Spatial characteristics of periodic structures formed as a result of interaction of laser radiation with metal and semiconductor surfaces

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A theoretical analysis is made of the correlation functions of periodic structures formed as a result of interaction of laser radiation with metal and semiconductor surfaces. Explicit expressions for the correlation functions are obtained for the linear stage of the interaction. A nonlinear stage is also considered. It is shown that nonlinear effects result in freezing of the correlation structure formed in the linear stage. The degenerate case is considered in detail for angles of incidence $\theta > \pi/4$.

1. INTRODUCTION

Over twenty years have passed since the discovery of formation of periodic structures on the surfaces of solids as a result of interaction with laser radiation, but the flood of work on this subject continues to grow (for a review and a bibliography see Ref. 1). A considerable quickening of the research has resulted from recognition of the fact that in most cases the process is due to resonant excitation of surface electrodynamic waves. An important consequence is that the period and orientation of the structures formed on metal and semiconductor surfaces are independent of the properties of the target material but are governed by the parameters of laser radiation such as the wavelength, polarization, and angle of incidence.

In spite of the common nature of the amplification of an electromagnetic field by the excitation of surface electrodynamic waves, the actual mechanism of formation of periodic structures may vary with the energy and time characteristics of the radiation and with the properties of the target material. We shall not specify this mechanism and consider the stage of the process which is linear for a given mechanism of formation of periodic structures.

Our aim will be to determine the spatial and time characteristics of periodic structures. We shall first consider the linear stage of the process and then discuss nonlinear effects due to the interaction of surface electrodynamic waves as they are scattered by the newly formed periodic structure. Following Ref. 2, we shall consider the specific case when the formation of periodic structures is due to evaporation. Similar results can also be obtained readily for other mechanisms of formation of periodic structures.

We shall specify the equation for the profile of a solid dependent on time t in the form $z = z_{\bullet}(\mathbf{r},t)$ on the assumption that z = 0 describes a plane obtained by averaging the real surface. The coordinate z is directed along the normal to this plane into the semiconductor and **r** is a two-dimensional vector in the plane.

A model of discrete modes (MDM) is adopted in Ref. 2: in this model the function $z_{\bullet}(\mathbf{r},t)$ is reduced to a finite number of harmonics with wave vectors satisfying the condition of a resonance in respect of surface electrodynamic waves, which is $|\mathbf{q} + \mathbf{g}_i| \approx k_0$. Here, \mathbf{q} is the tangential component (in the z = 0 plane) of the wave vector of the incident radiation; $|\mathbf{q}| = k_0 \sin\theta$; $k_0 = \omega/c$; θ is the angle of incidence; ω is the frequency and c is the velocity of light.

Among all the modes we shall consider separately a degenerate mode with a wave vector \mathbf{g}_+ satisfying the condition $\mathbf{q} \cdot \mathbf{g}_{+} = 0$, which ensures a simultaneous resonance of $\mathbf{g} = \mathbf{g}_+$ and $\mathbf{g} = \mathbf{g}_- = -\mathbf{g}_+$. The basis for this model is a strong time dependence of unstable modes resulting in their selection at extrema of the growth increment considered as a function of the wave vector g. However, the MDM in fact corresponds to an approximation of a real system with an infinite number of degrees of freedom by a system with a finite number. The question then arises of the accuracy of the results obtained in the MDM. There are many series of problems the solution of which cannot be obtained within the MDM framework and they include, for example, the problem of spatial characteristics of periodic structures. Therefore, there is a need to develop a more general approach which allows for the spatial degrees of freedom of the structures. This will be our main task.

Dropping the MDM, we shall write down the expressions for the function of the surface profile $z_{\star}(\mathbf{r},t)$ allowing not only for the discrete vectors \mathbf{g}_i but also for the contributions of their vicinities, which is equivalent to an allowance for a slow coordinate dependence of the amplitudes of discrete modes. We then obtain the following expansion for the function $z_{\star}(\mathbf{r},t)$:

$$\mathbf{z}.(\mathbf{r},t) = \sum_{i} b_{i}(\mathbf{r},t) \exp(i\mathbf{g}_{i}\mathbf{r}) + \sum_{i>j} b_{ij}(\mathbf{r},t) \exp(i\mathbf{g}_{ij}\mathbf{r}) + \text{c.c.}$$
(1.1)

A prime indicates, as in the corresponding MDM formula,² that the term corresponding to a degenerate mode is taken with half the weight $\mathbf{g}_{ij} = \mathbf{g}_i - \mathbf{g}_j$. Strictly speaking, Eq. (1.1) is multivalued in the sense that the wings of the spectral expansion in terms of the wave vectors overlap. However, during the linear stage we can assume that z_{\star} (\mathbf{r} ,t) can be expanded as a Fourier integral, which alters only slightly the later analysis. Before the nonlinear stage, for which the expansion (1.1) is important, the overlap of the spectral expansions is exponentially small and this justifies the adopted approach.

