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Design of surface microrelief with selective radiative properties

A. SENTENAC and J.-J. GREFFET

Laboratoire d'Energétique Moléculaire, Macroscopique, Combustion Ecole Centrale Paris, Centre National de la Recherche Scientifique, 92295 Châtenay-Malabry, Cedex, France

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Abstract—The radiative properties of a material can be modified by its surface microrelief. We show that if the typical length of the surface roughness is smaller than a fraction of wavelength, the roughness can be treated as a homogeneous equivalent layer. Using this model and standard optimization techniques, we have developed an algorithm for the design of surfaces of a given material with prescribed radiative properties. As examples, we propose a relief structure that decreases the reflectivity of glass between 8 and 12 μm and another one that increases the emissivity of platinum in the infrared.

1. INTRODUCTION

THE RADIATIVE properties of surfaces are highly dependent on various factors like oxidation, roughness or contamination. This fact is well known and accounts for the wide dispersion observed in the available data [1]. The control of the radiative properties of surfaces is of interest for the control of radiative fluxes. The greenhouse effect is an example of spectrally selective radiative behavior. They can be used either to increase the heat fluxes as in the case of solar absorbers, or to reduce them as in the case of insulating surfaces on satellites.

In order to control the radiative properties, one may choose a material having special emissive qualities. For example, in the case of insulation of a satellite, a flat surface of germanium reflects the incident visible light coming from the sun and its high emissivity in the infrared also contributes to cooling. However, for most applications, the materials are chosen for their mechanical properties and the problem arising from the control of the radiative fluxes has to be solved by other means. A possible solution is to design a composite material in order to modify its radiative behavior [2, 3].

The modification of radiative properties can be achieved in different ways. The development of oxidation generally increases the emissivity. This may be a simple solution in some cases. A method that has been widely used is the modification of the surface roughness on a macroscopic scale. This can be achieved by inducing a random roughness or periodic arrays of grooves of selected section. In all cases, these modifications increase the emissivity. This effect can be understood on the basis of a geometric optical image. An incident ray will bounce several times on the surface before leaving the surface. Hence, the absorptivity is increased. In other words, a sort of local blackbody effect has been created. Note that this approach cannot reduce the value of the emissivity and does not produce spectrally selective effects [4–6].

In the case of a roughness of a few wavelengths, geometrical optic concepts cannot be applied. A correct description of the interaction between the structure of the surface and the incident radiation requires an electromagnetic approach. A pioneering work by Sacadura has explored the effect of microrelief using either the Kirchhoff approximation or a first order perturbation calculation [7]. The theory of scattering by random rough surfaces either for radar or optical wavelengths has progressed and thorough reviews are available [8, 9]. Experiments using well controlled surfaces have shown good agreement with modern theories [10, 11].

The radiative properties of a surface may be modified by the use of surface coatings. Based on interference effects, they are currently used for the design of antireflection coatings either for visible or infrared components. The technique also makes it possible to obtain a prescribed selective absorptivity. Its efficiency may be remarkable. The main problem is the mechanical resistance of the coating.

The technique described in this paper lies between the two approaches presented above. It is based on the fact that a radiation of a given wavelength λ propagates in an inhomogeneous medium as if it were homogeneous, provided that the inhomogeneity length scale is much smaller than λ. Hence, the propagation of radiation can be studied, in a much simpler way, by introducing a homogeneous equivalent refractive index. As a consequence, a microrelief structure is equivalent to a pile of homogeneous layers, as shown in Fig. 1, and should behave as an interference coating. As compared with the blackbody effect produced by large scale roughness, microroughness effects are wavelength dependent and may also enhance the reflectivity.

In the next section, we will restrict ourselves to the

NOMENCLATURE

- d period of a grating
- F filling factor of a grating
- $f(n_1, \ldots, n_N)$ merit function of the optimization
- h thickness of a layer or height of a grating
- n refractive index
- n_1 refractive index of the layer 1
- r_{ij} Fresnel reflection factor between media (i) and (j)
- R reflectivity.

