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# Determination of rate of epitaxial growth by means of combined usage of atomic force microscopy and light scattering data

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**Abstract**. In the paper, a method of experimental estimation of epitaxial growth rate has been suggested. A corner stone of this method is obtaining image of initial shape of solid state surface by means of atomic force microscopy before the start of the process of epitaxial growth. These experimental data should be completed by measurements of special bistatic cross-section of visible light scattering on the sample surface both before the beginning of technological process and after its end. Mathematical model of epitaxial growth is based on simplification of the Kardar-Parisi-Zhang equation. The method of characteristics has been applied to solve this equation.

## 1. Introduction

Since the pioneer work [1] of Donald Eigler and Erhard Schweizer, words 'nanotechnology' and 'nanoengineering' were synonyms. But after appearance of paper [2], another sense of term 'nanoengineering' has taken place too; namely, one may understand nanoengineering as the problem of optimal control by distributed parameter system.

Let us describe epitaxial growth of solid state surface on the basis of the Kardar-Parisi-Zhang (KPZ) model [3]:

$$\frac{\partial H}{\partial t} = v + \frac{v}{2} \cdot (\nabla H)^2 + D \cdot \nabla^2 H + Q(\vec{x}, t), \quad (1)$$

where $H(\vec{x}, t)$ is the height of solid state surface, $\vec{x} = (x_1, x_2)$ is two-dimensional vector of transversal coordinates, $\nabla$ is two-dimensional gradient, $v$ is the rate of growth of the surface along the local normal to it, $D$ is the diffusion coefficient of sputtering matter and $Q(\vec{x}, t)$ is additional source of external particles near the surface.

The KPZ-equation (1) proves to be highly adequate to the physical experiment during the simulation of manufacturing process of multilayer mirrors and gratings employed in X-ray optics [4]; therefore, verification procedure for model (1) of surface growth with support of this one by means of atomic force microscopy (AFM) is not required.

On the domain $\Omega \subset R^2$ equation (1) ought to be equipped by initial condition:

$$H(\vec{x}, 0) = H_0(\vec{x}), \quad (2)$$

corresponding to initial shape of the surface under investigation.

If the source $Q(\vec{x},t) \geq 0$ on the cylinder $\Omega \times [0, t_*]$, then one can consider this function as a control [2]. At last, let one impose on the solution of equation (1) with initial condition (2) the requirement that at the moment of time $t = t_*$ function $H(\vec{x},t)$ must provide the best mean square approximation of fixed shape $H_*(\vec{x})$:

$$\int_\Omega [H(\vec{x},t_*) - H_*(\vec{x})]^2 d^2x \to \min . \tag{3}$$

It is obvious that equation (1) and restrictions (2)-(3) form the problem of optimal control by distributed parameter system [2, 5]. But to construct the numerical solution of this problem, one has to find parameters $v$ and $D$ in input equation (1).

If $\operatorname{diam}\Omega >> \sqrt{D \cdot t_*}$, then there is a way to estimate these values; namely, in the absence of additional source $Q(\vec{x},t)$ of sputtering particles the approximate solution of the Cauchy problem (1)-(2) is equal to [3]:

$$H(\vec{x},t) \approx v \cdot t + \frac{2 \cdot D}{v} \cdot \ln\left( \int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} \exp\left[ -\frac{(\vec{x}-\vec{\xi})^2}{4 \cdot D \cdot t} + \frac{v \cdot H_0(\vec{\xi})}{2 \cdot D} \right] \cdot \frac{d^2\xi}{4 \cdot \pi \cdot D \cdot t} \right). \tag{4}$$

Further height of the surface $H(\vec{x},t)$ on domain $\Omega$ must be measured by means of AFM both before the beginning of the technological process and after the finish of it. This information is enough to apply method of solution of inverse coefficient problem for determination of parameters $v$ and $D$ developed in paper [6]. The application of this method meets with difficulties in practice as it belongs to the advanced chapters of modern functional analysis [7]. Thus, more straightforward approaches for estimation of these parameters based on physical arguments are to be considered.

