Nonlinear strong-absorption wave in optically bistable medium. Problem of velocity selection

S. É. Esipov

Institute of Solid State Physics, Academy of Sciences of the USSR

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The integrodifferential equation which determines the shape of the strong-absorption wavefront in an optically bistable medium is analyzed. For an optically thick wave, this equation is similar to the familiar equation in the theory for the growth of a needle-shaped dendrite from a supercooled melt. The propagation of flames and laser combustion are described by similar equations. Within the laser pump beam, the shape of a wavefront in steady-state motion is degenerate; there is a family of fronts, which are elliptic paraboloids (or parabolas, in the two-dimensional case). A condition which relates the unknown wave velocity v and the unknown principal radii of curvature of the tip of the paraboloid, ρ_1 and ρ_2 (or v and ρ, in the two-dimensional case), is found from the solution. There is the problem of a velocity selection for a given pump intensity. The electron-hole plasma which is excited plays the same role for the wave as is played for a dendrite by an anisotropy in the kinetics of the solidification of a melt. The theory of dendrite velocity selection in an anisotropic medium as it exists today is not satisfactory.

INTRODUCTION

A strong-absorption wave in an optically bistable or very nonlinear extended medium can be excited by a laser pump in gases,[1] semiconductors,[2,3] and insulators.[4,5] This wave can propagate either independently or in association of waves of other types such as melting, evaporation,[4] combustion,[1] etc. The mechanism for the propagation of a strong-absorption wave might run as follows: The optical absorption coefficient in the bistable medium of interest depends on the temperature T. The absorption is strong if the temperature exceeds a certain critical or characteristic value T_0; in this case, an electron-hole (e-h) plasma forms (for definiteness we will discuss semiconductors below). The recombination of this plasma causes a heating of the medium, and this heating propagates as a result of (for example) thermal conductivity. Let us assume that, by virtue of the initial conditions, the temperature profile along the light beam, $T(z)$, is already nonuniform, so that the value T_0 is reached at a certain point z_1, and thereafter $T(z)$ is higher than T_0: $T(z) > T_0$, $z < z_1$. The front of the wave, which is initially at the point $z = z_1$, then moves opposite the beam, since the evolution and transfer of the heat raise the temperature ahead of the front. There is usually also a trailing edge $T(z_2) = T_0$, $z_2 < z_1$, if the beam is bounded in the transverse direction. A strong-absorption wave has a threshold in terms of the pump intensity, $J_\infty \geqslant J_{\rm th}$. The threshold value $J_{\rm th}$ is determined by the excitation conditions and the properties of the medium.

Experiments have been carried out on wave propagation in the interior of a medium,[1–3] on the localization of a wave at the front boundary[2,3] and the rear boundary[3] of a medium ("front" and "rear" from the standpoint of the light), and on localization near the focus of a laser beam.[1,3] Localization at boundaries gives rise to a variety of dynamic regimes: the ejection of kinks,[2] self-oscillations, autowaves,[3] a period-doubling of self-oscillations,[3] etc.

The theory of strong-absorption waves which has been derived for gases[1] and transparent solids[2,5,6] is semiphenomenological and essentially similar to the classical theory of combustion waves.[7,8] A one-dimensional description has been adopted for the fields of the temperature, $T(z)$, the light intensity $J(z)$, and the plasma density $n(z)$ along the beam. Because of energy conservation, a solution in the form of a one-dimensional wave which is moving at a constant velocity v has no threshold:

$$v=\bar{v},\quad \bar{v}=J_\infty/c(T_0-T_\infty). \tag{1}$$

Here c is the specific heat at constant volume, and T_∞ is the temperature of the medium far from the wave. A transverse relaxation of the temperature, which leads to a threshold, has been taken into account by means of a phenomenological term $(T - T_\infty)\tau_T$. This term is interpreted in the following way: The three-dimensional Laplacian is written as consisting of a longitudinal part $\partial^2/\partial z^2$ and a transverse part $r^{-1}(\partial/\partial r)[r(\partial/\partial r)]$. By virtue of an assumption of the model, the thermal field has a transverse dimension on the order of the beam radius R_l, so the transverse part of the Laplacian is replaced by a relaxation term, where $\tau_T = AR^2/\lambda$. The coefficient A is a number on the order of unity which is used as an adjustable parameter, and λ is the thermal diffusivity. Not infrequently, the size of the thermal field turns out to be considerably greater than that of the beam. The difference itself, of course, depends on the nature of the dynamic regime.[2,3]

Nevertheless, this one-dimensional description has made it possible to explain most of the experimental facts and to reach an agreement between semiquantitative estimates and measured quantities within an order of magnitude.[3] A more accurate agreement has been prevented by the choice of a single value of the coefficient A.

