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Diffraction of light by a bigrating: Surface polariton resonances and electric field enhancements

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We present a theory of the diffraction of light of arbitrary polarization incident on a doubly periodic dielectric grating. We use Rayleigh's method together with the vectorial equivalent of Kirchhoff's integral. The amplitudes of the diffracted spectral orders are calculated for a sinusoidal bigrating and for a square lattice of hemiellipsoids on a flat surface, for Ag. The enhancement of the electric field on and near the surface is calculated and is found to reach values of about 300.

In recent years much interest has been attached to optical interactions at corrugated surfaces.[1] In those cases where the corrugation is deterministic and periodic, the overwhelming majority of exact theoretical studies have been for singly periodic gratings (classical gratings). Here we outline a nonperturbative theory of light diffraction from a doubly periodic grating (a bigrating) of finite conductivity, and present results obtained from its numerical implementation.

The diffraction of light from such a bigrating has been studied theoretically in four recent papers. Derrick *et al.*[2] employed a coordinate transformation that maps the corrugated surface into a plane, and solved the transformed equations by an iterative scheme, while the other two approaches[3,4] involve the numerical integration of Maxwell's equations through the selvedge region. The efficiencies of the diffracted orders were found for crossed pyramidal and sinusoidal gratings.

The theory presented here is based on Rayleigh's method,[5] the vectorial equivalent of Kirchhoff's integral,[6] and the extinction theorem.[7] The utility of this approach in studying optical interactions at rough surfaces was appreciated first by Marvin.[8] Although Rayleigh's method is known to be limited in its applicability,[9–14] in the present study well-converged numerical results were obtained for all grating amplitudes within the region of physical interest with very rapid computation times.

In contrast with the emphasis in the earlier studies,[2–4] we are concerned with the reflectivity when incident light couples through the corrugations to surface polaritons, and with the enhancement of the electric field near the surface under such resonance conditions. This is of importance in connection with surface-enhanced Raman scattering.[15] The numerical calculations were carried out for sinusoidal bigratings and for bigratings formed by a square lattice of hemiellipsoids on a planar surface. The latter profile seems not to have been studied up to now, and so provides the first case to be treated exactly, in this context, of bumps separated from each other by regions of flat surface.

We consider vacuum in the region $x_3 > \zeta(\vec{x}_\parallel)$ and a lossy dielectric, of complex dielectric constant $\epsilon(\omega)$, in the region $x_3 < \zeta(\vec{x}_\parallel)$. Here $\vec{x}_\parallel = \hat{x}_1 x_1 + \hat{x}_2 x_2$ and $\hat{x}_1$, $\hat{x}_2$, and $\hat{x}_3$ are orthogonal unit vectors. The surface profile function $\zeta(\vec{x}_\parallel)$ is doubly periodic:

$$\zeta(\vec{x}_\parallel + \vec{a}_1) = \zeta(\vec{x}_\parallel + \vec{a}_2) = \zeta(\vec{x}_\parallel) ,$$

where $\vec{a}_1$ and $\vec{a}_2$ are noncollinear vectors in the plane $x_3 = 0$.

An exact expression for the electric field $\vec{E}^{>}(\vec{x}t) = \vec{E}^{>}(\vec{x}|\omega)\exp(-i\omega t)$, valid in the vacuum above the selvedge region, $x_3 > \zeta_{\max}$, that satisfies Maxwell's equations and the Bloch condition for the doubly periodic geometry is given by

$$\vec{E}^{>}(\vec{x}|\omega) = \vec{E}_i(\vec{k}|\omega)e^{i\vec{k}\cdot\vec{x}} + \sum_{\vec{G}_\parallel} \vec{E}_s(\vec{k}_\parallel + \vec{G}_\parallel|\omega)\exp[i\vec{K}_s(\vec{k}_\parallel + \vec{G}_\parallel)\cdot\vec{x}] , \tag{1}$$

where

$$\vec{k} = \vec{k}_\parallel - \alpha_0(k_\parallel\omega)\hat{x}_3, \quad \vec{K}_s(\vec{K}_\parallel) = \vec{K}_\parallel + \alpha_0(K_\parallel\omega)\hat{x}_3 , \tag{2}$$

$$\vec{E}_i(\vec{k}|\omega) = \left[\hat{k}_\parallel + \hat{x}_3\frac{k_\parallel}{\alpha_0(k_\parallel\omega)}\right]B_\parallel + (\hat{x}_3 \times \hat{k}_\parallel)B_\perp , \tag{3a}$$