The main equations describing the evolution of periodic structures in the continuum approach are obtained from the corresponding MDM equations if we assume that the amplitudes of the profiles b_i and b_{ij} and of the field ε_i are functions of the spatial variables and if in every case where the vector goccurs explicitly we replace it by the vector $\mathbf{g}_l \rightarrow \hat{\mathbf{g}}_l = \mathbf{g}_l - i\partial /\partial \mathbf{r}$, where l = i or *ij*. Consequently, these equations become

$$\hat{B}(\mathbf{q}+\mathbf{g}_{i})\varepsilon_{i}(\mathbf{r},t)$$

$$=iA(\hat{\alpha}_{i})k_{0}b_{i}(\mathbf{r},t)-i\sum_{i}\varepsilon_{j}(\mathbf{r},t)\sin^{2}\frac{\alpha_{ij}}{2}k_{0}b_{ij}(\mathbf{r},t), \qquad (1.2)$$

$$\frac{db_i(\mathbf{r},t)}{dt} = \frac{F(t)}{|\hat{\mathbf{g}_i}|} \bar{A}^{\cdot}(\hat{\alpha}_i) \varepsilon_i(\mathbf{r},t), \quad i \neq +, -,$$
(1.3)

$$\frac{db_{+}(\mathbf{r},t)}{dt} = \frac{F(t)}{|\hat{\mathbf{g}}_{+}|} \left[\bar{A}^{\cdot}(\hat{\alpha}_{+}) \varepsilon_{+}(\mathbf{r},t) + \bar{A}(\alpha_{-}) \varepsilon_{-}^{\cdot}(\mathbf{r},t) \right], \ b_{-} = b_{+}^{\cdot},$$

$$\frac{db_{ij}(\mathbf{r},t)}{dt} = \frac{F(t)}{t} \varepsilon_i(\mathbf{r},t) \varepsilon_j^*(\mathbf{r},t) \cos \hat{\alpha}_{ij}.$$
(1.5)

$$\frac{\partial \omega_{ij}(\mathbf{r}, t)}{\partial t} = \frac{1}{2|\hat{\mathbf{g}}_{ij}|} \varepsilon_i(\mathbf{r}, t) \varepsilon_j^*(\mathbf{r}, t) \cos \hat{\boldsymbol{\alpha}}_{ij}.$$
(1)

Here,

$$\hat{B}(\mathbf{q}+\hat{\mathbf{g}_{i}}) = \zeta + [1-(\mathbf{q}+\hat{\mathbf{g}_{i}})^{2}/k_{0}^{2}]^{\nu_{a}}, \quad (1.6)$$
$$A(\hat{\alpha}_{i}) = (|E_{us}|^{2} + |E_{up}|^{2})^{-\nu_{a}}[E_{up}(\cos\hat{\alpha}_{i} - \sin\theta)$$

$$- \left(\left| E_{os} \right| + \left| E_{op} \right| \right) = \left[\left| E_{op} \right| \cos \alpha_i - \sin \theta \right] - E_{os} \sin \alpha_i \cos \theta \right], \quad (1.7)$$

$$\bar{A}(\hat{\alpha}_i) = A(\hat{\alpha}_i) + (|E_{0s}|^2 + |E_{0p}|^2)^{-\gamma_b} E_{0p} \sin \theta, \quad (1.8)$$

$$\hat{\alpha}_{ij} = 2 \arcsin(|\hat{\mathbf{g}}_{ij}|/2k_0), \quad \cos \hat{\alpha}_i = (k_0^2 + q^2 - \hat{\mathbf{g}}_i^2)/2qk_0, \\ F(t) = c_0 \exp\left(-\frac{U}{T_0}\right) \frac{U}{T_0^2} \frac{c\zeta'}{2\pi\kappa} (|E_{0s}|^2 + |E_{0p}|^2),$$

 $\zeta = \zeta' + i\zeta''$ is the surface impedance (ζ' and ζ'' are real quantities), where $|\zeta| \ll 1$; the incident radiation seemed to be a plane linearly polarized wave; E_{0p} and E_{0s} are the projections of the amplitude of the intensity of the electromagnetic field on the plane of incidence and at right-angles to it; \varkappa is the thermal conductivity; T_0 is the time-dependent value of the temperature of the interface averaged over the whole of its area; c_0 is a constant of the order of the velocity of sound; U is a constant of the order of the energy of the interatomic interaction in the target conductor. The functions of the operator $i\delta/\delta \mathbf{r}$ and the asterisk is used to denote the Hermitian conjugate.

