal that is the solution of the equation

$$G_\omega(q|k) = G(q,\omega)2\pi\delta(q-k) + G(q,\omega)t(q|k)G(k,\omega). \tag{A.2}$$

In Eq. (A.2) $G(q,\omega)$, defined by $\langle G_\omega(q|k)\rangle = 2\pi\delta(q-k)G(q,\omega)$, is the averaged Green's function and is given by

$$G(q,\omega) = 2\pi\delta(q-k)\frac{1}{G_0^{-1}(k,\omega) - M(k,\omega)}, \tag{A.3}$$

where $M(k,\omega)$ is the averaged proper self-energy. The latter is given by

$$\langle M(q|k)\rangle = 2\pi\delta(q-k)M(k,\omega), \tag{A.4}$$

where the (unaveraged) proper self-energy $M(q|k)$ is the solution of [25]

$$M(q|k) = V(q,k|\omega) + \int_{-\infty}^{\infty}\frac{dp}{2\pi}M(q|p)G(p,\omega)W(p,k|\omega), \tag{A.5}$$

and we have introduced the notation

$$W(q,k|\omega) = V(q,k|\omega) - \langle M(q|k)\rangle . \tag{A.6}$$

The operator $t(p|r)$ in Eq. (A.2) was introduced to satisfy

$$\int_{-\infty}^{\infty}\frac{dp}{2\pi}W(q,p|\omega)G(p|k,\omega) = t(q|k)G(k,\omega), \tag{A.7}$$

and is the solution of the equation

$$t(q|k) = W(q,k|\omega) + \int_{-\infty}^{\infty}\frac{dp}{2\pi}\int_{-\infty}^{\infty}\frac{dr}{2\pi}W(q,p|\omega)G(p,\omega)t(p|k). \tag{A.8}$$

From Eq. (A.2) it follows that $\langle t(q|k)\rangle = 0$. The transmission amplitude $T(q,\omega)$ in terms of the averaged Green's function $G(p,\omega)$ and the operator $t_\omega(q|k)$ is then given by

$$T(q,\omega) = \mathscr{T}_0(k,\omega)H_0[2\pi\delta(q-k) + G(q,\omega)t(q|k)], \tag{A.9}$$

where

$$\mathscr{T}_0(k,\omega) = -2i\frac{\alpha_0(k,w}{\epsilon_0}t_{10}(k,\omega)e^{i\alpha_1(k,\omega)D}G(k,\omega). \tag{A.10}$$

Having in hand the solution for $T(q,\omega)$ we can calculate the fields of frequency ω in the prism and in the metal film and, consequently, the nonlinear driving term of frequency 2ω.

In the same manner we can seek the solution of the nonlinear scattering problem. If we now introduce a new unknown function $S(q|2k)$ by the equation

$$T(q,2\omega) = iG_0(p,2\omega)Q(q,2\omega) + iG_0(q,2\omega)\int_{-\infty}^{\infty}\frac{dq}{2\pi}S(p|q)G_0(q,2\omega)Q(q,2\omega), \tag{A.11}$$

it follows from Eq. (2.11) that the function $S(q|p)$ satisfies the equation

$$S(q|p) = V(q|p,2\omega) + \int_{-\infty}^{\infty}\frac{dr}{2\pi}V(q|r,2\omega)G_0(r,2\omega)S(r|p). \tag{A.12}$$

The amplitude $S(q|p)$ describes the multiple scattering of electromagnetic waves of frequency 2ω, including the scattering of surface plasmon polaritons of frequency 2ω. It can be rewritten in terms of the Green's function associated with the rough interface of the metal film exactly as this was done for the fundamental fields. As a result we obtain the following expression for the transmission amplitude

$$T(p,2\omega) = iG(p,2\omega)Q(p,2\omega) + iG(p,2\omega)\int_{-\infty}^{\infty}\frac{dq}{2\pi}t_{2\omega}(p|q)G(q,2\omega)Q(q,2\omega). \tag{A.13}$$

Appendix B.

In this appendix we present the explicit expression for the effective nonlinear coefficients $\gamma_i^{(p,v,b)}$.