$$\vec{E}_s(\vec{K}_\parallel|\omega) = \left[\hat{K}_\parallel - \hat{x}_3\frac{K_\parallel}{\alpha_0(K_\parallel\omega)}\right]A_\parallel(\vec{K}_\parallel\omega) + (\hat{x}_3 \times \hat{K}_\parallel)A_\perp(\vec{K}_\parallel\omega) , \tag{3b}$$

and where $\alpha_0(K_\parallel\omega)$ equals $(\omega^2/c^2 - K_\parallel^2)^{1/2}$ for $K_\parallel < \omega/c$ and $i(K_\parallel^2 - \omega^2/c^2)^{1/2}$ for $K_\parallel > \omega/c$. Here $\vec{k}$ and $\vec{E}_i$ are the wave vector and amplitude of the incident light; $\vec{k}_\parallel = \hat{x}_1 k_1 + \hat{x}_2 k_2$ with $k_1 = (\omega/c)\sin\theta_i \times \cos\phi_i$ and $k_2 = (\omega/c)\sin\theta_i \sin\phi_i$ for an angle of incidence θ_i, measured from the x_3 axis, and an azimuthal angle ϕ_i, measured from the x_1 axis. The sum is over all $\vec{G}_{\parallel}$, the translation vectors of the lattice reciprocal to the lattice defined by the bigrating [e.g., later we shall consider $\vec{a}_1 = a\hat{x}_1$ and $\vec{a}_2 = a\hat{x}_2$: then $\vec{G}_{\parallel} = (m,n)2\pi/a$ for integers m,n]. Each term in the sum represents either a diffracted beam $(K_{\parallel} < \omega/c)$ or an evanescent wave $(K_{\parallel} > \omega/c)$. $B_{\parallel}$ and $B_{\perp}$ determine the amplitudes of the components of the incident light that are p and s polarized, respectively. The $A_{\parallel}(\vec{K}_{\parallel}\omega)$ and $A_{\perp}(\vec{K}_{\parallel}\omega)$ determine the p and s polarization of each diffracted wave.

In the present method only the field in the vacuum enters the boundary conditions, thus halving the dimension of the matrix to be inverted. For this purpose we use the vectorial equivalent of Kirchhoff's integral, whose derivation in Ref. 6 we modify by choosing the integration volume to be the dielectric medium, with the observation point $\vec{x}$ in the vacuum (and $x_3 > \zeta_{\max}$), so that the left-hand side of Eq. (9.75) of Ref. 6 vanishes (extinction theorem). The fields in the medium are replaced by the field in the vacuum $E^{>}(\vec{x}|\omega)$ with use of the boundary conditions at the interface. The boundary condition for $E^{>}(\vec{x}|\omega)$ obtained in this way is finally

$$0 = -\frac{1}{4\pi}\int_S dS'\Big(\{\hat{n}'\times[\vec{\nabla}'\times\vec{E}^{>}(\vec{x}'|\omega)]\}G_{\epsilon}(\vec{x};\vec{x}'|\omega) + [\hat{n}'\times\vec{E}^{>}(\vec{x}'|\omega)]\times\vec{\nabla}'G_{\epsilon}(\vec{x};\vec{x}'|\omega) + \frac{1}{\epsilon(\omega)}[\hat{n}'\cdot\vec{E}^{>}(\vec{x}'|\omega)]\vec{\nabla}'G_{\epsilon}(\vec{x};\vec{x}'|\omega)\Big) , \tag{4}$$

where $\hat{n}$ is a unit vector outwardly normal to S, the interface $x_3 = \zeta(\hat{x}_{\parallel})$, at each point, and G_{ϵ} is the well known electromagnetic Green's function for a dielectric medium. Invoking the Rayleigh hypothesis, we substitute Eq. (1) for $\vec{E}^{>}$ into Eq. (4). After expanding $\exp[\alpha\zeta(\vec{x}_{\parallel})]$ in a Fourier series, we can project out of this integral boundary condition a doubly infinite set of simultaneous, linear, inhomogeneous equations for the $A_{\parallel}(\vec{k}_{\parallel}+\vec{G}_{\parallel})$ and $A_{\perp}(\vec{k}_{\parallel}+\vec{G}_{\parallel})$.