Experiments have been interpreted on the basis of the model of a thermal wave which was summarized at the beginning of this paper, and in which the motion of the front is associated with thermal conductivity. The ambipolar diffusion of the e-h plasma (as a transport process) and the plasma-density dependence of the optical absorption edge (as a nonlinearity) could in principle "create" similar nonthermal waves.[6]

In this paper we analyze a system of equations for the shape of the strong-absorption wavefronts for the three-di-

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mensional ($3D$) and two-dimensional ($2D$) cases. Further progress toward an analytic theory is possible (Secs. 1 and 2) for an optically thick wave, in which case the optical absorption length is short in comparison with the dimensions of the problem, and also for an optically thin wave, in which case the absorption (and refraction) of the light is ignored altogether. If the beam is not bounded in the transverse direction, the equation for the shape of the front of an optically thick wave is the same as that for a needle-shaped dendrite or combustion front. A solution in the form of a wave in steady-state motion (Sec. 3) yields only one condition on the three unknowns involved here: the velocity v and the radii of curvature, ρ_1 and ρ_2, of the tip of the wave (or v and ρ in the $2D$ case). A so-called Ivantsov continuum of possible steady states arises.[9-11] For a needle-shaped dendrite this condition corresponds to a functional dependence $v \propto \max(\rho_1^{-1}, \rho_2^{-1})$, apparently because as time elapses the front branches out, reaching small values of ρ and large velocities v. For a strong-absorption wave, the same condition yields $v = \bar{v}[1 - O(\rho_1^{-1}) - O(\rho_2^{-1})]$, and the highest velocity corresponds to the largest radii of curvature: those of a plane front. The difference in behavior stems from the circumstance that the heat evolution (the power of the sources) at the front of a dendrite is proportional to the normal velocity of the front at each point, while in an absorption wave the power of the sources does not depend on the velocity. The equations are identical because the motion of the front is uniform.

In Secs. 4 and 5, we call on the theory of velocity selection which has recently been derived for dendrites. Considerations regarding the maximum velocity $v(\rho_1, \rho_2)$ do not by themselves imply a selection of any sort, and the time-varying problem should be studied. It was thus a natural step to analyze the theory, which has recently been developing very rapidly. In this theory, the following assertions have been made, originally on the basis of local models[12,13] and later on the basis of a complete integrodifferential equation (Refs. 14–19; see also the review by Kessler *et al.*[20]).

a) The addition of perturbations (of the surface tension and/or the kinetics at the solidification front) to the equation completely erases the Ivantsov continuum. This and the following assertions are made on the basis of a singular perturbation theory.

b) The incorporation for these perturbations of a special anisotropy

$$B_m(\psi) = 1 - \varepsilon \cos(m\psi), \tag{2a}$$

where ε is the small parameter of the anisotropy, and ψ is the angle between the z axis and the normal to the front at the given point, makes it possible to reconstruct a countable set of solutions from the continuum which has been erased.

c) Among the solutions which are reconstructed, that which corresponds to the maximum growth rate is singularly stable (in the linear sense).

In this paper we show that incorporating the rapid diffusion of plasma in the wave over the recombination time τ_R leads to corrections of the same kind as in the nonequilibrium kinetics at a crystallization front. In this case the function $B(\psi)$ takes the form

$$B(\psi) = \frac{1}{1 + \varepsilon \cos \psi}, \tag{2b}$$

and at small ε we can use the approximation $B(\psi) \approx B_1(\psi)$. This approximation, incidentally, is valid only at the real axis. For complex ψ, expressions (2a) and (2b) have different singularities, while assertions a)–c) are associated with specifically the behavior of $B(\psi)$ near singularities.[1)] We show in Sec. 5 that for the anisotropy in (2a) there is a velocity selection, while for that in (2b) there is not.

