

# Dynamics of the one-dimensional nucleus in a decomposing solid solution

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The dynamics of the one-dimensional nucleus (layer) of the new phase in a decomposing solution is investigated within the Cahn–Hilliard equations. An analytic approach that makes it possible to obtain an equation of the dynamics of the one-dimensional nucleus, which is nonlocal with respect to the time, is developed. It is shown that the temporal evolution of the layer depends sensitively on its initial thickness. © 1996 American Institute of Physics.  
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## 1. INTRODUCTION

Decomposing solid solutions are characterized by the fact that two phases differing with respect to the concentrations of the components coexist below the phase-separation temperature. Let us consider a substitutional solid solution consisting of atoms of two kinds (*A* and *B*) with a concentration of atoms of type *A* equal to *C*. The free energy of the decomposing solution has the form of a Landau functional (see, for example, Ref. 1):

$$F = \int \left[ \frac{1}{2} \gamma (\nabla C)^2 - \frac{1}{2} \alpha (C - \bar{c})^2 + \frac{1}{4} \beta (C - \bar{c})^4 \right] d\mathbf{R}, \quad (1)$$

where  $\alpha$ ,  $\beta$ , and  $\gamma$  are positive coefficients. The stationary states of the solution are determined from the condition that the variational derivative  $\delta F / \delta C$  be equal to zero under the additional condition that the total number of particles of each kind be conserved:

$$\frac{1}{v} \int c d\mathbf{R} = c_0, \quad (2)$$

$$c = (C - \bar{c}) \sqrt{\beta / \alpha},$$

where *v* is the volume of the system. The variational procedure leads to the following equation in the dimensionless variables  $\mathbf{r} = \mathbf{R} \sqrt{\alpha / 2\gamma}$ :

$$\Delta c + 2(c - c^3) = h, \quad (3)$$

where *h* is a Lagrange multiplier, which can be determined from the condition (2). Among the solutions of Eq. (3) there are solutions that correspond to the maximum of the functional *F*. In the two- and three-dimensional cases they are radially symmetric solutions, which describe the critical nucleus of the new phase.<sup>2,3</sup> The size of this nucleus is inversely proportional to the constant *h*. The phase boundary in the variables selected has a width  $\sim 1$  and separates metastable and stable phases. The difference between the energies per particle in these phases is proportional to *h*, and, therefore, *h* determines the degree of metastability of the solution. When  $h > 0$ , the phase with  $c \sim 1$  is metastable due to the supersaturation of the *A* phase with component *B*. Therefore, the parameter *h* can also be called the degree of supersaturation in the system. The critical nucleus is known to be unstable against small changes in its size. In Refs. 2 and 3

the dynamics of the growth (dissolution) of the nucleus of the stable phase were investigated within the relaxation equation for the parameter *c*. The results obtained coincide with the results of the classical Zel'dovich–Volmer theory for the relaxation of a metastable phase,<sup>4,5</sup> which is based on the conception of a sharp phase boundary.

In the one-dimensional case the Zel'dovich–Volmer theory predicts the absence of a critical nucleus (a one-dimensional nucleus is understood to be an artificially created layer of the stable phase in a decomposing solid solution). However, the existence of the critical nucleus of the new phase follows from Eq. (3),<sup>6</sup> the dependence of its size on the degree of supersaturation being a logarithmic function rather than a power law, as will be shown below. For this reason, there should be interest in the problem of the dynamics of the boundaries of the one-dimensional nucleus with consideration of their finite thickness. To solve it we utilize the Cahn–Hilliard equation, which describes the dynamics of systems with a conserved order parameter.

In Sec. 2 of this paper we present an analytic solution for the one-dimensional critical nucleus, which is valid for arbitrary degrees of supersaturation of the solution. An equation which describes (for different initial conditions and degrees of supersaturation) the growth or dissolution of the layer of the new phase with consideration of the finite width of the phase boundary is derived and investigated in Sec. 3.