We have carried out numerical calculations for two forms of the surface profile function: (1) sinusoidal,

$$\zeta(\vec{x}_{\parallel}) = \zeta_1\cos(2\pi x_1/a) + \zeta_2\cos(2\pi x_2/a) ,$$

and (2) hemiellipsoidal,

$$\zeta(\vec{x}_{\parallel}) = h(1 - x_{\parallel}^2/c^2)^{1/2} ,$$

for $|x_{\parallel}| < c$ and $\zeta(\vec{x}_{\parallel}) = 0$ for $|x_{\parallel}| > c$ and $|x_1|$, $|x_2| < a/2$.

The numerical results were convergent to 0.1% $\rightarrow$1% for the (specular) reflectivity and 1% $\rightarrow$5% for the total field on the surface (at the surface maximum), when $\vec{G}_{\parallel} = (m,n)2\pi/a$ with $-5 \leq m,n \leq 5$ were kept.

We used $a = 8000$ Å, incident light of a wavelength $\lambda = 5145$ Å, $\phi_i = 0^{\circ}$, and $\epsilon = -11 + i0.33$ for Ag, to treat the sinusoidal profile with $\zeta_2 = 0$ up to $\zeta_2 = \zeta_1$ and with $\zeta_1/a = 0.01$, 0.02, and 0.03. We calculated the field enhancement $\mathscr{E} \equiv \vec{E}\cdot\vec{E}^*/\vec{E}_i\cdot\vec{E}_i^*$ near that surface and also the intensities of the diffracted beams. At $\theta_i = 24^{\circ}$ the wave $(m,n) = (1,0)$ is a resonantly excited surface polariton; and $(-1,-1)$, $(0,-1)$, $(-2,0)$, $(-1,0)$, (0,0), $(-1,1)$, and (0,1) are diffracted beams (but only the $n = 0$ orders appear when $\zeta_2 = 0$). The beams with $n = 0$ are p polarized; beams with $n \neq 0$ are of mixed polarization. The diffracted energy is nearly all in the (specular) beam (0,0)—its efficiency is at least 10^2 times that of the other beams (except right on resonance at $\zeta_1/a = 0.02$). The reflectivity minimum as a function of θ_i, at $\theta_i = 24^{\circ}$, falls to zero as ζ_1/a is increased up to $\zeta_1/a = 0.02$, while the peak in field enhancement (at a point above the surface maximum) increases up to $\mathscr{E} = 222$ for $\zeta_2 = 0$ and $\mathscr{E} = 283$ for $\zeta_2 = \zeta_1$ (Fig. 1 is for $\zeta_2 = \zeta_1$). Further increasing ζ_1/a causes the reflectivity minimum to increase and the $\mathscr{E}$ peak to decrease: at $\zeta_1/a = 0.02$ the incident light is fully coupled to the surface polariton; further increasing ζ_1/a only increases the polariton's corrugation-induced radiation damping and thus diminishes the field at the surface. For $\zeta_2 = \zeta_1 = 0.02a$ and $\theta_i = 24.03^{\circ}$, $\mathscr{E}$ was calculated throughout the surface region ($|x_1| \leq a/2$, $-2\zeta_0 \leq x_3 \leq 3\zeta_0$, for several values of x_2). Its largest

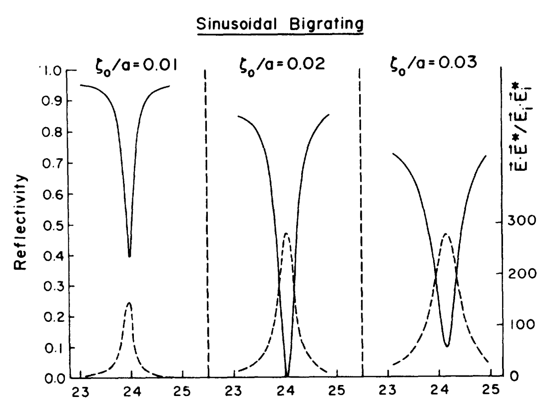

FIG. 1. (Solid curve) reflectivity for the (0,0)-diffracted order, and (dashed curve) the enhancement of the electric field $\vec{E}\cdot\vec{E}^*/\vec{E}_i\cdot\vec{E}_i^*$, at a point above the surface maximum, both vs angle of incidence θ_i, for sinusoidal bigratings of three corrugation strengths ζ_0/a, on Ag. ($a = 8000$ Å, $\lambda = 5145$ Å.)

value, $\mathscr{E} = 290$, was found at $\vec{x} = (a/4,\ 0,\ 1.7\zeta_1)$, just to the right of the surface maximum. A similar calculation for $\zeta_2 = 0$ gives a smaller maximum $\mathscr{E}$, and shows excellent agreement with the results of Garcia,[16] who used the formally exact extinction theorem method for the classical grating (further justifying our use of Rayleigh's method here).

