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Oscillatory Temperature-driven Morphological Relaxation of Surface Ripple Using Weak Pulsed Laser

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ABSTRACT

A continuum (Mullins-type) model is proposed for the non-isothermal, isotropic evolution of a crystal surface on which mass transport occurs by surface diffusion. The departure from constant temperature is assumed induced by low-energy incident pulsed radiation. It has been previously shown experimentally and theoretically that such heating mode gives rise to the quasistationary regime, in which the surface temperature of a thick solid film oscillates about the mean value with the pulse repetition frequency. The implications of oscillatory driving with high frequency values on relaxation of surface ripple are examined; in particular, the traveling wave solutions with decreasing amplitude are detected numerically. Pulsed heating also results in faster smoothing of the ripple, compared to the case when the surface is at constant temperature which is same as the mean temperature in the pulsed heating mode. Impact on ripple shape is minor for ripple amplitudes considered.

INTRODUCTION

Recently, experimental efforts have been undertaken on assessing impact of weak, interfering pulsed laser beams on nano-scale pattern formation in the course of crystal growth from vapor [1]-[4]. It was suggested that non-isothermal surface diffusion resulting from the rapid spatio-temporal variations of surface temperature is the cause of pattern formation.

In a separate line of research, the mesoscopic step-flow model was proposed in Ref. [5]. The adatom diffusivity, kinetic attachment coefficients and equilibrium concentration were assumed periodically oscillating in time about the respective mean values. Pulsed laser irradiation was suggested as one of possible methods to excite oscillations. Slope selection, surface metastability, and driving frequency-dependent surface stability have been found. Thus it seems that oscillatory driving of morphological evolution on crystal surfaces has the potential to demonstrate rich nonlinear phenomena similar to one found in other branches of physics; the inverted pendulum problem and pattern formation in oscillatory driven fluids and granular media are two of the many examples (see references in Ref. [5]).

Here one more step is taken towards understanding the impact of weak, non-intrusive pulsed laser beams on pattern formation. The macroscopic, continuum model is developed to study morphological relaxation of surface ripples; this framework could be easily extended to study thermodynamically unstable crystal growth which results in pattern formation.

THE MODEL

Considered is a 1+1 case corresponding to a two-dimensional crystal with a one-dimensional surface. The evolution of the crystal surface $z = h(x,t)$ is described by the following phenomenological partial differential equation [6] - [8]:

$$h_t = B\frac{\partial}{\partial x}\left(\frac{\exp\left(-E_d/\left\{kT\right\}\right)}{kT}(1+h_x^2)^{-1/2}\left[\frac{-h_{xx}}{(1+h_x^2)^{3/2}}\right]_x\right), \tag{1}$$

where $B = \Omega^2\nu D_0\gamma$ and the Arrhenius dependence of diffusivity on temperature is shown explicitly. D_0 and E_d are the pre-factor and activation energy, respectively; other parameters have usual meaning. Subscript x denotes differentiation.

Let the horizontal dimension of the spatially modulated (rippled) surface is L, and let ℓ is a constant integer number of ripple wave lengths per L. Then $\lambda = L/\ell$ is the wave length of the ripple and $q = 2\pi/\lambda$ is the wave number. Assume the entire surface is irradiated by laser pulses.

Eq. (1) admits trivial solution (equilibrium state)

$$h = h_0 = const. \tag{2}$$

at any temperature $T = T(x,t)$. Without loss of generality $h_0 = 0$ can be chosen. When pulsed laser heating is applied to the surface for sufficiently long period of time, a quasistationary regime develops, which is characterized by the mean temperature $T_0 = const. > T_a$ (where T_a is ambient temperature), and small temperature oscillations on this background [9, 10]. Consider perturbation of the equilibrium state $h = \tilde{h}(x,t)$, and simultaneous perturbation of the mean temperature field, $T = T_0 + \tilde{T}(x,t)$, such that

$$\left|\frac{\tilde{T}(x,t)}{T_0}\right| \equiv |\hat{T}(x,t)| \ll 1, \quad \tilde{h}(x,0) = H_0\cos qx, \tag{3}$$

and $|\tilde{h}(x,t)|$ is not necessarily small.