## 2. ONE-DIMENSIONAL CRITICAL NUCLEUS

Here we present an analytic solution of Eq. (3), which describes the one-dimensional critical nucleus and is needed for the ensuing treatment. We assume that the concentration in the solution depends on one variable *z*. Then, Eq. (3) has the integral

$$\frac{1}{2} \left( \frac{\partial c}{\partial z} \right)^2 + U(c) = I,$$

$$U(c) = c^2 - \frac{1}{2} c^4 - hc, \quad h > 0, \quad (4)$$

which is analogous to conservation of the energy when a classical particle moves in the potential *U*(*c*). The concentration distribution for the critical nucleus of the new phase is described by the solution of Eq. (4) corresponding to two

coupled kinks (see Ref. 6), which has the asymptote  $c(z) \rightarrow c_0$  as  $z \rightarrow \pm\infty$  and, therefore, satisfies Eq. (2). Integrating (4) with consideration of the equality  $I = U(c_0)$ , we obtain

$$c = c_0 + \frac{1}{2} \sqrt{(c_0 - c_1)(c_0 - c_2)} \left[ \tanh\left(\frac{x - 2a}{2}\right) - \tanh\left(\frac{x + 2a}{2}\right) \right],$$

$$x = \sqrt{(c_0 - c_1)(c_0 - c_2)} z,$$

$$\cosh(2a) = \frac{2c_0 - (c_1 + c_2)}{c_1 - c_2}, \quad (5)$$

where  $c_0 > c_1 > c_2$  are roots of the equation  $U(c) = U(c_0)$ . At small degrees of supersaturation  $h \ll 1$  we have

$$c = 1 - \frac{h}{4} + \tanh(z - a) - \tanh(z + a),$$

$$a = \frac{1}{4} \ln \frac{16}{h}, \quad c_0 = 1 - \frac{h}{4}, \quad c_{1,2} = -1 \pm \sqrt{h}. \quad (6)$$

Thus, a one-dimensional critical nucleus exists in the supersaturated solution. The existence of the critical nucleus is due to the exponentially weak [ $\sim \exp(-4a)$ ] interaction of the phase boundaries, which leads to a logarithmic dependence of its size on the degree of supersaturation. We note that the existence of the nucleus in the two- and three-dimensional case is associated with different dependences of the bulk and surface energies on its size and that at small degrees of supersaturation it scarcely depends on the width of the phase boundary.<sup>2,3</sup>

We next consider the problem of the growth (dissolution) of the layer of the new phase with consideration of the finite width of the phase boundary.

### 3. DYNAMICS OF THE GROWTH (DISSOLUTION) OF THE LAYER OF THE NEW PHASE

The relaxational dynamics in systems with a conserved order parameter are described by the Cahn-Hilliard equation.<sup>1</sup> In dimensionless variables it has the form

$$\frac{\partial c}{\partial t} = - \frac{\partial^2}{\partial z^2} \left[ \frac{\partial^2 c}{\partial z^2} + 2(c - c^3) \right]. \quad (7)$$

Consider the evolution of the initial distribution corresponding to the layer of the new phase with a concentration  $c \sim -1$ :

$$c(z, t=0) = \tanh[z - z_0(0)] - \tanh[z + z_0(0)] + 1 - h/4, \quad (8)$$

where the initial thickness of the layer  $2z_0(0)$  can be either greater or less than the critical value. The boundary conditions to Eq. (7) are the conditions that there be no diffusive fluxes at infinity:

$$j = \frac{\partial}{\partial z} \left[ \frac{\partial^2 c}{\partial z^2} + 2(c - c^3) \right] \Big|_{\pm\infty} = 0. \quad (9)$$

The second pair of boundary conditions has the form

$$c|_{\pm\infty} = 1 - h/4. \quad (10)$$

Thus, the mean concentration in the solution is equal to the concentration of the phase existing when  $|z| \rightarrow \infty$ . The analytic solution of the problem is based on the assumption that the phase fronts are weakly distorted as they move. We seek the solution of Eq. (7) in the form

$$c(z, t) = \tanh[z - z_0(t)] - \tanh[z + z_0(t)] + 1 + u(z, t). \quad (11)$$

Assuming that the velocity of the boundaries  $V = \partial z_0 / \partial t$  and the correction  $u(z, t)$  are small, we have

$$\frac{\partial u}{\partial t} - Vg = - \frac{\partial^2}{\partial z^2} \left( \frac{\partial^2 u}{\partial z^2} - 4u + 6ug + f \right), \quad (12)$$

where

$$g(z, t) = \frac{1}{\cosh^2(z - z_0)} + \frac{1}{\cosh^2(z + z_0)},$$

$$f(z, t) = 2\{\tanh^3[z - z_0(t)] - \tanh^3[z + z_0(t)] + 1 - [\tanh[z - z_0(t)] - \tanh[z + z_0(t)] + 1]^3\}.$$