This distinction, according to which the selection is critically sensitive to an arbitrarily small change in $B(\psi)$, is nonphysical. On the one hand, it is not amenable to experimental test. On the other, a microscopic calculation of $B(\psi)$ itself unavoidably involves perturbation theory. The use of perturbation theory should now become systematically singular, and that situation is essentially meaningless. Putting aside the question of the mathematical validity of the theory of anisotropic corrections, we will content ourselves with the assertion that this theory is not applicable to the existing anisotropy $B(\psi)$ in (2b). This theory is equally inapplicable to other nonlinear systems.

A strong-absorption wave is a new entity, used to study the formation of nonequilibrium structures similar to systems with phase transitions,[9,20] to systems with viscous liquids which displace each other,[21] etc.

1. DERIVATION OF EQUATIONS FOR THE SHAPE OF STRONG-ABSORPTION WAVEFRONTS. THREE-DIMENSIONAL (3D) WAVE

We consider the thermal wave of Ref. 6, in which the nonlinearity of the refractive index is associated with the temperature, $\alpha(T)$, while the wave motion is associated with the thermal conductivity. The fields of the temperature T and the intensity J obey the equations

$$\begin{aligned} T_t' &= \lambda \Delta T + \alpha(T) J/c, \\ J_z' &= -\alpha(T) J. \end{aligned} \tag{3a}$$

The light propagation direction coincides with the negative z direction of the cylindrical coordinate system $\xi = (z, \mathbf{r})$, $\mathbf{r} = (r, \varphi)$; refraction of the light is ignored; and c is the specific heat of the medium at constant volume. The boundary conditions on Eqs. (3a) are

$$T|_{z\to\infty} = T|_{r\to\infty} = T_\infty, \qquad J|_{z\to\infty} = J_\infty j_\infty(\mathbf{r}), \qquad j_\infty(0) = 1. \tag{4a}$$

Let us assume that $\alpha(T)$ is a very simple bistable function $\alpha(T) = \alpha H(T - T_0)$, where $H(x)$ is the unit step function. The connected spatial region in which the condition $T(\xi, t) \geqslant T_0$ holds is then called a "strong-absorption wave." We are interested in a solution $T(z - vt, \mathbf{r})$, $J(z - vt, \mathbf{r})$ which moves at a constant velocity v, under the assumption that the wave is formed by the two fronts $z_1(\mathbf{r})$ and $z_2(\mathbf{r})$, $z_1(\mathbf{r}) \geqslant z_2(\mathbf{r})$. We define the unit of length to be ρ, which is one of the radii of curvature of the front at the z axis; we define the unit of time as ρ/v; we define the unit of intensity as J_∞; and we put the origin of the temperature scale at T_∞ and express the temperature in units of J_∞/vc. In terms of these dimensionless variables, Eqs. (3a) and (4a) become

$$\theta_t' = (2p)^{-1} \Delta\theta + \gamma H(\theta - \theta_0), \qquad j_t' = -\gamma j H(\theta - \theta_0), \tag{3b}$$

$$\theta|_{z\to\infty} = \theta|_{r\to\infty} = 0, \quad j|_{z\to\infty} = j_\infty(\mathbf{r}), \tag{4b}$$

where $p = \rho v/2\lambda$ is the Peclet number, $\gamma = \alpha\rho$, $\theta_0 = (T_0 - T_\infty) vc/J_\infty$, and the fields $\theta(\xi, t)$ and $j(\xi, t)$ are the dimensionless temperature and the dimensionless intensity. Substituting the solution of the second equation in (3b) into the first, and transforming to the argument $z \to z - t$, we have the following analytic equation for $\theta(\xi)$:

$$(2p)^{-1}\Delta\theta+\theta_z'+\gamma j_\infty(\mathbf{r})\exp\{-\gamma[z_1(\mathbf{r})-z]\}H(\theta-\theta_0)=0, \quad (5)$$

Here we are assuming $\theta(\xi) = \theta_0$ at $\xi = [z_{1,2}(\mathbf{r}), r, \varphi]$. Using the Green's function for (3b) or (5), we can write explicit integral equations for the functions $z_{1,2}(\mathbf{r})$:

$$\theta_0=\gamma\left(\frac{p}{2\pi}\right)^{3/2}\int_0^\infty \frac{dt}{t^{3/2}}\iint d^2\mathbf{r}' j_\infty(\mathbf{r}')$$
$$\times\int_{z_2(\mathbf{r}')-t}^{z_1(\mathbf{r}')-t} dz' \exp\left\{\gamma[z'-z_1(\mathbf{r}')+t]-\frac{p}{2t}R_{1,2}\right\}. \quad (6)$$