Next, the expression $\exp\left(-E_d/\left\{kT_0(1+\hat{T})\right\}\right)/\left\{kT_0(1+\hat{T})\right\}$ in Eq. (1) is expanded in powers of small quantity $\hat{T}(x,t)$, and the expression in parenthesis is differentiated with respect to x. This yields:

$$h_t = BP_{n-1}\left(\hat{T}\right)\hat{T}_x(1+h_x^2)^{-1/2}\left[\frac{-h_{xx}}{(1+h_x^2)^{3/2}}\right]_x + BP_n\left(\hat{T}\right)\left\{(1+h_x^2)^{-1/2}\left[\frac{-h_{xx}}{(1+h_x^2)^{3/2}}\right]_x\right\}_x, \tag{4}$$

where $P_n\left(\hat{T}\right)$ and $P_{n-1}\left(\hat{T}\right) = dP_n/d\hat{T}$ are polynomials of degrees n and $n-1$, respectively. The tilde sign over $\tilde{h}(x,t)$ has been omitted. Numerical simulations demonstrated that taking $n = 2$ is sufficient to accurately compute evolutions of the shape. Thus $n = 2$ is assumed for the rest of the paper. The coefficients of the polynomials P_2 and P_1 are

$$a_2 = a_0\left(\frac{g_0^2}{2} - 2g_0 + 1\right), \quad a_1 = a_0\left(g_0 - 1\right), \quad a_0 = \frac{\exp\left(-g_0\right)}{kT_0}, \tag{5}$$

where

$$g_0 = \frac{E_d}{kT_0}. \tag{6}$$

The coefficients have dimension of inverse energy.

Eq. (4) is general in the sense that it admits any small perturbation of the mean surface temperature field T_0. The particular choice for the perturbation is discussed in the next Section.

Model for the temperature perturbation

Pulsed irradiation of a crystal surface gives rise to a quasistationary state in which temperature at the targeted spot fluctuates about the mean value T_0 with a frequency equal to the source pulse repetition frequency ω. With the goal of modeling the effects produced by pulsed irradiation the simplest model form for the perturbation $\hat{T}(x,t)$ is postulated:

$$\hat{T}(x,t) = (\cos \omega t)\left[Q_0 + Q_1 h_* \cos\left(qx + \beta\right)\right], \tag{7}$$

where $0 < Q_0,\ Q_1 h_* \ll 1$, h_* is nondimensionalized h and β is the phase shift of the modulation with respect to the ripple. The values of T_0, Q_0 and Q_1 are determined by the impulsive power density and the mean power density of the radiation, the absorbivity of the surface at the radiation wavelength, and the thermophysical and optical characteristics of the material [9]-[12].

The periodicity in time of the perturbation is the consequence of the well-developed quasistationary regime, as the result of the pulsed irradiation. The two terms in Eq. (7) model the quasistationary regime on the rippled, evolving surface of a solid film. Note that as $h_* \to 0$ only the first term remains, which describes the quasistationary regime on a flat, horizontal and stationary surface. The proportionality of the amplitude of the second term to first power of h_*, and the spatially-periodic form are assumed after [11, 12], where the related theories of formation of laser-induced surface ripples (LISR) were developed; see also the review paper [13], and [14].

The problem (4) - (7) has two time scales. These scales are the period of pulse repetition $t_p = 1/\omega$, and the characteristic time of ripple relaxation at constant $T = T_0$. This latter scale is the time it takes the surface diffusion to diminish the initial amplitude of the ripple by e times, and it is given by $t_s = \left(B a_0 q^4\right)^{-1}$. The case of high pulse repetition frequency will be considered, so that $t_p \ll t_s$. In this limit the ripple relaxation on the long time scale t_s can be studied using Eq. (4) where the time- and space-periodic functions $BP_{n-1}\left(\hat{T}\right)\hat{T}_x$ and $BP_n\left(\hat{T}\right)$ are averaged over the temporal period of oscillation. The averaging procedure reduces to zero such terms in the latter function that correspond to odd powers of $\hat{T}(x,t)$, and also such terms in the former function that correspond to odd sum of powers of $\hat{T}(x,t)$ and $\hat{T}_x(x,t)$. The averaged and non-dimensionalized evolution Eq. (4) reads:

$$h_t = -\bar{B}\left[\bar{\sigma}_x \bar{\sigma}(1+h_x^2)^{-1/2}\left[\frac{-h_{xx}}{(1+h_x^2)^{3/2}}\right]_x - \left(A_1 + \frac{\bar{\sigma}^2}{2}\right)\left\{(1+h_x^2)^{-1/2}\left[\frac{-h_{xx}}{(1+h_x^2)^{3/2}}\right]_x\right\}_x\right], \tag{8}$$

where

$$\bar{\sigma}(x,t) = Q_0 + Q_1 h \cos\left(\bar{q}x + \beta\right). \tag{9}$$

The non-dimensional parameters are $\bar{B} = a_2 B/(\omega L^4)$, $A_1 = a_0/a_2$, $A_2 = a_4/a_2$, $A_3 = a_6/a_2$ and $\bar{q} = 2\pi\ell$. $1/\omega$ is used for unit of time and L for unit of length. Note that in the case $n = 1$ the averaged evolution equation reduces to nonlinear, isothermal ($T = T_0$) Mullins equation.