This paper deals with estimation for the rate $v$ of epitaxial growth of crystal surface in the situation when $v \cdot H >> D$, where $H$ is typical value of height of crystal surface roughness. In this case, one can neglect in input equation (1) by term with Laplasian. And after putting $Q(\vec{x},t) = 0$ in equation (1), this one is reduced to the so-called equation simplified Kardar-Parisi-Zhang (sKPZ) equation:

$$\frac{\partial H}{\partial t} = v + \frac{v}{2} \cdot (\nabla H)^2 . \tag{5}$$

This equation differs sharply from the primordial KPZ one (1); moreover, equation (5) can be solved implicitly by means of the method of characteristics [8]:

$$H = v \cdot t + H_0(\vec{y}) - \frac{v \cdot t}{2} \cdot (\nabla H_0(\vec{y}))^2 , \qquad \vec{x} = \vec{y} - v \cdot t \cdot \nabla H_0(\vec{y}), \tag{6}$$

where $\vec{y} = (y_1, y_2)$ are Lagrange variables. To derive explicit dependence for height of the surface $H(\vec{x},t)$, one must find from the second equation of system (6) vector-function $\vec{y} = \vec{y}(\vec{x},t)$ and substitute it into the first equation of system (6). Generally speaking, it is impossible to obtain analytical formula connecting the initial shape of the surface (2) with the rate $v$. Nevertheless, absence of terms describing the surface diffusion of sputtering substance and the external source of sputtering particles in equation (5) gives one a possibility to determine the rate of growth of the surface.

In practice, initial condition (2) for equation (5) is often equal to superposition of smooth large scale function $h_0(\vec{x})$ and small perturbation $\mu \cdot u_0(\vec{x})$:

$$H_0(\vec{x}) = h_0(\vec{x}) + \mu \cdot u_0(\vec{x}) , \tag{7}$$

where $\mu$ is small parameter: $0 < \mu << 1$. As a rule, large scale function $h_0(\vec{x})$ is a result of preliminary technological processing of the surface before the start of epitaxial process and small perturbation is some technologically unremovable fluctuations. An example of such surface is shown in Figure 1.

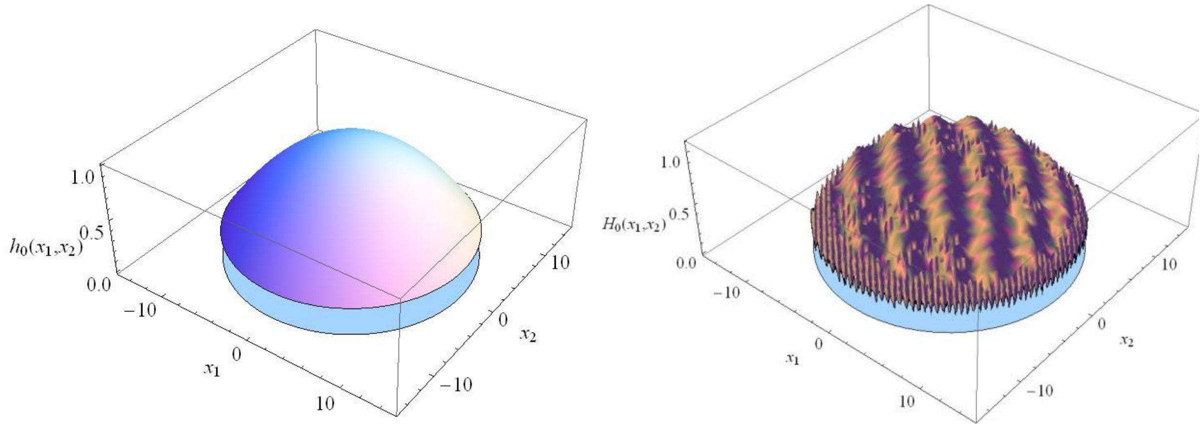


**Figure 1.** Initial condition as smooth large scale function (on the left) and as superposition of smooth large scale function and its small perturbation (on the right).

For further considerations, it is important that due to representation (7) of initial shape, specific bistatic cross-section of electromagnetic waves on this surface proves to be calculated.

The rest of the article is organized as follows: in Section 2 peculiarities of construction of asymptotic solution for the sKPZ-equation (5) for the special choice of function $h_0(\vec{x})$ are demonstrated. Section 3 deals with the estimation of specific bistatic cross-section of electromagnetic waves on this surface. Final Section is devoted to the discussion of results elaborated and perspectives of further investigations.