2. LINEAR STAGE OF THE PROCESS

Separation of degenerate and nondegenerate modes is meaningful only for angles of incidence $\partial \gg \zeta' |\zeta''|$. When these angles are low, the degenerate and nondegenerate modes merge, but this case requires a separate analysis; therefore, we shall begin by considering the angles of incidence obeying $\partial \gg \zeta' |\zeta''|$. We shall consider in greater detail the case of an s-polarized incident wave ($E_p = 0$). The results obtained can be generalized to the case of an arbitrarily polarized incident wave.

In this section we shall consider the linear stage of the process of formation of periodic structures when the following inequality is obeyed:

$$|b_l|k_0 \ll \zeta', \tag{2.1}$$

so that on the right-hand side of Eq. (1.2) we can ignore the second term. It then follows from Eqs. (1.2)-(1.4) subject to Eqs. (1.7) and (1.8) that

$$\frac{db_{\mathbf{i}}(\mathbf{r},t)}{dt} = iFH(\alpha_{\mathbf{q}+\hat{\mathbf{g}}_{\mathbf{i}}}) \hat{B}^{-1}(\mathbf{q}+\hat{\mathbf{g}}_{\mathbf{i}}) b_{\mathbf{i}}(\mathbf{r},t), \quad i \neq +, -,$$

$$(2.2)$$

$$\frac{db_{+}(\mathbf{r},t)}{dt} = iF\cos^{3}\theta[\hat{B}^{-1}(\mathbf{q}+\hat{\mathbf{g}}_{+})-\hat{B}^{-1}(\mathbf{q}-\hat{\mathbf{g}}_{+})]b_{+}(\mathbf{r},t),$$

(2.3)

where

(1.4)

dt

$$H(\alpha) = \cos^2 \theta \sin^2 \alpha (1 + \sin^2 \theta - 2 \sin \theta \cos \alpha)^{-1/2},$$

$$\alpha_{\mathbf{q}+\hat{\mathbf{g}}} = \arccos\left[(q^2 + \mathbf{q}\hat{\mathbf{g}})/qk_0\right],$$

and in Eq. (2.3) we retain the spatial dependence only for the quantities \hat{B} because for $\theta \gg \zeta' |\zeta''|$ it is these quantities that determine the spatial dependence $b_+(\mathbf{r},t)$.

Since $b_i(\mathbf{r},t)$ are amplitudes of periodic harmonics varying slowly with the coordinates and corresponding to the wave vectors \mathbf{g}_i , the initial conditions for $b_i(\mathbf{r},t)$ are

$$b_i(\mathbf{r}, 0) = \exp(-i\mathbf{g}_i\mathbf{r})b^0(\mathbf{r}). \qquad (2.4)$$

Here, $b^{0}(\mathbf{r}) = z_{\star}(\mathbf{r}, t = 0)$ is the function describing the profile of the surface at the very first moments of the interaction with the incident radiation, describing the real structure of the surface roughness of the target.

The formal solution of Eqs. (2.2) and (2.3) subject to the initial conditions (2.4) is

$$b_i(\mathbf{r},t) = \int d^2 \mathbf{r}' I_i(\mathbf{r}-\mathbf{r}',t) \exp(-ig_i \mathbf{r}') b^o(\mathbf{r}'), \qquad (2.5)$$

where

$$U_i(\mathbf{r},t) = \int \frac{d^2 \mathbf{x}}{(2\pi)^2} \exp[s\Lambda_i(\mathbf{x}) + i\mathbf{x}\mathbf{r}], \qquad (2.6)$$

$$\Lambda_{i}(\mathbf{x}) = i \frac{H(\alpha_{\mathbf{q}+\mathbf{g}_{i}+\mathbf{x}})}{B(\mathbf{q}+\mathbf{g}_{i}+\mathbf{x})}, \quad i \neq +, -, \qquad (2.7)$$

$$\Lambda_{+}(\boldsymbol{\varkappa}) = i \cos^{3} \theta [1/B(\mathbf{q}+\mathbf{g}_{+}+\boldsymbol{\varkappa}) - 1/B^{*}(\mathbf{q}+\mathbf{g}_{+}-\boldsymbol{\varkappa})], \quad (2.8)$$

$$s = \int_{0}^{\infty} F(t') dt', \qquad (2.9)$$

r, **r'**, and \varkappa are two-dimensional vectors and the quantity *s* introduced by Eq. (2.9) represents dimensionless time. Since the vectors \mathbf{g}_i correspond to harmonics with the maximum value of the instability increment (see Ref. 2), they are subject to the condition

$$\operatorname{Re}(\partial\Lambda_i(\varkappa)/\partial\varkappa)_{\varkappa=0}=0.$$
(2.10)