Greek symbols
- ε emissivity
- λ wavelength.

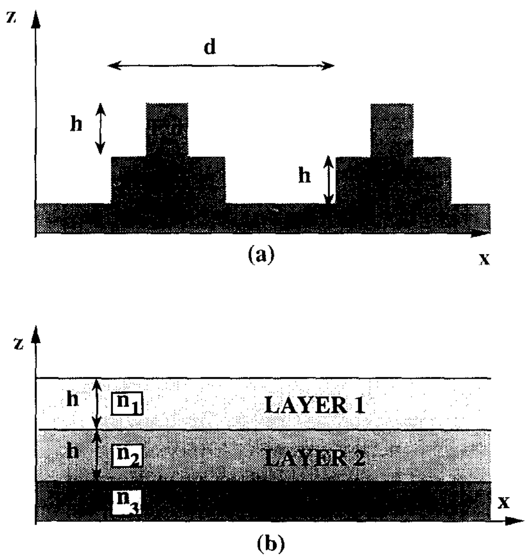


FIG. 1. (a) Geometry of a multilevel (2-level) stairstep grating. (b) The equivalent multilayer for the multilevel stairstep grating in the long-wavelength limit. The equivalent optical index n_i of the layer depends on the filling factor F_i of the grating at the same height.

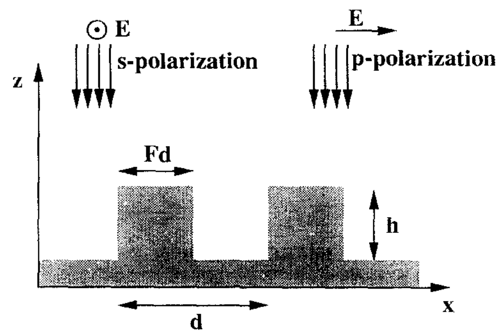


FIG. 2. Geometry of a lamellar grating of period d, filling factor $F = 0.5$ and height h, illuminated under normal incidence by an s- or p-polarized beam.

special case of periodic 1D surfaces, called gratings. We use an exact method previously developed [12] to compute the reflectivity of such surfaces. Thanks to this technique, we will investigate the range of validity of the concept of homogenization. In the third section, we will describe how the radiative properties of a given material can be modified by changing its surface microrelief. The design process is based on the equivalence of a multilevel stairstep grating with a pile of homogeneous layers, as seen in Fig. 1. Using a standard optimization technique, we obtain an index profile that meets the radiative constraints. Then, we translate this profile into an equivalent relief structure. In the fourth section we apply the technique to two different cases. The first example deals with glass. We have defined a grating profile that reduces the reflectivity of glass between 8 and 12 μm. As a consequence, the emissivity is increased in the range of wavelengths of the thermal emission. This is the typical behavior of an insulating glass. The second application presented is for a metal. We have designed a relief that increases the emissivity of a platinum sample. This should be useful for the realization of a standard material for emissivity measurement.

2. VALIDITY OF THE CONCEPT OF HOMOGENIZATION

In this section we examine the validity of the concept of homogenization by comparing the reflectivity of a lamellar grating with the reflectivity of the equivalent homogeneous layer. Let us first define the geometry of the system depicted in Fig. 2. We consider a homogeneous medium characterized by its complex index n. The surface is invariant along Oy and is described by a periodic function $z = S(x)$. We define the filling factor of the surface, F, as the ratio of material over vacuum within one period of the grating.

The surface is illuminated, under normal incidence, by a monochromatic beam described by its electric field in order to account for its phase. When the electric field is along the Oy axis, we will call it s-polarization whereas the p-polarized case corresponds to an electric field along the Ox axis.