The method of lines approach was used for computations, with the central, second-order finite differencing in space and the implicit Runge-Kutta integration in time [15]. Boundary conditions at $x = 0, 1$ are periodic. By closely examining Eq. (8) it become clear that values $\beta = 0, \pi/2, \pm\pi/4$ are special in the sense that they can either change signs of periodic coefficients, or turn the sine function into the cosine function (or vice versa). Thus only these values were tried in numerical simulation. At $t = 0$ the ripple shape is given by Eq. (3), where the initial amplitude H_0 is replaced by nondimensional initial amplitude $\bar{H}_0 = H_0/L$. The values of nondimensional parameters correspond to GaAs on GaAs diffusion [14]. $\ell = 4$ ensures that $t_s \gg t_p$; $L = 10\mu$m.

RESULTS

Small amplitude case

Figure 1 shows the nondimensional amplitudes of the ripple at crests (vs. time) for $\beta = 0, \pi/2$ and $\pm\pi/4$. For all four values of β the ripple shape is the cosine function at all times.

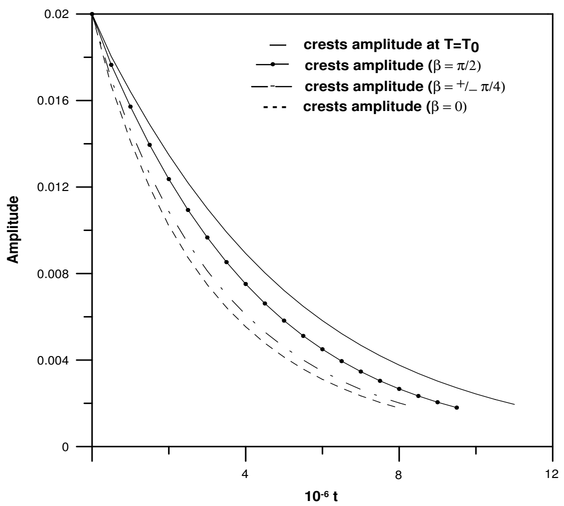

Figure 1: Amplitude of the ripple given phase shift in Eq. (7) either $\beta = 0$, or $\beta = \pm\pi/4$, or $\beta = \pi/2$. Solid curve: pulsed heating is off and surface is at constant temperature T_0. $Q_0 = 0.1$, $Q_1 = 5.0$.

It can be seen that for any value of the phase shift the ripple under the oscillatory driving about mean temperature T_0 relaxes faster than the ripple held at constant temperature T_0. The relaxation is fastest when $\beta = 0$, e.g. when the extrema of the spatially-periodic multiplier in Eq. (7) occur at ripple extrema.

Traveling Waves

In the numerical simulations with $\beta = \pi/2, \pm\pi/4$ and sufficiently large values of Q_0 and Q_1 the ripple undergoes the uniform translation to the left or right with small, but detectable speed. In other words, the traveling wave with the decaying amplitude appears on the surface. The direction of the traveling wave depends on magnitudes of Q_0 and Q_1, as well as on value of β. For instance, for $\beta = \pi/2$, $Q_0 = 0.1$ and $Q_1 = 2.5,\ 5.0$ the traveling wave direction is to the right, but the direction is opposite for $Q_0 = 0.5$ and $Q_1 = 2.5$. Waves always travel in the opposite directions for $\beta = \pi/4$ and $-\pi/4$. For $\beta = 0$ the traveling wave is absent for any values of Q_0 and Q_1.

Large amplitude case

The main difference from the small amplitude case is the shape of the ripple; it slightly deviates from the cosine function for amplitudes in the range 0.08 - 0.03 due to, primarily, strong nonlinearity. The contribution of the pulsed heating effect itself in shape deviation is present, but is very small for $\bar{H}_0 = 0.08$ (that is, almost same deviation occurs when the pulsed heating is off and the surface is held at constant temperature T_0). For smaller amplitudes the ripple shape is indistinguishable from the cosine function, as discussed above.

CONCLUSIONS

This paper suggests simple, nonlinear continuum model of driven morphological evolution by nonisothermal surface diffusion (such that the temperature oscillations about mean value T_0 are induced by weak pulsed laser beam) of the pre-existed surface morphology. The numerical simulations demonstrate that rates of smoothing are faster than classical rate for the isothermal, no oscillations case $T = T_0$. Also, the unexpected traveling wave mode of relaxation is detected for some values of the parameter governing the horizontal shift of the (time-oscillatory) temperature perturbation with respect to the ripple. For more information, see [14]. Work on the model for pattern-forming, thermodynamically unstable crystal growth is in progress.

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