The integration over $d^2\mathbf{r}'$ is carried out in the plane perpendicular to the z axis, over the (r,φ) region in which the relation $\theta(\mathbf{r})\geqslant\theta_0$ holds;

$$R_{1,2}^2=R_\perp^2+[z_{1,2}(\mathbf{r})-z']^2,$$
$$R_\perp^2=r^2+r'^2-2rr'\cos\varphi. \quad (7)$$

The right side of (6) is a constant when considered as a function of ξ. Consequently, a solution, if one exists, is not unique. System (6) for $z_{1,2}(\mathbf{r})$ is complicated. We will discuss two limiting cases in terms of the value of the parameter γ.

Optically thick wave

In this case all the incident light is absorbed just behind the front, and the parameter γ is large. In Eq. (6), we can carry out the integration over dz',

$$\theta_0=\left(\frac{p}{2\pi}\right)^{3/2}\int_0^\infty \frac{dt}{t^{3/2}}\iint d^2\mathbf{r}' j_\infty(\mathbf{r}')$$
$$\times\exp\left\{-\frac{p}{2t}[R_\perp^2+(z(\mathbf{r})-z(\mathbf{r}')+t)^2]\right\}, \quad (8a)$$

where $z(\mathbf{r})\equiv z_1(\mathbf{r})$, and then the integration over dt:

$$\theta_0=\frac{p}{2\pi}\iint d^2\mathbf{r}' j_\infty(\mathbf{r}')R^{-1}\exp[-pR-p(z(\mathbf{r})-z(\mathbf{r}'))],$$
$$R^2=R_\perp^2+[z(\mathbf{r})-z(\mathbf{r}')]^2. \quad (8b)$$

The position of the trailing edge is obviously not important now. In Sec. 3, for example, we discuss the situation with $j_\infty(\mathbf{r})\equiv 1$, in which there is no trailing edge at all. In this case the solution of Eqs. (8) is known from the theory of dendrite growth and was derived within the Ivantsov ansatz (Sec. 3).

Optically thin wave

We assume that the optical absorption length is larger than all the length scales which determine the behavior of the wave; the parameter γ is small. Integrating over dz', we find, for the front, $z(\mathbf{r})\equiv z_1(\mathbf{r})$,

$$\theta_0=\frac{\gamma p}{4\pi}\int_0^\infty\frac{dt}{t}\iint d^2\mathbf{r}' j_\infty(\mathbf{r}')\exp\left(-\frac{pR_\perp^2}{2t}\right)$$
$$\times\operatorname{erfc}\left[\left(\frac{p}{2t}\right)^{1/2}(z(\mathbf{r})-z(\mathbf{r}')+t)\right]. \quad (9)$$

The absorbed radiant energy is evolved as heat uniformly everywhere within the front $z(r)$, so the temperature differs from point to point within the front. This circumstance rules out the use of the Ivantsov ansatz,[10,22] which is valid for Eq. (8). Beginning in Sec. 3 below, we discuss an optically thick wave.

2. 2D WAVE

For a $2D$ wave we introduce the Cartesian coordinate system $\xi = (z,x)$, with z axis running opposite the beam direction, as before. The boundary condition on the light intensity now takes the form

$$J|_{z\to\infty}=J_\infty(x)=J_\infty j_\infty(x).$$

The equations analogous to (6) are

$$\theta_0=\frac{\gamma p}{2\pi}\int_0^\infty\frac{dt}{t}\int_{-\infty}^\infty dx' j_\infty(x')$$
$$\times\int_{z_2(x')-t}^{z_1(x')-t} dz'\exp\left[\gamma(z'-z_1(x')+t)-\frac{pR_{1,2}^2}{2t}\right], \quad (10)$$

where the quantities $R_{1,2}$ are as in (7), but with $R_\perp^2 = (x-x')^2$. For an optically thick wave we find the equation [cf. (8a)]

$$\theta_0=\frac{p}{2\pi}\int_0^\infty\frac{dt}{t}\int_{-\infty}^\infty dx' j_\infty(x')$$
$$\times\exp\left\{-\frac{p}{2t}[(z(x)-z(x')+t)^2+R_\perp^2]\right\} \quad (11a)$$

or [cf. (8b)]