Equation (2.5) and the function $I_i(\mathbf{r},t)$ are of interest at those times when the amplitudes of the periodic structures are large compared with their initial values. In this case the real part of the argument of the exponential function for a large part of the integration domain in Eq. (2.6) is considerably greater than unity, so that we can use the steepest-descent method. We then obtain the following expressions for the functions $I_i(\mathbf{r},t)$ which are valid in a wide range of variables:

$$I_{i}(\mathbf{r},t) = \frac{k_{0}^{2}}{\pi (k_{0} |\mathbf{r}|)^{\frac{1}{1}}} [DH_{i}s(1+i)]^{-\frac{1}{2}} \\ \times \exp\left[(1+i) \frac{sH_{i}}{2\xi'} \left(1 - \frac{h_{i}\varphi_{i}^{2}}{2} \right) \right]$$
(2.11)

$$-(1+i)\frac{(k_{0}|\mathbf{r}|-vH_{i}s)^{2}}{DH_{i}s}$$

$$+ik_{0}|\mathbf{r}|\left(1-\cos\varphi_{i}+\frac{1}{2}(\zeta'+|\zeta''|)^{2}\right)\right], \ i\neq -, -,$$

$$I_{+}(\mathbf{r},t)=\frac{k_{0}^{2}}{4\sin^{2}\theta}\frac{1}{Ds\cos^{3}\theta}$$

$$\times \exp\left[\frac{s\cos^{3}\theta}{\zeta'}-(1+i)\frac{(k_{0}r_{1}-vs\cos^{3}\theta)^{2}}{Ds\cos^{3}\theta}\right]$$

$$-(1-i)\frac{(k_{0}r_{2}-vs\cos^{3}\theta)^{2}}{Ds\cos^{3}\theta}+i\frac{k_{0}(r_{1}-r_{2})}{2}(\zeta'+|\zeta''|)^{2}\right].$$
(2.12)

We will introduce here the notation

$$v = \frac{1}{2\zeta'^{2}}(\zeta' + |\zeta''|), D = \frac{2}{\zeta'^{3}}(\zeta' + |\zeta''|)^{2}, \qquad (2.13)$$

$$h_{i} = \frac{1}{H(\alpha_{i})} \left(\frac{\partial^{2} H}{\partial \alpha^{2}} \right)_{\alpha = \alpha_{i}}, \quad r_{i,2} = \frac{1}{2} \left(\mathbf{r}, \frac{\mathbf{g}_{+}}{\cos^{2} \theta} \pm \frac{\mathbf{q}}{\sin^{2} \theta} \right),$$
(2.14)

where the angle α_i corresponds to a maximum of the function $H(\alpha)$ and φ_i is the angle between **r** and $\mathbf{q} + \mathbf{g}_i$.

We shall now consider the range of angles of incidence $\theta \leq |\zeta''| |\zeta'$. Equations (1.2), (1.4), and (1.5) remain valid if b_+ and b_- are understood to represent a mode obtained as a result of merging of degenerate and nondegenerate modes in the limit $\theta \rightarrow 0$ and if the angle α_g is between the vector g and the plane in which the incident wave is polarized at right angles. In Eqs. (2.5) and (2.6) we have to substitute $\Lambda_0(\varkappa)$ in the form

$$\Lambda_0(\boldsymbol{\varkappa}) = i \left[\frac{\sin^2 \alpha_{g_0 + \boldsymbol{\varkappa}}}{B(g_0 + \boldsymbol{\varkappa})} - \frac{\sin^2 \alpha_{-g_0 - \boldsymbol{\varkappa}}}{B^{\bullet}(-g_0 - \boldsymbol{\varkappa})} \right], \quad |g_0| = k_0, \quad \alpha_{g_0} = \frac{\pi}{2}$$

A formula similar to Eqs. (2.11) and (2.12) can now be written as follows:

$$I_{0}(\mathbf{r},t) = \frac{k_{0}^{2}}{\pi (k_{0} |\mathbf{r}|)^{\frac{1}{2}}} (2Ds)^{-\frac{1}{2}} \exp\left\{\frac{s}{\zeta'} (1-\varphi^{2}) -2\frac{k_{0}^{2} |\mathbf{r}|^{2}}{Ds} + \frac{ik_{0} |\mathbf{r}|}{2} [\varphi^{2} + (\zeta' + |\zeta''|)^{2}]\right\} + \text{c.c.}$$
(2.15)