## 2. Derivation of asymptotic solution for the simplified KPZ-equation

Decomposition (7) for initial condition means that in order to construct solution of equation (5) one has to use perturbation theory:

$$H(\vec{x},t) = h(\vec{x},t) + \mu \cdot u(\vec{x},t) + \dots . \tag{8}$$

Substitution of expansion (8) into the sKPZ-equation (5) gives rise to a chain of the Cauchy problems; namely, for zero order approximation on $\mu$ in asymptotic series (8) the Cauchy problem is:

$$\frac{\partial h}{\partial t} = v + \frac{v}{2} \cdot (\nabla h)^2, \qquad h(\vec{x},0) = h_0(\vec{x}), \tag{9}$$

and for the first order approximation on $\mu$ in asymptotic series (8) the Cauchy problem is:

$$\frac{\partial u}{\partial t} - v \cdot \nabla h(\vec{x},t) \cdot \nabla u = 0, \qquad u(\vec{x},0) = u_0(\vec{x}). \tag{10}$$

In order to do the next step in solution of the system of equations (9)-(10) one has to render concrete function $h_0(\vec{x})$.

If one chooses regular shape $h_0(\vec{x})$ in initial condition (7) as follows:

$$h_0(\vec{x}) = h_* - \frac{\vec{x}^2}{2 \cdot L_*}, \tag{11}$$

where $h_*$ and $L_*$ are constant positive values, then in the framework of the method of characteristics for the first-order partial differential equation [9] it is easy to check that in this case exact solution of the Cauchy problem (8) is equal to:

$$h(\vec{x},t) = h_* + v \cdot t - \frac{\vec{x}^2}{2 \cdot (L_* + v \cdot t)}. \tag{12}$$

Substituting this function into equation (10) one can find that it is reduced to the next equation:

$$\frac{\partial u}{\partial t} + \frac{v \cdot \vec{x}}{L_* + v \cdot t} \cdot \nabla u = 0\,. \tag{13}$$

This equation is linear partial differential equation with variable coefficients and one can derive exact solution of the Cauchy problem for equation (13) with initial condition $u_0(\vec{x})$ by the same method of characteristics [9]:

$$u(\vec{x},t) = u_0\left(\frac{L_* \cdot \vec{x}}{L_* + v \cdot t}\right). \tag{14}$$

Thus asymptotic solution of the sKPZ-equation (5) with regular part (11) of initial condition (7) is equal to:

$$H(\vec{x},t) = h_* + v \cdot t - \frac{\vec{x}^2}{2 \cdot (L_* + v \cdot t)} + \mu \cdot u_0\left(\frac{L_* \cdot \vec{x}}{L_* + v \cdot t}\right) + O(\mu^2)\,. \tag{15}$$

Schematic graphs of section of function (15) under the assumption $h_*/L_* << 1$ at initial moment of time (below) and at arbitrary moment of time $t > 0$ (higher) are presented in Figure 2. Temporal evolution of function (12) is presented on this figure by blue lines. From Figure 2 one can see that under growth of time roughness of surface shape becomes smaller in comparison with roughness of initial shape.

Such behaviour is determined by two circumstances. First of all, from formula (14) it is clear that spatial scale $l(t)$ of perturbation $u(\vec{x},t)$ grows as $l(t) = l_0 \cdot (1 + v \cdot t/L_*)$, where $l_0$ is spatial scale of initial perturbation $u_0(\vec{x})$. On the other hand, from formula (12) it is obvious that with increasing time function $h(x,t)$ becomes closer to plane $z = h_* + v \cdot t$. These planes are shown in Figure 2 by dashed lines. It means that if for some positive value $\lambda$ the following inequalities are true:

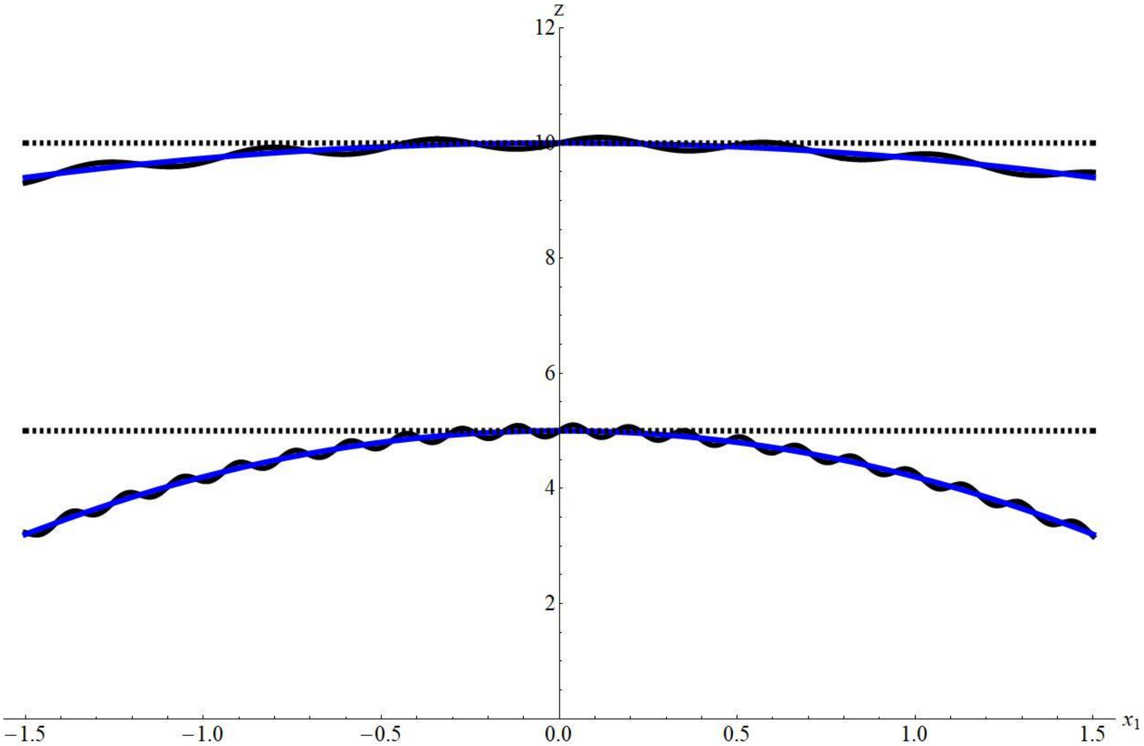


**Figure 2.** Temporal evolution of the section of the height by plane $x_2 = 0$.

$$\mu\cdot|u_0(\vec{x})|<<\lambda,\qquad \mu\cdot|\nabla u_0(\vec{x})|<<1,\tag{16}$$

then with increasing time inequalities:

$$\mu\cdot|u(\vec{x},t)|<<\lambda,\qquad \mu\cdot|\nabla u(\vec{x},t)|<<1,\tag{17}$$

are true too.

If one interprets value $\lambda$ as wavelength of monochromatic visible light: $\lambda = 380\div 780$ nm, then under $\max_{\vec{x}\in\Omega}|u_0(\vec{x})|\sim 10$ nm inequalities (16) are valid. Therefore, inequalities (17) are valid too. Hence, one can describe scattering of monochromatic visible light on the surface (15) in the framework of a model of small-scale surface [10].

## 3. Specific bistatic cross-section of electromagnetic waves on the shape evolving in accordance with the simplified KPZ-equation

The simplest formulation for a model of small-scale surface can be done under the conjecture that perturbation of smooth surface $u_0(\vec{x})$ is stationary stochastic two-dimensional field with zero average: $<u_0(\vec{x})>=0$.

In this case, it is obvious from formula (14) that $<u(\vec{x},t)>=0$. Therefore, from formula (15) one can easily find that $<H(\vec{x},t)>=h(\vec{x},t)$. It means that covariance function of total height $H(\vec{x},t)$ of solid state surface can be calculated directly:

$$B(\vec{\xi},t)=<(H(\vec{x},t)-<H(\vec{x},t)>)\cdot(H(\vec{x}+\vec{\xi},t)-<H(\vec{x}+\vec{\xi},t)>)>,\tag{18}$$

the two-dimensional Fourier-transform from function (18):

$$S(\vec{q},t)=\int B(\vec{\xi},t)\cdot\exp(-i\cdot\vec{q}\cdot\vec{\xi})\cdot d^2\xi\tag{19}$$

being its spectral density.

Further, let one consider scattering on the surface (15) of monochromatic visible light with horizontal (H) and vertical (V) polarizations:

$$\vec{E}_H^{(i)}=(0,1,0)\cdot A(x_1,z),\qquad \vec{E}_V^{(i)}=(\cos\theta_i,1,\sin\theta_i)\cdot A(x_1,z),\tag{20}$$

where $A(x_1,z)=E_0\cdot\exp(i\cdot k\cdot\cos\theta_i\cdot x_1-i\cdot k\cdot\sin\theta_i\cdot z)$ is complex amplitude of monochromatic wave, $k=2\cdot\pi/\lambda$ is wavevector corresponding to wavelength $\lambda$ and $\theta_i$ is input angle.

