

# Two-dimensional wetting on an elastic substrate

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The influence of substrate elasticity on the phenomenon of wetting in two-dimensional systems is considered. For the transition of two steps from a bound state to a free state it is shown that the interaction of the steps via elastic deformations of the substrate leads to a logarithmic growth of the activation barrier for the transition as  $T \rightarrow T_c$ . It is found that in a two-dimensional lattice of steps the corresponding phase transition should be first-order. For the problem of the wetting of a step by a two-dimensional phase it is found that the transition should occur discontinuously from incomplete to complete wetting, with a large activation barrier.

## 1. INTRODUCTION

The problem of wetting has attracted considerable attention in recent years. A review of the numerous papers on this theme can be found in Refs. 1–4. As a rule, the problem of wetting is understood as that of finding how the thickness of an adsorbed film depends on various parameters, e.g., the temperature and chemical potential. The wetting phase transition is understood as the transition from a thin (two-dimensional) to a thick (three-dimensional) adsorbed film. The work done in recent years has demonstrated the decisive influence of long-range interactions, e.g., van der Waals interactions on the wetting phenomenon.<sup>4–6</sup>

The problem of wetting can also be formulated in two dimensions. In this case we are concerned with the change in the width of a strip of some two-dimensional phase growing in the neighborhood of a line defect on a surface. In experiment, this phenomenon has been observed repeatedly on electron micrographs.<sup>7–10</sup> In all the cases investigated the role of the line defects has been played by steps on the surface of a single crystal, while the role of the two-dimensional phase has been played by the adsorbate or the reconstructed surface.

A similar problem is the question of the transition of two or more interacting linear entities from a bound state to a free state.<sup>11–14</sup> The decay of steps from heights of two or three lattice constants to heights equal to one lattice constant has been observed in the experiments of Refs. 15 and 16.

It is natural to expect that in the two-dimensional case, as in the three-dimensional case, interactions that fall off slowly will determine the behavior of the system in wetting. An example of such an interaction in two-dimensional systems is the elastic interaction via the substrate. Between the steps there is a repulsive interaction that falls off as the inverse square of the distance between them.<sup>17</sup> The energy of the elastic deformations that arise when a strip of the two-dimensional phase appears increases logarithmically with the width of the strip.<sup>18</sup> In this paper we shall consider the effect of such elastic deformations on the phenomenon of two-dimensional wetting.

## 2. TRANSITION OF TWO STEPS FROM A BOUND TO A FREE STATE

We shall describe this transition by a model of two interacting filaments, arranged, in the ground state, along the  $x$  axis. The Hamiltonian of the model has the form

$$H = \int dx \{ \frac{1}{2} J_1 [ (\nabla l_1)^2 + (\nabla l_2)^2 ] + U(l_1 - l_2) \}. \quad (1)$$

Here  $l_1$  and  $l_2$  are the displacements of the filaments in the direction perpendicular to the filaments. We shall not consider states in which the steps pass through each other, i.e., when “overhangs” are formed on the surface. For definiteness, we shall choose  $l_1 > l_2$ . The potential of the interaction between two steps at a distance  $l$  is conveniently chosen in the form

$$U(l) = \begin{cases} \infty, & l < 0 \\ -U_0, & 0 < l < a, \\ \alpha/l^2, & l > a \end{cases} \quad (2)$$

where  $a$  is a distance of the order of several interatomic spacings. The potential (2) describes attraction at short distances ( $U_0 > 0$ ) and repulsion on account of elastic deformations<sup>17</sup> at large distances.

The step is displaced as a result of the formation of kinks (a break in the step) with energy  $E_k$ . Considering the mean square displacements of a filament and a step it is easy to relate  $J_1$  to  $E_k$ :

$$J_1 = T \exp(E_k/T)/b, \quad (3)$$

where  $b$  is the lattice constant. Replacing  $l_1$  and  $l_2$ :

$$l_1 = \frac{1}{2}(l + l_0), \quad l_2 = \frac{1}{2}(l_0 - l), \quad (4)$$

we obtain the Hamiltonian describing the relative displacements of the filaments ( $J = \frac{1}{2}J_1$ ):

$$H = \int dx \left\{ \frac{1}{2} J \left( \frac{dl}{dx} \right)^2 + U(l) \right\}. \quad (5)$$