An analysis of Eqs. (2.11), (2.12), and (2.15) leads to the following conclusions. In the range of angles of incidence $\theta \ge |\zeta''|\zeta'$ an instability resulting in the formation of periodic structures is of convective nature: the rise of the \mathbf{g}_i th Fourier component of the initial perturbation $b^0(\mathbf{r})$ is accompanied by a simultaneous displacement on the surface.³ For a non-degenerate mode $(i \ne +, -)$ this displacement occurs in the direction of the vector $\mathbf{q} + \mathbf{g}_i$ and for a degenerate mode it occurs in the direction of the vector \mathbf{q} . The absolute dimensionless velocities obtained from the formula $\mathbf{V}_l = k_0 d\mathbf{r}_l/ds$, where \mathbf{r}_l is the spatial coordinate of the maximum of a perturbation packet, are given by

 $|\mathbf{V}_{i}| = vH_{i}, i \neq +, -; |\mathbf{V}_{+}| = 2v \cos^{4} \theta.$

When the angle of incidence obeys $\theta \ll |\zeta''|\zeta'$, the instability is absolute.³

Perturbation packets spread with time in accordance with diffusion laws. The order of magnitude of the dimensionless diffusion constant is given by Eq. (2.14). The shape of a two-dimensional packet depends strongly on the case

FIG. 1. Shapes of the packets $I_i(\mathbf{r},s)$ for the degenerate (a) and nondegenerate (b) cases with s-polarized incident waves; $\theta \ge |\zeta''|\zeta'$.

under discussion. For example, in the degenerate case the packet is an ellipse with the axes

$$a \sim \cos \theta (Ds \cos^3 \theta)^{\frac{1}{2}} / k_0, \quad b \sim \sin \theta (Ds \cos^3 \theta)^{\frac{1}{2}} / k_0,$$

where a is directed at right-angles to the vector **q** and b along the vector **q** (Fig. 1a). In the nondegenerate case a packet is a circular sector of radius $R \sim (H_1Ds)^{1/2}/k_0$ with a vertex angle $\varphi \sim (2\zeta'/H_1h_1s)^{1/2}$ and inside these packets the "memory" of the initial conditions decays with distance in accordance with the law $(k_0|\mathbf{r}|)^{-1/2}$ (Fig. 1b). This is the shape of a packet for angles of incidence satisfying $\theta \ll |\zeta''|\zeta'$, but the radius of the circular sector is now $\sim (\frac{1}{2}Ds)^{1/2}/k_0$, whereas the vertex angle is $\sim (\zeta'/s)^{1/2}$.

Since the initial conditions during the formation of surface structures are of random nature, it is desirable to consider the correlation functions of the amplitudes of the profile of the new surface structure:

$$G_{ij}(\mathbf{r}-\mathbf{r}', t, t') = \langle b_i(\mathbf{r}, t) b_j^*(\mathbf{r}', t') \rangle, \qquad (2.16)$$

where the angular brackets denote averaging over the realizations of the initial state $b^{0}(\mathbf{r})$ and, because of the statistical homogeneity of the system, the function G_{ij} depends on the difference between the coordinates $\mathbf{r} - \mathbf{r}'$. We shall rewrite G_{ij} in terms of the solutions of the linear problem (2.5) given above. We shall bear in mind that the correlation length of the initial surface irregularities is usually $l_0 \sim 10^{-2} - 10^{-5}$ cm (Ref. 4) and much less than the spatial scale of the functions $I_i(\mathbf{r},t)$, which is

$$L\approx (Ds)^{\prime\prime_2}/k_0=\lambda/\pi\zeta^{\prime\prime_3}(\zeta^\prime+|\zeta^{\prime\prime}|)^2.$$

After substitution of Eq. (2.5) in Eq. (2.16), and appropriate calculations, we find that

$$G_{ij}(\mathbf{r}, t, t') = \delta_{ij}G_0 \int d\mathbf{r}' I_i(\mathbf{r}+\mathbf{r}', t) I_i^{\bullet}(\mathbf{r}', t'),$$

$$G_0 = \int d\mathbf{r}' \exp(-i\mathbf{g}\mathbf{r}') \langle b^0(\mathbf{r}') b^{0\bullet}(0) \rangle.$$