For the square lattice of hemiellipsoids, with the same a, λ, ϵ, and ϕ_i, we studied the reflectivity and field enhancement, around resonance, while varying h, the axis perpendicular to the surface, and c, the axis in the surface (Figs. 2 and 3). Now, unlike in previous grating studies, one has bumps separated by flat regions, with the possibility of independently varying the bumps' height and separation. For a fixed c, the reflectivity dip around $\theta_i = 24°$ decreases continuously as h/a is increased from 0.01 to 0.08, and does not reach zero and begin increasing as for the sinusoidal gratings. At $c/a = 0.15$ the reflectivity is above 90%, with almost no dip even for the larger h/a; at $c/a = 0.25$ the reflectivity dip drops to 33% for $h/a = 0.07$, after which the curve is so broadened as to be almost flat; at $c/a = 0.40$ (Fig. 2) it reaches 3% for $h/a = 0.08$. The peak in the field enhancement [on the surface maximum $\vec{x} = (0, 0, h)$], for fixed c/a, was seen to increase with h/a, reach a maximum value at $h/a = 0.04$ or 0.05, and then decrease. Thus the field enhancement on the surface peak reaches its maximum value when the surface peak-to-valley distance, in the plane of incidence, is $0.04a \rightarrow 0.05a$ for the hemiellipsoids with various c/a values, just as for the sinusoidal bigrating and sinusoidal classical grating. The eventual decline in field enhancement caused by the surface polariton damping depends primarily on the height-to-period ratio, whereas the efficiency of the incident light coupling to the polariton, as measured by the depth of the reflectivity dip, depends strongly also on the fraction of the surface that is flat (i.e., on the distance between bumps). For the hemiellipsoids, even at $c/a = 0.5$, the largest possible value, 21% of the plane $x_3 = 0$ remains flat surface, and so the coupling efficiency is poorer than with a sinusoidal grating. For the grating of hemiellipsoids, the increase in radiative damp-

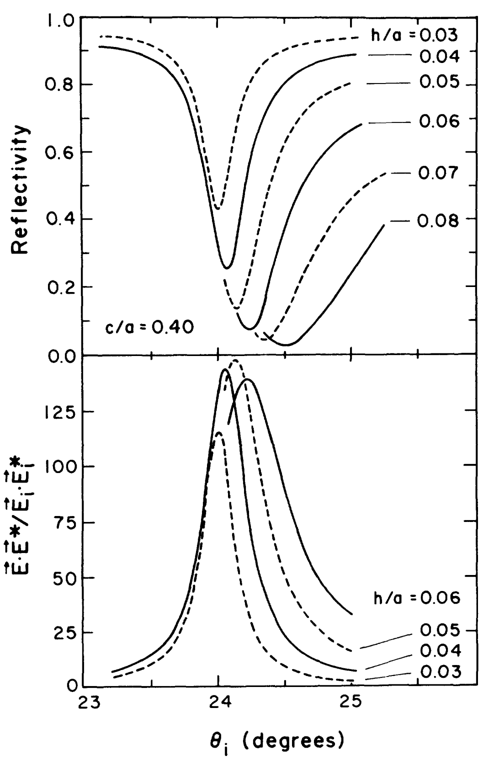

FIG. 2. Reflectivity for the (0,0)-diffracted order and the enhancement of the electric field, $\vec{E}\cdot\vec{E}^*/\vec{E}_i\cdot\vec{E}_i^*$, at a point above the surface maximum, vs angle of incidence θ_i, for a bigrating of hemiellipsoids on an Ag surface, with axes in the plane fixed, $c/a = 0.4$, and axes perpendicular to the plane varying, h/a. ($a = 8000$ Å, $\lambda = 5145$ Å.)