It follows from the conservation condition of the parameter  $c$  that

$$\frac{\partial}{\partial t} \int_{-\infty}^{\infty} u dz = 4V, \quad (13)$$

i.e., the variation of the concentration as the phase boundaries move must be "compensated" by the variation of the integral value of the correction  $u$ . It also follows from Eq. (12) that far from the phase boundaries the equation for  $u$  takes the form of an ordinary diffusion equation. Therefore, it is convenient to seek the solution of Eq. (12) in the form of the sum of the diffusion field  $\psi$ , which originates from the moving phase boundaries, and the correction to the stationary form of the phase boundary  $\varphi$ :

$$u(z, t) = \psi(z, t) + \varphi(z, t). \quad (14)$$

The functions  $\psi$  and  $\varphi$  satisfy the equations

$$\frac{\partial \psi}{\partial t} = 4 \frac{\partial^2 \psi}{\partial z^2} + Vg(z, t), \quad \psi|_{\pm\infty} = -h/4, \quad (15)$$

$$\frac{\partial \varphi}{\partial t} = - \frac{\partial^2}{\partial z^2} \left[ \frac{\partial^2 \varphi}{\partial z^2} - 4\varphi + 6g\varphi + f + \frac{\partial^2 \psi}{\partial z^2} + 6g\psi \right],$$

$$\varphi|_{\pm\infty} = 0. \quad (16)$$

Thus, both the interaction of the phase boundaries ( $f$ ) and the diffusion field ( $\psi$ ) are driving forces of the variation of the thickness of the layer. We expand the function  $\varphi$  in the eigenfunctions  $\varphi_n$  of the operator  $\hat{L}$ :

$$\hat{L} = - \frac{\partial^2}{\partial z^2} \left[ - \frac{\partial^2}{\partial z^2} - 6g + 4 \right],$$

$$\hat{L}\varphi_n = \lambda_n \varphi_n, \quad (17)$$

$$\varphi = \sum_n b_n \varphi_n.$$

The boundary conditions for the functions  $\varphi_n$  have the form

$$\frac{\partial}{\partial z} \left[ \frac{\partial^2 \varphi_n}{\partial z^2} + (6g-4)\varphi_n \right] \Big|_{\pm\infty} = 0, \quad \varphi_n|_{\pm\infty} = 0. \quad (18)$$

Multiplying (17) by the functions  $\chi_n$ , which are defined by the relations

$$\varphi_n = \frac{\partial^2 \chi_n}{\partial z^2}, \quad \int_{-\infty}^{\infty} \chi_n \varphi_m dz = 0 \quad \text{for } n \neq m \quad (19)$$

(see Ref. 1), and integrating over the entire  $z$  axis, we obtain

$$\frac{\partial b_n}{\partial t} = -\lambda_n b_n - \int_{-\infty}^{\infty} \varphi_n \left[ f + \frac{\partial^2 \psi}{\partial z^2} + 6g\psi \right] dz. \quad (20)$$

On the basis of the assumption that the correction  $u$  is small, it must be required that

$$\int_{-\infty}^{\infty} \varphi_n \left[ f + \frac{\partial^2 \psi}{\partial z^2} + 6g\psi \right] dz = 0 \quad (21)$$

for all the eigenfunctions corresponding to the negative or zero eigenvalues  $\lambda_n$ . The spectrum of the operator  $\hat{L}$  contains "dangerous" (in the sense indicated) values close to zero. In fact, if there is no interaction between the phase boundaries ( $z_0 \rightarrow \infty$ ), the eigenfunctions  $[\cosh^2(z \pm z_0)]^{-1}$  correspond to  $\lambda = 0$ . At large, but finite distances between the kinks the doubly degenerate zeroth eigenvalue  $\lambda$  splits. The eigenfunction that is odd with respect to  $z$

$$\varphi_a = \frac{1}{\cosh^2(z-z_0)} - \frac{1}{\cosh^2(z+z_0)} \quad (22)$$

corresponds, as before, to  $\lambda_a = 0$ . As is shown in the appendix, the eigenfunction that is even with respect to  $z$  corresponds to the negative value  $\lambda_s \sim -\exp(-8z_0)$  and has the form

$$\varphi_s = \frac{1}{\cosh^2(z-z_0)} + \frac{1}{\cosh^2(z+z_0)} + q(z). \quad (23)$$

Here the function  $q(z)$  has the asymptote  $q \sim A \exp(-\sqrt{-\lambda}|z|/2)$  as  $z \rightarrow \pm\infty$ , where  $A \sim -\sqrt{-\lambda}$ . It can be shown (see the appendix) that in our case  $\lambda_s$  is a unique negative eigenvalue, and, therefore, we must still require that the condition (21) hold only for the eigenfunctions  $\varphi_a$  and  $\varphi_s$ .