$$\gamma_0^{(p)}(k) = t_{10}(2k,2\omega)e^{i\alpha_1(2k,2\omega)D}\left\{\mu_3^{(p)}k\frac{\alpha_0(2k,2\omega)}{\epsilon_0}\frac{\alpha_1(k,\omega)}{\epsilon_1(w)}f_+(k)f_-(k)\right.$$
$$\left. -2k\left[\mu_1^{(p)}k^2f_+^2(k) + \mu_2^{(p)}\left(\frac{\alpha_1(k,\omega)}{\epsilon_1(\omega)}\right)^2 f_-^2(k)\right]\right\} \tag{B.1}$$

$$\gamma_1^{(p)}(q,k) = t_{10}(q,2\omega)e^{i\alpha_1(q,2\omega)D}\left\{\mu_3^{(p)}\frac{\alpha_0(q,2\omega)}{\epsilon_0}\left[k\frac{\alpha_1(q-k,\omega)}{\epsilon_1(w)}f_+(k)f_-(q-k)\right.\right.$$
$$\left.+(q-k)\frac{\alpha_1(k,\omega)}{\epsilon_1(w)}f_+(q-k)f_-(k)\right]-2q\left[\mu_1^{(p)}(q-k)kf_+(q-k)f_+(k)\right.$$
$$\left.\left.+\mu_2^{(p)}\frac{\alpha_1(k,\omega)}{\epsilon_1(\omega)}\frac{\alpha_1(q-k,\omega)}{\epsilon_1(\omega)}f_-(q-k)f_+(k)\right]\right\} \tag{B.2}$$

$$\gamma_2^{(p)}(q,p) = t_{10}(q,2\omega)e^{i\alpha_1(q,2\omega)D}\left\{\mu_3^{(p)}\frac{\alpha_0(q,2\omega)}{\epsilon_0}\left[p\frac{\alpha_1(q-p,\omega)}{\epsilon_1(w)}f_+(p)f_-(q-p)\right.\right.$$
$$\left.+(q-p)\frac{\alpha_1(p,\omega)}{\epsilon_1(w)}f_+(q-p)f_-(p)\right]-2q\left[\mu_1^{(p)}(q-p)pf_+(q-p)f_+(p)\right.$$
$$\left.\left.+\mu_2^{(p)}\frac{\alpha_1(q-p,\omega)}{\epsilon_1(\omega)}\frac{\alpha_1(p,\omega)}{\epsilon_1(\omega)}f_-(q-p)f_+(p)\right]\right\}, \tag{B.3}$$

with

$$f_\pm(q) = \frac{1 \pm r_{12}(q,\omega)e^{2i\alpha_1(q,\omega)D}}{t_{12}(q,\omega)e^{i\alpha_1(q,\omega)D}} \tag{B.4}$$

and

$$\gamma_0^{(v)}(k) = \mu_3^{(v)}k\frac{\alpha_1(2k,2\omega)}{\epsilon_1(2\omega)}\frac{\alpha_2(k,\omega)}{\epsilon_2(w)}g_+(2k) - 2kg_-(2k)\left[\mu_1^{(v)}k^2 + \mu_2^{(v)}\left(\frac{\alpha_2(k,\omega)}{\epsilon_2(\omega)}\right)^2\right], \tag{B.5}$$

$$\gamma_1^{(v)}(q,k) = \mu_3^{(v)}\frac{\alpha_1(q,2\omega)}{\epsilon_1(2\omega)}g_+(q)\left[k\frac{\alpha_2(q-k,\omega)}{\epsilon_2(w)} + (q-k)\frac{\alpha_2(k,\omega)}{\epsilon_2(w)}\right]$$
$$-2qg_-(q)\left[\mu_1^{(v)}(q-k)k + \mu_2^{(v)}\frac{\alpha_2(k,\omega)}{\epsilon_2(\omega)}\frac{\alpha_2(q-k,\omega)}{\epsilon_2(\omega)}\right] \tag{B.6}$$

$$\gamma_2^{(v)}(q,p) = \mu_3^{(v)}\frac{\alpha_1(q,2\omega)}{\epsilon_1(2\omega)}g_+(q)\left[p\frac{\alpha_2(q-p,\omega)}{\epsilon_2(w)} + (q-p)\frac{\alpha_2(p,\omega)}{\epsilon_2(w)}\right]$$
$$-2qg_-(q)\left[\mu_1^{(v)}(q-p)p + \mu_2^{(v)}\frac{\alpha_2(q-p,\omega)}{\epsilon_2(\omega)}\frac{\alpha_2(p,\omega)}{\epsilon_2(\omega)}\right], \tag{B.7}$$

with

$$g_\pm(q) = 1 \pm r_{01}(q,2\omega)e^{2i\alpha_1(q,2\omega)D}. \tag{B.8}$$

$$\gamma_0^{(b)}(k) = \frac{id_{11}}{\epsilon_2^2(\omega)}\left\{\frac{\alpha_1(2k,2\omega)}{\epsilon_1(2\omega)}\left(1 + r_{01}(2k,2\omega)e^{2i\alpha_1(2k,2\omega)D}\right)\right.$$
$$\left.+\frac{2\alpha_2(k,\omega)}{\epsilon_2(2\omega)}\left(1 - r_{01}(2k,2\omega)e^{2i\alpha_1(2k,2\omega)D}\right)\right\}F(2k,k), \tag{B.9}$$