Our first step is to define the rule of equivalence, i.e. a formula giving the equivalent index for a given surface roughness. This point has already been discussed in ref. [13]. These authors have found that the equivalent index depends solely on the index of the medium, the polarization and the filling factor F. For s-polarization, the equivalent index n_{eq} is given by

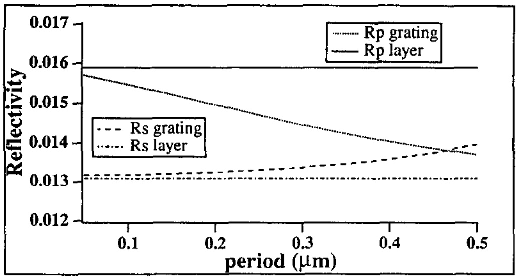


FIG. 3. Comparison of the reflectivity (R_s, R_p) of a lamellar grating made of glass with $F = 0.5$, $h = \lambda/8$ with that of an equivalent layer ($R_{s,p}$ layer) for both polarizations, as a function of the period of the grating. The wavelength can vary between 0.4 and 1.5 μm. The optical index $n = 1.5$.

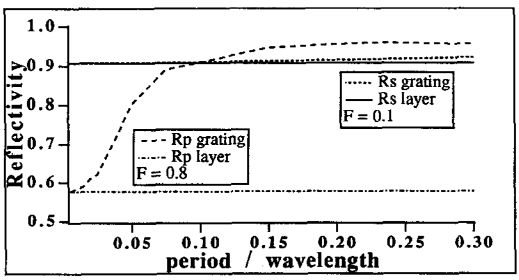


FIG. 4. As in Fig. 3, except that $h = \lambda/4$, $F = 0.8$ for p-polarization, $F = 0.1$ for s-polarization, $\lambda = 1$ μm and the substrate is platinum with an optical index $n = 0.1+i4$.

$$n_{eq}^2 = Fn^2+(1-F) \quad (1)$$

whereas for p-polarization the effective index is

$$1/n_{eq}^2 = (F/n^2)+(1-F). \quad (2)$$

In order to check the validity of the concept of homogenization, we compare the reflectivity of a grating obtained by a rigorous volume integral method [12] with that of the equivalent homogeneous layer whose optical index is given by equations (1) or (2) and whose thickness is the height of the grating. The system chosen for this comparison is a lamellar grating, illuminated under normal incidence, of height h, filling factor F, and period d varying from, $\lambda/80$ to $\lambda/3$. Note that the period is so small that the surface does not produce any grating order and reflects the light specularly as a homogeneous layer. Let us first examine the behavior of a typical dielectric. We have chosen glass with a value of the index $n = 1.5$. This value is valid between 0.4 and 1.5 μm approximately. Figure 3 shows that the agreement is better than 5% for d smaller than $\lambda/10$.

We now turn to the case of a metal whose index n is equal to $4i+0.1$; it corresponds to the index of platinum at 1 μm. We obtain an agreement between the exact solution and the equivalent system better than 5% for d smaller than $\lambda/30$ as seen in Fig. 4. This value is much smaller than that found in Fig. 3, indeed, the rate of convergence depends on the index of refraction of the substrate, i.e. on the wavelength *in* the medium. As a rule of thumb, equivalence between the two systems is reached when the period is smaller than $\lambda/10\,|n|$.

In the next section, we assume that the reflection properties of a lamellar grating, of given filling factor, with a small period compared to the wavelength *in* the medium, are accurately modelled by an equivalent homogeneous layer. In Fig. 1, we generalize this assumption to the case of a multilevel stairstep grating with different filling factors. Thus it is possible to evaluate the reflectivity R of such gratings by using standard techniques developed for a stack of layers. The emissivity is obtained as $\varepsilon = 1-R$.