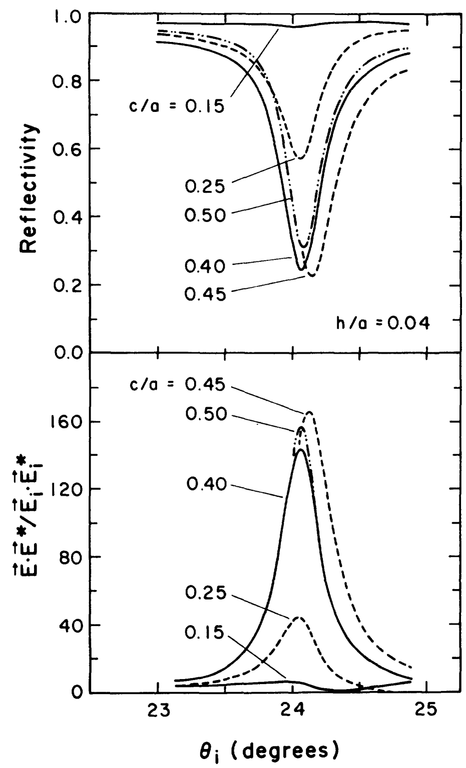

FIG. 3. Reflectivity for the (0,0)-diffracted order and the enhancement of the electric field, $\vec{E}\cdot\vec{E}^*/\vec{E}_i\cdot\vec{E}_i^*$, at a point above the surface maximum, vs angle of incidence θ_i, for a bigrating of hemiellipsoids on a Ag surface, with axes perpendicular to the surface fixed, $h/a = 0.04$, and axes in the surface varying, c/a. ($a = 8000$ Å, $\lambda = 5145$ Å.)

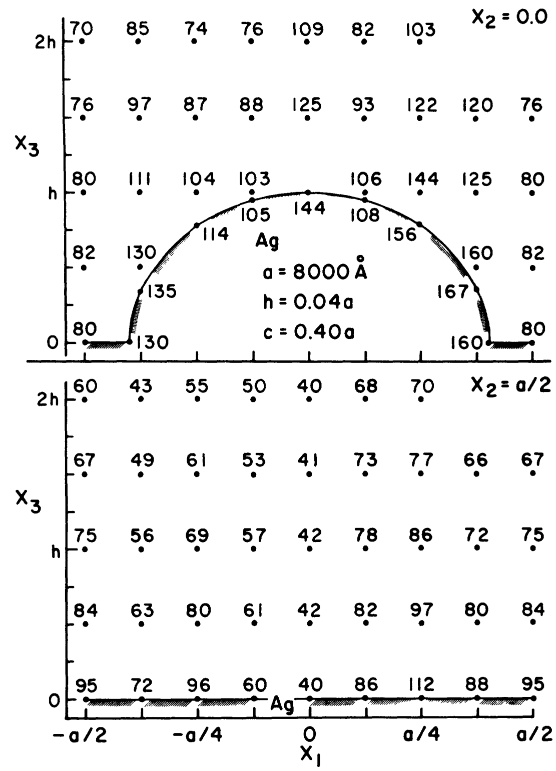

FIG. 4. Enhancement of the electric field, $\vec{E}\cdot\vec{E}^*/\vec{E}_i\cdot\vec{E}_i^*$, for light ($\lambda = 5145$ Å) incident at $\theta_i = 24.06^\circ$ on a bigrating of hemiellipsoids on Ag. Calculated in two planes $x_2 = 0$ and $x_2 = a/2$ ($a = 8000$ Å).

ing with increasing h/a wipes out the polariton faster than the increase in coupling with h/a can lower the reflectivity to zero.

For $\theta_i = 24.06^\circ$ and with $h/a = 0.04$ and $c/a = 0.4$, the field enhancement in the surface region, for $x_2 = 0$ and $x_2 = a/2$, is shown in Fig. 4.

For both the sinusoidal and hemiellipsoid bigrating, we also studied the effect of varying ϕ_i from 0° to 45°: the reflectivity minimum moves continuously to larger θ_i values, and increases and then decreases in depth. This behavior will be presented in more detail in a future, fuller, paper.

In conclusion, the present method is implemented quickly [to find the field throughout the surface region for a fixed x_2 with twice as many points (x_1, x_3) as in Fig. 4, with a 162×162 matrix, required 7 sec of execution time on the CDC-7600]; it gives convergent results for corrugation strengths of interest (i.e., beyond where the field enhancement attains a maximum); and it enables us to study bumps on a surface to see how their height and separation affect the coupling of light to surface polaritons and the attendant field enhancement.

ACKNOWLEDGMENTS

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