One-dimensional systems with a Hamiltonian of the type (5) are conveniently studied by going over from the one-dimensional statistical problem to the one-dimensional quantum problem.<sup>19–21</sup> Here the free energy of the system is  $F = TE_0$ , where  $E_0$  is the energy of the ground state of the Schrödinger equation

$$-\frac{T^2}{2J} \frac{d^2\Psi}{dl^2} + U(l)\Psi = E\Psi. \quad (6)$$

In the wetting problem it is customary to characterize the behavior of the system by the mean square fluctuation  $\xi_{\perp}^2$  and correlation length  $\xi_{\parallel}$ . These are expressed in terms of the wavefunctions  $\Psi_n$  as follows:

$$\xi_{\perp}^2 = \int dl \Psi_0^2(l) (l - \bar{l})^2, \quad (7)$$

where  $\bar{l} = \int dl \Psi_0(l)$ ;

$$\langle [l(x) - \bar{l}] [l(0) - \bar{l}] \rangle = \sum_n \exp[-(E_n - E_0) |x|] \int dl \Psi_0 \Psi_n l \propto \exp[-(E_1 - E_0) |x|] = \exp(-|x|/\xi_{\parallel}). \quad (8)$$

In the case of the potential (2) Eq. (6) can be solved exactly. For  $E < 0$  the substitution  $\Psi = l^{1/2} f(\beta l)$  brings Eq. (6) for  $l > a$  to the form

$$f'' + \frac{1}{\beta l} f' - \left( \frac{\nu^2}{\beta^2 l^2} + 1 \right) f = 0, \quad (9)$$

where

$$\beta^2 = |E| 2J/T, \quad \nu = (2J\alpha/T^2 + 1/\epsilon)^{1/2}.$$

The solution of (9) that decreases as  $l \rightarrow \infty$  is expressed in terms of the modified Bessel function  $K_\nu$  (Ref. 22). Thus, for  $l > a$  the wavefunction has the form

$$\Psi = B l^{1/2} K_\nu(\beta l). \quad (10)$$

The asymptotic forms of  $\Psi$  are

$$\Psi \approx B (\pi/2\beta)^{1/2} e^{-\beta l}, \quad \beta l \rightarrow \infty, \quad (11)$$

$$\Psi \approx 1/2 B \Gamma(\nu) (2/\beta)^\nu e^{1/2 - \nu}, \quad \beta l \rightarrow 0. \quad (12)$$

In the region  $0 < l < a$  the solution of (6) has the form

$$\Psi = A \sin ka, \quad k = [2J(U_0 + |E|)^{1/2}/T].$$

From the joining condition we obtain an equation for the dependence  $E(T)$ :

$$ka \operatorname{ctg} ka = 1/2 - \beta \alpha K_{\nu-1}(\beta a) / K_\nu(\beta a). \quad (13)$$

The transition point is determined from the condition  $E_0 = 0$ . In this case the solution of Eq. (6) can be expressed in terms of elementary functions. The solution that decreases as  $l \rightarrow \infty$  has the form

$$\Psi = B l^{1/2 - \nu}, \quad (14)$$

and the equation for the transition temperature is

$$ka \operatorname{ctg} ka = 1/2 - \nu \quad (15)$$

or

$$z \operatorname{ctg} z = 1/2 - (z^2 \delta + 1/\epsilon)^{1/2}, \quad z = (2JU_0)^{1/2} a / T_c; \quad \delta = \alpha/a^2 U_0.$$

The solution  $z_c$  of Eq. (15) as a function of  $\delta$  increases monotonically from  $\pi/2$  to  $\pi$ :

$$z_c = \begin{cases} 1/2\pi(1+\delta), & \delta \ll 1 \\ \pi(1-1/\pi\delta)^{1/2}, & \delta \gg 1 \end{cases} \quad (16)$$