$$\theta_0=\frac{p}{2\pi}\int_{-\infty}^\infty dx' j_\infty(x')K_0\{p[(z(x)-z_1(x'))^2+R_\perp^2]^{1/2}\}$$
$$\times\exp[-p(z(x)-z(x'))], \quad (11b)$$

where $K_0(z)$ is the modified Bessel function. A solution of Eqs. (11), like a solution of Eqs. (8), is known for $j_\infty(x)\equiv 1$ (Sec. 3). Finally, in the case of an optically thin wave we find from (10) an equation to which we can apply the same words that we applied to its $3D$ analog, (9):

$$\theta_0=\frac{\gamma p}{8\pi}\int_0^\infty\frac{dt}{t^{1/2}}\int_{-\infty}^\infty dx' j_\infty(x')\exp\left(-\frac{pR_\perp^2}{2t}\right)$$
$$\times\operatorname{erfc}\left[\left(\frac{p}{2t}\right)^{1/2}(z(x)-z(x')+t)\right]. \quad (12)$$

3. PARABOLIC SOLUTIONS FOR OPTICALLY THICK WAVE

We set $j_\infty(\mathbf{r})\equiv 1$ in Eq. (8b). Now, the solution may not be axisymmetric. As Horvay and Cahn have shown (Ref. 23; see also Ref. 14), there exists a solution of Eq. (8b) in the form of an arbitrary elliptic paraboloid $z(\mathbf{r}) = -x^2/2 - \nu y^2/2$, where $(x,y) = (r,\varphi)$, and the principal radii of curvature are 1 and ν^{-1} (in dimensional units, $\rho_1 = \rho$, $\rho_2 = \rho/\nu$). Substituting this solution back into Eq. (8b), we find a condition on the parameters θ_0, p, and ν:

$$\theta_0=\frac{4p}{\pi}\int_0^\infty\int_0^\infty\frac{dx\,dy}{x^2+\nu y^2+1}\exp[-p(x^2+y^2)]$$

$$=\begin{cases}1-(1+\nu)/2p, & p\gg\max(1,\nu),\\ -p\nu^{-1/2}\ln p, & -\ln p\gg\max(1,-\ln\nu).\end{cases}\qquad(13a)$$

For a circular paraboloid ($\nu=1$) we find

$$\theta_0=pe^pE_1(p)=\begin{cases}1-1/p, & p\gg1,\\ -p\ln p, & p\ll1.\end{cases}\qquad(14)$$

Let us analyze condition (13a) at large and small Peclet numbers. Using the explicit expressions for the parameters θ_0 and p, and solving the equations for the velocity, we find

$$v=\bar{v}\,[1-(1+\nu)\lambda/\rho],\qquad(13b)$$

$$v=(2\lambda/\rho)\exp[-2\nu^{1/2}\lambda/\bar{v}\rho].\qquad(13c)$$

In (13b) [and in (15b) below] the second term is a small correction to the first. The regions in which asymptotic expressions (13b) and (13c) are valid are the same as those for asymptotic expressions (13a).

In the $2D$ case there is the analogous solution $z(x)=-x^2/2$, for which the wavefront is a parabola. The corresponding condition on the parameters θ_0 and p is

$$\theta_0=(\pi p)^{1/2}e^p\,\mathrm{erfc}(p^{1/2})=\begin{cases}1-1/2p, & p\gg1,\\ (\pi p)^{1/2}, & p\ll1.\end{cases}\qquad(15a)$$

Hence

$$v=\bar{v}-\lambda/\rho,\ p\gg1,\qquad(15b)$$

$$v=\pi\rho\bar{v}^2/2\lambda,\ p\ll1.\qquad(15c)$$

Analyzing the convergence of the integrals in Eq. (8b), we find a convergence radius on the order of $[\max(1,\nu^{-1},p,p/\nu)]^{-1}$. On this basis we can assert the assumption $j_\infty\equiv1$ is valid near the "bow" of the wave if the radius of the light beam, R_l, is greater than the convergence radius. In dimensional units, this assumption is valid if the condition

$$R_l\gg\min[\rho_1,\ \rho_2,\ \lambda/\bar{v}]\qquad(16)$$

holds or (for the $2D$ case) if the condition

$$R_l\gg\min[\rho,\ \lambda/\bar{v}]$$

holds.

We see that Eqs. (13)–(15) determine only a certain combination of the velocity v and the radii of curvature ρ and ρ/ν, depending on the external conditions (v and ρ in the $2D$ case). There is the problem of the selection of a single velocity.