Using the form of the functions I_i , we can readily obtain the following expression for $G_{ij}(\mathbf{r},t,t')$:

$$G_{ij}(\mathbf{r}, s, s') = \delta_{ij}G_0 \frac{k_0^2}{\pi (k_0 |\mathbf{r}|)^{\frac{1}{2}}} \{DH_i[s(1+i)+s'(1-i)]\}^{-\frac{1}{2}} \\ \times \exp\left\{\frac{H_i[s(1+i)+s'(1-i)]}{2\zeta'} \left(1-\frac{h_i\varphi_i^2}{2}\right) -2\frac{[k_0 |\mathbf{r}|-vH_i(s-s')]^2}{DH_i[s(1-i)+s'(1+i)]} +ik_0 |\mathbf{r}| \left[1-\cos\varphi +\frac{1}{2}(\zeta'+|\zeta''|)^2\right]\right\}, \quad i \neq +, -, \quad (2.17)$$

$$G_{++}(\mathbf{r}, s, s') = \frac{k_0^2 G_0}{4 \sin 2\theta} \frac{1}{D \cos^3 \theta |s/(1+i)+s'/(1-i)|} \\ \times \exp\left\{\frac{(s+s')\cos^3 \theta}{\zeta'} - \frac{[k_0 r_1 - v \cos^3 \theta (s-s')]^2}{D \cos^3 \theta [s/(1+i)+s'/(1-i)]} - \frac{[k_0 r_2 - v \cos^3 \theta (s-s')]^2}{D \cos^3 \theta [s/(1+i)+s'/(1-i)]}\right\}$$

$$+\frac{i}{2}k_{0}(r_{1}-r_{2})(\zeta'+|\zeta''|)^{2}\bigg\}, \qquad (2.18)$$

$$G(\theta \sim 0, \mathbf{r}, s, s') = G_0 I_0(\mathbf{r}, s + s').$$
(2.19)

We must mention one further consequence of the fact that the initial condition length l_0 is much less than the spatial scale L. Irrespective of the statistics which is obeyed by the initial conditions $b^0(\mathbf{r})$, the functions $b_i(\mathbf{r},t)$ have the Gaussian statistics. Therefore, the correlation functions determine fully the functional of the distribution of the probability for a random field $b_i(\mathbf{r},t)$.

3. NONLINEAR STAGE OF THE PROCESS

In this section we shall consider in detail the nonlinear stage for a degenerate mode. We shall assume that an *s*-polarized wave is incident at an angle $\theta \gg |\zeta''| \zeta'$. As before, only the operators \hat{B} determine the spatial dependences of all the functions. The system (1.2), (1.4), and (1.5) then becomes

$$[\zeta'(1-i)+\hat{\beta}_{+}]\varepsilon_{+}(\mathbf{r},s)/\cos^{2}\theta = -\tilde{b}_{+}(\mathbf{r},s)-\tilde{b}_{+-}(\mathbf{r},s)\varepsilon_{-}(\mathbf{r},s),$$
(3.1)

$$[\zeta'(1-i)+\hat{\beta}_{-}]\varepsilon_{-}(\mathbf{r},s)/\cos^{2}\theta = \tilde{b}_{-}(\mathbf{r},s)-\tilde{b}_{+-}(\mathbf{r},s)\varepsilon_{+}(\mathbf{r},s),$$
(3.2)

$$d\tilde{b}_{+}(\mathbf{r}, s)/ds = \cos\theta[\varepsilon_{-}(\mathbf{r}, s) - \varepsilon_{+}(\mathbf{r}, s)], \quad \tilde{b}_{-} = \tilde{b}_{+}(\mathbf{r}, s)$$

$$\frac{db_{+-}(\mathbf{r},s)}{ds} = -\left(\cos 2\theta/2\cos\theta\right)\varepsilon_{+}(\mathbf{r},s)\varepsilon_{-}(\mathbf{r},s),\qquad(3.4)$$

where we have separated from \hat{B}_{\pm} the operator term $i\hat{B}_{\pm} = \zeta'(1-i) + \hat{\beta}_{\pm}$ and introduced the notation $\tilde{b}_{l} = k_{0}b_{l}$.