Taking into account the dependence  $J_1(T)$  given by (3), we obtain an equation for the transition temperature

$$T_c = E_k / \ln(\pi^2 c^2 b T_c / U_0 a^2), \quad (17)$$

where  $1/2 < c < 1$  is determined by the quantity  $\delta$ . The approximate solution of (17) for  $E_k/T_c \gg 1$  has the form

$$T_c \approx \frac{E_k}{\ln(E_k \pi^2 c^2 b / U_0 a^2)}. \quad (18)$$

Using the wavefunctions (10), it is easy to determine the dependence  $\xi_{\perp}(T - T_c)$  near the transition point:

$$\xi_{\perp}^2 \propto 1/|E| \propto 1/|T - T_c|. \quad (19)$$

For  $T > T_c$  the bound state is separated, because of the elastic repulsion, from the free potential barrier. We shall calculate how the height  $E_a$  of this barrier depends on  $T - T_c$ . The metastable states of the pair of steps in the quantum-mechanical analogy correspond to quasistationary levels with  $E > 0$ , and the probability of formation of a segment with free steps is proportional to the probability of a tunneling transition from the quasistationary to the free state. Therefore, knowing the dependence of the width of a quasistationary level on  $E$ , we can determine  $E_a(T - T_c)$ . Performing transformations analogous to those in the case  $E < 0$ , we represent the solution of (6) in the region  $l > a$  in the form

$$\Psi = \frac{l^{1/2}}{S^{1/2}} \left[ -\exp\left(-i\frac{\pi}{2}\left(\nu + \frac{1}{2}\right)\right) H_\nu^{(2)}(\beta l) + S \exp\left(i\frac{\pi}{2}\left(\nu + \frac{1}{2}\right)\right) H_\nu^{(1)}(\beta l) \right]. \quad (20)$$

Here  $H_\nu^{(1)}$  and  $H_\nu^{(2)}$  are Hankel functions<sup>22</sup> and  $S$  is the scattering matrix. The asymptotic form of the solution (20) as  $l \rightarrow \infty$  is

$$\Psi \approx (2/\pi S)^{1/2} (-e^{-i\beta l} + S e^{i\beta l}). \quad (21)$$

From the joining condition we find  $S$ :

$$S = (Z/Z^*) \exp(-i\pi(\nu + 1/2)), \quad (22)$$

where

$$Z = \left[ 1 - \frac{\operatorname{tg} ka}{ka} (\nu + 1/2) - \frac{\beta}{k} \operatorname{tg} ka \frac{Y_{\nu-1}(\beta a)}{Y_\nu(\beta a)} \right] + i \frac{J_\nu(\beta a)}{Y_\nu(\beta a)} \left[ 1 - \frac{\operatorname{tg} ka}{ka} (\nu + 1/2) - \frac{\beta}{k} \operatorname{tg} ka \frac{J_{\nu+1}(\beta a)}{J_\nu(\beta a)} \right],$$

and the symbol \* denotes complex conjugation. Here  $J_\nu$  and  $Y_\nu$  are Bessel functions of the first and second kind.<sup>22</sup> The poles of the scattering matrix (22) determine the energy  $E_0$  and width  $\gamma$  of the quasistationary states. We shall be interested in the neighborhood of the transition point, i.e.,  $E_0 \rightarrow 0$ , which corresponds to  $\beta a \ll 1$ . The Bessel functions  $J_\nu$  and  $Y_\nu$  have a simple form near zero<sup>22</sup>:

$$J_\nu \approx \left(\frac{z}{2}\right)^\nu / \Gamma(\nu+1), \quad Y_\nu(z) = \frac{1}{\pi} \Gamma(\nu) \left(\frac{z}{2}\right)^{-\nu}. \quad (23)$$

The metastability effects will be manifest in the case of a large potential barrier, i.e., for  $\alpha/a^2 U_0 \gtrsim 1$ . We shall consider the limiting case  $\alpha/a^2 U_0 \gg 1$ , which corresponds to  $\nu \gg 1$  for  $T \sim T_c$ . In this case, in the scattering matrix (22) we can neglect the terms containing  $Y_{\nu-1}(\beta a)/Y_\nu(\beta a)$  and  $J_{\nu+1}(\beta a)/J_\nu(\beta a)$ . The imaginary part of the denominator of (22) decreases like  $E^{2\nu}$  while the real part decreases like  $E$  as  $E \rightarrow 0$ . In these conditions the concept of a quasistationary level is fully applicable. It is not difficult to obtain expression for the dependences  $E_0(T - T_c)$  and  $\gamma(T - T_c)$ :

$$\frac{E_0}{U_0} \approx \frac{T - T_c}{T_c} \left(1 + \frac{E_k}{T_c}\right), \quad \gamma = \left(\frac{E_0}{U_0} \frac{\pi^2 \nu^2}{16e^2}\right)^\nu \propto \left(\frac{T - T_c}{T_c}\right)^\nu. \quad (24)$$