If we seek the maximum velocity by varying the radii of curvature, we find from (13)–(15) that the maximum is reached for a plane front, with $\rho_1,\rho_2=\infty$. In this case we have $v=\bar{v}$.

Comparison with the 1*D* model

Let us compare these results with the $1D$ model. For an optically thick wave we find the following from Eqs. (14) and (15) of Ref. 6:

$$\theta_0=(1+2/p\tau_0)^{-1/2}=\begin{cases}1-1/p\tau_0, & p\tau_0\gg1,\\ (p\tau_0/2)^{1/2}, & p\tau_0\ll1.\end{cases}\qquad(17a)$$

Here we have switched to the dimensionless variables of the present paper,

$$\tau_0=\tau_T v/\rho=2A\,(R/\rho)^2p,$$

$$p\tau_0=2Ap^2=Av^2R^2/2\lambda^2,$$

while in dimensional units we would have

$$v=\bar{v}\,(1-2\lambda^2/A\bar{v}^2R^2),\quad J_\infty\gg J_{th},\qquad(17b)$$

$$v=\frac{8^{1/2}\lambda}{A^{1/2}R}\left(1-\frac{2\lambda}{A^{1/2}Rv}\right),\quad 0\leqslant J_\infty-J_{th}\ll J_{th},\qquad(17c)$$

where the threshold intensity is $J_{th}=2\lambda c(T_0-T_\infty)/A^{1/2}R$. In this notation, applicability condition (16) becomes specifically $J_\infty\gg J_{th}$.

We thus see that until we solve this velocity selection problem we cannot decide how well the $1D$ model agrees with the exact solution. All that we can do is assert that the threshold in the $1D$ model arises at the same intensities (if the values of ρ_1 and ρ_2 are not too far apart) as those at which the validity of the derivation of conditions (13)–(15) breaks down. Far from the threshold, $J_\infty\gg J_{th}$, the $1D$ model yields the expression $v=\bar{v}$ for the velocity. This result corresponds to a plane front and a maximum velocity.

4. EFFECT OF EXCITED PLASMA. THEORY OF ANISOTROPIC CORRECTIONS

In this section of the paper we calculate the singular corrections for the finite lifetime of nonequilibrium carriers, τ_R. In general, the band gap of a semiconductor, E_g, which determines the optical absorption edge, depends on not only the temperature but also the density of the excited plasma; i.e., we have $E_g(T,n)$. In the (T,n) plane there is accordingly a line which divides regions of strong and weak absorption. In the linear approximation, the critical values T_0 and n_0 are related near the edge by the relation

$$\hbar\omega=E_g(T_\infty,n_\infty)+\frac{\partial E_g}{\partial T}(T_0-T_\infty)+\frac{\partial E_g}{\partial n}(n_0-n_\infty),\qquad(18)$$

where $\hbar\omega$ is the photon energy, which satisfies $\hbar\omega-E_g\ll E_g$. We are assuming that the plasma is nondegenerate and that the partial derivatives $\partial E_g/\partial T$ and $\partial E_g/\partial n$ are negative. Using (18), we can write the following expression for the parameter θ_0, which characterizes the dimensionless temperature at the front:

$$\theta_0=\frac{(T_0-T_\infty)vc}{J_\infty}=\frac{vc}{J_\infty}\left[\delta T-\left(\frac{\partial E_g}{\partial T}\Big/\frac{\partial E_g}{\partial n}\right)n\right].\qquad(19)$$

Here $n=n_0-n_\infty$, and δT is the heating which would be required if the effect of the plasma were ignored.

Let us now calculate the density n, so that we can use the parameter in (19) in Eq. (8b). We assume that an e-h plasma is excited in an optically thick wave and that this plasma is capable of propagation by ambipolar diffusion and also capable of recombination.[6] The wave has no trailing edge ($j_\infty\equiv1$). Since τ_R is short, the plasma is in a narrow layer near the front, and we are to find a description of its profile along the normal to the front. For this purpose we have the $1D$ equation

$$Dn''-n/\tau_R+(\alpha J_\infty/\hbar\omega)\exp(-\alpha l/\cos\psi)=0,\qquad(20)$$