3. DESIGN OF A SELECTIVE SURFACE

The design of surfaces, with expected radiative properties is an inverse problem. It can be greatly simplified by using the rule of equivalence between inhomogeneous and homogeneous media in the long wavelength limit. Indeed, the radiative reflectivity of a stack of layers is well known and easily computed whereas difficulties arise when dealing directly with rough surfaces. By adapting the index and thickness of the different layers, one can obtain a large range of radiative properties. However, the resulting index profile must be convenient enough to be transposed into a technically feasible multilevel stairstep grating, as seen in Fig. 1.

Let us first describe the approach used to calculate the reflectivity of a stack of layers. An incident radiation will produce a partly reflected and transmitted wave at each interface, which will in turn be partly reflected and transmitted. The interference of all these waves will eventually give the global reflectivity of the system.

In the case of one layer of index n_2 and thickness h, imbedded in two media of index n_1 and n_3, the reflectivity for a wavelength λ and for normal incidence is given by

$$R = |r_{12}+r_{23}\exp(2i\pi h/\lambda)|^2/|1+r_{12}r_{23}\exp(2i\pi h/\lambda)|^2$$

where $r_{ij} = (n_i-n_j)/(n_i+n_j)$ is the Fresnel reflection factors between media (i), (j). Note that R is a periodic function of the thickness h. The radiative behavior of such a system is strongly dependent upon the thickness and the index of the layer. This can be generalized to a pile of N layers. In this case, the computation of the reflectivity reduces to the product of N 2×2 matrices and is very fast. More details are given in ref. [14].

In order to find a system of layers with prescribed radiative properties, we use an optimization technique. First, we create a merit function which contains the expected reflectivities for certain wavelengths. For example, if M reflectivities R_i are required for M wavelengths λ_i, the merit function will be written in the form

$$f(n_1,\ldots,n_N) = \sum_{i=1}^{M} |R_{N,h}(n_1,\ldots,n_N,\lambda_i) - R_i|,$$

where n_j represents the wavelength-dependent optical index of the layer j and $R_{N,h}$ is the reflectivity of the pile of N layers of thickness h.

Then, we minimize f by adapting the indexes, while the number of layers and their thicknesses are fixed. The minimum is found by the modified Newton algorithm with simple inequality constraints of the library NAG (e04jaf). The optimization is carried out for different values of h, and a refinement on both thickness and indexes is performed around the minimum. For practical reasons, the multilevel stairstep grating should be designed with a filling factor monotonically decreasing from the substrate towards the vacuum. Hence, it is more advantageous to use directly, as unknown parameters for the optimization, the filling factors F_i of the layer i, for i varying from 1 to N, where the label 1 corresponds to the upper layer. Then, the index n_i of the layer is deduced from F_i thanks to equations (1) or (2). In this case, the constraints on F_i are

$$0 < F_1 < F_2 < \cdots < F_i < F_{i+1} < \cdots < F_N < 1.$$

One can also use, as unknown parameters, $a_i = F_i - F_{i-1}$, the amount of substrate added to the layer i compared to layer $i-1$. In this case, the constraints are $a_i > 0$ for i varying from 1 to N,

$$\sum_{i=1}^{N} a_i < 1.$$

The latter can be included in the merit function.

In all events, this method gives solutions that meet the first requirement for experimental feasibility, i.e. the decreasing filling factor. Although it may find local minima, that are strongly dependent on the initial values, it has solved several problems in a satisfactory way, as shown below.

4. APPLICATIONS

4.1. *An insulating glass*

The radiative transfer between glass and atmosphere can be modified by decreasing the emissivity of glass between 8 and 12 μm. In this region, the atmosphere is transparent whereas the reflectivity of glass has a maximum. Hence, by reducing the reflectivity, we increase the emitted flux without increasing the absorption. This property may be useful in order to lower the temperature of a system.