Thus, the probability of a transition from a metastable bound state to the free state tends to zero by a power law as  $T \rightarrow T_c$ , corresponding to a logarithmic growth of the activation energy of the process. The exponent in the dependence  $\gamma(T - T_c)$  is easily obtained in the quasiclassical approximation:

$$\begin{aligned} & \gamma^{-1} \propto \exp \left\{ -\frac{2}{T} \int_a^{(\alpha/E)^{1/2}} \left[ 2J \left( \frac{\alpha}{x^2} - E \right) \right]^{1/2} dx \right\} \\ & = \exp \left\{ -\frac{2(2J\alpha)^{1/2}}{T} \ln \left[ \left( \frac{\alpha}{E} \right)^{1/2} \frac{1}{a} \right] \right\} \propto E^{-\nu} \left( \frac{T - T_c}{T_c} \right)^{-\nu}. \end{aligned} \quad (25)$$

The condition for applicability of the quasiclassical approximation is the requirement  $\nu \gg 1$ . We note that in the general case of an interaction potential of the form  $\alpha/x^n$  between the steps the quasiclassical approximation is applicable only for  $n = 1, 2$  (Ref. 23). For  $n = 1$  the tunneling probability is proportional to  $\exp[-\text{const}/(T - T_c)^{1/2}]$ . For  $n > 2$  the quasiclassical formula gives a finite tunneling probability as  $E \rightarrow 0$  ( $T \rightarrow T_c$ ), in contradiction to the exact solution.<sup>23</sup>

As  $T \rightarrow T_c$  the size  $L_c$  of a critical nucleus of a state with steps that have dissociated, as follows from (25), has the behavior

$$L_c \approx (\alpha/E)^{1/2} \propto (T_c/|T - T_c|)^{1/2}. \quad (26)$$

The kinetics of the process by which the steps pass from a bound to a free state (and back) is determined by several factors. The results obtained above make it possible to distinguish two factors that lead to a growth of the relaxation time as  $T \rightarrow T_c$ . These are, first, the activation barrier (24), (25), and, second, the growth of the size  $L_c$  of a critical nucleus. The second factor is associated with the necessity of diffusion of surface atoms through distances at least of order  $L_c$  in the rearrangement of the steps. The probabilities of both processes have a power dependence on  $T - T_c$  and  $T \rightarrow T_c$ . Therefore, we may expect that the total probability of formation of a nucleus will vary as a power of  $T - T_c$  and the corresponding relaxation time will satisfy

$$\tau \propto |(T - T_c)/T_c|^{-\nu_1},$$

where  $\nu_1 > \nu$ . The exact relationship between  $\nu_1$  and  $\nu$  can depend on the details of the relaxation mechanism, about which little is known, and can differ, therefore, from the very simple relation  $\nu_1 = \nu + 1$ .

As noted above, metastable phenomena should be observed only for  $\nu \gg 1$ . At the same time, this condition should correspond to the parameters that obtain in experiment. In this case, the contribution to the exponent  $\nu_1$  from the kinetics of formation of the nucleus—a contribution connected with the fact that  $L_c \rightarrow \infty$  as  $T \rightarrow T_c$ , will be small in comparison with the contribution from the activation barrier.

This approach does not permit a rigorous investigation of the kinetics of the system, and the accuracy of the estimate of the kinetics corresponds essentially to the accuracy of the quasiclassical approximation (25). However, solving (13) exactly makes it possible, on the one hand, to investigate accurately the thermodynamics of the problem, and, on the other, to justify the results of the quasiclassical approximation.