The system (3.1)-(3.4) differs from the corresponding system for the MDM by the presence of the operators representing differentiation with respect to the spatial coordinates $\hat{\beta}_{\pm}$. We shall consider the solution obtained for $|\tilde{b}_{\pm}(s)|$ in the MDM. As shown in Ref. 2, when the angles of incidence obey $\theta > \pi/4$, the quantity $|\tilde{b}_{\pm}(s)|$ rises without limit with time. We shall consider this specific case. During the initial stage of the process the nonlinear effects can be ignored and $|\tilde{b}_{\pm}(s)|$ revolves in accordance with the law

$$\tilde{b}_{+}(s) = |\tilde{b}_{+}(0)| \exp(s\cos^{3}\theta/\zeta').$$
(3.5)

When $|\tilde{b}_{+-}(s)|$ becomes of the order of $2\zeta'/\cos^2\theta$, we can no longer ignore the nonlinear effects. From this condition we find the time s_0 when the nonlinear terms begin to play an important role:

$$s_0 \approx \frac{\zeta'}{\cos^3 \theta} \ln \frac{b_0}{|\tilde{b}^+(0)|}, \quad b_0 = \frac{4\zeta'}{\cos \theta |\cos 2\theta|^{\frac{\gamma_0}{\gamma_0}}}.$$
 (3.6)

If $|\tilde{b}_{+-}(s)|$ rises sufficiently to satisfy $|\tilde{b}_{+-}(s)| \ge 2\xi'/\cos^2\theta$, we can ignore the left-hand side in Eqs. (3.1)-(3.2); we then find that $|\tilde{b}_{+}(s)|$ is described by

$$|\tilde{b}_{+}(s)| \approx 4\cos^{2}\theta(s-s_{0})/|\cos 2\theta|^{\eta}.$$
(3.7)

Since the derivative $d |\tilde{b}_+(s)|/ds$ rises with time and by the moment $s \approx s_0$ becomes of the order of $2/\cos^2\theta$, the additional time Δs necessary for the transition from the regime of Eq. (3.5) to that of Eq. (3.7) is much shorter than s_0 :

$$\Delta s/s_0 \sim [\ln (b_0/|\tilde{b}_+(0)|)]^{-1} \ll 1.$$

The dependence of $|\tilde{b}_+(s)|$ on s obtained above is plotted in Fig. 2.

We shall now find the approximate solution of the system (3.1)-(3.4). The solution $\tilde{b}_+(\mathbf{r},s)$ for the linear stage is given by Eq. (2.15). We shall denote it by $\tilde{b}_+^0(\mathbf{r},s)$. It then follows from Eqs. (3.1)-(3.4) that since s lies within a region Δs near s_0 , the smallness of the region allows us to ignore the spatial evolution of the packets to within $[\ln(b_0/|\tilde{b}_+^0(0)|)]^{-1/2} \ll 1$. When s leaves this region, then—as in the case of the MDM—we can ignore the left-hand sides of Eqs. (3.1) and (3.2) so that the solution analogous to Eq. (3.7) becomes

$$\tilde{b}_{+}(\mathbf{r}, s) = \tilde{b}_{+}^{0}(\mathbf{r}, s_{0})(s-s_{0})4\cos^{2}\theta/|\tilde{b}_{+}^{0}(\mathbf{r}, s_{0})||\cos 2\theta|^{\gamma_{2}}.$$
(3.8)

We note that since the terms containing the derivatives can be ignored in Eqs. (3.1) and (3.2), the spatial structure is "frozen" and this is expressed by Eq. (3.8).

We thus find that the solution $\tilde{b}_+(\mathbf{r},s)$ can be written in the form

$$\tilde{b}_{+}(\mathbf{r},\mathbf{s}) = \begin{cases} \tilde{b}_{+}^{0}(\mathbf{r},s), & s < s_{0}, \\ \frac{4\cos^{2}\theta}{|\cos 2\theta|^{\frac{\gamma_{c}}{2}}} \frac{\tilde{b}_{+}^{0}(\mathbf{r},s_{0})}{|\tilde{b}_{+}^{0}(\mathbf{r},s_{0})|} (s-s_{0}), & s > s_{0}. \end{cases}$$
(3.9)

Similarly the solution $\tilde{b}_{+-}(\mathbf{r},s)$ is found to be as follows:

(0)

$$\tilde{b}_{+-}(\mathbf{r},s) = \begin{cases} 0, & \text{or } s_0 \\ 2\cos\theta \left(\frac{\tilde{b}_{+}^0(\mathbf{r},s_0)}{|\tilde{b}_{+}^0(\mathbf{r},s_0)|}\right)^2 & (s-s_0), & s>s_0. \end{cases}$$
(3.10)

s<sa.