Because of the symmetry of the problem, the condition (21) with the antisymmetric function  $\varphi_a$  is satisfied automatically. After taking the integrals, the orthogonality condition for  $\varphi_s$  takes the form

$$\int_{-\infty}^{\infty} \psi \left( \frac{1}{\cosh^2(z-z_0)} + \frac{1}{\cosh^2(z+z_0)} \right) dz = -16 \exp(-4z_0) \quad (24)$$

[the corrections that are proportional to  $q$  and have a higher order of smallness with respect to  $\exp(-z_0)$  are neglected here]. In fact, Eq. (24) implicitly specifies the velocity of the layer. We find the dependence of the value of the diffusion field  $\psi$  at the point where the phase boundary is located. This requires solving the diffusion problem (15). Taking into account only the terms that are linear with respect to the ve-

locity of the kinks, we assume that the coordinates of the phase boundaries are not time-dependent. After some ordinary manipulations, we obtain

$$\int_0^t \frac{V(\tau)}{\sqrt{t-\tau}} d\tau = \frac{\sqrt{\pi}}{4} [h - 16 \exp(-4z_0)]. \quad (25)$$

Equation (25) was obtained under the condition  $t \gg z_0^2$ , which corresponds to the establishment of a steady-state distribution of the diffusion field  $\psi$  within the layer. The equation of motion of the layer is significantly nonlocal, since the variation of the diffusive flux to the phase boundary is far more rapid than the variation of the coordinates of the boundaries. The interaction of the phase boundaries results in dissolution of the layer even at positive degrees of supersaturation. When the thickness of the layer is equal to the critical value [ $z_0 = a = (1/4) \ln(16/h)$ ], the velocity of the phase boundaries equals zero. If the initial thickness  $z_0(0)$  of the layer is greater than the critical value, the thickness increases. Going over in (25) to the new variables

$$\xi = 4[z_0(t) - z_0(0)], \quad (26)$$

$$\theta = \left\{ \frac{16 \exp[-4z_0(0)]}{\sqrt{\pi}} \right\}^2 t, \quad (27)$$

$$H = \frac{h}{16} \exp[4z_0(0)], \quad (28)$$

we obtain

$$\int_0^\theta \frac{\dot{\xi}(s)}{\sqrt{\theta-s}} ds = \pi [H - \exp(-\xi)], \quad (29)$$

where  $\dot{\xi} = d\xi/ds$ . The value  $H = 1$  corresponds to the critical thickness of the layer. Multiplying (29) by  $1/\sqrt{y-\theta}$  and integrating over  $\theta$  from zero to  $y$ , we obtain

$$\xi = \int_0^y \frac{H - \exp(-\xi)}{\sqrt{y-\theta}} d\theta. \quad (30)$$

It follows from (30) that the thickness of the layer increases when  $H > 1$  and that

$$2(H-1)\sqrt{y} < \xi(y) < 2H\sqrt{y}. \quad (31)$$

In the case of  $H \gg 1$ , the size of the nucleus varies according to the known square-root law.<sup>7</sup> Figure 1 presents the results of the numerical solution of Eq. (29). When  $H < 1$ , the nucleus dissolves. The formal solution of Eq. (30) has the property that  $\xi$  goes to  $-\infty$  during the finite times  $y^*$ . We seek the solution of Eq. (30) near  $y^*$  in the form

$$\exp(-\xi) = \frac{1}{(y^*-y)^\omega} \sum_{j=0}^{\infty} c_j (y^*-y)^j. \quad (32)$$

In order for the integral on the right-hand side of (3) to diverge, we must have  $\omega \geq 0.5$ . Since the left-hand side of (30) undergoes logarithmic divergence when  $y \rightarrow y^*$ , to obtain logarithmic divergence on the right-hand side we must have  $\omega = 0.5$ . Restricting ourselves to the first term in the sum in (32), we obtain