We have used the optimization procedure described above to design a coating that could reduce the normal reflectivity between 8 and 12 μm. The index of glass, as a function of the wavelength, was found in ref. [15]. Zero-reflectivity is demanded from 8 to 12 μm in the merit function. The resulting reflectivity of the optimized pile of layers is shown in Fig. 5 for s- and p-polarization. For comparison, the reflectivity of a flat

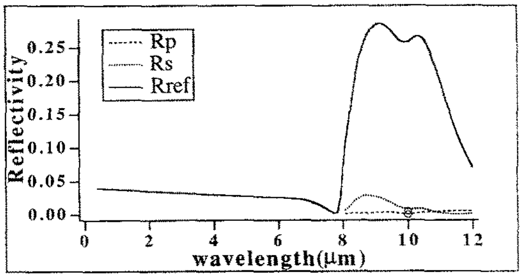


FIG. 5. Reflectivity (R_s, R_p) for s- and p-polarization as a function of λ, of a pile of 10 layers placed on a glass substrate, designed by the optimization method in order to get zero-reflectivity in the 8–12 μm region. The solid curve (R_{ref}) represents the reflectivity of the substrate alone. The markers indicate the s and p reflectivities calculated with the rigorous integral method for the equivalent multilevel stairstep grating whose profile is depicted in Fig. 6.

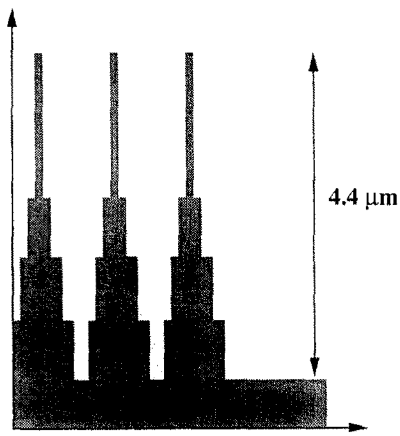


FIG. 6. Profile of the multilevel stairstep grating equivalent to the stack of 10 layers whose reflectivities are plotted in Fig. 5.

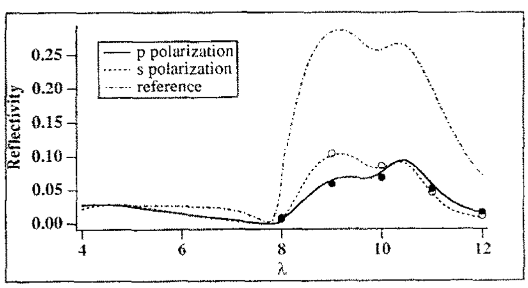
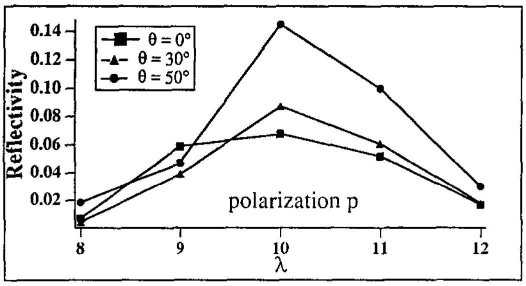


FIG. 7. Reflectivity for s- and p-polarization as a function of λ of one layer placed on a glass substrate, designed by the optimization method to minimize the reflectivity in the 8–12 μm range. The reference represents the reflectivity of a flat surface of glass. The markers indicate the reflectivities of the equivalent grating calculated with the rigorous integral method. $d = 1$ μm, $F = 0.37$, $h = 2$ μm.

interface of glass without coating is also plotted. The reflectivity values, given by the rigorous code applied to this grating at 10 μm, circled in Fig. 5, are in good agreement with those of the stack of layers. The equivalent multilevel stairstep grating profile is depicted in Fig. 6. Since the manufacture of such a grating is not easy, we present in Fig. 7 the reflectivity