$$\gamma_1^{(b)}(q,k) = \frac{id_{11}}{\epsilon_2^2(\omega)} \left\{ \frac{\alpha_1(q,2\omega)}{\epsilon_1(2\omega)} \left(1 + r_{01}(q,2\omega)\, e^{2i\alpha_1(q,2\omega)D}\right) + \frac{\alpha_2(k,\omega) + \alpha_1(q-k,\omega)}{\epsilon_2(2\omega)} \left(1 - r_{01}(q,2\omega)\, e^{2i\alpha_1(q,2\omega)D}\right) \right\} [F(q,k) + F(q,q-k)], \tag{B.10}$$

$$\gamma_2^{(b)}(q,p) = \frac{id_{11}}{\epsilon_2^2(\omega)} \left\{ \frac{\alpha_1(q,2\omega)}{\epsilon_1(2\omega)} \left(1 + r_{01}(q,2\omega)\, e^{2i\alpha_1(q,2\omega)D}\right) + \frac{\alpha_2(p,\omega) + \alpha_1(q-p,\omega)}{\epsilon_2(2\omega)} \left(1 - r_{01}(q,2\omega)\, e^{2i\alpha_1(q,2\omega)D}\right) \right\} F(q,p). \tag{B.11}$$

References

[1] F. Brown, R.E. Parks, A.M. Sleeper, Phys. Rev. Lett. 14 (1965) 1029.
[2] S.S. Jha, Phys. Rev. Lett. 15 (1965) 412.
[3] N. Bloembergen, R.K. Chang, S.S. Jha, C.H. Lee, Phys. Rev. 74 (1968) 813.
[4] A.R. McGurn, A.A. Maradudin, V. Celli, Phys. Rev. B 31 (1985) 4866.
[5] A.R. McGurn, T.A. Leskova, V.M. Agranovich, Phys. Rev. B 44 (1991) 11441.
[6] X. Wang, H.J. Simon, Opt. Lett. 16 (1991) 1475.
[7] H.J. Simon, Y. Wang, L.B. Zhou, Z. Chan, Opt. Lett. 17 (1992) 1268.
[8] O.A. Aktsipetrov, V.N. Golovkina, O.I. Kapusta, T.A. Leskova, N.N. Novikova, Phys. Lett. A 170 (1992) 231.
[9] Y. Wang, H.J. Simon, Phys. Rev. B 47 (1993) 13695.
[10] L. Kuang, H.J. Simon, Phys. Lett. A 197 (1995) 257.
[11] S.I. Bozhevolnyi, K. Pedersen, Surface Science 377–379 (1997) 384.
[12] E. Kretschmann, Z. Physik 241 (1971) 313.
[13] D. Sarid, Phys. Rev. Lett. 47 (1981) 1927.
[14] M. Fukui, J.E. Sipe, V.C.Y. So, G.I. Stegeman, Solid State Commun. 27 (1978) 1265.
[15] H.J. Simon, D.E. Mitchell, J.G. Watson, Phys. Rev. Lett. 33 (1974) 1531.
[16] C.K. Chen, A.R.B. de Castro, Y.R. Shen, Phys. Rev. Lett. 46 (1981) 145.
[17] R.T. Deck, R.K. Grygier, Appl. Opt. 23 (1984) 3202.
[18] C.K. Chen, A.R.B. de Castro, Y.R. Shen, Opt. Lett. 4 (1979) 393.
[19] I.V. Novikov, A.A. Maradudin, T.A. Leskova, E.R. Méndez, M. Leyva-Lucero (unpublished work).
[20] Lord Rayleigh, The Theory of Sound, 2nd ed, vol. II, MacMillan, London, 1895, pp. 89 and 297–311.
[21] R. Petit, M. Cadilhac, C.R. Acad. Sci. Paris B 262 (1966) 468.
[22] N.R. Hill, V. Celli, Phys. Rev. B 17 (1978) 2478.
[23] Y.J. Chen, E. Burstein, Nuovo Cimento 39B (1977) 807.
[24] H.J. Simon, R.E. Benner, J.G. Rako, Opt. Commun. 23 (1977) 245.
[25] G.C. Brown, V. Celli, M. Haller, A.A. Maradudin, A. Marvin, Phys. Rev. B 31 (1985) 4993.
[26] See, for example, V. Freilikher, E. Kanzieper, A.A. Maradudin, Phys. Rep. 288 (1997) 127.
[27] K.A. O'Donnell, R. Torre, C.S. West, Opt. Lett. 21 (1996) 1738.
[28] K.A. O'Donnell, R. Torre, C.S. West, Phys. Rev. B 55 (1997) 7985.
[29] K.A. O'Donnell, R. Torre, Opt. Commun. 138 (1997) 341.