At last, because of validity of inequalities (17) specific bistatic cross-sections with polarizations $a,b=H,V$ of waves (20) on the surface under investigation are equal to [10]:

$$\sigma^0_{ab}(\vec{q},t)=\frac{4\cdot k^4}{\pi}\cdot f_{ab}(\theta_i,\theta_s,\phi_s)\cdot S(\vec{q},t),\tag{21}$$

where

$$f_{ab}(\theta_i,\theta_s,\phi_s)=\begin{pmatrix}\cos^2\theta_i\cdot\cos^2\theta_s\cdot\cos^2\phi_s & \cos^2\theta_i\cdot\sin^2\phi_s\\ \cos^2\theta_s\cdot\sin^2\phi_s & (\sin\theta_i\cdot\sin\theta_s-\cos\phi_s)^2\end{pmatrix}\tag{22}$$

is matrix of polarization coefficients, $\theta_s$ and $\phi_s$ are scattering angles in spherical system of coordinate and arguments in spectral density in (21) are equal to:

$$q_1=k\cdot\sin\theta_s\cdot\cos\phi_s-k\cdot\sin\theta_i,\qquad q_2=k\cdot\sin\theta_s\cdot\sin\phi_s.\tag{23}$$

Using formula (14), it is easy to find that

$$B(\vec{\xi},t)=\mu^2\cdot K_0\left(\frac{L_*\cdot\vec{\xi}}{L_*+v\cdot t}\right)+o(\mu^2),\tag{24}$$

where

$$K_0(\vec{\xi}) = < u_0(\vec{x}) \cdot u_0(\vec{x}+\vec{\xi}) > \tag{25}$$

is autocorrelated function of stochastic field $u_0(\vec{x})$.

Thus, spectral density for covariance function (24) is equal to:

$$S(\vec{q},t) = \mu^2 \cdot \left( \frac{L_* + v \cdot t}{L_*} \right)^2 \cdot S_0\left( \frac{L_* + v \cdot t}{L_*} \cdot \vec{q} \right) + o(\mu^2), \tag{26}$$

where $S_0(\vec{q}) = \int K_0(\vec{\xi}) \cdot \exp(-i \cdot \vec{q} \cdot \vec{\xi}) \cdot d^2\xi$ is spectral density corresponding to autocorrelated function (25).

In particular, at $t = 0$ formula (26) is reduced to:

$$S(\vec{q},0) = \mu^2 \cdot S_0(\vec{q}) + o(\mu^2). \tag{27}$$

Due to validity of inequalities (16) at $t = 0$ specific bistatic cross-sections also can be calculated in accordance with formulae (21)-(23).

Combining under $\vec{q} = 0$ formulae (26) and (27) with formula (21) one can derive that:

$$\frac{\sigma^0_{ab}(0,t)}{\sigma^0_{ab}(0,0)} = \left( 1 + \frac{v \cdot t}{L_*} \right)^2 + O(\mu). \tag{28}$$

Due to requirement $\vec{q} = 0$, formulae (23) give one position of receiver of monochromatic light with respect to transmitter:

$$\phi_s = 0, \quad \theta_s = \theta_i. \tag{29}$$

Substituting values (29) into formulae (22) one can obtain that $\sigma^0_{VH} = \sigma^0_{HV} = 0$ and $\sigma^0_{HH} = \sigma^0_{VV}$.

## 4. Conclusion

In the article, algorithm of determination of epitaxial growth rate $v$ in the framework of the sKPZ-equation has been presented. In order to find this parameter, one has to do the following operations:

- to prepare initial shape $h_0(\vec{x})$ of the surface under investigation with regular part (10) under condition $h_*/L_* << 1$;
- to measure specific bistatic cross-section $\sigma^0_{HH}$ (or $\sigma^0_{VV}$) of visible light before the start of technological process;
- to measure by means of AFM the real height $H_0(\vec{x})$ of the sample before the start of technological process;
- to perform for real initial shape $H_0(\vec{x})$ denoising procedure with the help of thresholding [11];
- to find the parameter $L_*$ of smoothed shape (11) in the vicinity of point $(0,0)$ by means of mean-square fitting;
- to fix the duration $t_*$ of technological process;
- to measure specific bistatic cross-section $\sigma^0_{HH}$ (or $\sigma^0_{VV}$) of visible light after the end of technological process;
- to calculate epitaxial growth rate $v$ in accordance with formula (28).

Thresholding is known to be a kind of denoising procedure in the framework of wavelet analysis [11]. To adjust this procedure to real AFM data, one has to do a number of computational experiments in order to find optimal type of wavelet and optimal level of wavelet decomposition.

In conclusion, it is necessary to underline that formulae (12) and (14) are rigorously valid only under suggestion that process of epitaxial growth develops only in small neighbourhood of point $(0,0)$. More careful mathematical treatment with boundary conditions for equation (5) is required in order to estimate its influence on exactness of formula (28).

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