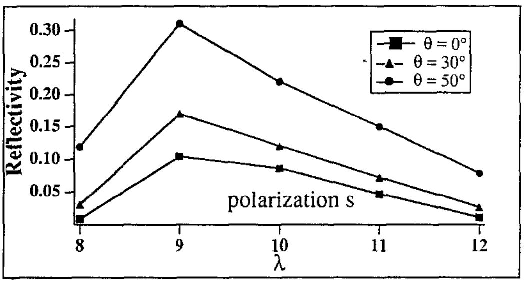


FIG. 8. Reflectivities of the grating depicted in Fig. 7 ($d = 1$ μm, $F = 0.37$, $h = 2$ μm) as a function of the wavelength for several angles of incidence. Rigorous calculations.

of a *lamellar* grating designed to increase the emissivity in the 8–12 μm range. Its filling factor F is equal to 0.37, and its height is 2 μm. Rigorous calculations, performed with periods of 1 and 2 μm (not shown) are in good agreement with the homogeneous layer model. Since the grating behaves more or less like an antireflection coating, one can expect the reflectivity to be smaller than that of the substrate alone whatever the angle of incidence [14]. In Fig. 8, the grating's reflectivity is plotted for several angles of incidence. Bearing in mind that for $\lambda = 9$ μm and $\theta_{inc} = 50°$, the reflectivity of the flat surface is equal to 0.19 for p-polarization and 0.49 for s-polarization, the previous statement is clearly verified.

4.2. *A standard for emissivity measurements*

In this section we show that this technique can also be applied successfully to metallic surfaces. We have considered the modification of the emissivity of a platinum sample. Indeed, the main problem when defining a material for intercomparison of emissivity measurements is the stability of the sample during several heating and cooling cycles. For this purpose, the best material is platinum but its emissivity is very low. So, it would be interesting to increase its emissivity by creating an appropriate microrelief structure on its surface.

In this case, the optimization technique has been applied to a platinum sample for p-polarization only. The wavelength-dependent values of the optical index are taken from [16]. We have used one constraint demanding an emissivity of 0.9 at 10 μm and we have looked for a solution using one layer. For a flat surface, the value of the emissivity at this wavelength is 0.03 for both s- and p-polarization at normal incidence. The reflectivity, for p-polarization, of the optimized layer is plotted in Fig. 9. The circles indicate the reflectivities of the lamellar grating that have been computed with the rigorous method [12]. Its filling factor F is equal to 0.98 and its depth is 0.94 μm. It appears clearly that the constraint is very well satisfied. For s-polarization the reflectivity does not change from that of a flat interface. Actually, this surprising result can be partly explained by noticing that the effective index for p-polarization of such a grating is that of a dielectric ($\lambda = 10$ μm, $n_{substrate} = 9.7+i37.5$, $F = 0.98$, $n_{eff^p} = 7+i0.06$), whereas it remains that of a metal for s-polarization ($n_{eff^s} = 9.6+i37$). Thus, for p-polarization, the grating behaves like a dielectric antireflection coating, which may account for the typical pseudoperiodicity of the reflectivity as a function of the wavelength as seen in Fig. 9. Note that a similar behavior can be obtained for s-polarization by taking a very small filling factor. In that case, the s-polarization effective index will be that of a dielectric (for example, $\lambda = 1$ μm, $n_{substrate} = 0.1+i4$, $F = 0.05$, $n_{eff^s} = 0.38+i0.06$).

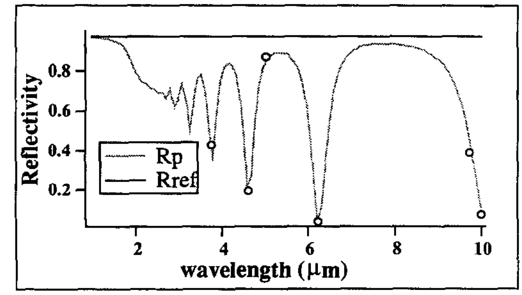


FIG. 9. Reflectivity of p-polarized light (R_p) as a function of λ for one layer, on a substrate of platinum, designed by the optimization method in order to get an emissivity of 0.9 for $\lambda = 10$ μm. The solid curve (R_{ref}) is the reflectivity of the substrate alone. The markers represent the p reflectivities calculated with the rigorous integral method for the equivalent lamellar grating with $d = 0.01$ μm, $h = 0.94$ μm, $F = 0.98$.