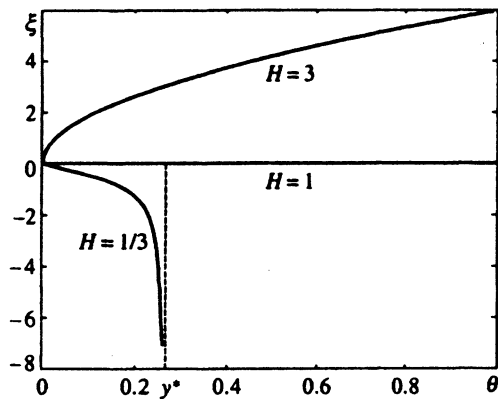


FIG. 1. Results of the numerical solution of Eq. (29).

$$c_0 = 1/2, \quad 2 \ln 2 = 2H \sqrt{y^*} - \ln \sqrt{y^*}. \quad (33)$$

These arguments are confirmed by the results of the numerical solution of Eq. (29) for  $H < 1$  (see Fig. 1). Of course, the solution has physical meaning only for  $\xi > -4z_0$  ( $\xi \sim -4z_0$  corresponds to complete dissolution of the nucleus). However, when  $z_0 \gg 1$ , the dissolution time  $T$  of the nucleus can be evaluated using the expression

$$T = \frac{\pi y^*}{2^8} \exp[8z_0(0)]. \quad (34)$$

The dissolution time is, thus, exponentially dependent on the initial thickness of the layer.

Thus, in this work we have investigated the nonlinear dynamics of the growth (dissolution) of the layer of the stable phase in a decomposing solution. The results are valid for layers whose thickness is significantly greater than the thickness of the phase boundary and small degrees of supersaturation in the solution. The analytic solution of the Cahn–Hilliard equation is based on the assumption that the distortion of the phase boundaries as they move is small. The consistency of this approach is evidenced by the small magnitude of the correction  $u(z, t)$ , which takes into account the height and width of the kinks. We note that our previous comparison of a numerical solution with a soliton-like description of the dynamics of kinks in restricted multilayered structures<sup>8</sup> also points to the accuracy of the analytic method considered here for solving this class of problems.

The existence of a one-dimensional critical nucleus results from the finite width of the phase boundary. One consequence of this is the logarithmic dependence of the size of the nucleus on the degree of supersaturation (6). An equation for the velocity of the phase boundaries (25), which is significantly nonlocal with respect to the time, has been obtained. The physical reason for this nonlocality is the diffusion-controlled supply of the excess component to the phase boundary. An analysis of the equation of motion for the phase fronts makes it possible both to obtain the known expression for the growth rate of the layer of the new phase (31) and to evaluate the dissolution time of the layer. Because of the weakness of this interaction, the dissolution time increases exponentially as the initial thickness of the layer increases [see (34)]. We note that the laws governing the

behavior of the one-dimensional nucleus described here are characteristic of the unrestricted problem. If, for example, the conditions for the absence of diffusive fluxes (9) are assigned at distances  $\sim L$  from the phase boundaries, the dynamics of the layer change. A stationary distribution of the diffusion field is established within a time  $t \gg L^2$ , and the thickness of the layer ceases to vary. In real systems the development of “transverse” instability of the planar phase boundaries is also possible. We believe that the approach developed here to describe the motion of phase boundaries in systems with a conserved order parameter can be generalized to the multidimensional case.

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#### APPENDIX A:

Let us consider the problem of finding the eigenvalue  $\lambda_s$  and the corresponding eigenfunction  $\varphi_s$  of the operator  $\hat{L}$  that is symmetric with respect to  $z$  for the case in which the value of  $\lambda_s$  is small. Integrating Eq. (17) over the entire  $z$  axis and using the boundary conditions, we find that for  $\lambda_s \neq 0$  the eigenfunction should satisfy the condition

$$\int_{-\infty}^{\infty} \varphi_s(z) dz = 0. \quad (A1)$$

Therefore,  $\varphi_s(z)$  cannot be a simple sum of the functions  $[\cosh^2(z \pm z_0)]^{-1}$ , which are solutions of the problem in the absence of an interaction between the phase boundaries ( $z_0 \rightarrow \infty$ ). However, at sufficiently large  $z_0 \gg 1$  it is natural to seek  $\varphi_s(z)$  in the form

$$\varphi_s = \frac{1}{\cosh^2(z - z_0)} + \frac{1}{\cosh^2(z + z_0)} + q(z), \quad (A2)$$

where  $q$  is small and vanishes when  $z_0 \rightarrow \infty$  (the validity of this assumption is confirmed below). For the fulfillment of the condition (A1) we should require that