In experiment the transition of steps from a bound state to a free state has been observed on vicinal faces with angular inclination  $5\text{--}15^\circ$  (Ref. 16). In this case the system is two-dimensional and one may speak of a phase transition. We shall consider the question of the order of the phase transition in the case when  $L \rightarrow \infty$ , where  $L$  is the period of the lattice of steps in the free state. We shall write a phenomenological expression for the free-energy density of the lattice of steps in the free state:

$$F_1 = A_1/L^3 + B_1 T^2/J_2(T)L^3. \quad (27)$$

Here  $A_1$  and  $B_1$  are certain constants. The first term in (27) describes the elastic repulsion, and the second describes the contribution to the free energy from thermal fluctuations of the steps. This term is simply the kinetic energy of a quantum particle in a box of width  $L$ . In the bound state the free-energy density can be represented in the form

$$F_2 = \frac{A_2}{(2L)^3} + \frac{B_2 T^2}{J_2(T)(2L)^3} + \frac{D T^2}{4A_1 a^2 L} - \frac{U_0}{2L}. \quad (28)$$

Here  $D \approx 1$ . The other constants can be related to the constants in (27). For simplicity we shall assume that  $A_2 = L_1 A_1$ , since the energy of the elastic interaction of the steps is proportional to the square of the step height,<sup>17</sup> and that  $B_2 = B_1$  and  $J_2 = 2J_1$ . The third and fourth terms in (28) describe the free energy of the steps in the bound state and correspond to the contribution of thermal fluctuations and the binding energy. Equating  $F_1$  and  $F_2$ , we can obtain an equation for  $T_c$ , analogous to (17):

$$(2JU_0)^{1/2} \frac{a}{T_c} = \left[ \frac{D(1 + A/2U_0 L^2)}{2(1 - 15Ba^2/16DL^2)} \right]^{1/2}. \quad (29)$$

The temperature dependence of the free energy is connected with the second term for  $F_1$  and with the second and third terms for  $F_2$ . These dependences differ only by a factor of order  $\alpha/L^2 \ll 1$ . Consequently, the entropies of the phases at the transition point will differ sharply. This implies that, in the framework of the model considered, the phase transition in the lattice of steps for large values of  $L$  will be first-order.

The reasoning given is based implicitly on the assumption of a large barrier between the bound state and free state of the steps. Such a barrier, as indicated above, arises for  $\nu \gg 1$ . In the opposite case of small  $\nu$ , pronounced fluctuation phenomena, which are not taken into account in the above phenomenological analysis, will be present in the system. In this case it is also possible that the transition of the decoupling of the steps is a second-order transition. Unfortunately, in the case of the second-order transition we have not succeeded in identifying the universality class.

For a system of two steps the relaxation time  $\tau$  tends to infinity as  $E \rightarrow 0$  [see (26)]. In a two-dimensional lattice of steps  $E$  is bounded by the quantity  $\alpha/L^2$ . Therefore, the maximum relaxation time attainable in experiment will behave as

$$\tau_{\max} \propto L^{2\nu_1}. \quad (30)$$

### 3. TWO-DIMENSIONAL WETTING

We turn to the problem of the wetting of steps by a two-dimensional phase. As shown by observations in an electron microscope,<sup>7-10</sup> in many cases the phase wetting a step forms a strip of width  $l$  to one side of the step. We shall consider such a strip on a terrace of width  $L$ . The Hamiltonian de-

cribing the system will also have the form (5), but with a different dependence  $U(l)$ , which can be chosen in the form

$$U(l) = \begin{cases} \infty, & l < a, \quad l > L \\ \alpha \ln(l/a) - \mu(l-a) - U_0, & a < l < L \end{cases} \quad (31)$$

where  $\mu$  is the chemical potential,  $\alpha > 0$ , and  $U_0$  describes the energy gain upon wetting of the steps. The logarithmic term in (31) is connected with the energy of the elastic deformations that arise because of the difference between the surface energy of the phase being formed and the surface energy of the remaining substrate.<sup>18</sup> Suppose, for definiteness, that we are concerned with the wetting of a step by a commensurate two-dimensional phase. Then the constant  $J$  can be represented in the form (3). Unfortunately, it has not been possible to obtain an exact solution in the case of the potential (31). In certain cases, however, one can obtain approximate solutions by using the expansion of the potential (31) about the boundaries of the terrace, and the quasiclassical approximation.