where l is the coordinate directed into the wave, opposite the outward normal to the front. The origin of the scale of this coordinate is at the front. The angle ψ is the angle between the z axis and the outward normal, and D is the diffusion coefficient. The drift term associated with the translational motion of the wave has been discarded on the basis of numerical estimates. For example, some experiments[3] with ZnSe involved the values $D \sim 10$ cm^2/s, $\tau_R \sim 10^{-10}$ s, $v \sim 10$–100 cm/s, and $\alpha \sim 10^4$ cm^{-1}, and the diffusion length $(D\tau_R)^{1/2} \approx 3 \cdot 10^{-4}$ cm$\sim \alpha^{-1}$ was comparable to the optical absorption length. The drift length $v\tau_R \sim 10^{-9}$–10^{-8} cm, on the other hand, was negligible.

Solving Eq. (20) with the boundary conditions $n(\pm\infty) = 0$, we find the density at the front,

$$n(l=0)=\frac{J_\infty}{2\hbar\omega}\left(\frac{\tau_R}{D}\right)^{1/2}\frac{\cos\psi}{1+\varepsilon\cos\psi},\qquad \varepsilon^{-1}=\alpha(D\tau_R)^{1/2},$$

and the parameter θ_0,

$$\theta_0=\bar\theta_0-\beta B(\psi)\cos\psi,\quad \bar\theta_0=vc\delta T/J_\infty,$$
$$\beta=\frac{vc}{2\hbar\omega}\left(\frac{\tau_R}{D}\right)^{1/2}\frac{\partial E_g}{\partial n}\Big/\frac{\partial E_g}{\partial T},\qquad (21)$$

where $B(\psi)$ has the form in (2b). In the theory of dendrite growth there is a correction $\beta B_m(\psi)\cos\psi$ to Eq. (11b), where $B_m(\psi)$ has the form in (2a) (see Refs. 24 and 25 in this connection). We find the same expression when we incorporate the anisotropic kinetics of the solidification at the front. Under the condition $\beta \ll 1$, we seek a solution in the form $\zeta(r) = z(r) + r^2/2$, where $\zeta(r)$ is a small correction to the Ivantsov paraboloid, for Eqs. (8b), (11b), with θ_0 from (21) (for simplicity we are treating the axisymmetric case, with $\nu = 1$; in the $2D$ case we would have to replace r by x). In the singular-perturbation method, the correction $\zeta(r)$ consists of a regular part and a singular part. The regular part is found from the solution of the equations with the derivatives ignored. From dendrite theory we know that it is an exceedingly complicated problem to derive the regular part of $\zeta(r)$. This problem has been solved in the $2D$ case for surface tension[26,27] and for kinetics.[24] No selection arises in this case. I would like to call attention to another circumstance related to the calculation of the singular correction. For this calculation, one studies an equation with derivatives near the singular point.[15,19] For example, in the limit of small Peclet numbers, $p \ll 1$, an equation with a single parameter arises near $r = i(x = i)$ for the $3D(2D)$ problem:

$$\frac{1}{(t+\varphi')^{1/2}}\left[\frac{1}{1+(t+\varphi')^{-1/2}}\right]=\lambda^*\varphi,\qquad \lambda^*=\frac{\theta_0\varepsilon^5}{4\beta},\qquad (22a)$$

where $r = i - i\varepsilon^2 t/2$ $(x = i - i\varepsilon^2 t/2)$, and $\zeta = \varepsilon^4\varphi/4$. For comparison, here is the equation near the singular point for the case $B_1(\psi) = 1 - \varepsilon\cos\psi$:

$$\frac{1}{(t+\varphi')^{1/2}}\left[1-\frac{1}{(t+\varphi')^{1/2}}\right]=\lambda^*\varphi.\qquad (23a)$$

The anisotropies $B(\psi)$ and $B_1(\psi)$ are approximately the same if $\varepsilon \ll 1$. Equations (22a) and (23a) are of course completely different, as are the results of the "selection."

5. ANALYSIS OF SINGULAR EQUATIONS

Equations (22a) and (23a) have a solution $\varphi_N \approx (\lambda^*)^{-1} t^{-1/2}$, at large t. This solution serves as an inhomogeneous solution if the equations are linearized. In the analysis we need to select those values of $\lambda^*(\nu)$ for which, by integrating from $+i\nu$ or $-i\nu$, $\nu \gg 1$, to the real axis in the plane of the complex variable t, with the initial condition $\varphi = \varphi_N$, we find there the real solution $\mathrm{Im}[\varphi(\mathrm{Im}\, t = 0)] = 0$. The unknown values $\lambda^* = \lim\lambda^*(\nu)$ are then found in the limit $\nu \to \infty$.