$$\int_{-\infty}^{\infty} q(z) dz = -4. \quad (A3)$$

In the region  $z - z_0 \gg 1$  we obtain an equation for  $q(z)$

$$4 \frac{\partial^2 q}{\partial z^2} + \lambda q = 0. \quad (A4)$$

To satisfy the boundary conditions, we select the solution that decreases exponentially as  $z \rightarrow +\infty$ :

$$q = A \exp(-\sqrt{-\lambda} z / 2). \quad (A5)$$

Here we introduce the assumption that  $\lambda < 0$ , which is confirmed below. To obtain the solution in the range  $0 < z \ll (-\lambda)^{-1/2}$  we substitute (A2) into (17), and neglecting the small terms  $\lambda q$  and  $\lambda / \cosh^2(z + z_0)$ , we obtain

$$\frac{\partial^2}{\partial z^2} \left[ \frac{\partial^2 q}{\partial z^2} - 4q + \frac{6q}{\cosh^2(z-z_0)} \right] = \frac{\lambda}{\cosh^2(z-z_0)} - \frac{\partial^2}{\partial z^2} \frac{12}{\cosh^2(z-z_0)\cosh^2(z+z_0)}. \quad (\text{A6})$$

The overall solution of this equation has the form

$$q = D_1 Q_1 + D_2 Q_2 + Q_2 \int Q_1 F dz - Q_1 \int Q_2 F dz, \quad (\text{A7})$$

$$Q_1 = \frac{1}{\cosh^2(z-z_0)},$$

$$Q_2 = Q_1 \left\{ \frac{3}{8}(z-z_0) + \frac{1}{4} \sinh[2(z-z_0)] + \frac{1}{32} \sinh[4(z-z_0)] \right\},$$

$$F(z) = \lambda \ln[\cosh(z-z_0)] - \frac{12}{\cosh^2(z-z_0)\cosh^2(z+z_0)} + B_1 z + B_2.$$

Matching the solutions (A5) and (A7) and utilizing the even character of  $q(z)$ , we can find the constants  $A$ ,  $B_{1,2}$ ,  $D_{1,2}$ , and  $\lambda$ . We present the expressions for  $A$  and  $\lambda$ :

$$A = -2^6 \exp(-4z_0), \quad \lambda = -2^{12} \exp(-8z_0). \quad (\text{A8})$$

We note that these results confirm the assumptions made above regarding the smallness of  $q$  and  $\lambda$  and the negative value of  $\lambda$ .

We now show that there are no other negative eigenvalues in the spectrum of the operator  $\hat{L}$  in our case. To prove this assertion we assume the opposite: let there be two negative eigenvalues  $\lambda_1$  and  $\lambda_2$  and two corresponding eigenfunctions  $\varphi_1$  and  $\varphi_2$ . We define the functions conjugate to them  $\chi_1$  and  $\chi_2$  in accordance with (19). We now introduce the linear combinations  $\varphi = c_1 \varphi_1 + c_2 \varphi_2$  and  $\chi = c_1 \chi_1 + c_2 \chi_2$ . It is not difficult to see that the quantity

$$\int \chi \hat{L} \varphi dz = -\lambda_1 c_1^2 \int \left( \frac{\partial \chi_1}{\partial z} \right)^2 dz - \lambda_2 c_2^2 \int \left( \frac{\partial \chi_2}{\partial z} \right)^2 dz \quad (\text{A9})$$

is positive in this case. On the other hand,

$$\int \chi \hat{L} \varphi dz = - \int \varphi \hat{H} \varphi dz, \quad (\text{A10})$$

where

$$\hat{H} = -\frac{\partial^2}{\partial z^2} - 6g + 4 \quad (\text{A11})$$

and, therefore,  $\int \varphi \hat{H} \varphi dz < 0$ . If  $E_1 < 0$  and  $\psi_1(z)$  are now the smallest eigenvalue and the corresponding eigenfunction of the Schrödinger operator  $\hat{H}$ , we can select (by adjusting the coefficients  $c_1$  and  $c_2$ ) a function  $\tilde{\varphi}$  such that it is orthogonal to  $\psi_1$ . Since the value of the energy functional

$$E = \frac{\int \psi^* \hat{H} \psi dz}{\int \psi^* \psi dz} \quad (\text{A12})$$

calculated for  $\psi = \tilde{\varphi}$  is negative, there is one more negative eigenvalue for  $\hat{H}$ . It is known, however, that in our case all the energy levels, except  $E_1$ , are positive for the operator (A11). Thus, we arrive at a contradiction, and, therefore, our original assumption that there is another negative value of  $\lambda$  is incorrect.

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