Unfortunately, the manufacture of the grating presented in Fig. 9 is not possible with current techniques, the filling factor being too important and the period allowing the homogenization assumption being too small. Hence, new optimizations were performed with added constraints on the filling factor and rigorous calculations were carried out for gratings of increasing periods. Figure 10 displays the grating's reflectivity as a function of the period for different optimized data. It is shown that the homogenization assumption is accurate when the period is smaller than one-tenth of the wavelength *in* the medium, which confirms the rule of thumb given in Section 2. However, some interesting results are obtained for larger periods. For example, for $\lambda = 0.6$ μm the reflectivity of the grating whose period is 53 nm, filling factor 0.7 and height 153 nm is equal to 0.74; for $\lambda = 5$ μm the reflectivity of the grating whose period is 200 nm, filling factor 0.9 and height 355 nm is equal to 0.77; for $\lambda = 10$ μm

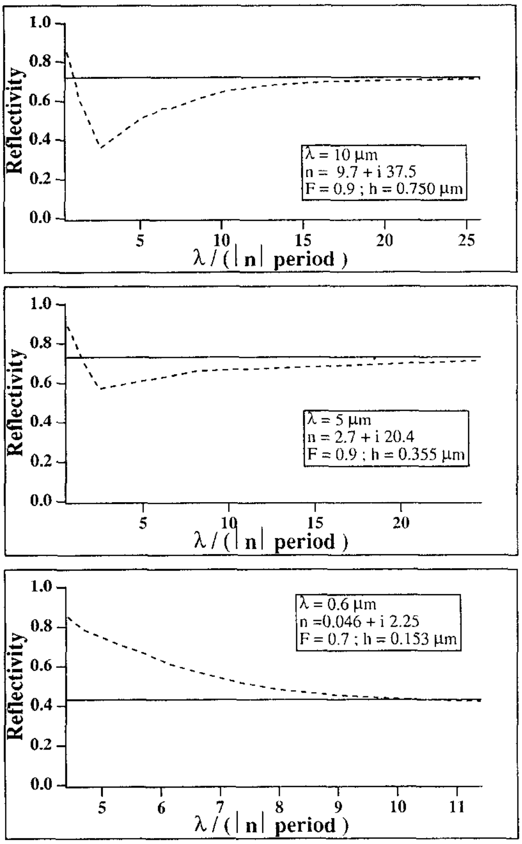


FIG. 10. Grating's reflectivity as a function of the ratio of wavelength in the medium over its period. Solid curve: reflectivity of the equivalent homogeneous layer. Dotted curve: reflectivity of the grating calculated with the rigorous approach.

the reflectivity of the grating whose period is 400 nm, filling factor 0.9 and height 750 nm is equal to 0.8. Currently, the ion etching process is able to create holes whose minimal width is about 20 nm, with a maximal ratio depth over width about 10. Hence, one can reasonably consider the manufacture of such gratings in the near future.

5. CONCLUSIONS

We have presented a method that allows us to design a surface microrelief with spectrally selective properties. The principle of the technique is based on the equivalence of a grating, with a period smaller than the wavelength, with a system of homogeneous layers. The validity of the concept of homogenization has been checked by comparing the reflectivity of a lamellar grating, obtained with a rigorous integral method, with that of the equivalent homogeneous layer. To illustrate the possibilities of the design technique, we have considered two different cases. First, we display a surface relief that significantly reduces the reflectivity of glass in the 8–12 μm region with possible applications to an insulating glass. Then, it is shown that the emissivity of platinum can be increased up to 0.3 with possible applications to the design of a reference surface for the calibration of emissivity measurement equipment.

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