We shall consider the case of a rigid interphase boundary ( $J \gg T^2/\alpha a^2$ ) and a broad terrace ( $L \gg a$ ). We approximate the potential at the right and left boundaries of the terrace by a linear potential ( $\mu > 0$ );

$$U_l = \begin{cases} -U_0 + (\alpha/a - \mu)(l-a), & l \geq a \\ \infty, & l < a \end{cases}$$

$$U_r = \begin{cases} -U_0 + \alpha \ln(L/a) - \mu(L-a) + (\mu - \alpha/L)(L-l), & l \leq L \\ \infty, & l > L \end{cases} \quad (32)$$

Expressions for the energy levels in a field of the form (32) are given in, e.g., Ref. 23. Using them, we obtain for the ground-state energies in the wells

$$E_{0l} = -U_0 + (T^2/2J)^{1/2} |\lambda_1| (\alpha/a - \mu)^{3/2},$$

$$E_{0r} = -U_0 + \alpha \ln(L/a) - \mu(L-a) + (T^2/2J)^{1/2} |\lambda_1| (\mu - \alpha/L)^{3/2}. \quad (33)$$

Here  $\lambda_1 = -2.338$  is the first zero of the Airy function. At the transition point, as will be seen below, the wells are separated by a large potential barrier. Neglecting the tunneling splitting of the levels, we obtain, by equating  $E_{0l}$  and  $E_{0r}$ , an equation for the equilibrium curve  $\mu(T)$ . We seek the solution in the form

$$\mu = gL^{-1} \alpha \ln(L/a).$$

Neglecting  $\mu$  in the second term in the expression for  $E_{0l}$  and in the last term in  $E_{0r}$ , we obtain

$$\left(\frac{T^2}{2J}\right)^{1/2} |\lambda_1| \left(\frac{\alpha}{a}\right)^{3/2} = \alpha \ln \frac{L}{a} - g \alpha \ln \frac{L}{a} + g \frac{\alpha a}{L} \ln \frac{L}{a}. \quad (34)$$

Neglecting the last term in (34), we obtain an approximate expression for  $g$  and  $E_0$ :

$$g \approx 1 - |\lambda_1| \left[\frac{T^2}{2J\alpha a^2}\right]^{1/2} / \ln\left(\frac{L}{a}\right), \quad 1 - g \ll 1, \quad (35)$$

$$E_0 + U_0 = |\lambda_1| \left(\frac{T^2}{2J\alpha a^2}\right)^{1/2} \alpha \ll \alpha. \quad (36)$$

The inequalities in (35) and (36) follow from the condition that the interphase boundary be rigid. The latter inequality confirms the applicability of the linear expansion used for the potential. We now calculate in the quasiclassical approximation the tunnel integral determining the activation energy  $E_a$  for the appearance of a region with the two-dimensional phase covering the whole terrace, i.e., for the transition of the interphase boundary from the left well to the right well:

$$E_a = (2J)^{1/2} \int_{a_1}^{L_1} \left[ -\mu(x-a) + \alpha \ln \frac{x}{a} - U_0 - E_0 \right]^{1/2} dx$$

$$= (2J)^{1/2} \int_{a_1}^{L_1} \left[ -\mu(x-a) + \alpha \ln \frac{x}{a} - \alpha \left(\frac{T^2}{2J\alpha a^2}\right)^{1/2} \right]^{1/2} dx, \quad (37)$$

where  $a_1$  and  $L_1$  are the zeros of the expression under the square root. The integral (37) can be estimated by the method of steepest descent. Finally, we obtain the approximate result

$$E_a \approx \frac{(2J\alpha)^{1/2}}{T} L \ln^2\left(\frac{L}{a}\right). \quad (38)$$

By virtue of the assumptions made,  $E_a \gg T$ . This implies that on the transition curve the time of establishment of equilibrium can be too long. In experiment the transition itself will then be observed from a metastable state.

Thus, elastic deformations lead to the result that the width of the strip of adsorbate on the terrace should change discontinuously, from an almost empty to an almost filled terrace. In addition, in experiment pronounced hysteresis phenomena should be observed. When elastic deformations are not taken into account, the width of the strip should grow continuously as a function of  $\mu$  (Ref. 14).

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<sup>23</sup>V. M. Galitskii, B. M. Karnakov, and V. I. Kogan, *Problems in Quantum Mechanics* [in Russian], Nauka, Moscow (1981), p. 648. [First ed. transl. published by Prentice-Hall, Englewood Cliffs (Fig. 3)].

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