We can determine the correlation functions during the linear stage if we average Eqs. (3.9) and (3.10). Bearing in mind that the random field $b_{+}^{0}(\mathbf{r},s)$ obeys the Gaussian statistics with the correlation function $G_{++}(\mathbf{r},s,s')$, which is given by Eq. (2.32), and carrying out standard calculations, we find that for $s,s' > s_0$, we have

$$G_{++}(\mathbf{r}, s, s') = \frac{16 \cos^{4} \theta}{k_{0}^{2} |\cos 2\theta|} (s - s_{0}) (s' - s_{0}) \frac{G}{|G|^{2}} \times \{E(|G|) - (1 - |G|^{2}) K(|G|)\},$$
(3.11)



FIG. 2. Dependence of Eq. (3.7) representing the amplitude of a grating $|\tilde{b}_+(s)|$ as a function of time s.

$$G_{+-,+-}(\mathbf{r},s,s') = \frac{4\cos^2\theta}{k_0^2}(s-s_0)(s'-s_0)\frac{G^2}{|G|^2} \times \{|G|^2 + (1-|G|^2)\ln(1-|G|^2)\},$$
(3.12)

where $G = G_{++}(\mathbf{r}, s_0, s_0)/G_{++}(0, s_0, s_0)$; K and E are complete elliptic integrals of the first and second kind, respectively. It follows from Eq. (3.11) that the spatial dimensions of a packet formed during the linear stage remain unaltered (Fig. 1a). The shape of the packet changes in the nonlinear stage and the change becomes greater on approach to the center of the packet ($\mathbf{r} = 0$), whereas at the edges the shape of the packet is the same as that of $G_{++}(\mathbf{r}, s_0, s_0)$. We can say qualitatively that a packet becomes flatter as a result of non-linear interactions, but its spatial dimensions are not affected. It should be noted that during the nonlinear stage the instability becomes absolute.

These conclusions are valid also, though only qualitatively, in the nondegenerate case which can be considered in a similar manner.

4. CONCLUSIONS

We shall now consider how well the above theory describes the experimental results. Gratings observed in experiments do not usually have a perfect regular spatial structure. Quite the opposite, in many cases such gratings are so distorted that even their very existence is in doubt.¹ The degree of distortion of the gratings can be described by correlation functions $\langle b_l(\mathbf{r},t)b_l^*(\mathbf{r}'t')\rangle$, which have the form of packets that depend strongly on the parameters of the problem and the nature of the grating. Therefore, the spatial structure of the distortions of the gratings varies within wide limits.

It follows from our discussion that in the degenerate case the area of a correlation packet represents an ellipse (Fig. 1a) and s grows linearly with time. At the end of the linear stage the parameters of the ellipse may become $a \sim b \sim (10^2 - 10^3)\lambda$, where λ is the wavelength of the incident radiation. Therefore, in this case we can speak of the formation of a "good" grating. It follows from the results of Sec. 3 that when nonlinear effects become important, this correlation structure is frozen and subsequently only the amplitude of the grating increases with time. In our opinion, this conclusion is interesting from the point of view of experimental generation of regular gratings. It is clear from Fig. 1a and from Eq. (3.6) that the area of a correlation packet at the end of the linear stage is an ellipse with the parameters

$$\frac{a}{\sin\theta} = \frac{b}{\cos\theta} \approx \frac{\left[2\ln\left(b_0/\left|\widetilde{b}_+(0)\right|\right)\right]^{\frac{1}{2}}}{2\pi\zeta'\left|\zeta''\right|}\lambda,\tag{4.1}$$

which depends strongly on the properties of the target material: the smaller the product of the real and imaginary parts of the surface impedance, the better the correlation of the resultant gratings. It should be noted that the parameters of surface irregularities before irradiation occur in Eq. (4.1) in the form $[\ln(b_0/|\tilde{b}_+(0)|)]^{1/2}$ and have practically no influence on the correlation length.

In the nondegenerate case two gratings are formed during the linear stage and they are well correlated along the vector $\mathbf{q} + \mathbf{g}_i$, but in the perpendicular direction the correlation length is $\varphi \sim (2\zeta'/sH_1h_1)^{1/2}$ times less. It follows from Fig. 1b that the transverse correlation length varies slowly with time and has the value $\sim \lambda(\pi |\zeta''|\zeta')^{-1}$. The correlation length along the vector $\mathbf{q} + \mathbf{g}_i$ increases with time as $s^{1/2}$ and at the end of the linear stage it has a value of the order of that given by Eq. (4.1). Therefore, nondegenerate gratings in the direction perpendicular to the vector $\mathbf{q} + \mathbf{g}_i$ are correlated much less, in agreement with qualitative considerations put forward in Ref. 5.

A more detailed comparison of the theory and experiment becomes possible when quantitative results on the correlation properties of the structures found experimentally are obtained.

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