A numerical integration yields the spectrum of unknown λ^* for Eq. (23a); the smallest positive value is $\lambda^* = 0.016$ ... There are no such values of λ^* in the case of Eq. (22a). To see just what happens, we consider the linearized versions of Eqs. (22a) and (23a), which have the same properties:

$$\varphi'+2\lambda^* t^{1/2}(t^{1/2}+1)^2=2t^{1/2}(t^{1/2}+1),\qquad (22b)$$

$$\varphi'+\frac{2\lambda^* t^2\varphi}{t^{1/2}-2}=2t\frac{t^{1/2}-1}{t^{1/2}-2}\qquad (23b)$$

A solution of (23b) is

$$\varphi(t)=e^{-\eta(t)}\left[C+2\int_0^t dt'\,t'\frac{t'^{1/2}-1}{t'^{1/2}-2}e^{\eta(t')}\right],\qquad (24)$$

$$\eta(t)=2\lambda^*\int_0^t dt'\frac{t'^2}{t'^{1/2}-2},\qquad (25)$$

This solution is defined in the t plane, with a cut along the negative part of the real axis. In order to join with the regular solution, we need to suppress the exponential growth of $\varphi(t)$ in (24) along the lines $\arg(\eta) = \pm\pi$. For this purpose, we choose the integration constant C to be

$$C=-2\int_0^{\infty\cdot e^{2\pi i/5}} dt'\,t'\frac{t'^{1/2}-1}{t'^{1/2}-2}e^{\eta(t')}\qquad (26)$$

and we impose the requirement of symmetry. In other words, we require $\varphi(t)$ b real on the positive part of the real axis at $t > 4$ [the integral $\eta(t)$ in (25) has a singularity at the point $t = 4$]. This requirement selects the parameter λ^*. The integrals are evaluated explicitly under the assumption $\lambda^* \gg 1$. The integral in (26) is then governed by the region $|t| \ll 1$, in which we have $\eta(t) = -\lambda^* t^3/3$, with $\arg(\eta) = \pi$ along the ray $2\pi/3$, and $\arg(C) = 4\pi/3$. On the real axis, the expression in square brackets in (24) is therefore proportional to $[-e^{4\pi i/3} + 1] \propto e^{\pi i/6}$; taking into account the imaginary part of $\eta(t)$ in (25), which stems from the circumvention of the pole at the point $t = 4$, we find the selected values $\lambda^* = (k + 5/6)/64$, $k = 0, 1, 2, \ldots$.

In the case of Eq. (22b), the selection condition is $\mathrm{Im}(C) = 0$, since $\eta(t)$ has no singularities at $|t| \neq 0, \infty$ [the notation C, η has been introduced for (22b) by analogy with (23b); see (24) and (25)]. We find

$$\mathrm{Im}(C)=\mathrm{Im}\left\{-2\int_0^{\infty\cdot e^{2\pi i/5}} dt\,t^{1/2}(t^{1/2}+1)\right.$$
$$\left.\times\exp\left[2\lambda^*\int_0^t dt'\,t'^{1/2}(t'^{1/2}+1)^2\right]\right\}=0$$

or

$$I(\sigma)=\mathrm{Im}\left\{\int_0^{\infty\cdot e^{\pi i/5}} dx\,x^2(x^2+1)\right.$$
$$\left.\times\exp\left[\sigma\left(\frac{x^5}{5}+\frac{x^4}{2}+\frac{x^3}{3}\right)\right]\right\}=0,\qquad (27)$$

where $\sigma = 4\lambda^*$. We make the further assertion that expression (27) cannot hold at any positive value of σ. We have been able to prove that $I(\sigma)$ is positive at $\sigma \sim 1$ only by numerical methods. The asymptotic expressions for $I(\sigma)$ are

$$I(\sigma)=\begin{Bmatrix} 5^{-1/5}\Gamma(4/5)\sin(4\pi/5)\sigma^{-4/5}, & \sigma\ll 1 \\ 2^{-1}\cdot 3^{-1/6}\Gamma(1/3)\sigma^{4/3}, & \sigma\gg 1 \end{Bmatrix} > 0,$$

and have the same sign.

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